Investigating Vortex Behaviour in Superconducting Thin Films through Magnetic Microscopy Techniques

Frederick Steven Wells
University of Wollongong

Follow this and additional works at: https://ro.uow.edu.au/theses1

Recommended Citation
Investigating Vortex Behaviour in Superconducting Thin Films through Magnetic Microscopy Techniques

Frederick Steven Wells

Supervisor:
Prof. Alexey V. Pan

This thesis is presented as required for the conferral of the degree:

Doctor of Philosophy (Physics)

The University of Wollongong
School of Physics and
Institute for Superconducting and Electronic Materials

October 24, 2017
Declaration

I, Frederick Steven Wells, declare that this thesis submitted in fulfilment of the requirements for the conferral of the degree Doctor of Philosophy (Physics), from the University of Wollongong, is wholly my own work unless otherwise referenced or acknowledged. This document has not been submitted for qualifications at any other academic institution.

Frederick Steven Wells

October 24, 2017
Abstract

Magnetic microscopy is the process of creating accurate images of magnetic flux, to map its variations over a small region. In this thesis, magnetic microscopy was used to investigate the distribution and behaviour of magnetic flux (in the form of vortices) in superconductors. Two different magnetic microscopy techniques were used: Magneto-Optical Imaging and Scanning SQUID Microscopy. From each image of magnetic flux, the local distribution of supercurrent was also calculated using an inversion of the Biot-Savart law.

This work centres on several separate yet related experimental investigations into the static and dynamic properties of magnetic vortices and supercurrents in thin film YBa$_2$Cu$_3$O$_{7-\delta}$ superconductors. These are as follows:

Firstly, the magneto-optical imaging technique was extended to ultra-fast magneto-optical video. This was used to investigate the dynamics of magnetic field and current in superconducting thin films under a variety of non-equilibrium conditions. One such condition was an external field increasing from zero, causing magnetic flux penetration. Novel behaviour was observed during penetration, with transient fields having a strongly peaked shape and exceeding the critical current limit, followed by a dynamic transition into magnetic relaxation behaviour.

Secondly, distributions of vortices were measured directly under very low magnetic field conditions using scanning SQUID microscopy. The particular distributions seen were characterised as isotropic vortex glass phases, but some vortices had uncharacteristically close spacings. This phase was also unexpectedly found to lose its orientational order with increasing field.

Finally, a novel two-dimensional ratchet pattern was proposed, as a step towards more robust and efficient superconducting diodes. The aim was to provide rectification of vortex motion (therefore allowing current to flow in one direction only) over a much greater temperature range than previous designs of a similar nature. The pattern has shown excellent rectification in simulations. Ratchets were also created experimentally using thickness modulation of YBa$_2$Cu$_3$O$_{7-\delta}$ thin films through a newly-developed partial ion-beam etching procedure.
Acknowledgments

First and foremost, this work was made possible by my abundantly patient supervisor Professor Alexey V. Pan, who not only gave me the inspiration and an evolving framework for most of the research that would eventually go into this thesis, but also gave me a great deal of constructive rebuke and motivation in those times when my progress was slow or even stopped. Thanks Alexey for being a mentor who I can talk to about any issue, academic or otherwise, this is something I will never forget.

And when it comes to giving motivation, I must mention my biggest motivation (and my biggest distraction) in the final years of my PhD, my partner Becky. The hundred little times she pushed me and forced me to focus on my work have culminated in lots of completed papers, and a finished thesis, which honestly might not have happened without her.

I am thankful for the initial decisions back in 2012 by the University of Wollongong to accept me into a PhD programme and to grant me an Australian Postgraduate Award, which allowed me to fund my study (and my life). Without this income in my first three and a half years, I would have struggled immensely with time and focus to complete my PhD. The struggle was real in my final years of study, as I balanced paid work, research and writing this thesis.

In the most concrete way, this work was made possible through my use of the laboratories and equipment at the Institute for Superconducting and Electronic Materials at the University of Wollongong. And so I must again thank Alexey for the provision of most of this equipment, and other laboratory supervisors whose equipment I used on occasion. I also thank the technical staff at ISEM for their invaluable skills and creativity in fixing equipment and solving technical problems large and small. Thanks to Dr. Sergey A. Fedoseev for tirelessly working to create high-quality superconducting samples. Those superconductors are the real star of this work, without them it would be nothing.

Thank you to Professor Hans Hilgenkamp and the Integrated Correlated Electrons group at the University of Twente for welcoming me into their group and allowing me access to the scanning SQUID microscope, which broadened the scope of my work beyond one imaging technique, as well as broadening my life experiences by showing me their unique part of the world.
# Contents

Abstract iii

List of Figures ix

List of Symbols xi

List of Symbols and Abbreviations xii

1 Introduction 1

1.1 Research Objectives . . . . . . . . . . . . . . . . . . . . . . . . . . . 3
1.2 Structure of the Thesis . . . . . . . . . . . . . . . . . . . . . . . . . . 3

2 Theory 5

2.1 Basic Principles of Superconductivity . . . . . . . . . . . . . . . . . . 6
   2.1.1 Cooper Pairing and BCS Theory . . . . . . . . . . . . . . . . . . 6
   2.1.2 The Meissner Effect . . . . . . . . . . . . . . . . . . . . . . . . . 7
   2.1.3 Magnetic Flux Quantisation . . . . . . . . . . . . . . . . . . . . 9
   2.1.4 Type I and II superconductors . . . . . . . . . . . . . . . . . . . 10
2.2 Yttrium Barium Copper Oxide (YBCO) . . . . . . . . . . . . . . . . . 13
2.3 Vortices in Type II Superconductors . . . . . . . . . . . . . . . . . . 15
   2.3.1 Forces on Vortices . . . . . . . . . . . . . . . . . . . . . . . . . 15
   2.3.2 Vortex Phases . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 18
   2.3.3 Vortices in Thin Films . . . . . . . . . . . . . . . . . . . . . . . 23
   2.3.4 Vortex Dynamics . . . . . . . . . . . . . . . . . . . . . . . . . . . 24
2.4 Vortex Ratchet Devices . . . . . . . . . . . . . . . . . . . . . . . . . . 25
   2.4.1 The Ratchet Effect . . . . . . . . . . . . . . . . . . . . . . . . . . 25
   2.4.2 Applications of Ratchets . . . . . . . . . . . . . . . . . . . . . . 28
   2.4.3 Vortex Diodes . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 28
2.5 Macroscopic Magnetic Properties . . . . . . . . . . . . . . . . . . . . 29
   2.5.1 Field Penetration and Field-Cooling . . . . . . . . . . . . . . . . 29
   2.5.2 Bean and Kim Models . . . . . . . . . . . . . . . . . . . . . . . . 31
   2.5.3 Dynamic Magnetic Properties . . . . . . . . . . . . . . . . . . . . 33
CONTENTS

2.6 Magnetic Microscopy Techniques ........................................... 35
  2.6.1 Magneto-Optical Imaging ............................................. 36
  2.6.2 High-Speed Magneto-Optical Video .................................. 37
  2.6.3 Scanning SQUID Microscopy ....................................... 39
  2.6.4 Comparative Study of MOI and SSM ................................... 41

2.7 Calculations and Analysis Techniques ................................. 43
  2.7.1 Current Calculation (The Inverse Problem) ....................... 43
  2.7.2 Stray Field and Stray Current Error ............................... 47

3 Techniques & Sample Preparation ........................................... 50
  3.1 Pulsed Laser Deposition ................................................ 50
  3.2 Patterning of YBCO films .............................................. 52
    3.2.1 Spin Coating ..................................................... 52
    3.2.2 Laser Lithography ............................................... 54
    3.2.3 Ion Beam Etching ............................................... 56

3.3 Optimisation of Patterning Procedures ............................... 58
  3.3.1 Spin Coating Optimisation ........................................ 59
  3.3.2 Laser Lithography Optimisation .................................. 60
  3.3.3 Ion Beam Etching Optimisation .................................. 62
  3.3.4 Original Patterning Procedure ................................... 64

3.4 Preliminary Measurements ................................................ 66
  3.4.1 MPMS Magnetometry ................................................ 67
  3.4.2 Profilometry ....................................................... 67
  3.4.3 Atomic Force Microscopy ........................................ 69
  3.4.4 Preliminary Tests using MOI ...................................... 70

3.5 Film Properties .......................................................... 70

4 Magneto-Optical Imaging & High-Speed Video ......................... 71
  4.1 Magneto-Optical Imaging Apparatus at ISEM .......................... 71
    4.1.1 Cold Finger Cryostat ........................................... 72
    4.1.2 Magnetic Field System ......................................... 73
    4.1.3 Polarising Microscope Setup .................................. 74
    4.1.4 High-Speed Camera (Photron Fastcam SA3) ...................... 75
    4.1.5 Indicator Films ................................................ 76
    4.1.6 Modifications to the Apparatus ................................ 79

  4.2 Image Acquisition and Analysis Software ........................... 81
    4.2.1 LabVIEW Program for Image Acquisition ...................... 81
    4.2.2 Current Calculation ........................................... 83
    4.2.3 Video Analysis ................................................ 84

  4.3 Experimental Results and Discussions .............................. 87
CONTENTS

4.3.1 Static Magneto-Optical Images and Current Profiles  ........  87
4.3.2 MOI for Locating Defects in YBCO  .................  89
4.3.3 Magneto-Optical Videos of Flux Penetration and Depenetration  92
4.3.4 Dynamic Evolution of Current During Penetration  ........  93
4.3.5 Validity of Dynamic Current Calculation  ...............  103
4.3.6 Vortex Velocity during Flux Penetration  .............  104
4.3.7 Analysis of Current-Carrying Properties by MOV  ....  106
4.3.8 Simulation of MOV Flux and Current Dynamics  ....  113

5  Scanning SQUID Microscopy of Vortex Glass  ..............  116
5.1 Scanning SQUID Apparatus at UTwente  .................  116
5.1.1 Magnetic Shielding  .....................  118
5.1.2 Preparation of SQUID tips  ..................  119
5.1.3 Scanning SQUID Microscopy Procedure  ............  120
5.2 SSM Image Analysis Software  .....................  121
5.2.1 Image Generation and Current Calculation Programs  ....  121
5.2.2 Vortex Phase Characterisation  .................  121
5.2.3 Autocorrelation  .........................  122
5.2.4 Delaunay Triangulation  ....................  123
5.3 Experimental Results and Discussions  ...............  125
5.3.1 Preliminary Background Field Measurement  ..........  125
5.3.2 Analysis of Vortex Glass  ...................  127
5.3.3 Field Dependence of Vortex Glass  ..............  134
5.3.4 Vortex Groups Observed under SSM  ..........  140

6  Vortex Ratchets  ..............  147
6.1 Ratchet Design  .........................  147
6.1.1 Existing Mirrored-Sawtooth Channel Geometry  ....  148
6.1.2 Proposed Geometries and Their Improvements  ..........  149
6.2 Ratchet Device Fabrication  .....................  152
6.2.1 Initial Macroscopic Patterning  ...............  153
6.2.2 Ratchet Patterning by Partial-Thickness Etching  ....  154
6.2.3 Alignment  .........................  155
6.3 Quality Control and Testing  .....................  156
6.3.1 MPMS  .........................  156
6.3.2 Optical Microscopy  .....................  156
6.3.3 Profilometry  .........................  157
6.4 Optimisation of Ratchet Patterning  .............  157
6.4.1 Depth Optimisation  ......................  158
6.4.2 Shape Optimisation  ......................  159
6.4.3 Variations to Etching Method ...................................... 161
6.5 Results So Far ................................................................. 162
  6.5.1 Magneto-Optical Imaging and Video ................................. 163
  6.5.2 Simulation .................................................................. 164

7 Additional Magnetic Microscopy Experiments .......................... 169
  7.1 Critical Analysis of In-Plane Field Correction ......................... 169
    7.1.1 Origin and Effect of In-Plane Field Components ................ 170
    7.1.2 Mathematical Analysis of the Correction Procedure ............. 173
    7.1.3 Experimental Verification ........................................... 176
    7.1.4 Conclusion ................................................................ 176
  7.2 SSM of Superconducting Rings with Half-Integer Flux Quanta .... 176
    7.2.1 Theory of Half-Integer Flux Quanta ................................. 177
    7.2.2 Observation of Integral and Half-Integral Flux Quanta .......... 179

8 Conclusion ........................................................................... 183

List of Publications .................................................................. 185

Bibliography ........................................................................... 186

A The Inverse Problem .............................................................. 201
  A.1 The Inverse Problem in One Dimension ............................... 203
  A.2 The Inverse Problem in 2D ............................................... 206
  A.3 Conclusion .................................................................... 210
## List of Figures

2.1 Penetration Depth and Coherence Length ................................. 9  
2.2 Meissner Effect and Vortices ............................................. 10  
2.3 Magnetisation in Type I and II Superconductors ......................... 12  
2.4 YBCO Atomic Structure .................................................... 14  
2.5 Vortex Phase Diagrams ..................................................... 19  
2.6 “Perfect” Lattice Phase ..................................................... 20  
2.7 Glass Phase ...................................................................... 21  
2.8 Liquid Phase ...................................................................... 22  
2.9 Vortex Diode Forward and Reverse Directions ......................... 29  
2.10 SQUID Diagrams .............................................................. 40  
2.11 Stray Fields and Currents .................................................. 48  

3.1 Lithographically Patterned Sample ......................................... 54  
3.2 Laser Lithography Apparatus ............................................... 55  
3.3 Ion Beam Etching Apparatus ............................................... 57  
3.4 Samples After Etching ......................................................... 59  
3.5 Mechanism of Etching Processes .......................................... 66  
3.6 Profilometry ...................................................................... 68  
3.7 AFM of Domains on YBCO Film Surface ................................. 69  

4.1 Magneto-Optical Imaging Apparatus ...................................... 72  
4.2 Field Ramping Characteristic ............................................... 74  
4.3 Indicator Film Structure & Light Path ..................................... 77  
4.4 Indicator Film Test .............................................................. 78  
4.5 Temperature Sensor Calibration ............................................ 81  
4.6 Indicator Film Calibration .................................................... 83  
4.7 Current Calculation Block Diagram ........................................ 84  
4.8 Static Magneto-Optical Images for Various Field Conditions .... 87  
4.9 Current and Magnetisation from Static MOI ............................ 89  
4.10 Location of Defect-Free Regions by MOI ............................... 90  
4.11 Critical Current in Defect-Free Regions ................................. 91
<table>
<thead>
<tr>
<th>Figure</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.12</td>
<td>Field and Current from MO Videos</td>
<td>93</td>
</tr>
<tr>
<td>4.13</td>
<td>Timeline of Field Penetration and Depenetration</td>
<td>94</td>
</tr>
<tr>
<td>4.14</td>
<td>Time-Evolution of Current</td>
<td>95</td>
</tr>
<tr>
<td>4.15</td>
<td>Motion of Flux Fronts and Current Peaks</td>
<td>97</td>
</tr>
<tr>
<td>4.16</td>
<td>Ramping Current Profiles</td>
<td>98</td>
</tr>
<tr>
<td>4.17</td>
<td>Relaxation Current Profiles</td>
<td>99</td>
</tr>
<tr>
<td>4.18</td>
<td>Transitional Current Profiles</td>
<td>101</td>
</tr>
<tr>
<td>4.19</td>
<td>Flux Front and Current Peak Positions</td>
<td>105</td>
</tr>
<tr>
<td>4.20</td>
<td>$J_c$ in High- and Low-Quality Samples</td>
<td>108</td>
</tr>
<tr>
<td>4.21</td>
<td>MOV for High- and Low-Quality Samples</td>
<td>109</td>
</tr>
<tr>
<td>4.22</td>
<td>Flux Penetration into High- and Low-Quality Samples</td>
<td>110</td>
</tr>
<tr>
<td>4.23</td>
<td>Current Evolution in High- and Low-$J_c$ Samples</td>
<td>111</td>
</tr>
<tr>
<td>4.24</td>
<td>Simulation Results</td>
<td>114</td>
</tr>
<tr>
<td>5.1</td>
<td>Scanning SQUID Microscopy Apparatus</td>
<td>117</td>
</tr>
<tr>
<td>5.2</td>
<td>SQUID Pickup Loop</td>
<td>119</td>
</tr>
<tr>
<td>5.3</td>
<td>Autocorrelation of Hexagonal Lattice</td>
<td>123</td>
</tr>
<tr>
<td>5.4</td>
<td>SSM of Vortices and Circulating Currents</td>
<td>129</td>
</tr>
<tr>
<td>5.5</td>
<td>SSM of Vortices and Circulating Currents</td>
<td>130</td>
</tr>
<tr>
<td>5.6</td>
<td>Field Profile of a Vortex</td>
<td>130</td>
</tr>
<tr>
<td>5.7</td>
<td>Autocorrelation of Vortex Distribution in Glass Phase</td>
<td>131</td>
</tr>
<tr>
<td>5.8</td>
<td>Delaunay Triangulation of the Vortex Glass</td>
<td>132</td>
</tr>
<tr>
<td>5.9</td>
<td>Number of Nearest Neighbours to each Vortex</td>
<td>133</td>
</tr>
<tr>
<td>5.10</td>
<td>Vortex Distributions over $\mu$T Field Range</td>
<td>135</td>
</tr>
<tr>
<td>5.11</td>
<td>Circulating Currents over $\mu$T Field Range</td>
<td>136</td>
</tr>
<tr>
<td>5.12</td>
<td>Delaunay Triangulation on $\mu$T Distributions</td>
<td>137</td>
</tr>
<tr>
<td>5.13</td>
<td>Neighbour Distribution vs. Field</td>
<td>138</td>
</tr>
<tr>
<td>5.14</td>
<td>Field Variation of Hexatic Order Parameter</td>
<td>139</td>
</tr>
<tr>
<td>5.15</td>
<td>Profiles of Vortex Pairs</td>
<td>142</td>
</tr>
<tr>
<td>5.16</td>
<td>SQUID Image of Linear Defect</td>
<td>144</td>
</tr>
<tr>
<td>6.1</td>
<td>Ratchet Geometry: Mirrored Sawtooth Channel</td>
<td>149</td>
</tr>
<tr>
<td>6.2</td>
<td>Ratchet Geometry: Offset-Sawtooth Channel</td>
<td>151</td>
</tr>
<tr>
<td>6.3</td>
<td>Ratchet Geometry: Channel with Linear Protrusions</td>
<td>152</td>
</tr>
<tr>
<td>6.4</td>
<td>Vortex Lensing Sample Geometry</td>
<td>153</td>
</tr>
<tr>
<td>6.5</td>
<td>Bridge Diode Geometry</td>
<td>154</td>
</tr>
<tr>
<td>6.6</td>
<td>Microscope Images of Ratchet Sample</td>
<td>157</td>
</tr>
<tr>
<td>6.7</td>
<td>Profilometry of Ratchet Sample</td>
<td>158</td>
</tr>
<tr>
<td>6.8</td>
<td>Ratchet Patterns in Layout Editor</td>
<td>159</td>
</tr>
<tr>
<td>6.9</td>
<td>Etched Ratchet Patterns on YBCO Sample</td>
<td>161</td>
</tr>
<tr>
<td>Figure</td>
<td>Description</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>-------------</td>
<td>------</td>
</tr>
<tr>
<td>6.10</td>
<td>MOI of Vortex Lens Sample</td>
<td>163</td>
</tr>
<tr>
<td>6.11</td>
<td>Simulated Pinning Potential</td>
<td>165</td>
</tr>
<tr>
<td>6.12</td>
<td>Simulation Results</td>
<td>166</td>
</tr>
<tr>
<td>7.1</td>
<td>In-Plane Stray Fields</td>
<td>171</td>
</tr>
<tr>
<td>7.2</td>
<td>Magnetisation and Applied Field</td>
<td>171</td>
</tr>
<tr>
<td>7.3</td>
<td>S- and D-wave Coupling</td>
<td>178</td>
</tr>
<tr>
<td>7.4</td>
<td>Quantum and Half-Quantum Flux Rings</td>
<td>180</td>
</tr>
<tr>
<td>7.5</td>
<td>SSM of Spontaneous Flux in Ring Samples</td>
<td>181</td>
</tr>
<tr>
<td>7.6</td>
<td>3D Plot of Spontaneous Flux</td>
<td>182</td>
</tr>
<tr>
<td>7.7</td>
<td>Integer and Half-Integer Quantised Flux Values</td>
<td>182</td>
</tr>
<tr>
<td>A.1</td>
<td>1D Inverse Problem Geometry</td>
<td>203</td>
</tr>
</tbody>
</table>
## List of Symbols and Abbreviations

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>Sample Half-Width</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Angle between Magnetic Induction and Indicator Film</td>
</tr>
<tr>
<td>$\alpha_F$</td>
<td>Faraday Rotation Angle</td>
</tr>
<tr>
<td>AFM</td>
<td>Atomic Force Microscopy</td>
</tr>
<tr>
<td>$B$</td>
<td>Magnetic Induction Field (Flux Density)</td>
</tr>
<tr>
<td>$b$</td>
<td>Bottleneck Width</td>
</tr>
<tr>
<td>$B_a$</td>
<td>Applied Magnetic Induction Field</td>
</tr>
<tr>
<td>$B_A$</td>
<td>Magnetic Anisotropy Field</td>
</tr>
<tr>
<td>$d$</td>
<td>Sample Thickness</td>
</tr>
<tr>
<td>$d_{ind}$</td>
<td>Thickness of Faraday-Active Layer of Indicator Film</td>
</tr>
<tr>
<td>$\delta$</td>
<td>Doping Level</td>
</tr>
<tr>
<td>$E_{int}, E_A$</td>
<td>Interaction Energy, Anisotropy Energy</td>
</tr>
<tr>
<td>$\eta$</td>
<td>Vortex Viscosity</td>
</tr>
<tr>
<td>$F$</td>
<td>Force</td>
</tr>
<tr>
<td>fps</td>
<td>Frames per Second</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full Width at Half Maximum</td>
</tr>
<tr>
<td>$h$</td>
<td>Height of Indicator Film</td>
</tr>
<tr>
<td>$H$</td>
<td>Magnetic Intensity Field</td>
</tr>
<tr>
<td>$H_{c_1}, H_{c_2}, H_{c_3}$</td>
<td>Critical Field (Type I), 1st, 2nd and 3rd Critical Fields (Type II)</td>
</tr>
<tr>
<td>HOP (or $</td>
<td>\psi_0</td>
</tr>
<tr>
<td>$i$</td>
<td>Matrix Row Index, Vortex Label</td>
</tr>
<tr>
<td>$I, I_0, I_{max}$</td>
<td>Light Intensity, Background Intensity, Maximum Intensity</td>
</tr>
<tr>
<td>ICE</td>
<td>Integrated Correlated Electronics (research group)</td>
</tr>
<tr>
<td>ISEM</td>
<td>Institute for Superconducting and Electronic Materials</td>
</tr>
<tr>
<td>$j$</td>
<td>Matrix Column Index, Neighbouring Vortex Label, Iteration Number</td>
</tr>
<tr>
<td>$J$</td>
<td>Current Density</td>
</tr>
<tr>
<td>$J_c$</td>
<td>Critical Current Density</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>Ginzburg-Landau Parameter</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Penetration Depth</td>
</tr>
<tr>
<td>Acronym</td>
<td>Definition</td>
</tr>
<tr>
<td>---------</td>
<td>------------</td>
</tr>
<tr>
<td>LHe</td>
<td>Liquid Helium</td>
</tr>
<tr>
<td>LN₂</td>
<td>Liquid Nitrogen</td>
</tr>
<tr>
<td>M</td>
<td>Magnetisation</td>
</tr>
<tr>
<td>Mₛ</td>
<td>Spontaneous Magnetisation</td>
</tr>
<tr>
<td>MO, MOI, MOV</td>
<td>Magneto-Optical, Magneto-Optical Imaging, Magneto-Optical Video</td>
</tr>
<tr>
<td>MPMS</td>
<td>Magnetic Property Measurement System</td>
</tr>
<tr>
<td>n</td>
<td>Frame Number</td>
</tr>
<tr>
<td>n(i)</td>
<td>Number of Nearest Neighbours to Vortex i</td>
</tr>
<tr>
<td>nₙ</td>
<td>Number of Free Vortices per Ratchet Cell,</td>
</tr>
<tr>
<td>Nₙ</td>
<td>Number of Internal Vortices</td>
</tr>
<tr>
<td>p</td>
<td>Position of Current Peak</td>
</tr>
<tr>
<td>P</td>
<td>Ratchet Period</td>
</tr>
<tr>
<td>φ</td>
<td>Angle between Magnetisation and Plane of Indicator Film</td>
</tr>
<tr>
<td>Φ, Φ₀, Φₙ</td>
<td>Magnetic Flux, Magnetic Flux Quantum, Fluxoid Vector</td>
</tr>
<tr>
<td>PLD</td>
<td>Pulsed Laser Deposition</td>
</tr>
<tr>
<td>R</td>
<td>Rectification Factor</td>
</tr>
<tr>
<td>ρ</td>
<td>charge density</td>
</tr>
<tr>
<td>RF</td>
<td>Radio Frequency</td>
</tr>
<tr>
<td>s</td>
<td>Size of Protrusions</td>
</tr>
<tr>
<td>SPM</td>
<td>Scanning Probe Microscopy</td>
</tr>
<tr>
<td>SSM</td>
<td>Scanning SQUID Microscopy</td>
</tr>
<tr>
<td>STO</td>
<td>Strontium Titanium Oxide (SrTiO₃)</td>
</tr>
<tr>
<td>SQUID</td>
<td>Superconducting QUantum Interference Device</td>
</tr>
<tr>
<td>t, tₙ, Δt</td>
<td>Time, Time at nth Frame, Time Step</td>
</tr>
<tr>
<td>T</td>
<td>Temperature</td>
</tr>
<tr>
<td>Tₖ</td>
<td>Critical Temperature</td>
</tr>
<tr>
<td>TFT</td>
<td>Thin Film Technology (research group)</td>
</tr>
<tr>
<td>θ</td>
<td>Polariser-Analyser Angle, Angle of Protrusions</td>
</tr>
<tr>
<td>θᵢᵢ</td>
<td>Angle between Vortices i, j</td>
</tr>
<tr>
<td>U</td>
<td>Pinning Potential</td>
</tr>
<tr>
<td>v, V</td>
<td>Vortex Velocity, Vortex Hopping Rate</td>
</tr>
<tr>
<td>V</td>
<td>Verdet Constant</td>
</tr>
<tr>
<td>W</td>
<td>Hanning Window Function, Ratchet Channel Width</td>
</tr>
<tr>
<td>ξ</td>
<td>Coherence Length</td>
</tr>
<tr>
<td>YBCO</td>
<td>Yttrium Barium Copper Oxide (YBa₂Cu₃O₇₋₅)</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

“What is a superconductor?”

This is a question that researchers like myself are asked on a regular basis. The off-the-cuff answer is usually something like:

“A material that conducts electricity with zero resistance.”

This definition is neat and concise and is often sufficient to satisfy the curiosity of students and the general public. It is certainly interesting to consider that an electrical current can flow through a superconducting wire without losing any energy, without heating the wire at all, and that this current could continue to flow forever in a simple superconducting loop.

However, this definition does little justice to the vast and deep field of research which has captured the passion of researchers, innovators and engineers who for more than 100 years have worked to understand this complex phenomenon and unlock the boundless potential of superconductors for electronic and magnetic applications.

The disappearance of electrical resistance was first noticed by Heike Kamerlingh Onnes in 1911 [1], who was measuring the resistivity of mercury as it was cooled to temperatures as low as could possibly be reached at that time. When the temperature was reduced below about 4K, the resistance suddenly and completely dropped to zero. This effect, later called “superconductivity”, was confirmed in many materials, with each having a different “critical” temperature $T_c$, below which the effect was seen.

In fact, almost any material can be made superconducting by making the temperature extraordinarily low. But for most materials the critical temperature is impractically close to absolute zero, making their superconductivity difficult to investigate or utilise. The critical temperature also decreases with magnetic field, limiting the field range over which low $T_c$ superconductors may be used. For these reasons, high-temperature superconductors are the most sought-after and promising for practical application.
CHAPTER 1. INTRODUCTION

To understand why superconductors have such potential, we must first consider why the simple definition given above is inadequate. Yes, superconductors have zero resistance - this may be a unique property of superconducting materials, but this fact alone is not sufficient to define them. The most blatant oversight in this definition is a second unique and interesting property: the fact that magnetic flux cannot exist inside a superconductor.

This property, called the Meissner effect, is most famously demonstrated by placing a small superconductor just above the surface of a larger magnet (or placing a small magnet above a larger superconductor), and watching it levitate in mid-air. This demonstration relies on the fact that the field around the magnet cannot enter the superconductor. When the magnetic flux is pushed out, a force is created on the superconductor itself (and on the magnet), a force which in this case opposes the force of gravity.

Therefore, a material is superconducting when it has two zeroes: zero resistance and zero magnetic flux inside of it. The material experiences a phase transition on dropping below $T_c$, where the resistance drops immediately to zero, and any magnetic field that was inside the material is expelled$^a$.

So what is a superconductor?

A more precise definition would be as follows:

“A material that is in a state in which it conducts electricity with zero resistance and also expels magnetic flux from its interior. The material will exist in this state over a certain temperature and magnetic field range specific to that material.”

Finally, we have a definition that explains the fundamental facts that are true of all superconductors$^b$. However, there is still a wealth of information yet to be discussed, and the most interesting effects occur only in a particular type of superconductors.

In certain superconducting materials known as ‘type II superconductors’, magnetic flux can in fact enter the material, but only by making small regions of it non-superconducting. These regions along with the small units of magnetic flux within them are called ‘vortices’ (singular ‘vortex’).

Now that several terms have been defined, the focus of this thesis can be properly introduced:

This thesis attempts to give a better glimpse of one area of superconductivity - the behaviour of vortices in thin film superconductors. It is certainly not an exhaustive study of even this specific topic, but enough to explain the motivation

---

$^a$The implications of field expulsion on passing $T_c$ are discussed in section 2.1.2
$^b$Yet another revision must be made in order to give a rigorous theoretical definition of superconductivity, but this will not be introduced until the theory chapter.
for the experimental investigations undertaken during the course of my research; to present the new and interesting behaviour that these have revealed; and to discuss how this work might influence future research on vortex behaviour.

1.1 Research Objectives

The aim of this research was to further the understanding of the behaviour of magnetic vortices in thin film superconductors. Three different aspects of vortex behaviour were investigated, using three distinct techniques. The objectives of each of these were as follows:

- To characterise the phase and order of vortices under a very small external magnetic field, using scanning SQUID microscopy.
- To observe ultra-fast vortex dynamics occurring during and after a change in the external magnetic field, using high-speed magneto-optical video.
- To improve the ability for rectification of vortex motion in superconducting diodes, through the design, simulation and creation of novel microstructures for vortex ratchet devices.

Each of these major investigations constitutes a separate chapter in the experimental part of this thesis.

1.2 Structure of the Thesis

This thesis presents research undertaken on the investigation and manipulation of magnetic vortices in superconducting \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) thin films.

This work centres on magnetic microscopy techniques for investigating the magnetic field around a thin film superconductor. It also describes the creation of vortex diodes by surface thickness modulation using laser lithography, as well as a range of other experimental works carried out in relation to magnetic microscopy. The theory section in chapter 2 explains the physical principles behind the most fundamental parts of this research - namely superconductivity, vortex phases and behaviour, and magnetic microscopy techniques (focussing on Magneto-Optical Imaging and Scanning SQUID Microscopy).

Chapter 3 describes the way in which the superconducting films used in this research were created, tested and shaped into the desired patterns. It also outlines the general properties of samples created in this way.

The experimental part of this thesis begins in chapter 4, which describes the magneto-optical imaging technique and its extension to high-speed video. It also
presents the novel results obtained in many magneto-optical imaging and video experiments. The results of publications 1 and 2 are presented in chapter 4.

Chapter 5 details the scanning SQUID microscopy technique and its use for investigation of vortices in a novel low-field-cooled glass phase. The results of publications 3 and 4 are presented in chapter 5.

Chapter 6 presents two novel designs for superconducting vortex ratchet patterns, along with simulation and experimental investigation of these. These ratchets could be used as diodes that operate over a greater temperature range than similar superconducting diodes.

Additional magnetic microscopy works are listed in chapter 7. These include the analysis of a procedure for correction of magneto-optical images; and the investigation of half-integer-quantised flux in superconducting loops using scanning SQUID.

While some sections have their own individual conclusions, a summary of the entire work and an overall conclusion is given in chapter 8.
Chapter 2

Theory

This chapter aims to introduce some of the theory of superconductivity and other topics that will be required for unfamiliar readers to understand the experimental part of this thesis.

Superconductivity will first be introduced in broad strokes, then the discussion will quickly narrow to focus specifically on the material (YBCO) and properties that were investigated in my own experiments. A description of the theory behind some magnetic microscopy techniques and the subsequent techniques for analysis of their results are given at the end of the chapter.

The discussion is predominately qualitative, as this chapter is intended for those who have little familiarity with the concept of superconductivity. Many of the figures presented in this chapter are explicitly labelled as ‘sketches’, since they do not represent mathematically accurate variation of the quantities described, but are only intended as visual aids for conceptualisation.

The use of equations and symbols is minimised, although some familiarity with electromagnetic theory is assumed. For example, the important distinction is made between magnetic induction field \( B \) and magnetic intensity field \( H \), which are related by

\[
B = \mu_0 (H + M)
\]

(2.1)

where \( M \) is the magnetisation of the material in which these fields are present, and \( \mu_0 \) is the permeability of free space. In later chapters this distinction is relaxed, and \( B \) is referred to simply as the magnetic field.

This chapter begins with quite general theory relating to superconducting materials, but becomes increasingly specific, with the last few sections of this chapter describing the theoretical basis for the experimental part of this thesis.
2.1 Basic Principles of Superconductivity

This section details the very basics of the fundamental theory of superconductivity. It begins with the origin of the zero resistance property, then discusses the Meissner effect, the quantisation of magnetic flux, and finally the origins of the different types of superconductors. Only a brief overview of each topic is given, for a much more comprehensive theoretical examination of superconductivity, see for example [2–4].

2.1.1 Cooper Pairing and BCS Theory

Since resistance-free current is not normal, the way that current flows in a superconductor must be fundamentally different to the way it flows through a normal material. Rather than a movement of individual disassociated electrons, the electrons in a superconductor exist in loosely bound pairs, which are called Cooper pairs.

Unpaired electrons in a normal material would experience zero resistance only in a perfect crystal lattice at zero temperature. Single electrons encounter resistance due to collisions with ions in the imperfect crystal lattice. These occur when ions are displaced from their equilibrium lattice positions. However, paired electrons may pass through the lattice unhindered even in the presence of crystalline defects, phonons and thermal oscillations.

Bardeen, Cooper and Schriefer proposed that the existence of Cooper pairs was made energetically favourable by a phonon-mediated attraction [5]. This made up part of their 1957 microscopic theory of the origin of superconductivity (now called BCS theory). This theory was able to explain an observed energy gap between the ground state of a material in the normal state and the energetically favourable superconducting state. This gap is related to the energy required to break apart the Cooper pair, and this was shown to be provided by thermal fluctuations at $T_c$. This theory therefore explained why superconductors could only exist at low temperatures.

Order Parameter

The wavefunction of Cooper pairs is called the superconducting order parameter. As with any wavefunction, the order parameter is a complex-valued field whose magnitude at any point determines the probability density of finding a Cooper pair at that point.

On the microscopic scale, it is not truly the zero resistance or zero field property that defines a material as being in the superconducting state, but the existence of an order parameter that is phase-coherent over a macroscopic length scale [3]. At
CHAPTER 2. THEORY

the edge of a superconducting region, the order parameter decreases as shown in figure 2.1. The characteristic length of this approximately exponential decrease is called the coherence length, \( \xi \). \( \xi \) varies significantly for different materials, from several micrometres to less than one nanometre.

The Josephson Effect

Superconductors in close proximity experience a coupling which is known as the Josephson effect [6]. Cooper pairs can travel from one superconducting region to another across a small gap by tunnelling.

Josephson predicted that a current will flow between two superconducting regions, with current density proportional to the sine of the phase difference of the order parameter from one region to the other [3, 6, 7]. Such a phase difference can only exist when the superconducting regions are separated by a non-superconducting gap (otherwise the phases of all Cooper pairs become coherent). The current that flows across this gap is called the “Josephson current”. For the overlap of wavefunctions to occur, and therefore for the Josephson current to flow, this gap must be of the order of \( \xi \).

The system of two superconducting regions separated by a gap is referred to as a “Josephson junction”. A Josephson junction has a maximum current that can pass through it, when the phase difference is \( \pi/2 \) radians. This maximum current is referred to as the critical current \( J_c \) of the junction. The critical current of a Josephson junction is always smaller than the critical current of the bulk material, due to weakened superconductivity at the junction [7].

2.1.2 The Meissner Effect

The Meissner effect is the experimentally observed phenomenon whereby magnetic fields are ‘expelled’ from a superconducting bulk [8].

No magnetic field can exist in the bulk of a superconducting sample when it is in the Meissner state. \( \mathbf{M} \) is equal and opposite to \( \mathbf{H} \) everywhere inside the superconductor, which means the total induction field is zero, by equation 2.1. Since magnetisation is a dipole effect, the field lines due to magnetisation curve around the outside of the sample, leading to the induction field \( \mathbf{B} \) appearing to “bend” around a superconducting sample [9].

It should be stressed that the Meissner effect is not simply a result of the zero resistance property of superconductors. That is, it is not simply perfect diamagnetism.

To understand why, consider the experimental evidence for what happens to the field as a material first becomes superconducting: When the temperature is reduced
below $T_c$, at the same time as resistance drops to zero, any magnetic field that is already inside the material is pushed out. The Meissner currents therefore appear spontaneously, not by electromagnetic induction, since there is no change in external field. If a superconductor were simply a perfect conductor, it would indeed resist the entry of any new magnetic field, but a magnetic field already existing within the sample would not be expelled when it became superconducting.

The observation that field is spontaneously expelled when temperature drops below $T_c$ leads to two important conclusions:

1. The Meissner effect is a separate phenomenon to the zero resistance property of superconductors. In fact, these two properties are equally important in defining superconductivity.

2. The drop to zero resistance is not just a change in one property of the material, but a true transition from one thermodynamic phase to another.

Penetration Depth

Magnetic field is only expelled from the bulk of a superconducting material. The Meissner effect allows the presence of an exponentially decaying field within a small distance of the edge of the sample, which is known as the penetration depth.

The penetration depth depends on the properties of a particular superconducting material, and typically lies in the range of tens to hundreds of nanometres. The behaviour of field near the edge of a superconductor is shown in figure 2.1.

Since a field gradient is therefore present at the edge of a superconducting sample, Maxwell’s equations state that a current must flow parallel to this edge. Currents flow in a region near each sample edge with thickness approximately $\lambda$. These are known as “Meissner currents” or “screening currents” since they are necessary to screen the magnetic field from the superconducting bulk.

London Theory

The penetration depth was first proposed theoretically in 1935 by Heinz and Fritz London, who developed a phenomenological theory describing the variation of both electric and magnetic fields in a superconductor and in the surrounding non-superconducting material [10].

The London theory was based on simple experimental observations:

- Firstly, that magnetic fields cannot exist in the bulk of a superconductor.

- and secondly, that electrons flow with zero resistance in a superconducting bulk.
Figure 2.1: Sketch of the spatial variation of magnetic field and order parameter near the edge of a superconductor. These properties both drop off approximately exponentially, with characteristic lengths $\lambda$ (penetration depth) and $\xi$ (coherence length) respectively.

By applying these assumptions to the Lorentz force law on superconducting electrons, along with Maxwell’s equations and some intuitive logic, the London brothers proposed equations describing the variation of the $E$, $B$ and $J$ fields within a superconducting material. These equations will not be given here, but are available in many textbooks [3].

The London theory led to the experimentally verifiable prediction that an externally applied field does not immediately drop to zero at the edge of a superconducting region. Instead, it decreases exponentially with distance into the sample over a short characteristic length $\lambda$, which is called the “penetration depth”. The penetration depth of a particular material can be derived from the properties of the superconducting charge carriers along with some universal constants [3].

2.1.3 Magnetic Flux Quantisation

It has been made clear that magnetic flux is completely expelled from the interior of a superconductor. To clarify mathematically: no magnetic flux passes through any closed loop within a superconducting bulk, provided that the interior of this loop contains no non-superconducting region. But what if this loop encircles a hole?

Interestingly, whenever a hole passes completely through the superconductor (from one side to the other), magnetic flux can indeed pass through it, but this flux only exists in quantised units of $\Phi_0$. $\Phi_0$ is called the “magnetic flux quantum”
or “fluxoid”, and it is not material-dependant. It has a constant value of $2.067 \times 10^{-15}$ Wb, which can be derived from other fundamental constants of nature [3].

![Figure 2.2: Sketch of the interaction of magnetic field lines (blue arrows) with a superconducting film in different magnetic states. Light grey indicates normal state, and dark grey superconducting state. (a) In the normal state, most superconducting materials have very little magnetic response. (b) In the Meissner state, the film expels all field from its interior. (c) In the vortex state, magnetic field is not completely expelled, but is present in the sample within cylindrical normal-state regions called vortices. These images are illustrative only, in reality the field lines do not bend so sharply.](image)

### 2.1.4 Type I and II superconductors

Superconductors are divided into two types based on their response to increasing magnetic fields. While all superconductors have the same response to very small magnetic fields (complete expulsion), radically different behaviour is seen as the field is increased, particularly as it surpasses certain material-specific thresholds. Each type is discussed in detail below:

- In type I superconductors, the Meissner effect is complete up to a critical magnetic field value $H_c$, below which no flux can exist in the interior of the sample. When the external field exceeds $H_c$, the expulsion of field cannot be maintained and the material returns to a normal (non-superconducting) state.

- Type II superconductors show qualitatively different behaviour, the magnetic field is completely expelled only up to a much lower value $H_{c1}$, referred to as the first critical field. For fields above $H_{c1}$, magnetic flux does enter (or ‘penetrate’) into the sample in quantised units of $\Phi_0$. Flux density $B$ in the
sample increases with field $H$, with diamagnetic magnetisation therefore decreasing in the manner shown in figure 2.3(b), up to a second critical field $H_{c2}$. At $H_{c2}$, the bulk of the sample is completely filled with magnetic flux as shown by the zero point of magnetisation in figure 2.3(b). This means that the bulk of the sample has returned to the normal state. Superconductivity can, however, persist in a surface layer up to a much higher third critical field $H_{c3}$ (not shown in the diagram).

Each quantised unit of flux that enters a type II superconductor between $H_{c1}$ and $H_{c2}$ creates a small isolated cylindrical region of normal state material in equilibrium with the surrounding superconducting state. This allows a single fluxoid to pass through the material without violating the Meissner condition of zero flux in the superconducting region, as shown in figure 2.2(c) [3, 11]. Field is maximum at the centre of the fluxoid and decreases radially, while order parameter increases near the edge of the normal core. It is therefore apparent that this structure emerges only when $\xi < \lambda$. The field gradient leads to screening currents flowing in a circular pattern with maximum current near the edge of the normal core, and zero in the centre. The pattern of current flowing around a fluxoid is given the name ‘vortex’ (a name which can also be used loosely to refer to the associated fluxoid and normal core as well). The behaviour and interaction of vortices is discussed in section 2.3.2.

When a type II superconductor does not contain any vortices, it is said to be in the Meissner state. When it does contain vortices, it is in the vortex state. The transition between these states is marked by a sharp change in the magnetic response of the superconductor, as seen in figure 2.3(b). While the sample still expels magnetic flux in the vortex state, this expulsion is no longer perfect, so the sample does not experience the true Meissner effect. Since some flux is still expelled, screening currents continue to flow around the sample edges when it is in the vortex state.

**Origin of Type I and II Superconductivity**

The sharp distinction between superconductors of type I and II finds its origin in the variation of a single parameter $\kappa$ (the Ginzburg-Landau parameter), which is defined as the ratio of penetration depth to coherence length of a superconducting material:

$$\kappa = \frac{\lambda}{\xi}$$

At any boundary between normal and superconducting material, there will be a surface energy resulting from the variation of both magnetic field and order parameter. This arises from the energetically unfavourable expulsion of magnetic field...
Figure 2.3: Diamagnetic response of magnetisation to external field in (a) type I and (b) type II superconductors. The dotted line in (b) shows type I response for comparison. Curves are sketches only and critical field values are not to scale.

(positive diamagnetic energy), which is offset by the energetically favourable Cooper-pairing of electrons (negative condensation energy) in the superconducting bulk.[3]

An increase in $\lambda$ will decrease the positive diamagnetic contribution to the surface energy, since it reduces the region over which magnetic field must be expelled. Conversely, an increase in $\xi$ leads to an increase in positive surface energy, since Cooper-paired electrons are allowed to flow in a larger non-superconducting region. The variation of $\kappa$ therefore leads to significant differences in surface energy [3]:

- If $\kappa$ is small ($0 < \kappa < 1/\sqrt{2}$), then the surface energy at any boundary between normal and superconducting regions will be positive due to the small $\lambda$ and large $\xi$.

The existence of such boundaries is therefore energetically unfavourable, and a mixed normal-superconducting state only exists under very specific conditions (as in the intermediate state at $H \simeq H_c$). This therefore leads to type I superconductivity.

- Conversely, the surface energy is negative for $\kappa > 1/\sqrt{2}$, making it energetically favourable for boundaries between normal and superconducting regions to exist. This is the condition for type II superconductivity.

In this case, when normal-superconducting boundaries exist (in a mixed state) the flux-filled normal regions will tend to divide into smaller regions to maximise boundary area. The configuration that maximises boundary area while maintaining quantisation of flux is a well-separated array of cylindrical normal-state regions that each contain one flux quantum $\Phi_0$, from whence arise vortices [3]. In this case, the mixed state is no longer merely an intermediate special
case for $H \simeq H_c$, but exists (as the vortex state) over a much larger range of fields, from $H_{c1}$ to $H_{c2}$ - see figure 2.3(b). This field range is much larger than that of the Meissner state, and allows type II superconductors to withstand much higher fields than type I before superconductivity is lost.

The sketch in figure 2.1 therefore represents a type II superconductor, since $\lambda$ is significantly larger than $\xi$.

The remainder of this thesis focuses solely on type II superconductors in the vortex state. They can have much higher critical temperatures and magnetic fields than type I, making them significantly more useful for application. The vortex state also possess interesting and unique properties, which will be discussed in section 2.3. The study of vortices will be the focus of the majority of this thesis.

2.2 Yttrium Barium Copper Oxide (YBCO)

The superconductors used in this research are thin film samples of Yttrium Barium Copper Oxide, which has the chemical formula YBa$_2$Cu$_3$O$_{7-\delta}$ (often shortened to YBCO). It is a type II superconductor that can have a critical temperature as high as 92 K.

A 7 – $\delta$ subscript is given to oxygen in the chemical formula to indicate that YBCO is almost always subjected to oxygen-reduction doping, the reduction of oxygen atoms from the true stoichiometric ratio. The doping level $\delta$ of a particular YBCO sample has a very significant effect on the superconducting and electronic properties of that sample [12]. The highest $T_c$ is found at the so-called “optimal doping”, which is generally taken as $\delta \sim 0.07$.

YBCO is a ceramic compound in the cuprate family of superconductors. The undoped YBa$_2$Cu$_3$O$_7$ has a perovskite structure consisting of layered planes of different atomic composition. The unit cell, as shown in figure 2.4, contains one Y plane, two BaO planes, two CuO$_2$ and one CuO plane. The CuO plane consists of linear chains of copper and oxygen atoms. When $\delta$ is increased, the oxygen vacancies tend to appear at sites on these CuO chains.

YBa$_2$Cu$_3$O$_{7-\delta}$ is highly sensitive to water, which can cause it to decompose into a non-superconducting phase [13, 14]. Samples experience rapid degradation of both $T_c$ and $J_c$ if immersed, or gradual degradation over time when exposed to moisture in the air. To control this, YBCO samples are often stored in a low humidity environment.

No high-temperature superconductor is naturally occurring, and this includes YBCO. The method employed for production of YBCO samples in this research was pulsed laser deposition (PLD)[15, 16]. Using PLD, YBCO samples were produced
CHAPTER 2. THEORY

Figure 2.4: Diagram of YBa$_2$Cu$_3$O$_7$ unit cell showing copper oxide planes and chains. Yellow circles are oxygen, red are copper, green are barium and the blue circle is yttrium, as labelled. Lattice directions are also indicated.

as thin films with the $c$-axis oriented perpendicular to the plane of the film, the process of which will be described in section 3.1.

Other cuprate superconducting materials also exist such as La$_2$CuO$_4$ [17], and Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ [18]. These compounds are collectively referred to as “high-temperature superconductors”, along with any other superconductor with critical temperature above 77K (though the term is sometimes loosely applied to other compounds with a lower $T_c$). To emphasise the difference in properties between high-temperature superconductors and other superconducting materials, other materials with simpler atomic structures are referred to as “conventional superconductors”.

Consideration of the superconducting properties of YBCO and other cuprate materials shows that superconductivity is strongest in the CuO$_2$ planes. This leads to strongly anisotropic critical current in all cuprates, with much higher currents
able to flow in the $ab$-plane than in the $c$-direction.

### 2.3 Vortices in Type II Superconductors

As introduced in the section 2.1.4, a vortex is a pattern of current encircling a cylindrical normal region within a superconducting bulk, containing one quantum of magnetic flux. The normal core, flux quantum and circulating current are inextricably linked, and none can exist without the others (except in the case of Josephson vortices, as described in a subsection below).

Vortices in type II superconductors are often not static. They are subject to a number of motion-influencing forces including the driving force, vortex-vortex repulsion, viscous drag, pinning forces and thermal motion. Each force on a vortex contributes uniquely to their overall behaviour, making the dynamics of vortex motion a rich and complex field of study.

#### Josephson Vortices

A Josephson vortex is a special type of vortex that is formed on a Josephson junction. A quantum of flux can pass through the material more easily at the location of a Josephson junction due to the existence of the non-superconducting region. No normal core exists for Josephson vortices. Instead, the fluxoid is centred at a position in the normal part of the junction. By contrast, a vortex that does have a normal core is referred to as an Abrikosov vortex\(^a\).

Since a Josephson junction is highly non-isotropic, the magnetic flux profile is not circularly symmetric but elongated along the direction of the junction. Circulating currents have a similarly elongated profile.

#### 2.3.1 Forces on Vortices

Firstly, when a current flows around a vortex, the vortex experiences a driving force due to the interaction of this external current with its own circulating current. This superposition of currents creates an increase in current density on one side of the fluxoid, while it is decreased at the other side. The fluxoid tends to move toward the region of greater current density. This force is very similar to the Magnus force on spinning objects moving through the air. The driving force is given by [3]:

$$
F_{\text{driv}} = J \times \Phi_0
$$  \hspace{1cm} (2.3)

\(^a\)However, in some cases, mixed Abrikosov-Josephson vortices have been observed [19].
Where the fluxoid vector $\Phi_0$ is defined to have magnitude $\Phi_0$ and direction matching that of the magnetic field in the vortex core. The magnitude and direction of the driving force on vortices is therefore equivalent to the Lorentz force of classical electromagnetism. This has led to the term ‘Lorentz force’ being commonly used to describe the force that vortices experience in the presence of a current, although this is not technically correct. The field of the vortex remains confined to the normal core, while current flows only outside of this core, meaning that a direct Lorentz interaction of the field and current does not truly occur.

As a result of the driving force, when two or more similarly-directed vortices exist in a superconductor, they have a natural tendency to repel one another. This is due to the interaction of the circulating current around each vortex with the other. The current due to each vortex will create a force on the other in such a way that they mutually repel one another. This is called “vortex-vortex repulsion”. An equation is not given here for vortex-vortex repulsion, since it is complex and depends on the type of vortices and the properties of the superconducting material\textsuperscript{b} [20, 21].

Conversely, two oppositely-directed vortices have a tendency to attract one another. When a vortex contains a quantum of magnetic flux in the opposite direction to that of the original external field, it is referred to as an “antivortex”, and the two oppositely-directed vortices are therefore referred to as a “vortex-antivortex pair”\textsuperscript{c}. The current around an antivortex circulates in the opposite direction, which creates an attractive force on the regular vortex, and vice-versa. Mutual attraction can cause vortex-antivortex pairs to meet, and when this occurs they will annihilate, since both the fields and currents cancel one another out.

Pinning forces arise due to the attraction of vortices to regions of weakened superconductivity known as “pinning sites”. Pinning sites may be crystal lattice defects, holes or “antidots”, or regions in which the sample has been deliberately damaged (eg. by irradiation). In most cases, many pinning sites exist throughout a superconducting material, and these define a potential landscape. The pinning force at any position in the material is given by the gradient of the pinning potential $U$.

$$F_{\text{pin}} = \nabla U$$  \hspace{1cm} (2.4)

In Bean’s critical-state model, which is discussed in section 2.5.2, vortices remain static in their pinning sites whenever the driving force is less than the pinning force. If the current density around a vortex exceeds a critical value ($J_c$), then the driving

\textsuperscript{b}See equation 6.1 for an example of a vortex-vortex repulsion equation that holds under specific conditions

\textsuperscript{c}The choice of which field direction to label ‘regular’ vortices and which to label ‘antivortices’ is entirely arbitrary.
force exceeds the pinning force and the vortex begins to move. The critical current density required for de-pinning individual vortices is therefore given by

\[ J_c \times \Phi_0 = -\nabla U \]  \hspace{1cm} (2.5)

When vortex motion is induced by a current exceeding \( J_c \), a voltage is also induced. This implies that the effective resistance of the superconducting material is no longer zero for \( J \geq J_c \).

The de-pinning of an individual vortex does not always mean that it is free to move. When the vortex density is high, each vortex is also held in place by the repulsion of many neighbouring vortices. Since some or all of these neighbours may also be pinned, a much stronger “collective pinning” force prevents the motion of the entire lattice of vortices (see section 2.3.2). When currents are large enough to overcome collective pinning, large numbers of vortices tend to move together, while retaining a relatively fixed distance from one another due to vortex-vortex repulsion.

The superconducting material provides viscous drag on vortices when they are in motion [22]. This drag force tends to slow down vortices by an amount proportional to their velocity, for both individual and collective motion. By analogy to viscous forces on particles moving through a fluid, the equation for this drag force is:

\[ F_{\text{drag}} = -\eta v \]  \hspace{1cm} (2.6)

Where \( \eta \) is the viscosity, which is material dependant, and \( v \) is the vortex velocity.

Finally, vortices are subject to thermal motion in much the same way as molecules or other small particles. Vortices in a superconductor at a temperature above absolute zero experience a vibrational force proportional to the temperature. The thermal force is randomly directed at any instant, but has a zero time average.

**Penetration and De-penetration**

The main mechanism by which vortices enter a superconducting sample is by penetration. “Penetration” refers to the formation of a vortex at the edge of a sample as flux moves from the non-superconducting region into the superconducting region. The flux in the non-superconducting region tends to be higher near the sample edge due to the “demagnetisation effect”, which is the shape-dependent increase of magnetic field around the sample edges since this field has been expelled from the interior of the sample. Thus, the superconducting region closest to the edge experiences a higher field than the bulk of the sample. When the field near the sample edge increases, the sample can no longer continue to expel it, and a quantum of flux (or several) moves into the sample. The currents flowing at the sample edge
redistribute into circulating currents around the newly-formed vortex, which then usually moves further into the sample due to the existence of an inward-sloping field gradient\textsuperscript{d}.

De-penetration is the reverse of penetration: it occurs as flux leaves the superconducting region by the destruction of vortices at the edges. Vortices experience a short-range attraction toward edges of the sample, and tend to exit the sample as flux moves outside the sample edge. This short-range attraction occurs because circulating current around a vortex is altered by proximity to the sample edge. Since supercurrent cannot flow outside of the superconducting region (beyond $\xi$), circulating currents on the side of the vortex closest to the edge are condensed into a smaller area \[2\]. The higher current density on the side of the vortex closest to the sample edge causes a driving force on the vortex, which is directed toward the edge. The magnitude of this force increases as the vortex moves closer to the edge.

The entry and exit of vortices is made less energetically favourable by the existence of potential barriers at the edges. These edge barriers can arise due to pinning, which is enhanced by roughness of the edge, or it can be a geometrical effect resulting from the cross-sectional geometry of flat samples \[23,24\].

### 2.3.2 Vortex Phases

The larger-scale distribution of vortices in a type II superconductor is referred to as the “vortex phase” by analogy to the phases of matter. Just as the arrangement of atoms in a material is vastly different for the solid, liquid and gas states, and further differences arise between different phases of material in a given state, the arrangement of vortices with respect to one another occurs in vastly different ways.

A superconducting material can exist in the Meissner state, mixed (vortex) state, or normal (non-superconducting) state. These states are discussed in section 2.1.4, and shown in figure 2.5(a). Of these, the most complex is the vortex state, which is further divided into many different phases, depending on the distribution and motion of vortices. An illustrative example of some of the possible phases is shown in figure 2.5(b). The three main phases are: lattice, glass and liquid, and each of these will be described in detail in the following subsections.

Differences in vortex distribution between phases arise due to experimental conditions such as field strength, temperature and magnetic history; properties of the superconducting material such as anisotropy and dimensionality; and the type(s) of available effective pinning centres \[25-29\]. Transitions from one phase to another arise due to changes in the aforementioned conditions.

A vortex phase diagram shows the different phases seen in a particular super-\textsuperscript{d}The field gradient will be explained in section 2.5.2
Figure 2.5: Sketches of: (a) A typical temperature/field phase diagram for a type II superconductor, (b) An example of a vortex phase diagram. The size and shape of the phases in the diagram are illustrative only. The thin spike in the glass and lattice phases close to the Meissner phase is referred to as the “re-entrant” part of the phase diagram. In reality this spike is generally much smaller and closer to $H_{c1}$.

conductive material for given temperature and magnetic field conditions. However, the phase diagram may vary greatly under changes in the properties of the superconducting material\cite{9, 25}.

**Lattice Phase**

In a very pure superconductor at temperature far below $T_c$, the dominant force on vortices is vortex-vortex repulsion. If the influences of defects, thermal fluctuations and driving forces were removed completely, the most stable distribution of vortices would be a regular lattice taking the hexagonal shape shown in figure 2.6a. This distribution is described as a “regular” or “perfect” vortex lattice, and is highly stable because each vortex lies as far from each of its neighbours as possible. In this perfect lattice each vortex has six nearest neighbours and the distribution has sixfold rotational symmetry.

Figure 2.6(b) shows Delaunay triangulation applied to the regular lattice. Delaunay triangulation is used to identify the nearest neighbours to any vortex, and to determine the angles of the bonds between these neighbours. It is described in detail in section 5.2.2. For the perfect lattice shown in figure 2.6(b), each vortex has exactly six nearest neighbours, with an angle of 60 degrees between each of them.

The perfect lattice described above is rarely, if ever, seen in experiment. Even very regular lattices have deviations in the form of topological defects. Specifically, a topological defect is a change in the distribution that cannot be reversed by a continuous deformation of the Delaunay triangulation. Such defects therefore do
not include small movements of vortices around their equilibrium positions, but occur when the lattice restructures such that it no longer shows the symmetry seen in figure 2.6(b). Topological defects can take the form of dislocations or disclinations [30, 31].

A disclination can be defined as occurring when a vortex has a number of nearest neighbours other than 6, this is described as a positional defect [31]. A dislocation is defined as a pair of adjacent disclinations occurring in such a way that one vortex has more nearest neighbours while a nearby vortex has fewer neighbours, this is described as an orientational defect [31]. Dislocations and disclinations can therefore be easily identified computationally from the Delaunay triangulation.

Provided that the total proportion of topological defects is small, the overall distribution of vortices is still considered to be in the lattice phase.

Topological defects usually arise due to underlying variations in the superconductor’s potential landscape. For example, an isolated topological defect can be formed due to a strong pinning site, which causes a vortex to be trapped at a location far from its position in the perfect lattice, which in turn causes other vortices to redistribute themselves around it.

Glass Phases

When the density of topological defects is great, a vortex distribution becomes highly disordered and is considered to be in a glass phase [25, 26, 32]. Glass phases arise when pinning is the dominant force influencing the position of vortices. Since the arrangement of vortices in a glass phase is stable (they are not mobile as in the liquid phase), it is described as a state of quenched disorder. A very detailed description
of glass phases is given in a review by Natterman [26]. An example of vortices in a glass phase is given in figure 2.7.

![Figure 2.7:](image)

There are several classifications of glass phases, characterised by the degree and type of order found in the vortex distribution. As may be expected in a largely-disordered system, there is great variation among these glass phases. Commonly identified glass phases include the Bragg glass [33], positional vortex glass [26], Bose glass [27, 28] and others. The phase diagram of a given superconductor may in fact contain many different glass phases, the relative size of each phase being determined by a multitude of material properties [26, 34, 35]. Some glass phases may only be seen under very specific conditions in a given material.

In a vortex lattice, the majority of vortices are unpinned or only weakly-pinned, and hence vortex-vortex repulsion is the dominant mechanism determining the positions of vortices. As the number of pinned vortices increases, the density of topological defects increases and the vortex distribution becomes more glass-like. Therefore the pinning force is said to be the dominant mechanism in a glass phase.

The investigation of vortex distributions in glass phases has been a subject of intense interest in the literature [26, 32], with transitions to glass phases observable through macroscopic changes in magnetisation and current, in two-dimensional [35] and three-dimensional [34] superconductors. Vortex glass phases can also be directly observed and analysed, and they show a variety of interesting features [36], and dependences [37].

In this research, investigations were made into vortex glasses through direct measurements of vortex positions using scanning SQUID microscopy. These are described in sections 5.3.2 and 5.3.3.
CHAPTER 2. THEORY

Liquid Phase

As the temperature of a vortex lattice or glass is increased, thermal motion results in a “shaking” of the vortices about their equilibrium position. When the temperature is high enough for thermal fluctuations to remove vortices from their equilibrium positions, the distribution enters the liquid phase. This transition is generally not sharp, and may occur more quickly in some parts of the sample than others. In the absence of external (driving) forces, vortices in the liquid phase will experience a random walk similar to physical particles in a liquid or gas phase. This is illustrated in figure 2.8.

![Figure 2.8: Random vortex distribution in the liquid phase, with arrows showing thermal motion](image)

For strongly-pinned vortices (most vortices in the glass phase), the equilibrium position is the centre of the pinning site. For weakly-pinned or unpinned vortices (most vortices in the lattice phase), the equilibrium position arises due to vortex-vortex repulsion. For vortices in the liquid phase, thermal motion is dominant over both pinning and vortex-vortex repulsion. Vortices in this phase possess enough thermal energy to leave pinning sites and the equilibrium positions between neighbouring vortices.

The vortex liquid part of the phase diagram is generally seen at relatively high temperatures and under high applied field. Since the vortices are in motion, and a temperature close to $T_c$ must be maintained, the distribution of vortices in the liquid phase could not be investigated in a direct manner as was carried out for the vortex glass phase.

A vortex liquid phase can also be induced by the application of a driving force, which leads to quite different behaviour in this phase. The motion of vortices under a driving force will be discussed further in section 2.3.4.
2.3.3 Vortices in Thin Films

Vortices in thin films are different in structure due to their low dimensionality, but the forces on them are similar to those found in bulk (three-dimensional) superconductors.

Vortices in thin films have a strong tendency to orient themselves normally to the film’s surface\[38\], even if field is applied at some angle to the normal. If this angle is large, the component of field parallel to the film’s surface can create another array of vortices lying along the film. Therefore two perpendicularly oriented arrays of vortices are often seen in thin films, with one array parallel to its surface (in-plane), and the other normal to it (out-of-plane). These arrays can influence one another or be decoupled \[39\].

Since thin film superconductors are often produced with their surface parallel to the \textit{ab}-plane (see sections 2.2 and 3.1), these two vortex orientations therefore align with the crystalline directions in the film. The distinction between them is therefore heightened, as in-plane vortices will preferentially lie along the planes of weakened superconductivity between Cu-O$_2$ planes.

Vortices in each of these orientations behave in a manner quite different from one another and from vortices in a bulk sample \[40\]. Since field is usually applied perpendicular to thin film \textit{c}-oriented YBCO in this research, the vortices observed are out-of-plane.

Vortex Phases in Thin Films

Different vortex phases are realised in the two-dimensional (2D) case of out-of-plane vortices in superconducting thin films. While the lattice and liquid phases are similar, glass phases show different characteristics and ordering with the lower dimensionality.

Many 2D glass phases show short-range sixfold point symmetry\[26\], similar to the sixfold symmetry of the lattice phase. Glass phases showing this type of short-range orientational order are broadly referred to as “hexatic” \[36, 41\]. In a hexatic glass, it is common for most vortices to have six nearest neighbours, and for these to have roughly even angular spacing. On the other hand, the distances between neighbouring vortices are usually irregular (i.e there is no positional order), and there is no long-range symmetry.

By contrast, a 2D glass phase that does not show this type of ordering usually shows very little orientational order at all, and is therefore referred to as an “isotropic glass” \[36, 41\]. Many simulations have shown the emergence of isotropic and hexatic glass phases under different conditions\[31, 34\], but a direct transition from isotropic to hexatic or vice versa has not been observed.
The experimental investigations in sections 5.3.2 and 5.3.3 focus specifically on identification of hexatic and isotropic vortex glasses.

2.3.4 Vortex Dynamics

When vortices are in motion, the changing magnetic field leads to a changing electric field, and therefore a voltage. This non-zero voltage creates an effective resistance in a superconducting sample [3]. The movement of vortices can be considered to result in dissipation of energy, and hence reduces the energy of the superconducting electrons.

Therefore the superconductor does exhibit resistance when it is in the liquid phase, and this means that the transition from liquid phase superconductivity to the normal state is often smooth. High-temperature superconductors often have a large liquid phase and therefore no longer show such sharp transitions in resistance as seen for conventional superconductors. The sharpness of the transition from the normal to superconducting state can also be used to characterise the quality of a superconducting material, as discussed in section 3.4.1.

Vortices can be removed from their pinning sites by application of a driving force, as described in section 2.3.1. However, when vortices are in the lattice phase, a motion of vortices can be induced only if the driving force is greater than the collective pinning force of the vortex distribution [25, 42]. This is because each individual vortex is not only held in place by pinning, but by repulsion from all other pinned vortices. The effect of several vortices being pinned can also fix the position of many nearby vortices since the lattice structure is maintained. The collective pinning effect is generally much stronger than the pinning of any individual vortex. Therefore a driving force greater than the pinning force for individual sites may not cause any vortices to move.

The collective pinning effect also exists in glass phases but is less significant since vortex-vortex repulsion is generally less strong than the pinning force in these distributions. In this case, a driving force less than the collective pinning force may cause individual vortices to “hop” from one pinning site to another. A very small resistance is induced by vortex hopping, due to the short-lived motion of each vortex, but the overall distribution is mainly static and is therefore not considered to be in the liquid state. Vortex hopping is discussed further in section 2.5.3 in relation to the macroscopic flux creep phenomenon.

If the driving force is greater than the collective pinning force, a vortex lattice could theoretically move as a bulk, but due to the highly irregular potential landscape created by pinning forces in real samples, this is not commonly observed.

Instead, some regions of the sample tend to have weaker pinning while other
regions have stronger pinning. Therefore a driving force greater than the collective pinning force for the weak-pinning regions leads to the mobilisation of only the vortices within these regions. The mobilised vortices then flow as “rivers” of vortex liquid in the direction of the driving force [25]. Vortices in the driven liquid state move in a turbulent fashion due to thermal motion and vortex-vortex repulsion from static pinned vortices. The river of vortices in the liquid state tends to meander around regions of higher pinning where vortices remain in a lattice or glass state.

2.4 Vortex Ratchet Devices

Rectification of vortex motion is defined as the suppression of vortex motion in one direction, while they remain relatively free to move in the opposite (anti-parallel) direction. A “vortex ratchet” is a device designed to maximise rectification of vortex motion in a superconducting system. A very thorough introduction to vortex ratchets is given in a recent review by Plourde [43].

The vortex ratchet effect is introduced and explained in section 2.4.1, and applications of this effect are listed in section 2.4.2. The specific application of superconducting diodes is discussed in section 2.4.3.

2.4.1 The Ratchet Effect

Before ratchet devices can be understood, the effect that allows rectification of vortex motion must be discussed.

Rectification is generally achieved by creating an asymmetric potential landscape for vortices, though there are some exceptions. The asymmetric potential is achieved through the creation of an asymmetric pinning landscape, which strongly pins some vortices, leading to an asymmetric vortex-vortex repulsion potential on those vortices that are not so strongly pinned. This will be explained below.

The asymmetric potential is designed to give rise to a single direction in which vortices are preferentially allowed to move. This is referred to as the “easy-flow” direction, while the direction in which they cannot so easily move is the “hard-flow” direction. For most ratchets, an oscillating driving force is applied that forces vortices in both the easy- and hard-flow directions by an equal time-averaged amount. The asymmetric potential then tends to move the time-averaged vortex position in the easy-flow direction.

Several methods for producing vortex ratchets have been proposed and tested:

- Local thickness modulations: The free energy of a vortex decreases with reduced sample thickness [43, 44]. This reduction in free energy leads to an increase in pinning potential, meaning that vortices are preferentially pinned
in thinner regions of the film. Daldini et al. created periodic modulation of a sample’s thickness by applying a photosensitive substance to the films and then applying spatially-periodic illumination [45]. This created symmetric grooves in the sample running along one direction, which act as pinning sites. This was seen to create asymmetry in vortex motion in perpendicular directions, though it may also lead to ratchet motion if the grooves were made to be asymmetric [46].

Lee et al. simulated an effective vortex ratchet using sawtooth-shaped thickness modulation [47], and a similar ratchet was produced experimentally by Sabatino et al. [48].

- Voids and antidots: Ratchets produced using arrays of triangular antidots have been investigated through experiment [49] and simulation [50, 51]. In this geometry, one point of each triangle is directed toward the base of the next in the array. Vortices can hop across the gaps between triangles more readily in the direction from point to base, since they are moving toward an area with larger pinning potential. This effectively creates a sawtooth potential.

No asymmetry in critical current was seen for an equivalent lattice of square antidots, showing that it is the asymmetric shape of pinning sites that produces the ratchet effect [44].

Alternatively, Wu et al. varied the pinning potential across a film by patterning symmetric antidots, but with a graded density [52]. This also induced rectification.

- Magnetic dots: Asymmetric pinning sites could be created by magnetic dots. In this case, the ratchet effect can be created in a similar way to the way it is produced by asymmetric antidots [43, 53].

- Superconducting-magnetic heterostructures: Ainbinder and Maksimov found that the presence of a magnetic strip may produce a diode effect by reducing one edge barrier to vortex penetration\textsuperscript{6}. Whenever an edge barrier is present, a certain threshold current must be applied before vortices begin to form. If a magnetic strip is present at one edge of a superconducting tape, this threshold value becomes dependent on the direction of transport current, leading to a ratchet effect. This also leads to an earlier transition from the Meissner state into the vortex state and then into the resistive state for one current direction [54].

\textsuperscript{6}see the “Penetration and De-penetration” subsection of section 2.3.1
• Non-uniform Josephson Junctions: Krasnov et al. found that Josephson vortices behave differently in different parts of a non-uniform junction [55]. Junctions with non-uniform critical current distributions or with temperature gradients were shown to favour vortex motion in a particular direction, which can lead to the presence of a voltage across the junction at zero current, and to a preferential Josephson current in one direction.

• Time-asymmetric driving force: An alternative technique proposed by Cole et al. uses an asymmetric driving force rather than an asymmetric potential [56, 57]. This technique relies on the interaction between Josephson (in-plane) vortices and pancake (out-of-plane) vortices in bulk CuO$_2$-based superconductors. Pancake vortices are attracted toward Josephson vortices and it has been shown that the movement of Josephson vortices (induced by changing the in-plane magnetic field) can drag pancake vortices in the same direction. The amount of dragging decreases as the speed of the Josephson vortex is increased. Therefore, when an asymmetric AC in-plane field such as a sawtooth waveform is applied to the sample, the Josephson vortices move slowly in one direction, dragging pancake vortices with them, before moving quickly in the opposite direction and not dragging the pancake vortices back by the same amount. Therefore a ratchet effect is achieved for the pancake vortices.

Many of these methods rely on only some of the vortices in the material being strongly pinned, with the unpinned vortices (called “free” or “interstitial” vortices) experiencing a ratchet potential created through repulsion from the pinned vortices [44, 49, 58, 59]. Whenever this is the case, the ratchet would only operate over a certain field range - specifically, the range in which there are enough vortices to fill the strong pinning sites and still leave other vortices free to experience the ratchet potential [49].

Vortex ratchets relying on interstitial vortex effects also tend to have complex temperature and magnetic field dependences, since these affect all of the forces on vortices in different manners [58, 59]. These ratchets are also dependent on the amplitude and frequency of the driving force, since these affect the dynamics of vortex motion and can therefore limit their response to the potential landscape [58].

Many vortex ratchets rely on a pinning potential landscape that is finely tuned in two dimensions, rather than a simple one-dimensional sawtooth potential [60–62]. These two-dimensional vortex ratchet potentials will be discussed in much greater detail in chapter 6, since the novel ratchet geometries proposed are based on this method.
2.4.2 Applications of Ratchets

The ratchet effect is very promising for applications. A variety of devices have been proposed, simulated and/or created that put this effect to use:

- **The most straightforward application is the flux pump**, whereby suitably-oriented vortex ratchets can be used in conjunction with a driving force to direct vortices from one part of a superconducting device to another, or out of the device entirely. This is particularly useful for SQUIDs, which are very sensitive to interference by vortices. A practical system to remove vortices from a realistic SQUID device was simulated using a ratchet potential by Lee et al. [47].

- **Flux lenses** can be created using suitably-oriented ratchets. Flux lensing is the local increase in vortex density in some regions of the sample and decrease of vortex density in others. This is used to preferentially direct vortices from one region of the sample to another, effectively “focussing” magnetic flux into one area [49]. Flux lenses differ from flux pumps in that they are generally passive, operating without a deliberately-applied driving force.

- Finally, since movement of vortices in a particular direction is associated with dissipation of current in a perpendicular direction, the ratchet effect can be used to create vortex diodes. Lossless current flows more freely in the direction for which vortex motion is suppressed, leading to an electrical diode effect. This application is discussed in more detail in the following section.

2.4.3 Vortex Diodes

The rectification of vortex motion can be used to create diode devices. These devices aim to restrict the flow of electric current in one direction while allowing it in the other. Diodes can be created based on the principle of current dissipation by vortex motion, as introduced in section 2.3.4. Since vortex motion in a ratchet occurs for a lower driving force in a given direction, there is increased vortex motion for one particular current direction, leading to a lower critical current and increased resistance in that direction.

Current flows more freely in a certain “forward” direction, since this drives vortices in the hard-flow direction, in which their motion is heavily suppressed. Current in the opposite (“reverse”) direction drives vortices in the easy-flow direction, which leads to dissipative vortex motion at much lower currents - in other words, a reduced critical current. The directionality of a vortex diode is illustrated in figure 2.9.

An applied current that is larger than $J_c$ in the forward direction but smaller than $J_c$ in the reverse direction will experience dissipative resistance only in the
2.5 Macroscopic Magnetic Properties

Since superconductors are rarely observed at the vortex scale, it is important to describe their magnetic behaviour on a larger scale. This will be done in this section, with reference to the microscopic behaviour that gives rise to each observable macroscopic effect.

2.5.1 Field Penetration and Field-Cooling

On a macroscopic scale, penetration of vortices into a superconductor can be observed by the growth of flux-filled regions from the sample edges. As vortices enter the sample, vortex-vortex repulsion tends to push the vortices that had entered previously further into the interior of the sample. The line dividing the flux-filled region from the flux-free region (sometimes called the Meissner region) further from the edges is called the “flux front”. As external field is increased, the flux front moves
inward from each edge. Current flows through the flux filled region in a direction parallel to the nearest edge.

Flux does not penetrate from the corners of the sample, and in fact the flux fronts from adjacent edges tend not to cross the diagonal line extending from any corner. These lines represent a discontinuity in current, as it turns sharply to remain parallel to the edge. These discontinuity-lines (or “d-lines”) tend to remain flux-free during penetration. “Full” or “complete” penetration is reached when the flux fronts meet at the sample centre and the sample is filled with vortices (though flux-free regions may persist on the d-lines). If the flux fronts are some distance from the sample centre, the sample is in the “partial penetration” state.

Magnetometry measurements of superconductors\(^7\) show that they exhibit significant magnetic hysteresis. That is, the magnetisation of a sample decreases as a previously-applied external field is reduced, but it does not return to zero when the external field is reduced to zero. The magnetised sample in zero external field is said to be in the “remnant state”. A further decrease in field to negative values (i.e. application of field in the opposite direction) results in this remnant magnetisation decreasing and eventually passing through zero to negative values.

This occurs because some flux remains trapped in the centre of the sample (or in the region closest to the penetration front if the sample was not fully penetrated) due to vortex pinning. The mechanism for the decrease in flux can be either vortices overcoming the pinning forces and exiting the sample from the edges, or by antivortices penetrating from the edges and annihilating with vortices pinned in the sample. These two processes are equivalent under the method of images, differing only in the location of the annihilation event \(^8\).

When the applied field value moves through zero (external field direction is reversed), an antivortex flux front penetrates from the sample edges. When vortices in the original direction are still present in the sample, this flux front is an annihilation front. Antivortices must annihilate with all vortices before the sample can be fully penetrated in the reverse direction. Along with these antivortices, a current in the opposite direction to the original screening current penetrates the sample \([54, 63]\).

This hysteretic behaviour is repeated on successive increase and decrease of the applied field. The penetration behaviour is different only for superconductors in the virgin state - that is, for superconducting samples that have not had any field applied since transitioning into the superconducting state. In this case, the penetration of vortices occurs in a simpler manner, since there is no remnant field to oppose them. The virgin sample therefore reaches complete penetration more quickly than a sample that has been previously penetrated.

\(^7\)such as those obtained by MPMS, as described in section 3.4.1

\(^8\)Both antivortices and the method of images are defined in section 2.3.1
For this reason, experimental investigations often specify whether superconductors are studied in the zero-field-cooled or field-cooled state. This refers to the value at which the applied field is held during cooling of the sample from above $T_c$ to the measurement temperature (generally the field is kept constant during cooling). Only zero-field-cooled samples can be studied from the virgin state. Field-cooled samples are also important for study, as the distribution of flux throughout the sample is quite different in this case to that of zero-field-cooled samples that have had flux added through penetration.

The differences between the vortex matter under the two types of cooling can be explained by considering the phase diagram of the material. Consider the phase diagram given in figure 2.5: to field-cool this sample to the lattice phase from above $T_c$, it must first pass through a large temperature range in which it is in the liquid phase, and then similarly for the glass phase. Other samples may pass through the liquid phase only, in which case the vortices are mobile, and will coalesce steadily into the equilibrium positions of the lattice phase as thermal fluctuations decrease. A zero-field-cooled sample remains in the Meissner state throughout the entire temperature range, then since only a few vortices are able to penetrate when field increases just above $H_{c1}$, it does not truly experience the low-field liquid and glass phases, but instead the flux-filled regions will have a lattice-like vortex distribution that is characterised by penetration history rather than phase. The vortex distribution during penetration is discussed in the next section. For this reason, the phase of vortex matter close to the Meissner state transition can only be investigated by field cooling.

### 2.5.2 Bean and Kim Models

To expand on the last paragraph in the previous section: the distribution of vortices during penetration, and in partially-penetrated samples, can be described by either the Bean or Kim critical-state models. While they differ in their specifics, each of these models describes the variation of both field and current across a superconducting sample, with a particular focus on penetration.

**Bean Critical-State Model**

The Bean model is based on the principle that current density in a superconducting sample cannot exceed a critical value $J_c$, which is fixed for a particular superconducting sample. It was devised by C. P. Bean in 1962, to explain the observed field gradient from the edge of a sample to the flux front after penetration [64].

In this model, $J_c$ is dependent on material properties of the sample, but has no
dependence on magnetic field. For comparison with other models, this is stated as:

\[ J_c(B) = J_{c0} \]  

(2.7)

The Bean critical-state model relies on the assumption that the maximum allowable current flows throughout the penetrated region. A one-dimensional “profile” of current across the film therefore shows a flat “plateau” at \( J_c \) in the penetrated region, which quickly drops to zero at the flux front. Since current flows parallel to the sample edge at a constant magnitude in this model, the magnetic field profile has an inward-sloping gradient from the edge, to the flux front where it reaches zero. Since the macroscopic field at any point in a superconductor is given by the density of vortices, this field slope means that the vortex distribution is much denser toward the sample edge and sparser approaching the flux front.

**Kim Critical-State Model**

Within a year, the Bean model was extended by Kim et al., who proposed that the critical current at each point in the film should vary according to the local field value at that point [65, 66].

By viewing the critical current as the current required to induce a field larger than \( H_{c2} \), it is sensible to suggest that the critical current would be lower for part of the sample in which the field is higher.

Critical current is not fixed but decreases with magnetic field, with a hyperbolic dependence given by:

\[ J_c(B) = J_{c0} \frac{1}{1 + B/B_0} \]  

(2.8)

Here \( J_{c0} \) is the critical current measured at zero field, while \( B_0 \) is a material-specific constant with dimensions of magnetic field.

This model retains the assumption that maximum current flows throughout the penetrated region, but the critical current varies throughout this region. Since field is largest at the sample edge, this is where the critical current is smallest. The largest critical current in the Kim critical-state model is seen close to the flux front. The current profile in this model has a plateau that slopes upward from the edge of the sample to a peak just before the flux front, after which it decreases quickly to zero.

**Discussion of Critical-State Models**

Other critical-state models have also been introduced following these pioneers. Most simply propose different field dependences of \( J_c \) on \( B \), such as a hyperbolic dependence [67]. This thesis will largely focus on the Bean and Kim descriptions, though
most calculations are model-independent.

Experimental\cite{41, 63} and numerical\cite{67} investigations have found that the hyperbolic and Kim models are both quite accurate in predicting the current distributions in superconducting samples.

Several mechanisms for the existence of a maximum current were suggested in the years following Bean’s prediction:

- A simple example that holds for a superconducting wire (even of type I) is that since current flowing through a wire induces a circumferential magnetic field, this induced field will eventually exceed the critical field and be large enough to destroy the superconductivity \cite{3}.

- A suggestion for the origin of critical current in bulk superconductors was that large currents induce de-pairing of the electrons in Cooper pairs, thus destroying the superconducting state \cite{68}. Again, this also holds for type I superconductors.

- In practice, the current required to remove vortices from their pinning sites (referred to as the “de-pinning current”, given by equation 2.5) is generally much lower than that required for de-pairing, so this provides a lower effective $J_c$. The shape of field and current profiles in the Bean and Kim critical-state models is attributed to the de-pinning critical current.

Currents larger than the de-pinning current do not destroy superconductivity, but merely induce resistance due to vortex motion. A further increase in current, past the de-pairing critical current, then causes the material to transition into the normal state.

### 2.5.3 Dynamic Magnetic Properties

The discussion up to this point has only considered the magnetic properties of superconductors in a state of pseudo-static equilibrium. These equilibrium states are well understood, but there remains much to be discovered by considering the behaviour of time-varying fields and currents within superconductors.

A very simple, but crucial, example of a dynamic effect is the penetration of magnetic flux into a sample. This is a complex process resulting from the motion of many individual vortices each behaving as described in section 2.3.4.

The first dynamic description of macroscopic flux behaviour during penetration was provided by Anderson, extending Kim’s critical-state model by proposing a model explaining the behaviour of vortices when current exceeds $J_c$ \cite{69}. This model, which is now referred to as the Kim-Anderson model, is as follows: when the driving
force on vortices is much larger than $J_c$, a large number of vortices become de-pinned and the vortex distribution enters the liquid phase (see section 2.3.2). These de-pinned vortices move together in the direction determined by the driving force, and this motion is referred to as “flux flow”. However, when the current is only slightly above $J_c$, as experienced when field is increased slowly, then only a few isolated vortices are removed from their pinning sites. Since these vortices are being weakly driven, they are quickly pinned in another pinning site. This results in vortices “hopping” from one pinning site to another at a rate $V$ that is determined by the driving force, and in the direction of this force. This motion is referred to as “flux creep”.

During penetration, both of these processes move flux inwards, and serve to reduce currents above $J_c$ back down to the critical value. In the case of flux creep, this reduction is exponential, and is therefore referred to as “relaxation” of current. The notions of flux flow and flux creep are still used to describe the fast initial motion of flux into a sample during penetration, which is followed by a much slower relaxation [70].

Anderson further proposed that even the critical state is not truly static, but is only defined when flux creep has slowed below an observable threshold [69]. Because of this, a value for $J_c$ cannot be uniquely determined experimentally for a given superconducting sample.

It was found that the penetration depth of flux into YBCO films after an external field increases from zero is dependent on the rate of increase of this field[71, 72]. This history-dependence shows that flux dynamics during penetration have a major effect on the final magnetic state of zero-field-cooled samples.

A large part of the experimental part of this thesis describes investigation into both magnetic flux dynamics and current dynamics in superconducting films. In particular, section 4.3.4 shows that transient current profiles under dynamic field conditions are radically different to the static current profiles and to those predicted by the Bean and Kim critical-state models.

There are also many other interesting magnetic effects that occur in superconductors at high speed, a few examples are:

- Dendritic flux avalanches, which are spectacular finger-like features occurring during penetration that appear to occur nearly instantaneously. Precision attempts to measure the speed of these events have yielded results varying from 5 km/s [73] to 180 km/s [74].

- Localised non-dendritic flux jumps, which have been shown to occur in less than 0.1 s [75].
• The dissipative response of superconducting wires to high-frequency AC driving currents [3].

2.6 Magnetic Microscopy Techniques

Many precise techniques have been developed to allow high-precision measurements of the local and global magnetic field around a sample. Since the magnetic field is generally observed on a small scale and is often displayed visually, these techniques are collectively referred to as “magnetic microscopy”. When applied to superconductors, magnetic microscopy is used to observe the position, density and dynamics of vortices, as well as the macroscopic behaviour of magnetic field and supercurrent.

Information about the local magnetic field in and around a sample is desired in many fields of materials science, and this can be measured through a variety of physical means. Hence, a diverse and ever-increasing range of magnetic microscopy techniques are used for this purpose.

Such techniques include but are not limited to: Bitter decoration (which was the first method to visualise the positions of vortices) [76–78], magnetic force microscopy [79]; magneto-optical imaging (MOI) [80, 81] and magneto-optical video (MOV) [41]; scanning SQUID microscopy (SSM) [36, 82] and the recently developed SQUID-on-tip microscopy [83, 84], μHall-probe microscopy [81, 85, 86], scanning magneto-resistive microscopy [87], scanning electron microscopy with polarization analysis (SEMPA) [88] and electron holography [89].

Many related magnetic imaging techniques are also used in biology and medicine, where they are valued for their non-contact and non-destructive nature. These applications include: optical magnetic imaging of living cells [90], magnetoencephalography (MEG) of brain activity [91–93], magnetocardiography (MCG) for magnetic heart signals [91, 94], and Magnetic Resonance Imaging (MRI) for soft tissue and the brain [95].

Magnetic microscopy techniques have a limitation arising from the distance between the scan height and the sample. This leads to the stray field being observed rather than the actual field at the sample surface. This will be explained in section 2.7.2.

In addition, most magnetic microscopy techniques are based on the principle of scanning probe microscopy (SPM)[96]. SPM techniques are very widely used for the investigation of micro- and nano-scale phenomena[97], but their point-by-point scanning mechanism leads to a fundamental limitation on the speed of measurement[36]. In order to investigate the dynamic magnetic properties of superconductors (such as those discussed in section 2.5.3), high-speed magnetic measurement techniques are required. High-speed magneto-optical video (MOV) was therefore developed, al-
lowing time-dependent magnetic fields to be observed and recorded at up to 30,000 frames per second in superconducting samples [98].

This thesis focusses on experimental work undertaken using two such techniques: Magneto-Optical Imaging/Video and Scanning SQUID Microscopy. These are the techniques that allow the highest temporal resolution and the highest magnetic field sensitivity respectively.

### 2.6.1 Magneto-Optical Imaging

Of all the magnetic microscopy techniques described above, magneto-optical imaging gives the most intuitive insight; it visualises the magnetic field around a superconductor, allowing field to be seen in real time through an optical polarising microscope. Such images allow a fast qualitative determination of a sample’s magnetic properties, as well as the acquisition of accurate quantised magnetic field data. Hence, magneto-optical imaging has become a widely used technique for the analysis of thin film superconductors [99].

Applications of MOI for thin film superconductors include: simple non-contact analysis of the quality of films [63, 99–101] and tapes [99, 102–105]; visual identification of cracks [102, 105], weak inter-granular links [100, 103, 104] and other sample defects[63, 106], step-wise visualisation of magnetic penetration, depenetration and hysteresis [35, 80]; visualisation of current flow in samples of novel geometry and composition [63, 107–109]; and observation of individual vortices [110].

The magneto-optical technique is based on the transmission of linearly polarised light through a layer of Faraday-active material, which is part of a magneto-optical indicator film. This film makes use of the Faraday Effect to rotate the polarisation of the light through an angle \( \alpha_F \) that is proportional to the local magnetic field at any point within the film. With a suitably oriented analyser, local variations in magnetic field at the film (which is the stray field above the sample) are therefore made visible as variations in light intensity.

In this research, bismuth-doped yttrium iron garnet (Bi:YIG) is used as the Faraday-active layer\(^h\). The film is produced with an optical axis perpendicular to the plane of the film - this is taken as the \( z \)-direction. When polarised light is incident normal to this film, its rotation is therefore proportional to the \( z \)-component of field at the point of incidence.

The rotation angle \( \alpha_F \) (also known as the Faraday angle) is ideally given by:

\[
\alpha_F = \mathcal{V} B_z d_{\text{ind}}
\]  

(2.9)

Here \( d_{\text{ind}} \) is the thickness of the Faraday-active layer and \( \mathcal{V} \) is a constant of pro-

\(^h\)The complete structure of the indicator films used is given in section 4.1.5
portionality called the Verdet constant, which depends on temperature, wavelength, and the properties of the particular indicator film used. In practice, the rotation angle is doubled, since light passes twice through the film.

Since magneto-optical imaging is carried out over a range of temperatures from room temperature to less than one Kelvin, the indicator material chosen must have a near-flat temperature response of the Verdet constant $V(T)$.

Equation 2.9 holds for fields with negligibly small $xy$-components. A more complete version of this equation is derived in section 7.1.

After rotation the light passes through an analyser which is oriented at a fixed angle $\theta$ with respect to the polariser. Invoking Malus’ law then gives the intensity at the analyser as:

$$I = I_0 + I_{\text{max}} \cos^2(2\alpha_F + \theta)$$ (2.10)

Where $I_0$ is the background light intensity (which is usually subtracted from all measurements), and $I_{\text{max}}$ is the maximum light intensity (equivalent to the intensity of light through the polariser).

Light intensity at each point in the microscope’s field of view is recorded using a camera. By application of equation 2.10, the magnetic field at each point on the indicator film is therefore determined directly from the measured light intensity.

### 2.6.2 High-Speed Magneto-Optical Video

Magneto-optical imaging is described as a real-time technique [98, 111], meaning that information on magnetic field can be acquired almost instantaneously. This is because indicator films respond to changes in magnetic field at an extremely fast rate, and this technique does not rely on a scanning mechanism. Therefore, the magneto-optical video technique arose through a simple replacing of the still camera with a high-speed video camera. This allows high-precision dynamic measurements of the changes in local magnetic field over time, with the only practical speed limitation being the frame rate of the camera used.

Prior to the development of MOV, several investigations into dynamic effects in superconductors were carried out using static MOI. This could be achieved by taking multiple images of samples under AC fields [112, 113], or by observing the differences in penetration when the external field ramp rate was varied [71].

However, these static imaging experiments could never fully illuminate the dynamic nature of vortices and fields in superconductors, especially the unique transient effects that occur during a change in external field. Since dynamic field and current effects had been seen to occur on extremely short time-scales [9, 71, 109, 114], these effects were often considered too difficult to observe in detail.

This is true for most magnetic microscopy techniques, with many taking minutes
or hours to create a single magnetic field image [36], but not for the magneto-optical technique. Typical indicator film materials have been shown to have un-dampened sensitivity to alternating magnetic fields up to at least $10^6$ Hz, although the upper bound on frequency may be as high as $10^9$ Hz [115]. This response speed is much faster than the frame rate of any high-speed camera currently in existence, and therefore the indicator film provides no practical limit on the speed of measurements [41].

Several issues do arise, however, in the implementation of high-speed MOV, many of which revolve around the fact that high-speed magnetic field systems are desired in order to investigate dynamic effects in the fastest regime possible. One major issue is that when fields are applied rapidly, eddy currents must be accounted for in any metallic parts of the cryostat, and even in the aluminium reflective layer of the indicator film [116, 117].

The processing of high-speed video requires significant computation power, due to the large number of data points in each measurement. The files produced have a large size, and calculations on these files often take a significant amount of time. This will be further discussed in section 2.7.1.

There are a rich variety of dynamic and transient magnetic effects that have yet to be experimentally investigated in superconducting samples. The dynamic magnetic properties of superconductors are introduced in section 2.5.3, and an experimental investigation of some of these effects using MOV is discussed in section 4.3.4. Other groups have applied high-speed MOV to investigate the behaviour of field during penetration [41, 72, 98], the dampening of high frequency transport currents [114], current relaxation following an overcritical current pulse in a superconducting strip [109], and the magnetic field dynamics during a thermal quench [118].

The high-speed MOV technique is also very promising for future applications, including observation of individual and collective vortex motion in response to external driving forces, further study into magnetic penetration behaviour, and for analysis of vortex ratchets (as discussed in section 6.5.1).

Some modifications have been proposed to improve the sensitivity of the MOV technique, including laser scanning [119] and high-speed polarimetry [120]. However, both of these reduce the image acquisition speed, and were not used in the current research. Conversely, laser-pulsed MOI is another variation that has produced pairs of images only nanoseconds apart [74], recording very fast dynamics, but this technique does not allow for continuous video capture and hence cannot fully investigate changes in the magnetic state over time.
2.6.3 Scanning SQUID Microscopy

Scanning SQUID Microscopy (SSM) provides mapping of magnetic fields with the highest precision currently attainable. This is achieved using a SQUID (Superconducting Quantum Interference Device) magnetometer, which has extremely high sensitivity to small changes in magnetic field [82].

The scanning SQUID technique is usually used to study a range of magnetic materials [82, 91], as well as geological [121, 122] and biological samples [122]. Applications for superconducting samples include: observation of vortex lattices and distributions [36, 37], vortex matter in micro-scale superconducting devices [82], and novel superconducting phenomena such as half-flux quanta [123–126] (see section 7.2).

The scanning SQUID technique works on the simple principle of raster scanning a SQUID magnetometer at a small distance from the sample surface, as shown in figure 2.10(a). This is similar to the operation of many other scanning probe microscopy techniques, and is described in detail in the following subsection.

SQUID Principles of Operation

A Superconducting Quantum Interference Device magnetometer can be created using a simple superconducting loop that is split into two halves connected by Josephson junctions. The type of device described here is a DC SQUID [82, 127].

Since flux through a superconducting loop is quantised in integer multiples of $\Phi_0$, application of an external field much smaller than $\Phi_0$ will not result in any field through the loop, but in a measurable screening current. As the external field is increased, this screening current will increase. Once the current exceeds the critical current of the Josephson junctions, a measurable voltage will be induced, which increases with increasing field.

SQUIDs are generally designed to minimise the critical current of the Josephson junctions to ensure that any change in screening current results in a change in voltage. This ensures that even very small changes in field can be detected by the magnetometer.

SQUID measurements are complicated by flux jumps, which occur when the external field grows large enough that the flux in the loop will increase by an integral multiple of $\Phi_0$. The first flux jump generally occurs when the external flux is larger than $\Phi_0/2$ but still smaller than $\Phi_0$. Therefore, the screening currents in the loop flow in the opposite direction, since the flux inside the loop is larger than that outside. The voltage also changes direction, but the relative change in voltage with changing field remains identical to what it was before the flux jump.

Therefore, SQUIDs can be used to measure extremely small changes in magnetic
field, and can operate over a large field range, but they cannot determine the absolute magnitude of magnetic field.

The circuit geometry used for scanning SQUID microscopy is shown in figure 2.10(b). The SQUID loop is extended with long wires that have a so-called “pickup loop” at the end. Field through the pickup loop will result in a screening current that flows along these wires to the main SQUID loop. The intention is that only the field at the position of the pickup loop is measured.

In this circuit, current is applied to the SQUID, and small changes in voltage
are measured in response to the changing field. A modulation field can be applied to the SQUID to prevent unwanted flux jumps.

### 2.6.4 Comparative Study of MOI and SSM

The two magnetic microscopy techniques employed in this work, Magneto-Optical Imaging and Scanning SQUID Microscopy, are similar in their purpose: both are techniques for visualising the magnetic field at a small height above a thin film sample. In practice, however, the differences between these techniques far outweigh their similarities, so much so that it is very difficult to obtain similar images from a given sample using these two different techniques. The most significant difference is in the scale of features that can be observed with each technique. Also, the different mechanisms involved in generating images lead to fundamentally different imaging artefacts and complications.

Magneto-Optical Imaging and Scanning SQUID Microscopy operate over quite different physical scales, with the scale of SSM measurements generally being much smaller. That is to say: the resolution of SSM is much higher, though it is generally limited to a smaller field of view than MOI. Scanning SQUID techniques can have resolution of a few hundred nanometres, and this is determined by how small the pickup loop can be made [84]. MOI has a resolution generally of several microns, though the resolution is not so easily defined, as it is determined by the extent of influence of a point magnetic field on the surrounding area of the indicator film.

Therefore there is an order of magnitude difference in the physical size of features seen by each technique. This difference is immensely significant in the study of superconductors due to both the size of vortices and the spacing between vortices being on a similar scale to the limits of resolution of MOI. The ability to resolve individual vortices was in fact a major milestone for MOI [110], while the resolution of SSM is so much higher that individual vortices were not only resolved, but their circular shape could be seen in much earlier published images obtained using SSM [128].

In addition to the difference in physical scale, magneto-optical imaging and scanning SQUID microscopy also operate within different field regimes. A SQUID is the most sensitive field detector in existence, with most commercial SQUIDs able to detect fields down to the nanoTesla regime. The most sensitive SQUIDs even have sensitivity down to a few picoTesla. The higher field sensitivity of SSM means that in an environment that is not perfectly shielded (such as the apparatus used here), the Earth’s magnetic field is a very significant factor, while it has proven to be quite negligible for MOI.

MOI images are direct photographs of the indicator film above a sample, which
visualises the magnetic field. However, the field measurement is indirect, with light intensity being the directly measured quantity. On the other hand, SSM images are computer-generated false-colour representations of the magnetic field measured at each point on the sample surface. The fact that the visual images are computer-generated makes SSM less like traditional microscopy, but these images generally give a more precise representation of the field at the sample surface.

Being an indirect measure of magnetic field, MO images are much more difficult to accurately quantify. In order to quantitatively determine the magnetic field at a point from an MO image, one must either know the incident light intensity, angle between the polarisers and several other parameters related to the indicator film, or use a calibration with known field values. A scanning SQUID, however, outputs a voltage which is directly proportional to the magnetic field through the pickup loop at each point. Quantitative field values are given by a simple multiplicative factor. One issue arising from SSM, however, is that the device is not insensitive to magnetic field through parts other than the pickup loop, and this error cannot be easily accounted for.

Artefacts arise in each of these methods. For SSM, artefacts are very common and generally occur due to the high sensitivity to magnetic noise and sample vibrations. Line averaging errors produce line-to-line defects (which are similar to those that occur in many other scanning probe microscopy techniques). Shading errors may occur due to drift of the SQUID voltage over time, leading to an apparent field gradient from parts of the image that were scanned earlier to those that were scanned later. For MOI, artefacts are more easily avoided for very precise measurements. This technique is mainly sensitive to optical features in the field of view of the microscope such as dirt and scratches on the indicator film, background light, and inhomogeneous illumination of the sample.

Magneto-optical imaging also displays an unwanted sensitivity to fields in the direction perpendicular to the optical axis of the film, as discussed in section 7.1, while SQUIDs are highly directional-specific. However, the SSM apparatus used for this research has a SQUID loop oriented at 30 degrees to the plane of the sample, so it is in fact sensitive to one of the in-plane field components.

Finally, the biggest advantage of the MOI technique for the purposes of this research is that it is very fast. As described above, high-speed video of magnetic flux can be obtained, with individual images being captured in less than one millisecond. By comparison, SSM is extraordinarily slow, taking several hours to obtain a single scan. Flux jumps in the SQUID loop can also occur, which may further slow down the scan as it re-takes each line of the scan for which a flux jump is detected. Particularly large flux jumps may also prematurely end a measurement run by invalidating the data for all points scanned after the flux jump.
2.7 Calculations and Analysis Techniques

While it is interesting to look at visual pictures of the magnetic field in superconductors, this is not the final goal of this research. The data obtained from magnetic microscopy of superconducting samples can be analysed to provide much more applicable information about the sample, such as the flow of current through the sample in the static and dynamic cases; the speed of motion of magnetic features; and analyses of the distribution of vortices.

2.7.1 Current Calculation (The Inverse Problem)

The “inverse problem” of magnetic microscopy refers to the calculation of two-dimensional currents \( (J_x \text{ and } J_y) \) in a thin film sample from measurements of magnetic field in the perpendicular direction \( (B_z) \), where the magnetic field is measured in a flat plane at a fixed distance above the plane of the currents.

In other words, the aim of this problem is to calculate current in a superconducting thin film from the magnetic fields observed with MOI or SSM. It is called the inverse problem since this calculation is achieved by an inversion of the Biot-Savart law (along with a few simple assumptions).

This calculation can be carried out independently for each magnetic field image acquired from magnetic microscopy to produce an accurate model-independent mapping of the distribution of current in the sample.

The procedure begins with the Biot-Savart law, which calculates the magnetic field \( B \) at a position \( r' \) that arises due to current \( J \) distributed over a range of other positions \( r \).

\[
B(r') = \frac{\mu_0}{4\pi} \int \frac{J(r) \times (r - r')}{|r - r'|^3} d^3r
\]  

(2.11)

This equation cannot generally be inverted, i.e. it is not possible to give unique values for each current component at every point \( r \) from an arbitrary field distribution.

However, this equation can be inverted for the specific geometry used for magnetic microscopy, with only a few modest assumptions. The geometric constraints are as follows: Firstly, we consider currents existing only in a film lying in the \( xy \)-plane, within a small thickness \( d \). This limits the range of \( r \). In addition, field is applied in the \( z \)-direction only (perpendicular to the film), and therefore only the \( z \)-component of field is measured. This measurement is performed over a flat plane that is parallel to the plane of the film at some distance \( h \). Therefore, the range of \( r' \) is over this plane, and the field is generally considered to be purely \( z \)-directed\(^1\).

\(^1\)Actually, a small \( xy \)-component will be induced by the currents, this is discussed in section 7.1
Mathematically, the geometrical constraints on the field can be expressed as:

$$\mathbf{B}(x', y', z') = B_z(x', y', h)\mathbf{\hat{z}} \quad (2.12)$$

Since $d$ is small and field is in the $z$-direction, the current can be assumed to have negligible $z$ component. It is also reasonable to assume minimal variation of the $x$ and $y$ current components in the $z$-direction due to the small thickness $d$. Therefore, the current vector at $\mathbf{r}$ takes the simple form: [80, 107]

$$\mathbf{J}(\mathbf{r}) = J_x(x, y)\mathbf{\hat{x}} + J_y(x, y)\mathbf{\hat{y}} \quad (2.13)$$

This is essentially a two-dimensional approximation, although the finite thickness $d$ of the film is still taken into account in the inversion process.

It is also reasonable to assume that the film is locally uncharged, since magneto-optical images are generally taken in the equilibrium state, and no static charge separation should occur. Any charge density $\rho$ built up in one area of the film would very quickly dissipate in the superconducting material. So $\rho \simeq 0$ and $\frac{\partial \rho}{\partial t} \simeq 0$.

With this assumption, the general charge continuity equation:

$$\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} = 0 \quad (2.14)$$

becomes an equation of current continuity:

$$\nabla \cdot \mathbf{J} = 0 \quad (2.15)$$

This implies that currents flow only in closed loops in the film. A two-dimensional Fourier transformation of equation 2.15 gives: [107]

$$-ik_x\tilde{J}_x(k_x, k_y) - ik_y\tilde{J}_y(k_x, k_y) = 0 \quad (2.16)$$

$$: \tilde{J}_x(k_x, k_y)k_x + \tilde{J}_y(k_x, k_y)k_y = 0 \quad (2.17)$$

The tilde represents a Fourier-transformed quantity. $k_x$ and $k_y$ are the frequency (Fourier) components in the $x$ and $y$ directions respectively.

By applying two-dimensional Fourier transformation to equation 2.11 and considering 2.17 along with the geometric constraints in 2.12, an invertible form of the Biot-Savart equation is acquired: [63, 80, 107, 129–131]

$$\tilde{B}_z(k_x, k_y, h) = i\mu_0 e^{-hk}k_y \sinh \left( \frac{d}{2}k \right) \tilde{J}_x(k_x, k_y) \quad (2.18)$$

Here $k = \sqrt{k_x^2 + k_y^2}$. Note that primes have been suppressed for the fre-
frequency components of \( B_z \), since each component is determined directly from the corresponding frequency component of \( \tilde{J}_x \). The inverse Biot-Savart equations are therefore given by:

\[
\tilde{J}_x(k_x, k_y) = \frac{ik_y}{\mu_0} e^{kh} \text{cosech}\left(\frac{kd}{2}\right) \tilde{B}_z(k_x, k_y, h, d) \quad (2.19)
\]

\[
\tilde{J}_y(k_x, k_y) = \frac{-ik_x}{\mu_0} e^{kh} \text{cosech}\left(\frac{kd}{2}\right) \tilde{B}_z(k_x, k_y, h, d) \quad (2.20)
\]

Where (2.20) is derived from equations 2.17 and 2.19. An inverse Fourier transformation can then be used to give the current components \( J_x(x, y) \) and \( J_y(x, y) \) at every point in the film. These components can in turn provide the magnitude of current at each point in the film using the simple relation:

\[
J(x, y) = |J(x, y)| = \sqrt{J_x(x, y)^2 + J_y(x, y)^2} \quad (2.21)
\]

This procedure has been shown to determine the magnitude and direction of current in each pixel-sized element of the film with a high degree of accuracy.

A more rigorous mathematical solution to the inverse problem is given in appendix A. The solution is based on other solutions previously given in the literature but also includes several novel steps in its derivation.

**High-frequency Filtering**

Currents calculated using the inverse Biot-Savart equations (2.19 and 2.20) were particularly sensitive to high-frequency noise. Any noise occurring at high spatial frequency \( k \) was heavily amplified due to the dependence of \( \tilde{J}_x \) on \( e^{hk} \). High-frequency Fourier components were therefore removed in order to avoid spurious results.

High-frequency shot noise is often present in magneto-optical images due to the random nature of photon capture in the camera. The noise level is greater when light intensity is low. High-frequency noise may also be present in scanning-SQUID images due to vibrations.

Since the magnitude of the calculated current is proportional to \( e^{hk} \) as mentioned above, any small error in \( B_z \) for large \( k \) can lead to large errors in \( \tilde{J}_x \) and \( \tilde{J}_y \). This effect is worsened with increasing measurement height \( h \).

Several low-pass filtering functions were tested in attempts to remove the high-frequency components of \( B_z \) before applying the inversion. A Hanning window function was found to give the most accurate results for current, while also preserving
CHAPTER 2. THEORY

the accuracy of field data. This window function was defined as follows[108]:

\[ W(k) = \begin{cases} \frac{1}{2} \left( 1 + \cos \left( \frac{\pi k}{k_{\text{max}}} \right) \right), & k < k_{\text{max}} \\ 0, & k > k_{\text{max}} \end{cases} \] (2.22)

Where the cut-off wavevector \( k_{\text{max}} \) is a parameter that must be optimised to reduce noise while preserving spatial resolution. The larger the value of \( k_{\text{max}} \) used, the more spatial resolution is retained, but this also retains more high frequency noise. Despite the importance of spatial resolution, it is therefore limited by the optimal value of \( k_{\text{max}} \) that can be taken, and so ultimately is determined by the noise in the image.

Creating Images to Represent the Current

For each magneto-optical image, the raw output of the current calculation procedure is three two-dimensional matrices containing the elements of \( J_x \), \( J_y \) and \( J \) at every point in the film.

These matrices are represented as images for simplicity of qualitative analysis of the current distribution.

Since the current is calculated in pixel-sized elements, it is simple to convert each current matrix into an image with the same scale and resolution as the original magnetic field image.

The current value in each element is converted into a grey scale value (GSV) to be represented as a 256 grey level image. A GSV of 255 produces a white pixel, and a value of 0 gives a black pixel. Therefore the current magnitude \( J \) is represented as an image using:

\[ \text{GSV} = \frac{(J - J_{\text{min}}) \times 255}{(J_{\text{max}} - J_{\text{min}})} \] (2.23)

\( J_{\text{min}} \) is the minimum current calculated from a magneto-optical image, which is represented as black, and \( J_{\text{max}} \) is the maximum current, which is represented as white. Similar images are created for \( J_x \) and \( J_y \).

The direction of current in the film can also be represented as an image, but this is done in an indirect manner using the magnetisation: The currents that flow in a superconductor are magnetisation currents, since neither polarisation currents nor resistive currents are expected in YBCO films [133]. Current is therefore locally related to the magnetisation \( \mathbf{M}(x,y) \) at each point in the sample by:

\[ \mathbf{J} = \nabla \times \mathbf{M} \] (2.24)

Since the assumption has been made that currents flowing in the thin film sample
have only $x$ and $y$ components, the magnetisation vector must lie in the $z$ direction. Therefore, the component form of equation 2.24 is: [132]

\[ J_x = \frac{\partial M_z}{\partial y} \]  
\[ J_y = \frac{\partial M_z}{\partial x} \]  

In this two-dimensional case, a contour plot of the magnetisation $M_z$ is equivalent to a map of current “streamlines”. Each streamline shows the two-dimensional path that current takes around the superconducting sample, with the density of lines giving an indication of the magnitude of current in each part of the sample [80].

The Fourier transformation of equations 2.25 and 2.26 is:

\[ \tilde{J}_x = -ik_y \tilde{M}_z \]  
\[ \tilde{J}_y = -ik_x \tilde{M}_z \]  

Which means that $M_z$ can be simply determined in Fourier space from the calculated Fourier components of $J$.

### Dynamic Current Calculation

For the purpose of this study, the current calculation procedure was extended into the dynamic regime. A major part of this research was the development and implementation of a method for determining the dynamic current distribution in a superconducting thin film over time.

Dynamic current calculation was achieved through an independent and sequential application of the Biot-Savart inversion procedure to each frame of a magneto-optical video. Since each frame of a magneto-optical video gives an accurate indication of the instantaneous field distribution at a particular time, the dynamic current calculation simply treats each frame as an independent magneto-optical image. This method is explained further in section 4.2.3.

#### 2.7.2 Stray Field and Stray Current Error

One inaccuracy in the currents calculated by this procedure arises from the spreading of “stray” fields, which occurs in the space between the surface of the sample and the measurement position. While the calculation procedure does take into account the finite measurement height, it is much more difficult to factor into these calculations the amount by which the field lines have spread over this short distance.
Therefore, current features on a very small scale such as those seen in figure 5.4 suffer from what will be referred to as “stray current error”, which tend to make small features appear larger in the calculated distribution of current. This can lead to an apparent overlap of magnetic features and currents that is not physical [134].

![Image of YBCO sample with vortices](image1)

**Figure 2.11:** The spread of “stray” magnetic fields above the sample surface, showing the enlargement of magnetic features at the height of the measurement plane. (a) shows vortices with a large spacing, the field size appears larger and current appears to travel in a larger loop. (b) shows closely spaced vortices, whose fields interact significantly at the measurement height, causing more significant errors in current.

**In-Plane Field Effects**

One inaccuracy that may arise in the magneto-optical imaging technique is due to the limited directionality of magnetic field detection. Magneto-optical indicator films have a non-negligible sensitivity to field components perpendicular to their optical axis. This means that a magneto-optical image does not correspond solely to the component of magnetic field in the z-direction, but any components in the xy-plane also have an influence on the image produced.

One may reasonably ask: *how do these fields in the xy-plane arise?* Since field is applied in the z-direction (perpendicular to the plane of the superconducting films) in the magneto-optical imaging procedure, and the field in each vortex should also point in the direction of the external field, it may be natural to think that no field should exist in the xy-plane.
The answer comes from the subtle realisation that the plane in which magnetic field is measured is some distance above the plane of the film. It is true that in the plane of the superconducting sample, field only exists in the z-direction. However, in the small distance between the sample and the indicator film, a bending of the field lines occurs.

The field lines bend for two physical reasons: Firstly, field outside the sample edges experiences an inward bending due to the demagnetisation effect (see section 2.3.1). Secondly, the field passing through the sample in the form of vortices experiences a spreading as flux exits the sample surface. Above the sample, the flux from each vortex is no longer confined to a small cylinder of width $2\lambda$, but may spread to form a continuous distribution some distance from the sample. The partially spread-out distribution of magnetic field at some small distance from the ends of the vortices is referred to as the “stray field”. This is shown schematically in figure 2.11, which also shows the effects of field spreading on the calculation of current. The origin of stray field is described in more detail in section 7.1.1.

It is not the field at the sample surface, but this stray field that is actually measured with magnetic microscopy. In both magneto-optical imaging and scanning SQUID microscopy, this leads to a broadening of small magnetic features, which is a significant source of error for analysis of superconducting films. However, further inaccuracies arise in MOI due to the stray field generally having some component in the xy-plane. An experimental measurement of the spread of stray fields is shown in figure 5.6.

A procedure was developed by Laviano et al. to correct for the errors caused by in-plane fields on the calculation of currents in superconducting films [135].
Chapter 3

Techniques & Sample Preparation

The high-quality YBa$_2$Cu$_3$O$_{7-\delta}$ thin films used in this research were created using pulsed laser deposition, the process of which is described in section 3.1. Samples were generally deposited onto 5 $\times$ 5 mm substrates of SrTiO$_3$. Most YBCO films used had a nominal thickness of $d = 200$ nm, and critical temperature $T_c \geq 90$ K.

The YBCO films were then patterned using the laser lithography and ion beam etching procedure described in section 3.2. Such patterning was necessary to avoid defects at the substrate edges, and was also useful to shape films into different geometries for specific purposes.

The patterned films were tested to ensure that they were of high quality, as discussed in section 3.4. This testing was usually by MPMS magnetometry, but sometimes also involved profilometry, atomic force microscopy and magneto-optical imaging. The general properties of the resulting YBCO films are given in section 3.5.

3.1 Pulsed Laser Deposition

The YBCO samples used in this research were produced by Pulsed Laser Deposition (PLD) [15, 136]. The PLD technique employs a powerful laser to ablate solid targets of YBCO in a low-pressure oxygen environment. This results in a plume of particles being expelled at high energy from the target. The majority of expelled particles will hit a suitably-placed substrate, where they form a thin YBCO film that coats the top surface of the substrate.

The samples used in this research were deposited using a purpose-built PLD chamber at the Institute for Superconducting and Electronic Materials [15, 136]. Deposition was generally carried out by Dr. Sergey A. Fedoseev, with some samples also deposited by Dr. Igor A. Golovchanskiy.

The PLD process begins with the substrate, which is chosen to have the same crystal structure as deposited compound, and with similar lattice parameters. This is to ensure that the first particles deposited align with the lattice positions on the
surface of the substrate, so that the lattice direction of the substrate matches that of the sample. This is known as epitaxial growth, and the epitaxy of a film refers to the quality of crystalline matching with the substrate. The majority of samples used for this research were deposited onto substrates of strontium titanium oxide (SrTiO$_3$), which is commonly referred to as STO. The STO substrate was usually produced with its top surface parallel to the \textit{ab}-plane, to ensure that the YBCO film would be produced with its \textit{ab}-plane in the plane of the film.

The substrates are placed within the PLD chamber, which is then evacuated of air and filled with oxygen to a lower pressure. The initial pre-evacuation was generally to a pressure lower than $10^{-4}$ Pa, then the oxygen pressure was generally 40 Pa [15, 136]. For optimal results, the temperature is then increased to 780° C. This temperature had been found to produce films with the highest critical current [136].

Once optimal conditions were reached, the ablation of YBCO targets could begin: A krypton fluoride excimer laser with wavelength 248 nm was directed toward each target with an effective fluency of $\sim$2 J/cm$^2$, and pulse frequency of generally 5 Hz [15, 137].

During deposition, the film slowly grows as particles are deposited. The locations in which particles tend to attach during deposition translates to a preferential growth mechanism, which depends on the sample material and crystal structure; laser energy and pulse frequency; substrate material and epitaxy; and the appearance of defects in the film (which may occur during the deposition process). Different growth mechanisms occur under different conditions, which can vastly affect the properties of the resulting film [15, 136, 137].

The growth mechanism generally seen in the YBCO films deposited for this research was island growth, which occurs when particles preferentially adhere to regions of already-deposited YBCO than to the substrate. Therefore, in the early stages of deposition, isolated regions (islands) of crystalline YBCO tend to form, and these are scattered across the substrate surface. These islands then grow vertically (along the \textit{c}-axis) and laterally (across the \textit{ab}-plane) as the deposition continues. Lateral growth is isotropic, giving the islands a circular shape. As this growth continues, the edges of neighbouring islands meet, though they do not merge due to mis-orientation of the crystalline direction in each island. Instead, domain boundaries are formed along these island edges. Vertical growth continues after the substrate surface is covered, and the islands tend to form into pillar-shaped domains of YBCO, oriented along the \textit{c}-axis. The size and structure of islands could be tuned by varying the laser pulse frequency [15].

The final YBCO film produced covers the surface of the substrate with quite even thickness and adheres very strongly to the substrate due to epitaxy. PLD-
deposited films are fairly smooth, though surface roughness does arise depending on growth conditions [15].

All samples produced were subjected to preliminary tests, and only those with suitably large $T_c$ and $J_c$ were selected for further use - these factors were found to be a good indication of the quality of a deposition. The testing process is discussed in section 3.4.1.

### 3.2 Patterning of YBCO films

After the samples were produced, a photolithographic patterning and ion beam etching procedure was applied to remove unwanted areas of the YBCO film, shaping the sample to the desired geometry.

This process was necessary to improve the sharpness of a film’s edges, since the penetration of magnetic flux into superconducting films is highly sensitive to the geometry of the sample edges.

Since the substrates are cut from a larger STO ingot, some edge roughness is unavoidable, leading to crystalline defects during epitaxial growth. By contrast, a very sharp edge can be produced using laser lithography and ion-beam etching.

The same patterning process is also used to create samples of different geometry. Most samples tested in this research were squares or circles, although others with much more complex geometries were produced including superconducting bridges [138], samples with arrays of antidots[139], and the novel geometry used for creation of vortex diodes, as discussed in chapter 6.

The entire patterning process was carried out in a clean room environment in order to ensure that samples were not contaminated by dust, which might affect the pattern produced.

The steps of the patterning process are described in each of the following sub-sections:

#### 3.2.1 Spin Coating

The first step in the patterning process was the application of a very thin and smooth layer of photoresist to the surface of the superconducting sample. This was achieved using a Laurell Technologies WS-400B-6NPP/LITE spin coater.

The principle of spin coating is very simple: a small droplet of a photoresist liquid is applied to the flat surface of a sample, which is then rotated at high speed. A thin layer of the photoresist adheres to the sample, while the excess is quickly removed by the centrifugal inertial force.

This photoresist (or simply ‘resist’) is a light-sensitive substance which forms a
protective coating over selected parts of the sample. AZ1518 photoresist was used for this research. It is a positive-tone resist, meaning that regions of the resist exposed to ultraviolet light become weakened and soluble in an appropriate developer solution (in this case AZ726 MIF), while regions not exposed remain insoluble.

Since photoresist is designed to be highly sensitive to light, caution must be taken at all times to avoid any unnecessary exposure. AZ1518 resist is most sensitive to ultraviolet light, so yellow-filtered lighting was used in all laboratories in which photoresist was used. The resist was always stored in a dark place when not in use.

**Spin Coating Method**

The spin coating procedure is as follows:

The sample must be cleaned before photoresist is applied. This begins with removal of the silver paste from the rear surface of the substrate using a scalpel. The paste is necessary to fix the substrate in place during PLD, but must be removed at this point to ensure that the sample is level for the laser lithography procedure. The sample surface is then cleaned with acetone in an ultrasonic bath, then rinsed with acetone and ethanol and dried with N\textsubscript{2} gas. It is vitally important that the sample surface is dirt-free before the photoresist is applied - the effect of a dirt particle is shown in figure 3.1.

The clean sample is placed onto the suction ring of the spin coater, and vacuum is applied from below to hold it in place. A small droplet of photoresist is applied to the centre of the sample using a clean disposable pipette. For optimal coating, this droplet should be spread into the corners of the sample using the pipette tip. Care must be taken not to touch the sample surface with the pipette tip, as this may cause scratches.

The lid is then closed, and the spin coater is initiated. Rotation occurs with a pre-programmed speed and duration, using the optimised parameters described in section 3.3. As the sample rotates, photoresist spreads into an even layer across the sample surface by the centrifugal effect.

When the rotation process is complete, the sample is removed from the spin coater and placed onto a hot plate with a temperature of 105°C for 1 minute. This step is referred to as pre-baking since it is carried out before the lithography process. Pre-baking hardens the resist uniformly to form a solid layer across the whole sample. The unused resist is then stored below 5°C to avoid degradation.

After the spin-coating and pre-baking process is complete, samples are immediately placed into a particle-free light-proof box for transport to the laser lithography instrument.
CHAPTER 3. TECHNIQUES & SAMPLE PREPARATION

3.2.2 Laser Lithography

Laser lithography was carried out on the photoresist-coated samples in order to “draw” the desired pattern onto the sample. The Heidelberg Instruments μPG 101 laser lithography instrument applies a focussed 405 nm ultraviolet laser with 1 μm spot size to the surface of the photoresist on the sample. This instrument applies the laser in a pre-determined pattern by raster scanning a stage containing the sample, and selectively opening and closing a shutter in the path of the beam for each $xy$-position on the sample surface.

Laser Lithography Method and Apparatus

The laser lithography process was carried out using the method described below. Descriptions of each component of the lithography apparatus are also given.

The sample is first placed onto a small hole at the centre of the mobile stage and held in place by negative pressure from a small vacuum pump. The stage is driven in the $x$- and $y$-directions by stepper motors over a large range in the horizontal plane. The sample orientation is carefully aligned to a raised edge of the stage, to

Figure 3.1: 10x magnified view of a lithographically patterned sample which had not yet been etched. A $1 \times 1$ mm ratchet patterned area is shown in the centre of the image. A large dirt particle is visible as a dark circular spot within a second patterned area at the top right, having a major effect on the pattern. The lighter halo around this spot is a thicker region of resist surrounding the particle, indicating that this particle lies beneath the photoresist.
ensure that these \( x \)- and \( y \)-directions are parallel to the sample edges.

![Key components of the \( \mu \)PG 101 laser lithography apparatus.](image)

Figure 3.2: Key components of the \( \mu \)PG 101 laser lithography apparatus.

The laser is mounted on a tower which can be adjusted in the \( z \)-direction using a stepper motor for coarse control, and a piezoelectric unit for fine control. A camera is also mounted on the tower, with its focal point matched to that of the laser.

Using the camera, the vertical position of the tower is adjusted to achieve the sharpest focus. The \( x, y \) co-ordinates of each corner of the sample are then located using the stepper motors and the camera view. If the edges are found not to lie along the \( x \)- and \( y \)-directions within a small tolerance, then the sample must be re-aligned. The focus is checked at each of these corners using the piezoelectric fine control. If a significant variation is found, the sample cannot be patterned as it is not level. This is usually due to excess silver paint, which must then be removed from the rear surface of the substrate, as discussed in section 3.3.

Once the lithography process begins, the stage raster scans over a pre-set range which is generally slightly larger than the dimensions of the sample.

After lithography is complete, a good patterning is assured using an optical microscope to ensure sharpness of the edges and to identify any defects. The microscope used has a green filter to prevent any unwanted exposure of the photoresist. If any issue with the patterning is found, the patterning process is halted and the photoresist is completely removed from the sample using acetone in a sonic bath. After the resist is removed, the patterning process may be restarted from the spin-coating step, or the sample may be discarded.

Provided that no such issue is found, the correctly-patterned sample is heated to 125°C for 1 minute. This second heating of the sample is referred to as post-baking, since it occurs after some parts of the resist have been exposed to light. Post-baking
is carried out at a higher temperature than pre-baking, and its purpose is to further harden those parts of the resist which are not to be removed.

After sample has cooled, it is immersed in AZ726 MIF developer solution to dissolve the light-exposed parts. The sample must be constantly agitated to ensure an even dissolution of all undesired parts of the photoresist. After development has continued for 20 seconds, the sample is rinsed using deionised water and dried using compressed nitrogen gas.

For samples with a thicker resist layer it was necessary to place the dish containing sample and developer into an ultrasonic bath for greater agitation, in order to fully remove the exposed photoresist. However, for samples coated using the optimal parameters, sufficient agitation could be achieved simply using tweezers and hand motion.

3.2.3 Ion Beam Etching

Ion beam etching is the process of removal of parts of a sample through the directing of a high-energy stream of ionised particles toward the sample surface. The photoresist protects some parts of the sample, while the undesired parts are removed. Etching of YBCO samples for this research was carried out using argon ions accelerated with a Mantis RFM30 ion gun within a custom-built vacuum chamber. The sample is cooled with liquid nitrogen during etching to ensure that the hot plasma does not heat the YBCO above $\sim 700^\circ$C, to avoid a structural phase change [140]. The ion beam etching apparatus is shown in figure 3.3.

Etching Method

The etching procedure begins with the mounting of samples on a titanium table, which is magnetically attached to a rotatable arm within the etching chamber. The arm is initially rotated to face the viewing window, away from the ion gun. The chamber is then pumped down to a pressure of $10^{-6}$ to $10^{-7}$ Hz. Liquid nitrogen ($\text{LN}_2$) is slowly poured through the $\text{LN}_2$ funnel to fill a small reservoir behind the table mount. An overflow tube allows the excess liquid and boiled-off nitrogen gas to escape.

Once the temperature and pressure of the sample space have stabilised, the argon ion flow (in the form of a fully ionised plasma) can be initiated (fired) as follows: Firstly, a valve is opened to allow argon gas flow through the ion gun into the chamber with a fairly high initial flow rate of typically $20 \text{ cm}^3/\text{min}$. The argon gas is then excited into the plasma state by applying radio frequency (RF) power of typically $100 \text{ W}$ to the gas in the ion gun, this causes electrons to disassociate from their nuclei and oscillate at the same frequency as applied. A voltage of $\sim 50 \text{ kV}$ is
also applied between a positive electrode at the back of the ion gun and a negative grid plate across the front, in order to accelerate the positive ions into the chamber. Successful plasma initiation is confirmed by observing a bright purple glow from the front of the ion gun.

Once a stable plasma is achieved, the conditions must be adjusted for optimal etching. The operating conditions for the plasma are quite different from the initiation conditions. The flow rate can be significantly reduced, while RF power and accelerating voltage must be increased. However, each of these parameters must be adjusted slowly in order to preserve the plasma state, which can be unstable.

The most critical parameters for etching are the partial pressure of argon, the accelerating voltage and the current passing through the ion gun. The optimisation of these parameters is discussed in section 3.3. The accelerating voltage correlates with the average energy of ions leaving the gun, while the current gives an indication of the flux of ions leaving the gun. Of these variables, only the voltage can be directly controlled by the operator, while argon pressure and current are dependent on other

**Figure 3.3:** Ion Beam Etching apparatus, with key components labelled. The etching chamber also functions as the load lock for a larger vacuum chamber, the edge of which is visible at the right of the image. The RFM30 ion gun is mounted on the far side of the etching chamber, opposite to the door.
controllable variables.

As soon as the operating conditions are reached and seen to be stable, the table can be rotated into the etching position, with the sample surface facing toward the ion gun. In this position, the stream of accelerated ions is incident normally to the sample surface, causing an even exposure of the entire sample as long as it is positioned in the centre of the ion beam. The unprotected parts of the sample will be progressively ablated by the stream of high-energy ions, while any ions incident on the photoresist should ideally not penetrate through to the protected sample beneath. This mechanism is illustrated in figure 3.5(b).

After the etch time has elapsed, the table is rotated away from the ion beam. The plasma can then be safely extinguished as follows: The power must not be simply switched off immediately, as this would cause rapid cooling of the inner tube of the ion gun, which could catastrophically lead to a crack. Instead, the RF power is reduced at a rate of 10W/min in order to slowly reduce the plasma energy and allow gradual cooling. The accelerating voltage can be simultaneously reduced, but this must also occur slowly to avoid premature extinguishing. Once the RF power has been decreased back to the initiation value of 100W, both the RF and the accelerating voltage can be shut off, extinguishing the plasma, and the argon flow can be reduced to zero.

At this point, the etching process is complete. Before removal of the sample, the chamber must be left at low pressure for several hours (usually overnight) to allow all liquid nitrogen to boil off and for the table and sample to warm up to close to room temperature. If atmosphere were immediately vented into the chamber, condensation would form on the cold sample, leading to rapid degradation due to oxidisation.

When the sample has sufficiently warmed and chamber vented, the sample can be removed. The table and sample are cleaned with acetone to remove the remaining photoresist and residue from the etch. The final etched pattern is checked under the microscope to ensure that all undesired parts of the YBCO were removed, and that the YBCO surface has not been damaged during any part of the patterning process. A typical undamaged etched pattern and a damaged pattern are shown in figure 3.4.

3.3 Optimisation of Patterning Procedures

Almost every part of the patterning procedure was optimised during the course of this research. This optimisation was carried out by myself and several other members of the TFT research group.
Figure 3.4: 10x magnified view of two areas of the same sample shown in figure 3.1. (a) shows a square section of patterned YBCO with minimal damage, while (b) shows a similar section in which some parts of the YBCO have been damaged during the patterning process. These parts appear to have lifted off during the final acetone wash.

3.3.1 Spin Coating Optimisation

The speed and duration of spin coater rotation were optimised empirically. For this research, the optimal parameters were found to be 5000 revolutions per minute (rpm) for 35 seconds, and these parameters were used for all subsequent samples. This speed and duration was found to give a fairly even coating with sufficient thickness.

A faster spin speed was found to give a coating that was too thin, which led to damage of the protected parts of the sample due to high energy ions penetrating the photoresist.

A slower spin speed or shorter duration was found to give a larger lip (thicker region) at each edge of the photoresist, which sometimes led to large regions being underexposed during lithography and therefore not being removed. The problem of photoresist lips at the edges of the sample was persistent and these undesirable features were still often present in samples coated using the best determined parameters. Since the photoresist is thicker within the lip, only the top part of the resist would be exposed by the laser if the lips were too large. Therefore the photoresist would not be fully removed in these regions, and this would leave some unwanted parts of the YBCO intact around the edges of the substrate.

If a significant photoresist lip was seen or suspected after the spin-coating procedure, it could be removed from the sample edges using a scalpel. Since the lip was generally found at the edge of the sample, careful use of the scalpel would not scratch the desired part in the centre. However, it was discovered that removal of the photoresist lip created a sharp edge in the photoresist layer, which occasionally led to lifting off of this entire layer during the development and rinsing process. Therefore, use of the scalpel was avoided for production of high quality samples,
and those with a significant lip were simply re-coated. For re-coating, the photore sist would be removed using acetone, the sample dried and new resist applied by repetition of the spin coating procedure with improved parameters. The lip would usually be significantly reduced or not present after a second spin-coating process.

Another problem was occasionally caused by samples being left in the light-proof box for an extended period, while waiting for the lithography instrument to become available. It was discovered that photoresist left on the sample surface for an extended period would undergo a change in developer solubility and produce a less sharp pattern when developed. Therefore, if any sample were left longer than about two hours, it would need to be re-coated as described above.

The optimal spin-coating parameters were found to be dependent on the quality of the particular batch of photoresist used, particularly on the age of the photoresist. Photoresists tend to degrade over a lifetime of a few months or years, and this lifetime is reduced with heat and light exposure. A photoresist which has degraded beyond usability is identified by increased viscosity, the length of time it has been kept in a vial, and the presence of solid flakes which may be visible in solution or under a microscope on the coated sample’s surface. The exposed parts of an older and more viscous photoresist may not be fully dissolved in developer, leading to incomplete etching. This could be countered simply by using a longer etch time or higher ion current for a slightly more viscous resist. However, whenever the resist was found to contain solid flakes, the entire batch would have to be discarded. These flakes would affect the smoothness of the spin-coated photoresist layer, and more importantly they would remain on exposed areas of the sample after development, leading to microscopic spots of superconducting YBCO remaining in the unwanted areas after etching.

To combat degradation of photoresist, the resist was stored in a light-proof bottle in a refrigerator with no internal light, and was transferred into small vials for use. Of course, these vials would never leave the yellow-filtered lighting of the spin-coating lab. Only one vial would be removed from the refrigerator for each spin coating procedure, and the resist in the vial would be checked visually for flakes and viscosity before use. If the signs of degradation were seen, the resist in the vial would be discarded and a new vial used.

3.3.2 Laser Lithography Optimisation

Some optimisation was also necessary for the laser lithography process. The optimal laser parameters were determined empirically by David Oakden, for best resolution and for complete removal of the desired parts of the photoresist and therefore of the sample beneath. The variable parameters were laser power, aperture size and
“energy factor”, and the optimal values were found to be 10mW, 100%, and energy factor x2 respectively. The energy factor was a manufacturer parameter that affected the duration of exposure of each exposed point on the surface. It may have been possible to produce patterns with smaller features if the aperture were less than 100% open, however this was found to produce asymmetric features in the pattern due to the imperfect geometry of the aperture itself.

The most sensitive factor in the lithographic process was the focus of the laser. If the laser were not focussed correctly, the laser spot size at the sample surface could increase significantly, which was found to produce patterns with blurred edges and loss of features up to 5 μm in size. Levelling of the sample was also found to be quite important for achievement of a good focus across the whole surface. A sample with some silver paint on the rear side could have a height difference of up to 0.5 mm between one edge and the other (which is much larger than the thickness of the film or the photoresist layer, as discussed in section 3.4.2). When the silver paint had been properly removed, a height difference less than 0.05 mm could be easily achieved, and this was found to be sufficient for high-quality patterning.

While severe misalignment of the sample with the x- and y-directions of the lithography stage occasionally produced a pattern that overlapped the sample edge, small misalignments did not greatly affect the pattern and could generally be ignored. These would simply lead to the drawing of a pattern that was tilted with respect to the sample edges. However, even small misalignments became a significant issue for samples that were to be patterned a second time (as required for the production of vortex ratchets). This is discussed further in section 6.4.

As mentioned in section 3.2.2, the spin-coating process was sometimes repeated for samples that were found to have been patterned incorrectly. The criteria for determining whether the patterning process could be repeated for incorrectly etched samples were: that the resist layer could be fully removed and laser pattern no longer visible under optical microscope, and that the YBCO surface did not appear to have been damaged. In general, it was found that any errors occurring during the lithography process could not be corrected by a second spin-coating, as the laser pattern would still be visible. Re-coating was attempted for some such samples, but it was discovered that the final sample produced would still show some signs of the original pattern, even after lithography was applied a second time and etching was carried out. Therefore, it was eventually decided that any incorrectly patterned samples would be immediately discarded, there was no way to reclaim them while still producing a high-quality etched sample.
3.3.3 Ion Beam Etching Optimisation

The parameters of the ion beam etching process were rigorously tested to ensure a lack of damage to parts of the sample protected by photoresist, while the unprotected parts were completely removed. The optimal operating conditions for etching, including the critical parameters and the total etch time, were determined empirically. These optimal conditions varied depending on the thickness of the sample, the properties of the photoresist, and the desired etch depth (if partial etching was required).

The optimal parameters for complete removal of 200 nm YBCO with optimal photoresist coating were found to be: an accelerating voltage of 300kV, ion current of 35mA, argon partial pressure of $5 \times 10^{-3}$mBar and total etch time of 30 minutes.

A higher accelerating voltage would produce higher-energy ions, which were sometimes seen to penetrate through the resist layer and caused spots of damage on the protected YBCO surface. A lower accelerating voltage produced lower-energy ions, and this would generally result in a longer etch time being required for complete etching. If the voltage was set much lower, the ions would not have enough energy to remove the unprotected parts of the sample, and the entire substrate surface would still be uniformly covered with YBCO after etching. This was seen several times in very early etching attempts.

The ion current is directly proportional to the number of ions incident on the sample per second. Therefore a larger ion current would generally reduce the required etch time.

The argon partial pressure was the least sensitive of the critical parameters. It was generally monitored and kept constant during etching to ensure that the etch rate was consistent. However, a small increase or decrease of this value was not found to have any significant effect on the etched samples produced. It was predicted that if the pressure were much too high, the etching process may have been slowed due to collisions of the high-energy ions with other particles before reaching the sample surface, but this was not observed in the current research.

For a complete etching, the etch time was not a particularly sensitive parameter, since there would be no ill effects if the sample were etched for longer than necessary. Once all of the unwanted YBCO had been removed, the substrate below would begin to ablate, but this did not cause any problems. If the sample were not etched for a long enough time, the unwanted parts of the YBCO may not be completely removed. On the other hand, when partial-thickness etching was desired, the etch time suddenly became an extremely sensitive parameter, as discussed in section 6.4.1.

Even for a complete etching, shorter etch times were generally desired. This was
not only to save time, but to reduce the amount of liquid nitrogen required to cool the sample during the etching procedure. Only a few litres of LN$_2$ could be brought into the clean room at a time, and this was enough to cool the sample for up to 1 hour if used sparingly. Since continual monitoring of the sample was necessary (as discussed in the following subsection), it was generally impractical to leave to collect more LN$_2$ once the etching process had started - it was preferable to simply reduce the etching time.

**Problems Arising During the Etching Procedure**

The argon plasma could be unstable and careful monitoring was necessary throughout the etching process, especially when the plasma was first initiated. Several of the most common plasma issues were as follows:

- The plasma may fail to fire, in which case the power could simply switched off and firing could be attempted again. Occasionally, several attempts were required to successfully initiate the plasma.

- After initiation, the plasma must be visually checked for homogeneity across the width of the ion gun, since small dots of brighter and higher energy plasma might appear. This is referred to as “pinning”. Pinning could be corrected by simply switching off and on the RF power in order to extinguish and re-fire the plasma.

- The front grid panel of the ion gun through which the plasma passes into the chamber can become charged, causing arcing between this grid and neighbouring metal surfaces and leading to a large spike in plasma current. This generally occurs if the accelerating voltage is increased beyond a stable operating level, and can usually be stopped by rapidly decreasing this voltage.

- The plasma may extinguish during the etching process, which is marked by a sudden drop in the accelerating voltage. This issue was generally more difficult to resolve and would add some uncertainty to the overall etch time. The first response to an extinguished plasma was to close and re-open the argon valve while maintaining RF power at its operating level, which would sometimes allow the plasma to immediately re-fire and quickly re-stabilise under operating conditions. However, if this did not cause the plasma to fire after a couple of attempts, then the etching timer would be paused, the table rotated away and the plasma initiation repeated using the standard initiation conditions. In this case, more liquid nitrogen was often required to complete the etching process as it would take extra time to restore the operating conditions.
The first attempts at ion beam etching were carried out using a Mantis RFM100 ion gun, which had a much larger beam diameter than the RFM30. An ion gun with large beam diameter was chosen to ensure beam homogeneity at the sample position, and to allow consistent etching of a larger number of samples distributed around the table.

However, this ion gun was particularly prone to both pinning upon plasma initiation and arcing when the plate voltage and current were increased beyond a certain point. Of these, the arcing was the most significant issue as it restricted the range of variation of the critical operating parameters.

The RFM100 ion gun was also more sensitive to rapid cooling due to its larger tube becoming hotter during operation. This ion gun eventually suffered from a crack in the tube, and it was replaced. The RFM30 was chosen for replacement in order to overcome the arcing issue, and the gradual-cooling shut down procedure was enforced more closely following the breakdown of the first ion gun.

Both arcing and pinning were found to be rare occurrences with the RFM30. Higher operating voltages and currents were possible using this ion gun, which allowed shorter etch times with comparable results. It was also found that extinguishing of the plasma occurred much less often after replacement - this was attributed to a smaller crack which may have formed some time before the RFM100 became unusable.

3.3.4 Original Patterning Procedure

The patterning procedure has been subject to continuous revision throughout the course of this research. The procedure has been described in the preceding sections as it stood during the final year of my research at ISEM.

During the first year of this study, the clean room at ISEM had not yet been constructed, and neither the laser lithography nor ion beam etching apparatus were yet in use. An alternate sample patterning procedure was in use at this time; Masked photolithography was used rather than laser lithography, and chemical etching instead of ion beam etching. The spin coating procedure was unchanged (up to optimisation).

This original procedure allowed samples to be patterned into predefined macroscopic shapes, with a precision of approximately 50 μm. The major differences between the original and new patterning procedures will be briefly described in the following subsections:
CHAPTER 3. TECHNIQUES & SAMPLE PREPARATION

Masked Photolithography

Before laser lithography was implemented, the lithography process was carried out using pre-printed glass masks to cover part of the photoresist-coated sample, while the undesired parts were exposed.

After spin-coating and pre-baking as described in section 3.2.1, the sample was placed into a UV-exposure box with the resist facing upward, and the mask was carefully placed on top with the printed side facing downward.

The sample and mask were held in place using a plastic film which was vacuum-sealed in order to create downward pressure on the top of the mask. The lid of the UV-exposure box contained powerful fluorescent lamps which were then activated, shining ultraviolet light through the glass mask onto the uncovered parts of the photoresist.

The masked lithography procedure was simpler and much faster than laser lithography, requiring only about one minute for exposure. There were, however, several disadvantages:

- Alignment of the sample with the mask could not be easily controlled. The pattern could therefore be produced off-centre and at a significant angle to the sample edges. Occasionally, parts of the pattern would lie off the edge of the substrate, and the sample could not be used.

- The edges of the pattern could not be produced as sharply as with a laser. The edge of the exposed area would be slightly blurred due to diffraction at the edges of the mask. This effect was minimised by reducing any gap between the sample and the mask, and by ensuring that the printed side of the mask was facing toward the sample surface.

Chemical Etching

Etching of the samples was originally quite rough, and was carried out using a simple chemical procedure.

After development, the unprotected parts of the sample were removed using nitric acid (HNO$_3$). The sample was simply immersed in a small vial of HNO$_3$, and held for a carefully-measured time of a few seconds. Agitating the sample, one could watch the YBCO quickly dissolve from the unprotected areas while the pattern remained.

When the chemical etch was complete, the substrate could be clearly seen in those places where the sample had been removed. If some dark YBCO was still visible in these areas, the sample was re-immersed for a very short time to fully
remove it. The sample was then quickly rinsed with deionised water to remove the nitric acid and prevent damage to the protected parts of the sample.

As with masked lithography, the chemical etching process was quick and easy, but had some major disadvantages compared to the new method:

- Firstly, and most importantly, sharp edges could not be precisely formed using a chemical technique. Since this etching technique works by eroding away the sample in a wet environment, the protected parts of the sample could not be fully shielded from the nitric acid. The acid had a tendency to seep under the edges of the photoresist layer and remove or damage these parts of the sample, as shown in figure 3.5(a).

- This technique was also very sensitive to split-second changes in timing: A slightly shorter immersion in nitric acid would often lead to incomplete removal of the unwanted YBCO, while a slightly longer immersion (or even a delay before rinsing with deionised water) could lead to further damage to the YBCO beneath the photoresist layer.

The effects of chemical and ion beam etching on YBCO samples are compared in figure 3.5.

![Diagram comparing chemical and ion beam etching](image)

**Figure 3.5:** Diagrams comparing the mechanisms of the two etching processes, showing the effects at the edge of the photoresist layer. Not to scale. (a) Chemical etching creates a rough edge since nitric acid seeps beneath the photoresist. (b) Ion beam etching creates a smooth edge, and may also ablate part of the substrate. High-energy ions are absorbed by the photoresist layer, though some may penetrate through to the YBCO beneath.

### 3.4 Preliminary Measurements

Preliminary measurements were necessary for quality assurance at several points during preparation of YBCO samples:

Firstly - and most crucially - magnetometry was used to test the global magnetic properties of the superconductor such as critical temperature and current. These tests were carried out when samples were first deposited, and again after patterning.
Samples found to have good superconducting properties could be immediately used for experimental purposes, but some were subjected to further testing to determine other properties of the YBCO films. Film thickness and etch depth could be tested with profilometry, atomic force microscopy was used to examine surface features such as domain size, and magneto-optical imaging was carried out before patterning for some samples to identify any current-limiting defects.

### 3.4.1 MPMS Magnetometry

The quality of YBCO films was analysed using a Quantum Design Magnetic Property Measurement System (MPMS) magnetometer. For every sample produced, MPMS was used to determine the superconducting transition temperature $T_c$ and the sharpness of this transition $\Delta T_c$. The critical current $J_c$ was also calculated using hysteresis measurements, as described in [138, 144].

The necessary parameters for a YBCO sample of suitable quality were $T_c > 90$ K, $\Delta T_c < 1$ K and $J_c > 10^9$ A/m$^2$ (measured at 77 K). Any samples not meeting these criteria were excluded from further measurements and processing.

Quality testing using the MPMS was necessary at two points during thin film preparation: after the sample had first been deposited and after the patterning process was completed. An example of these measurements is given in fig. 4.20, and the surrounding discussion.

The first test for each sample was carried out immediately after it was produced by pulsed laser deposition, before it was used for any purpose. This test was necessary due to the variation of superconducting properties between samples deposited in different PLD runs, arising from changes in both random and controlled deposition variables.

The second test was necessary because of the possibility of degradation of superconductivity during the etching procedure. Such degradation can be caused by penetration of high energy ions through the photoresist layer, causing both point defects (which may enhance $J_c$) and larger defects (which may suppress superconductivity). Degradation could also occur during removal of photoresist: extended exposure to acetone can degrade YBCO, or the sample could become mechanically scratched.

The samples may also become degraded if exposed to atmosphere for an extended period of time, as discussed in section 2.2.

### 3.4.2 Profilometry

The thickness of deposited YBCO films and the depth of etching were tested using a Dektak profilometer at the University of Wollongong’s Intelligent Polymer Research
Institute (IPRI).

The profilometer measures roughness and the height of any steps on a sample’s surface. It achieves this by continuously measuring the vertical position of a stylus as it is scanned linearly along the surface of a sample. The stylus used had a 12.5 μm tip, and the instrument can determine the vertical position of the point of contact between tip and sample to a resolution of 167 nm.

The instrument therefore cannot measure sample thickness directly, but it was possible to determine all relevant physical depths and thicknesses using measurements of step height after appropriate sample preparation. For example, the thickness of films deposited by PLD could be determined by applying a mask over part of the substrate during deposition, and then using profilometry to measure step height from the film to the bare substrate in the area that had been masked.

The majority of profilometry measurements in this research were used for the depth optimisation of partial etching. A series of measurements were taken for samples etched over a range of exposure times in order to calibrate the relationship between etch time and depth of YBCO removed. Partial etching was used for the creation of ratchet samples, as discussed in section 6.2.2.

![Figure 3.6: Raw profilometry data for a YBCO sample on STO substrate, after etching several grooves of depth ~300 μm. This was a completely etched sample, and the profilometry results show that the etching process also removed about 100μm of the substrate in each groove. The noise results from both surface roughness and small dirt particles.](image)

Profilometry was also used to test the thickness of the applied photoresist layer and the dependence of thickness and smoothness of the photoresist on the spin coating parameters. This was achieved by measuring the height of step edges after the development step of the patterning process, when only some parts of the resist
had been removed. The thickness of resist after spin coating using the optimal parameters (see section 3.3) was generally (600 ± 40) nm.

### 3.4.3 Atomic Force Microscopy

The surface morphology of YBCO films was examined using atomic force microscopy (AFM), in order to determine the size of domains in the material and other features. This was important for comparison of the size and location of surface features to the observed magnetic features, particularly those seen with scanning SQUID microscopy, as discussed in sections 5.3.2 and 5.3.3. The films examined were deposited to a thickness of ~200 nm, and had been subject to the full patterning procedure.

Atomic force microscopy measures the height of surface features on a sample on an atomic scale, relying on the Van der Walls forces between atoms on the surface and those on an atomically fine scanning tip. The deflection of the tip is measured as it scans across the sample, and this deflection is recorded in a similar manner to that employed in the profilometry technique, though it occurs on a scale several orders of magnitude smaller.

Typical atomic force microscopy images of a YBCO film are shown in figure 3.7. The sample shown in figure 3.7 is the same sample as was used for in-depth analysis of vortex arrangement in section 5.3.2.

![Atomic Force Microscopy images](image)

**Figure 3.7:** Atomic Force Microscopy images of two 10 × 10μm regions of a PLD-deposited YBCO film, showing the granular structure. Domain boundaries are seen as dark regions, while the interior of the domains appears grey.

Films deposited using the procedure described in section 3.1 develop a domain-like structure due to the island growth mechanism. Since YBCO is preferentially deposited onto the island-domains rather than the boundaries, the domain boundaries tend to be lower on the sample surface. These boundaries therefore appear darker in the images.
3.4.4 Preliminary Tests using MOI

Magneto-optical imaging (MOI) was carried out after deposition but before patterning for some samples, to test for current-limiting defects such as scratches or other physical damage. MOI was only used at this preliminary stage for those samples suspected to have incurred minor physical damage or for important samples requiring high-precision patterning.

Current-limiting defects and other factors influencing the quality of superconducting thin films can be clearly and easily identified using magneto-optical imaging and video.

Since magneto-optical imaging comprises the entire of chapter 4, discussion will be reserved for there. An example of the identification of localised physical defects and application of appropriate patterning to avoid them is detailed in section 4.3.2, while the use of magneto-optical video to analyse overall sample degradation is discussed in section 4.3.7.

3.5 Film Properties

Since many TFT group researchers have used similar YBCO thin films, produced as described in section 3.1, the properties of these films have been investigated quite extensively. The previous sections describe only a select few tests that were carried out by myself. Many other relevant properties and features of these films were examined by other group members, and these will be outlined below.

The critical current of YBCO films varies for one deposition to another, but can be higher than $J_c = 3 \times 10^{10}$ A/m$^2$ at 77K, as determined from MPMS tests [15, 16]. This $J_c$ generally increases by one to two orders of magnitude when the temperature is reduced to 10K.

The effective penetration depth for TFT YBCO films was measured to be $\lambda \approx 180$ nm [141, 142].

The dominant pinning sites in these films were strong columnar defects in the $c$-direction, occurring within domain boundaries [99, 143, 144]. The pinning potential was found to have a field dependence that was approximately hyperbolic [70, 143]. The individual vortex de-pinning radius for each pinning site is approximately 0.5 $\mu$m [141, 142].

These boundaries arise between domains that have low mis-orientation angles and are separated by domain walls consisting of edge-dislocations [99, 143, 144]. This differs from other superconducting films that often instead possess grains connected by weak Josephson links - such grains are not found in the YBCO films deposited in this research [99].
Chapter 4

Magneto-Optical Imaging & High-Speed Video

This chapter begins the experimental part of this thesis, presenting the results of several investigations into the magnetic properties and dynamic magnetic behaviour of superconducting films using the magneto-optical imaging (MOI) and high-speed magneto-optical video (MOV) techniques.

The magneto-optical imaging apparatus is described in section 4.1, and the software that was developed to control the imaging procedure and to apply calculations to the resulting data is given in section 4.2. The results of these investigations are given in section 4.3, with several novel results that were obtained using MOV forming the latter part of that section.

4.1 Magneto-Optical Imaging Apparatus at ISEM

The magneto-optical imaging apparatus at the Institute for Superconducting and Electronic Materials (ISEM) consists of:

- a continuous-flow cold finger cryostat which houses the sample at a controlled temperature in a raised position;
- a solenoid placed around the cold finger to apply magnetic field;
- an indicator film placed on top of the sample within the cryostat;
- a polarising microscope to view the visualisation of magnetic field; and
- a high-speed camera to record the images.

Each of these components will be discussed in more detail in the following subsections. Most components of the apparatus were individually designed by Professor
Alexey Pan and purpose-built for magneto-optical imaging. A diagram of the apparatus is given in figure 4.1, showing light path and data processing steps.

![Diagram of magneto-optical imaging apparatus](image)

**Figure 4.1:** Diagram of the magneto-optical imaging apparatus, showing light path, helium flow and data flow. The geometrical factors $d$ and $h$ are shown, representing the thickness of the sample and height of the indicator film respectively. A solenoid is placed around the cold finger to apply field, not shown for simplicity.

### 4.1.1 Cold Finger Cryostat

Sample temperatures of $\leq 4K$ were maintained using a custom Janis continuous-flow helium cryostat [41]. The cryostat contains a cold finger, which thermally connects the sample to a heat exchanger, while elevating the sample to the centre of the solenoid.

The heat exchanger consists of a coil of brass tubing in solid contact with a thick brass disk, which has high thermal mass in order to function as an effective heat sink. Liquid helium flows through the brass tube at a controlled rate, being vaporised in
The heat exchanger and exhausted to a recovery system. Liquid nitrogen can also be used for quick measurement of samples with $T_c > 77K$.

The cold finger is a long, thin brass cylinder which extends vertically from the heat exchanger. It has high thermal conductivity but much lower thermal mass in order to efficiently transfer heat downward from the sample to the heat exchanger. Cernox temperature sensors at the base and top of the cold finger confirm minimal temperature gradient between the sample and the heat exchanger. The sample is mounted at the top of the cold finger using a small amount of vacuum grease to ensure good thermal contact.

The cryostat is mounted inside a vacuum-sealed housing, which is pumped down to a pressure less than $10^{-5}$ mBar. A brass heat shield in the shape of a thin cylindrical shell is placed between the cold finger and the housing to absorb radiated heat from the walls. A 10 mm diameter window above the cold finger allows the sample to be viewed.

The heat exchanger also contains an internal resistance-heater, which is used to maintain controlled, steady temperatures above 4K. The heater is also useful for quickly heating samples above $T_c$ and re-cooling in order to re-measure penetration from the virgin state. A LakeShore 331 temperature controller monitors temperature at the two sensors with 0.01 K precision, and allows programmable control of the heater.

Connecting wires must pass from the temperature controller at room temperature to the heater and temperature sensors inside the cryostat. These wires must therefore be thermally anchored by wrapping around the helium tubes in order to prevent heat conduction from the part of the wire outside the cryostat. This is particularly important for the temperature sensors, where wire heat conduction could lead to false readings. For the sensor at the top of the cold finger, the wire is also wrapped tightly around the length of the cold finger, so that the temperature gradient in the wire is approximately the same as along the cold finger.

### 4.1.2 Magnetic Field System

Magnetic field was applied to the samples via a solenoid that encircles the sample area. Field was varied over a range of $\pm \sim 0.1T$ by adjusting current in the solenoid.

Current is passed from a Xantrex XKW 40-75 power supply to the solenoid, producing a very linear field response [21, 80], which was empirically found to be:

$$B(T) = 0.01095 \ I(A)$$  \hspace{1cm} (4.1)  

where $B$ is the magnetic field measured at the centre of the coil, where the sample lies.
For the measurements of flux penetration into superconducting films, the solenoid current was switched from zero to 10A, causing field to ramp up in the manner shown in figure 4.2. For this measurement, the field increased linearly with time for the first few milliseconds, then the ramp speed began to decrease after approximately 16ms. The field then slowly stabilised to its final value, reaching 90% of this value within 27ms but taking significantly longer to truly level.

This slow stabilisation is a result of the power supply slowly stabilising the current in a manner that avoids overshoot.

Averaging many similar curves, the maximum magnetic ramping speed of the coil was determined to be \((8.1 \pm 2.3)\text{T/s}\), and the field was seen to reliably reach \(>90\%\) of its maximum value at \((27 \pm 3)\text{ms}\) after current switching, with no overshoot.

![Figure 4.2: Typical characteristic curve of the solenoid used for external field ramping, measured using a Hall sensor. The zero time-point is taken from when the field first starts to increase after it is switched on. Calibration data courtesy of Stephen Wilson.](image-url)

4.1.3 Polarising Microscope Setup

To acquire magneto-optical images, superconducting samples were placed in the field of view of a purpose-built polarising microscope.

Within the microscope, light is passed through a polariser and then directed through an objective lens onto the indicator film (which is described in section 4.1.5).
The light is then reflected back into the microscope’s objective lens and passes through an analyser before reaching the eyepiece and/or camera. The full light path is shown in figure 4.1.

Movement of the sample inside the cryostat would be impractical, as would movement of the entire cryostat. Therefore, to allow adjustment of the viewing position, the microscope was mounted on an \(xy\)-mobile stage, while the sample position was fixed.

The light source used was a high-intensity Nikon Intensilight C-HGFI mercury lamp, which has emission lines in the region of the spectrum corresponding to the sensitive range of indicator films. Since the mercury lamp takes a few minutes to warm up, a lower-intensity incandescent bulb could also be used for quick alignment of the optics.

Two cameras were used with the MOI microscope: a Leica DC 300F camera for still images in colour, and a Photron Fastcam SA3 camera for still images and high-speed video in high-resolution greyscale.

### 4.1.4 High-Speed Camera (Photron Fastcam SA3)

The Photron Fastcam SA3 high-speed camera at ISEM was used to capture high-resolution magneto-optical images for quantitative analysis. This camera is capable of capturing very high-speed video, although the high-speed capabilities have been utilised only for the first time in this research.

The Fastcam SA3 is capable of capturing video at up to 120,000 frames per second (fps) or \(1.2 \times 10^6\) Hz. However, the actual frame rate at which MO video was captured was 250 to 2000 fps, which was sufficient for investigation of the fastest field changes seen using the samples and apparatus at ISEM. The reasons for this were as follows:

- The camera has a limited buffer size, which limits the overall length of videos to 2725 frames. Since full penetration into the films from the zero field state can take close to a second, the video capture time could not be shorter than about two seconds to ensure that the event is fully captured. Also, due to triggering issues, penetration events were occasionally missed completely when a shorter video capture time was used.

- In addition to this restriction, the speed of penetration of vortices into the film was limited by the ramping speed of the external magnet, which was \((8.1 \pm 2.3)\) T/s as discussed in section 4.1.2. Taking this into consideration, an increase in frame rate from those used would not significantly benefit the analysis of flux penetration in this experimental setup. Higher frame rates
would only become useful if a specialised fast-ramping or pulsed magnetic field system were used.

- Higher frame rates also require shorter exposure times, which leads to a reduction in light intensity. However, this limitation was overcome by using a very bright light source.

The exposure time of the camera could be selected, up to a maximum of $1/(\text{fps})$. An exposure time of 0.5 to 2 ms was used for most MOV. Since the camera is designed for precision high-speed imaging, and the indicator film response is linear with time at these speeds, each image could be interpreted as a direct mapping of the magnetic field averaged over the exposure time. The exposure time therefore corresponds to the temporal resolution of dynamic field measurements. The physical resolution using the most common magnification was calibrated to be 150 pixels/mm, as discussed in the following subsection.

The camera bias (shading) was corrected programmatically before any imaging procedure, and whenever the exposure time was changed. This simply required a shutter to be placed in front of the camera to calibrate for zero incident light.

### 4.1.5 Indicator Films

The core element of magneto-optical imaging is the indicator film, which visualises the magnetic field by rotation of polarised light. A description of the theory behind indicator films and Faraday rotation is given in section 2.6.1.

The Faraday-active part of the indicator films used is a thin, single-crystalline, ferrimagnetic layer of bismuth-doped yttrium iron garnet (Bi:YIG). It is grown by liquid phase epitaxy on a transparent gadolinium gallium garnet (GGG) substrate[98, 106, 145, 146]. A 125nm thick Aluminium reflective layer is deposited on top of the Bi:YIG layer, followed by a 120nm capping layer.

Indicator films are produced with spontaneous magnetisation $M_s$ in the plane of the film. The optical axis is directed normally to the plane of the film, to align with the direction of applied field and of incident light during the MOI procedure.

These films were purchased as disk-shaped ingots that are cut to a usable size, between $3 \times 3$ mm and $6 \times 6$ mm.

The film is placed face-down onto the sample for the MO imaging procedure. It is generally assumed that a gap of about $10\mu$m exists between the sample and the Faraday-active Bi:YIG layer due to roughness of the sample and indicator film surfaces, though the true width of this gap is difficult to measure[63, 107]. The field measurement height is therefore taken as $h = 10\mu$m for all current calculations (see section 2.7.1).

The light path through the indicator film during MOI is as follows:
Figure 4.3: The structure of the indicator film and its placement above the YBCO sample. Not to scale. Optical paths are indicated by yellow arrows, with incident beams directed normally downwards. Smaller black arrows indicate polarisation direction. Rotation of polarisation is shown in response to a large field $B_1$ on the left and small field $B_2$ on the right. In reality, this rotation occurs in a plane perpendicular to the optical path, but this could not be easily shown in a two-dimensional diagram.

- Polarised light is incident on the back of the GGG substrate, where the majority is transmitted through to the Bi:YIG layer.

- On transmission through the Bi:YIG, the polarisation rotates through an angle $\alpha_F$, which is proportional to the thickness of the layer and local magnetic field strength.

- The light is reflected from the surface of the aluminium.

- It passes through the Bi:YIG layer again and the polarisation is again rotated by $\alpha_F$. These rotations are additive.

- The light passes back through the GGG to the objective lens.

The structure of the indicator film is sketched in figure 4.3. This figure also shows the rotation of polarisation by an angle $\alpha_F = \alpha_1$ in response to a field $\mathbf{B}_1 = B_1 \hat{z}$, and $\alpha_F = \alpha_2$ in response to a smaller field $\mathbf{B}_2 = B_2 \hat{z}$.

Bi:YIG indicator films have been shown to have a very fast linear response to changing magnetic fields up to at least $10^6$ Hz [115], which is significantly higher than the frame rate of the high-speed camera used in this apparatus. Hence these films can be taken to respond practically instantaneously to changes in magnetic field.
Indicator Film Degradation

Some of the indicator films used exhibited significant degradation over time. Preliminary testing showed that identical magnetic features appeared sharper in indicator films that were less frequently used.

Degradation was investigated with a resolution test on each film using the magnetic strip of a credit card as a reference. A credit card was chosen for its easily-identified linear magnetic features, which have a suitably small thickness. Each film produced an image of these features that was Gaussian along a profile taken across the linear features. The quality of films was determined by the width (full width at half maximum or FWHM) and peak intensity of the resultant field profiles.

Notable variation was found between different films, despite the fact that they were cut from the same ingot. Two films are shown side by side on the same credit card in figure 4.4, showing significant broadening of the magnetic features in the more frequently used film.

![Figure 4.4: Resolution test of frequently used (left) and less frequently used (right) indicator films using the magnetic strip of a credit card. Brightness profiles are plotted across the linear features, with the fitted curves used to calculate FWHM shown in red, and baseline in green. Small scratches are seen in the bottom left corner of the film on the right.](image)

Spatial resolution was not truly determined by this simple test, but was found to be better than $\sim 40 \, \mu\text{m}$ for the least degraded films, by considering the smallest
line widths seen. The width of similar features could be more than twice this in very frequently-used films, which indicates a twofold reduction in spatial resolution. The lower brightness of the peaks also shows a slight reduction in contrast, which indicates a reduction in magnetic field sensitivity in the more frequently used films. This test therefore shows that significant degradation of the films must occur during regular use.

In addition, frequent use can lead to physical defects such as scratches and stains on the surface of the films, which are difficult to remove. All of these features are visible under the microscope and therefore produce artefacts in the magnetic field measurements, which carry through to the calculated current distribution.

It was therefore critical to examine indicator films carefully before choosing a high quality film for imaging. The procedure employed to maximise image quality was to use the same large indicator films (which showed moderate degradation and were relatively defect-free) for imaging of the majority of samples, while preserving the highest quality films for use with the most important samples, when accuracy of results was crucial.

4.1.6 Modifications to the Apparatus

Several modifications were made to the magneto-optical imaging apparatus at ISEM in order to facilitate the current research. Some of these modifications were necessary to resolve issues with the apparatus such as excessively high sample temperatures, while others allowed new functionality, with a particular focus on allowing the effective capture of magneto-optical video. The two most significant modifications will be detailed in the subsections below.

Light Source

Prior to this work, only static images of the magnetic fields were obtained by magneto-optical imaging experiments at ISEM. An existing Nikon 12V 50W Halogen lamp was suitable for obtaining still images, but not for video capture at high speed.

High-speed video requires a brighter light source because the exposure time is limited by the frame rate, as discussed in section 4.1.4. A shorter exposure time reduces the brightness of the image, since it allows less time for photons to reach the light sensitive area. Therefore a higher light intensity was required to produce high-speed images of a suitable brightness.

In order to increase light intensity, a super-bright Nikon Intensilight C-HGFI helium lamp was added to the system. A sliding light source mount was also added, which allowed the user to switch between this new light source and the existing
Measuring and Reducing Temperature Gradient

The temperature measured at the heat exchanger was found to be inconsistent with the temperature of the sample. Therefore a new temperature sensor was installed near the top of the cold finger to determine the temperature gradient along the cold finger.

A gradient was first suspected while attempting to image the field around superconducting Niobium samples ($T_c = 9\, K$). The temperature sensor read less than $5\, K$ and the samples had been repeatedly tested showing good superconducting properties including critical temperature, yet no superconductivity was seen through the magneto-optical microscope. This led to the suggestion that the sample location may be several Kelvin warmer than the position of the temperature sensor.

To test for this temperature gradient, the mounting of a new sensor (model Cernox SD) closer to the sample position was required. This sensor was first calibrated by mounting on top of the old sensor (Cernox CU) and cooling through the entire temperature range (approximately $300\, K$ to $4\, K$). The calibration curve is shown in figure 4.5, and is accurately modelled by a power law fitting at low temperatures. Deviation from the fit curve is seen around $90\, K$, but this did not affect results since the temperature gradient is negligible at such high temperatures, the SD sensor was only used for measurements below about $30\, K$.

Once calibrated, the sensor was mounted near the top of the cold finger, where it was both physically and thermally closer to the sample. The previous sensor had been mounted on the heat exchanger near the base of the cold finger. Upon cooling, the temperature of the heat exchanger quickly reached a steady value of $4.2\, K$ when cryogen was flowing. The temperature at the top of the cold finger was some ten kelvin higher than this, but reduced steadily, reaching $12\, K$ after the base temperature had been stable for one hour.

Despite attempts to resolve this problem, measurements could not be carried out below $12.5\, K$ for some time, and the sample temperature could not be kept stable. The source of the temperature gradient was eventually found to be a dirty screw-thread connection between the heat exchanger and the cold finger, which led to poor thermal contact. After cleaning this thread, the temperature of the samples in the MOI cryostat could once again reach close to $4\, K$. 

Halogen lamp.
4.2 Image Acquisition and Analysis Software

The magneto-optical imaging and analysis process was controlled using LabVIEW-based programs which were created during the course of this research. These programs allow acquisition of images and video of magnetic field under carefully controlled sample conditions; calculation of supercurrent distribution under static and dynamic conditions; and quantitative analysis of the field and current data obtained.

4.2.1 LabVIEW Program for Image Acquisition

The image acquisition program controlled sample conditions and captured magneto-optical images.

Specifically, its functions were:

- To monitor temperature in the sample space using temperature sensors, by communicating with a temperature controller. Limited temperature control was also possible using the resistance heater.

- To adjust magnetic field in the sample space by communicating with a variable power supply, in order to apply appropriate current to the solenoid.

**Figure 4.5:** Calibration data for the Cernox SD temperature sensor. Temperature values were taken from the pre-calibrated Cernox CU sensor. A power-law fit that gives a higher weighting to lower temperatures is shown as a guide.
• To display live video feed of the sample from either the Leica DC 300F or Fastcam SA3 camera.

• To capture and save images from the camera in a format that could be used by the analysis programs.

• To quantify the magnetic field information in these images for further analysis, as discussed in the following subsection.

The creation and functionality of this program is described in greater detail in [80].

Quantitative Imaging (Field Calibration)

In order to accurately calculate currents from magneto-optical images, quantitative information on the magnetic field distribution in the sample was required. This was obtained by calibration of image brightness with respect to magnetic field, using a calibration sub-program.

The calibration procedure was carried out with the indicator film in position on top of the sample and camera correctly aligned, but with the sample in the non-superconducting state (i.e. at high temperature). The brightness of a defect-free region of the indicator film was measured over the full field range. The resulting calibration data was used to convert the brightness of each pixel to a magnetic field value. An independent calibration was required before each imaging run to account for differences in illumination, polariser position, exposure time and indicator film properties including degradation.

An exemplar calibration is shown in figure 4.6. Under appropriate illumination, the response is fairly linear for most fields applied, but some curvature occurs close to 0.1 T due to indicator film saturation. The curve plateaus in the case of 13% illumination, due to saturation of the camera.

Routine calibrations used only a single illumination level, unless saturation was seen, in which case the illumination was reduced and another calibration carried out. They were generally taken using fewer data points, but resulted in similar curves.

The camera calibration sub-program gathers this data automatically, by taking several images at pre-defined field values over the full field range and determining the average brightness over the selected region for each field value. A preview plot of brightness against field allows the user to quickly check the accuracy of the calibration. Calibration data is then saved as a .csv file which can be interpreted by the current calculation program.
4.2.2 Current Calculation

The function of the current calculation program was exactly as its name suggests: using the magneto-optical images that were output from the image acquisition program, it could calculate the current at each point in the sample and then produce a variety of images to represent the current distribution.

This program follows the current calculation procedure outlined in section 2.7.1, which is based on inversion of the Biot-Savart law. This algorithm is applied to the magnetic field distribution from an MO image, giving current in the $x$- and $y$-directions along with the total current magnitude at every point. For each MO image, therefore, three current images were created ($J_x$, $J_y$, and $|J|$) as well as a current streamline map. Current streamlines could also be superimposed onto the original magneto-optical image to show both field and current flow in a concise way.

This program also provided the following options which could be carried out before the current calculation is initiated:

- Quantification of field images, which could be done in two ways: The first was to use calibration data obtained using the calibration sub-program, provided that this data had been obtained. The second was a linear approximation option using two data points in the MO image, for images taken under non-

![Figure 4.6: Calibration curves for a typical indicator film taken with an exposure time of 17 ms. The average brightness of the film is measured over a large area for a range of applied fields, at various illumination levels.](image)
zero applied field. The first data point uses the brightness of a defect-free region of the indicator film far from the superconducting sample, which is taken to correspond to the applied field. The second uses the brightness of a dark region in the centre of the superconducting sample, which is taken to be zero field.

- Averaging of several magnetic field images to eliminate noise, if required for low-light images.
- Subtraction of a background image (usually an image taken of the indicator film immediately before MO imaging, with all conditions identical but zero applied field).
- Inversion of the MO image, which was occasionally useful for fields in the reverse direction.
- Selection of a filtering function to be used (usually the Hanning window - see the high-frequency filtering subsection of 2.7.1).
- Selection of a region of interest over which to calculate, whenever the entire image was not required.

### 4.2.3 Video Analysis

The programs discussed so far were created to process static MO images. In order to allow capture and processing of high-speed magneto-optical video, several new LabVIEW programs were created:

- A video acquisition program to vary the magnetic field in a controlled manner, and capture video while monitoring sample conditions.
• A modified program to calculate current at each point in the superconducting film over time, which was output in video format.

• A further analysis program to produce graphs of current or field versus time in appropriate regions of interest.

Video Acquisition

The LabVIEW program for video acquisition was a heavily modified version of the image acquisition program described in section 4.2.1. In addition to monitoring and controlling temperature and magnetic field and capturing still images (snapshots), this program could also capture video with appropriate timing. The program could trigger video recording and a change of applied field in sequence, so that MOV would be recording at the moment the field began to change, in order to capture dynamic flux motion.

Video Current Calculation

The video current calculation program was used to create videos showing the evolution of current in the sample over time. Each frame represents a snapshot of the supercurrent in the film as calculated from the corresponding frame of magneto-optical video. The current images are computer-generated with brighter regions showing higher current magnitude. The brightness of each of these images is scaled relative to the maximum current for that time step.

This program considers the nth frame of magneto-optical video as a snapshot of the magnetic field at time $t_n$. Therefore, the brightness of a pixel at position $(x, y)$ in the nth frame is taken to correspond to the $z$-component of the magnetic field at the corresponding position and time, $B_z(x, y)|_{t_n}$.

Hence, the video current calculation program used the original current calculation program as a starting point, and operates in a similar fashion. The dynamic current calculation at each time-step is performed as for the static current calculation procedure, and this process is repeated frame-by-frame over the full measurement time (the length of the video).

The calculation begins with a Fourier transform applied to the field distribution in frame $n$ of the MOV, to give $\tilde{B_z}(k_x, k_y)|_{t_n}$. Equations (2.18) and (2.17) are then applied as in the static case, to give $\tilde{J_x}(k_x, k_y)|_{t_n}$ and $\tilde{J_y}(k_x, k_y)|_{t_n}$. An inverse Fourier transformation gives $J_x(x, y)|_{t_n}$, $J_y(x, y)|_{t_n}$ and $J(x, y)|_{t_n}$ for every point in the film.

The current distribution for each frame is output as both a matrix and an image for each of $J_x|_{t_n}$, $J_y|_{t_n}$ and $J|_{t_n}$, as in the static case. However, since most magneto-optical videos in this research were shot at a frame rate around 1000 fps, it was not practical to save an array of data points for each of the thousands of output
frames. Instead, for the purpose of conserving hard drive space and allowing the results to be easily observed and interpreted, video files were created to represent the variations in current distribution in the sample over time.

The dynamic current calculation program creates a .avi video file for each of $J_x$, $J_y$ and $J$. As frame $n$ of the input MOV is processed, the output image of $J_x|\ell_n$ is written to frame $n$ of the .avi file for $J_x$, and a similar process is used for $J_y$ and $J$.

The program was altered to accept .avi video input and carry out calculation of the current in each frame of the video independently. The inverse Biot-Savart procedure was then applied to give a current map over the sample for each frame (i.e. at a given point in time). These individual current maps could then be combined as frames of a new .avi video showing the evolution of current in the sample. Calibration data would be saved along with each video to preserve information on the magnitude of field, and allow further quantitative analysis of the current data.

**Further Video Analysis**

The videos of $J_x$, $J_y$ and $J$ contain all the information about the evolution of current in the sample, although each individual video contains an enormous amount of data and it is difficult to analyse in this format.

In order to quantitatively analyse the current data, linear slices were taken through each sample. A slice was generally taken through the centre of the sample, bisecting the two edges through which it passes (for square samples) in either the horizontal or vertical direction, since this is the region over which dynamic current was calculated most accurately (see section 2.7.1). The time-dependant current profiles in each of these one-dimensional slices could then be considered, rather than the time dependence of currents in two dimensions over the entire area of the sample.

The information on current at each point along this line represents a linear current profile. When applied to a video, this analysis therefore shows the variation of current profiles over time.

The average current over a region could also be measured over time. The regions investigated were usually: a penetrated part of the sample; an un-penetrated part; the entire sample; and a region of the indicator film far from the sample (which should ideally have zero current).

While this program was designed to analyse the time-variation of current, it could also be applied to the magneto-optical videos themselves to provide quantitative information on magnetic field dynamics. The time-variation of field profiles, and of average field in a region, could be plotted in the same way as for current. The field of part of the indicator film far from the sample was used to check the variation of external field for each video.
4.3 Experimental Results and Discussions

This section describes several experimental investigations into the static and dynamic magnetic features of YBa$_2$Cu$_3$O$_{7-\delta}$ films using magneto-optical imaging and high-speed magneto-optical video. Each subsection below will give the results and discussion for investigation into one such feature.

Sections 4.3.1 and 4.3.2 describe fairly common magneto-optical imaging experiments, then sections 4.3.3 and 4.3.4 move on to novel investigations into flux and current dynamics using magneto-optical video, and sections 4.3.5, 4.3.6 and 4.3.7 describe further results obtained by mathematical analyses of the flux and current dynamics. Finally, section 4.3.8 describes the comparison of these dynamic effects to simulation.

4.3.1 Static Magneto-Optical Images and Current Profiles

The first results obtained during my doctoral research were simple static magneto-optical images of YBCO films under different field conditions. The current distributions in the films were calculated for many of the images.

Many films were studied throughout the course of this research for a variety of purposes. This section shows the full analysis and investigation of one typical YBCO square film, which has a large scratch on the left side. This example is used to give an indication of the general behaviour of field and current in PLD-deposited YBCO films, while also showing the influence of a current-limiting defect.

![Figure 4.8: Magneto-Optical Images of the field distribution in a typical YBCO film, taken at (a)1 mT, (b) 33 mT, (c) 66 mT, (d) 106 mT, (e) the remnant state after the 106 mT field had been removed, and (f) the remnant state when the film had been heated to 50 K. A higher illumination was used for images (a) and (f) to show weaker fields.](image-url)
The film pictured in figure 4.8 was deposited under standard PLD conditions to a thickness of 200 nm, and was patterned using masked photolithography and chemical etching as explained in section 3.3.4. It was imaged using the Leica camera to provide colour pictures, as described in section 4.1.3.

While the colour images are sub-optimal for quantitative measurements, they are sufficient for visual examination. Many films created for this research appear identical at low magnification. While the higher resolution of the Photron camera makes a significant impact on calculations, it makes little difference to the naked eye.

For a complete magneto-optical investigation of this sample, the following process was carried out, as was standard for many samples:

- The film was cooled in zero field to 12 K, until the temperature was seen to have stabilised.
- Field was increased in many small steps from zero to 106 mT (the full range of the magnetic field system), with an MO image captured at each step. A selection of these images are shown in figure 4.8(a-d).
- The field was then reduced to zero, and the residual flux remaining in the film due to pinned vortices was imaged in 4.8(e). This residual flux is generally stable, and the film would be permanently magnetised provided a low temperature were maintained.
- The temperature was then incrementally increased, with the diminishing flux imaged at several \( T \) values. As temperature increases, thermal activation tends to de-pin vortices and the trapped flux is reduced. The remaining flux at \( T = 50 \text{K} \) is shown in 4.8(f).

Variations to this procedure for other samples included: increasing the field in small steps again after it had been reduced to zero to show non-virgin penetration; applying field in the opposite direction after the flux had been reduced to zero, to show antivortex penetration; and occasionally, cooling samples in a non-zero field, then reducing this field in small steps.

Following these measurements, the flow of current in the film was calculated and mapped for each of the images captured during this process.

The images in figure 4.9 show a number of quantities that were routinely calculated from a single magneto-optical image using the program described in section 4.2.2: The current components in the \( x \)- and \( y \)-direction were calculated at each point in the film, as mapped in 4.9(a),(b) then these were combined to give the total magnitude of current in the film as shown in 4.9(c). The magnetisation at each
CHAPTER 4. MAGNETO-OPTICAL IMAGING & HIGH-SPEED VIDEO

Figure 4.9: Computer-generated images mapping (a) \( J_x \), (b) \( J_y \), (c) \( |J| \) and (d) \( M \) over the area of a YBCO film. These were all calculated from the same 99 mT MO image of the film shown in 4.8. (e) Contour map of \( M \). (f) Superposition of this contour map (in red) onto the original MO image (in greyscale).

point in the film was simultaneously calculated, and is mapped in part (d) of the figure. Contours of the magnetisation are shown in 4.9(e), and these were superimposed onto the original MO image in 4.9(f) after automatic scaling to match the size of the film. This contour map shows the current streamlines. Since the current calculation program converts magneto-optical images to greyscale, the superimposed image no longer appears green.

4.3.2 MOI for Locating Defects in YBCO

The use of magneto-optical imaging for the identification and extraction of defect-free regions in a sample is demonstrated in this section. This work was jointly undertaken with Alexey V. Pan and Olga Shcherbakova, and is currently being finalised for publication.

Arguably the most important industrial application of magneto-optical imaging is identifying the physical locations of defects and defect-free regions in superconducting thin films and tapes. This is important for identification and elimination of current-limiting defects during large-scale manufacturing of superconducting tapes, which are used to transport large currents for commercial power distribution.

Also, at the laboratory scale, it was often important to identify any current-limiting defects before samples were patterned, as discussed in section 3.4.4. If samples were found to contain defects, defect-free regions could be exclusively selected
for extraction by patterning, as explained below.

Figure 4.10 shows the process by which MOI was used to consistently identify and extract defect-free regions of a requisite size and shape within larger samples: Magneto-optical imaging was applied to three $5 \times 5$ mm samples (labelled 1, 2, 3 in the figure) of YBCO which had been mechanically damaged. Defect-free rectangular regions were successfully extracted from these films, as shown in the lower part of this figure.

![Figure 4.10: Magneto-optical images of three scratched YBCO thin films (upper images), and of smaller films (lower images) that have been cut from the defect-free regions of the larger films indicated by dotted rectangles. All images taken at $B_a \approx 99\text{mT}$. [147]](image)

The YBCO films were created using the PLD technique described in section 3.1, to a thickness of 200 nm. The three films were deposited simultaneously onto substrates cut from the same larger piece of SrTiO$_3$ to ensure identical properties. Some of the films contained macroscopic current-limiting defects in the form of long “scratches”, occurring due to imperfections in the substrate during deposition.

Current-limiting defects are easily seen in the MOI images of the $5 \times 5$ mm films in figure 4.10, since regions of the sample with weakened superconductivity allow flux to penetrate more readily and therefore appear bright in these images. The scratches on these samples are seen as curved white or grey lines, with bright auras showing magnetic flux penetrating from the damaged areas.

Sample 1 contains minimal damage, although inhomogeneous flux penetration is seen at the edges due to crystalline defects arising from the chipped edges of the STO substrate, as discussed in section 3.2. A weak scratch is seen just below and to the left of the dotted region in the image of this sample. A scratch can be seen at the bottom middle of sample 2, extending up and to the left, and a few smaller scratches are seen at the bottom left. A much larger scratch extends horizontally from the right side of sample 3 and across its centre, which is crossed by another
large scratch along the right edge.

The current streamlines (indicated in red in the images) show that these chips and scratches are indeed current limiting defects. Current cannot flow freely through these regions of the film and is seen to curve around them. The curvature of current is seen most clearly above the scratch at the bottom of sample 2.

For each 5 × 5 mm sample, a 2 × 1 mm defect-free region was selected, as shown by white dotted rectangles in the images. These regions were selected as they contained no scratches and were furthest from any damaged regions of each film. The position of each defect-free region was recorded and translated into a lithographic pattern specific to that film. These patterns were applied to the films using the laser lithography technique described in section 3.2.2, and all parts of the film outside of these regions were removed using ion beam etching as described in 3.2.3.

![Critical current in the larger samples before patterning by laser lithography and in the defect-free smaller samples after lithography.][147]

The extracted samples were then tested using MOI and magnetometry measurements, the latter being shown in figure 4.11. This figure shows that the extracted defect-free samples have a higher critical current density at all fields at both 77 K and 10 K, with an increase of zero field $J_c$ from $3.8 \times 10^{10}$ A/m$^2$ to $5.5 \times 10^{10}$ A/m$^2$ at 77 K. It also shows that the samples have greater uniformity after extraction, which is most clearly seen by the closeness of $J_c$ curves taken at 10 K after lithography.

It is therefore seen that the total critical current density in YBCO films is strongly affected by macroscopic current-limiting defects such as scratches. Scratches
can be identified by magneto-optical imaging, and then the critical current can even be increased by appropriate patterning with reference to these images.

### 4.3.3 Magneto-Optical Videos of Flux Penetration and Depenetration

The first magneto-optical videos captured using the newly developed high-speed magneto-optical video procedure were of penetration of magnetic field into zero-field-cooled samples, then subsequent depenetration.

Measurements for each sample were taken as follows: Calibrations were first carried out at high temperature with the sample and indicator film in place, using the procedure outlined in section 4.2.1. The sample was then cooled in zero field, so that the first video would start from the virgin state (see section 2.5.1). A penetration video was then captured as field was increased from zero to a set maximum field following the ramping curve shown in figure 4.2. This maximum field was maintained while the penetration video was processed and saved (generally a couple of minutes). A depenetration video was then captured as the applied field was decreased to zero following a similar ramping curve in reverse. For some samples, a re-penetration video was then captured for similar ramping to the same maximum field, but from the non-virgin state.

Current was then calculated for each frame in each video independently using the video current calculation program described in section 4.2.3.

Figure 4.12 shows typical field and current evolution in a zero-field-cooled sample during penetration and depenetration. Each field image is a single frame of magneto-optical video, which represents a “snapshot” of the magnetic field distribution integrated over the exposure time of the camera, which in this case was 1 ms. This is essentially equivalent to a magneto-optical image of the dynamic flux at a particular point in time.

Each current image in figure 4.12 is an independent visualisation of the current distribution calculated at one point in time. For each time-step, the maximum current calculated for that time is mapped to a grey scale value of 255 (white), and the minimum to a GSV of 0 (black). This means that brighter regions of the image represent higher relative current, but an overall brighter image does not imply higher absolute current than a darker image (in contrast to the field images, for which brighter images do represent a higher absolute field).

In the case of penetration, the maximum current is seen to increase over time as the external field is ramped, then relax to a steady state. This behaviour was investigated in detail, and novel features were observed, as described in the section 4.3.4.
CHAPTER 4. MAGNETO-OPTICAL IMAGING & HIGH-SPEED VIDEO

4.3.4 Dynamic Evolution of Current During Penetration

This experiment involved investigation of the time-variation of current in YBCO thin films, calculated from MO videos taken when the external field was quickly ramped up from zero to a stable value of 0.1 T. The focus was on the dynamics of transient current distributions during and immediately following the external field ramp (i.e. during field penetration). This represents a novel analysis of the evolution of supercurrents during penetration, and reveals several novel features including transient currents greater than the static critical current.

Samples were zero-field-cooled to 7 K, then MO video was captured as the magnetic field was ramped up to 0.1 T at a rate of (8.1 ± 2.3) T/s [41]. MOV was taken at 500 frames per second, and the current density was calculated at each time-step by the video current calculation procedure described in section 4.2.3.

It was found that while the external field was increasing, the transient current profile was dominated by large peaks, greatly diverging from static current profiles measured for many similar samples (see section 4.3.1), and from the profiles predicted by the Bean and Kim models (see section 2.5.2). These peaks develop close to the sample edges, initially resembling screening currents but quickly growing in intensity as the external field increases. As the external field stabilises, a discontinuity is seen in the field and current behaviour. This discontinuity was seen to occur at the point when the magnetic behaviour of the sample transitions from strongly peaked currents increasing with time, toward magnetic relaxation. At this point, the current peaks begin to migrate toward the sample centre, and their magnitude decreases with time. The speed of migration of the current peaks decreases with time in an exponential manner, and the shape of the current profiles approaches...
that predicted by the Kim critical-state model\(^a\) as the motion comes to a stop [65, 66, 148, 149]. Each of these transient features will be described in detail throughout this section.

The first significant feature is that during flux penetration, currents are seen that are larger than \(J_c\) for the static sample. The profile of these currents shows large peaks near the sample edges. Similar dynamic current distributions have been previously observed in simulations: firstly when pulsed fields were applied to bulk samples, where this field evolution was explained by heat propagation due to vortices quickly entering the sample [150, 151]; and secondly when an applied field was varied in a high-frequency sinusoidal manner [112].

Finally, these measurements are seen to be consistent with static magneto-optical images since the current profile after relaxation approaches a static profile similar to that predicted by the Kim critical-state model. Relaxation from the dynamic current distribution toward this static state occurs in an exponential manner.

**Results and Discussion**

The time evolution of field and current during the observed penetration event is shown in figure 4.13.

---

\(^a\)(see section 2.5.2)
One-dimensional profiles were extracted from the calculated current data at fixed times, at the position shown by the red line in this figure. This position was chosen since it lies in the area that does not become charged during penetration [152, 153] (see the discussion of validity of dynamic current calculations in section 2.7.1). A selection of these current profiles are plotted in figures 4.16, 4.17 and 4.18, showing the evolution of current in each of the three stages described below.

The edges of the sample are indicated as approximate in these figures, since the edges of the sample are hidden from view by the indicator film, and their exact position cannot be seen. Their positions can, however, be determined. The edges must lie at a distance less than $\lambda$ from the positions at which the first non-zero currents are seen, since these are screening currents induced when the external field is below $H_{c1}$. The effective penetration depth $\lambda$ is 180 nm, for YBCO films produced by the TFT group [141, 142] (see section 3.5).

![Figure 4.14](image)

**Figure 4.14:** Time-evolution of current density at several fixed points along the central line, along with the evolution of the peak value of current density (which also changes position over time). Each curve shows a sharp increase followed by relaxation toward a final stable value. The position of each point is indicated by “depth”, with 0 depth being the edge of the film, and 100% depth being the centre. Unexpected maxima are indicated by vertical lines, these show brief increases in current during relaxation.

Figure 4.14, shows the change in current over time at several points along the central line. The peak value of current is also plotted, note that the peak current is defined as the maximum current on the central line, and its position moves with
The position of the current peaks are plotted in figure 4.15, along with the position of the flux front, showing that the point of maximum current is always just behind the flux front. Velocity and acceleration were calculated in Origin by averaging the slope around each point, using a quadratic Savitzky-Golay filter with a 20-point window, and assuming that the position of each flux front and current peak is zero (at the sample edge) at all times before penetration begins.

The data in figures 4.14 and 4.15 is analysed in detail in the subsections below, where the discussion is split into three time-domains.

The reason for this split is that the evolution of current in the film was very different before and after a transition. The initial behaviour is labelled as the “ramping” stage, and final behaviour the “relaxation” stage, due to their respective timing in relation to the external field ramp. The “transitional” stage shows behaviour that has similarities to each of the other stages, and is marked by significant changes in field and current dynamics in the film. Each of the observed stages is described in greater detail in the following subsections.

The timing of each stage was determined precisely from the MOV data, with the zero time-point being defined as the first moment at which there was a measured increase in external field. The true beginning of penetration is probably slightly earlier, but the increase in brightness of the indicator film in response to the smallest initial field increase would be below the camera’s threshold.

**Ramping Stage**

During the first 14 ms, the external field was increasing, and most of the current in the sample was seen close to the edges. The time-variation of the current profile during this stage is shown in figure 4.16. The ramping stage can be identified as the initial period in figure 4.14, before the peak current has reached its maximum value.

As mentioned previously, the first currents in the film are Meissner screening currents, flowing during the brief period in which the external field is smaller than \( H_{c1} \). Over the next few milliseconds the external field increases, causing the screening currents to increase in magnitude. The current peak also moves inward by a small amount, and the current begins to flow in a region deeper than the penetration depth, showing that vortex entry has begun.

At 6 ms a long “tail” of current forms, beginning at the current peak and reaching right to the sample centre. This tail represents screening currents that begin to flow throughout the film in response to the horizontal component of the field. This horizontal component arises due to the high demagnetising factor of YBCO thin films [154] causing bending of the external field. The majority of the
Figure 4.15: (a) External field ramping curve (see figure 4.2). (b) Position, (c) Velocity and (d) Acceleration of the flux fronts and current peaks originating from the left and right edges of the sample. Position is measured relative to the closest sample edge, with the left front and peak offset by 0.2mm for clarity. The dashed lines indicate the approximate time of maximum acceleration, and of maximum velocity, averaged over all curves.

Film is still in the Meissner state at this point, with only the outside edges being penetrated up to a flux front just beyond the current peak.
Figure 4.16: Evolution of the current profile in a 3 mm YBCO film during the ramping stage, as field ramps from zero to 0.01T over (27 ± 3) ms. Vertical lines show the approximate positions of sample edges.

The upward curvature of current peak plots in figure 4.15 shows that the peaks move with inward acceleration during the ramping stage. Flux fronts also move inward, but their motion does not reflect that of the current peaks during this early stage.

**Relaxation Stage**

When the external field has reached a stable value, magnetic motion in the sample does not stop immediately, but experiences a slow relaxation. The relaxation stage began at ~ 30 ms after field ramping for these measurements [41]. The peak in current continues to move away from the edge of the sample and its speed decreases exponentially, as shown in figure 4.17. The current distribution as a whole approaches its final static profile with exponentially decreasing speed.

The changes in current distribution during this stage are driven by the inward motion of vortices. The vortices move in response to a large field gradient, but their motion is damped due to pinning.

During the relaxation stage, the position $p$ of the current peak is seen to move toward the centre of the sample. A shoulder forms close to the original peak position $p_0$, which is near the edge of the sample. A current plateau exists between the peak and shoulder, and it slopes downward toward the shoulder.

The current plateau can first be clearly distinguished 30 ms after external field
Figure 4.17: Current evolution in the YBCO film during the relaxation stage, when external field is stable and the sample approaches equilibrium. The small dip around 2.1 mm is due to a sample defect. Vertical lines show the approximate positions of sample edges.

switching, but at this time its gradient is very steep. The gradient of the plateau decreases with time up to about 80 ms, after which the shape of the plateau remains stable, but the current density throughout this region decreases with time. Only the left plateau could be analysed here, since the right side of the film contained a scratch, which is visible at the top right corner of the field and current frames in figure 4.12(a).

The position of the shoulder at the end of the current plateau remains fairly constant throughout this time, as does the sharp drop in current from the shoulder to the sample edge. The height of this shoulder decreases as the current peak moves away from it. The magnitude of current over the entire plateau region decreases over time throughout the relaxation stage.

The speed of the current peak decreases exponentially until it reaches its final position, as shown in figure 4.15. The height of this peak also decreases with time as seen in figure 4.14. The motion of each of the current peaks during the relaxation stage was fitted using an exponential decay equation:

$$ p = A e^{-t/\tau} + p_0 $$

$A$, $\tau$ and $p_0$ are fit parameters, and the time $t$ is taken from when the first non-zero current is seen in the film. Each of the fitting curves shown in figure 4.15(a)
have an $R^2$ value better than 0.996.\textsuperscript{b} The time constant $\tau$ indicates how quickly the current relaxes, and would probably increase for films with stronger pinning. In this case, $\tau$ was determined to be $(43.7 \pm 0.6)\text{ms}$ for the right peak and $(37.5 \pm 0.4)\text{ms}$ for the left peak. The difference in relaxation between these peaks was unexpected, but may originate from the scratch on the right side.

Comparison of the flux front and current peak motion in figure 4.15 reveals that the speed of the flux front also decreases exponentially, and the current peak is just behind the front at all times. The distance between the flux front and current peak varies over a small range throughout relaxation. This variation is different for the left and right sides of the sample, and no systematic variation could be found.

During the relaxation stage, the current density at each point relaxes over time toward a stable value, as seen in fig. 4.14. This value is taken to be the static $J_c$ for each point, although the relaxation is complicated by the fact that the field at each point also varies with time, which affects $J_c$. The initial period of current increase is longer for points closer to the centre due to the constant inward motion of the flux front. The largest measured reduction in current density is at the 10% depth, where the maximum transient current is almost twice the asymptotic stable value. This can be attributed to a local field increase at this point during relaxation.

The current relaxation behaviour is made clearer by considering the magnitude of the current peak, since the field near the flux front is always small and fairly constant. This magnitude has a decreasing trend with time, asymptotically approaching a value which is taken to be close to $J_{c0}$.

The relaxation of the current peak value was expected to be monotonic, but two “bumps” were unexpectedly observed in the graph. These bumps are reproduced at every position measured, with the current density noticeably increasing universally between 72 and 82 ms and between 90 and 116 ms. While these bumps may be due to variations in external field, they may also represent B-field oscillations due to the flux wave phenomenon, which was observed by Kalisky et al. [155] in the transient dynamic regime after switching on a magnetic field smaller than the order-disorder transition field (i.e. in the low-field liquid state). This transition field should indeed be much higher than 0.1 T for YBCO samples [156]. Importantly, the oscillations occur some time after the external field has stabilised both in this case and in Kalisky’s results [155]. The period of these oscillations is shorter than those seen by Kalisky [155], which is expected since the measurement temperature is lower.

\textsuperscript{b}$R^2$ is the coefficient of determination, and $R^2 = 1$ describes a perfect fit
Transitional Stage

It is clear from the above results that the evolution of current during penetration is vastly different during the relaxation stage as compared to the ramping stage, and that some transition in behaviour occurs between these two stages. However, the current evolution begins to show features of the relaxation stage before the external field has reached a stable value (i.e. while ramping is still occurring), and hence, a brief but finite transitional stage was defined. During the transitional stage, the evolution of current in the sample seems to show features of both the ramping and relaxation stages.

For the period between 18 and 26 ms after field switching, the current peak begins moving away from the sample edge and the magnitude of the peak no longer increases, as seen in figure 4.18. The shoulder near the sample edge also forms over these few milliseconds.

![Figure 4.18: Current evolution in the YBCO film during the brief transitional stage, when external field has not reached its maximum value but peak position begins to shift as in the relaxation stage. Vertical lines show the approximate positions of sample edges.](image)

Several explanations for this transitional behaviour are proposed:

- the instrumental fact that the external field does not simply ramp linearly to its maximum value, but instead ramps at its fastest rate for the first $\sim 16$ ms, before stabilising more slowly to its final value to avoid overshoot[41];

- an intrinsic transition in the film as it enters the critical state, where it responds
to field ramping not by increasing screening-like currents which peak at the film edge, but allowing an inward shifting of the peak current; or

- a combination of these external and intrinsic factors.

The notion of a finite transitional stage is supported by changes in the motion of current peaks and flux fronts during this time, as indicated in figure 4.15. Each current peak and flux front has a maximum in acceleration at a time close to 12 ms. Then, the velocity of each current peak and flux front reaches a maximum around 32 ms, after which all velocities decreases in the exponential manner described above. These maxima occur earlier for the flux fronts than for the current peaks in all cases. These two time-points of changing flux front and current peak behaviour are marked as dashed lines in fig. 4.15, and may be taken as rough indications of the beginning and end of the transitional stage. These times are reasonably consistent with the timing of transitional behaviour seen in figure 4.18, and with the stabilisation of external field.

Conclusion

High-speed magneto-optical video was used to investigate the dynamic behaviour of field and current in YBCO thin films under a time-varying external field. This led to an analysis of the different stages of evolution of current density during flux penetration with very good temporal resolution.

The experimental data showed a large peak in the current distribution very close to the sample edge which appeared within 2ms of the emergence of the first non-zero external field. This peak increases in magnitude during the time period that the external field is increasing (this is labelled the ‘ramping stage’). During this stage, the current profile deviates strongly from both the Bean and Kim models, and indeed from any plateau-like currents. These current peaks should not be confused with the un-physical peaks discussed in section 7.1, which are static and very different in origin.

The ramping stage ends with a transition in penetration behaviour (which was a novel observation during the course of this research). At this time, the current peak stops increasing in magnitude and begins to move toward the sample centre. This transition occurs as the external field stabilises to its maximum value, and is marked by changes in the motion of peak current position and flux front.

After this transition, the current distribution relaxes toward equilibrium; the peak in current moves toward a final position closer to the sample centre, slowing exponentially and gradually reducing in intensity. A plateau in current forms which is initially very steep, but the gradient reduces over time.
The final current distribution after relaxation has occurred qualitatively matches the profile predicted by the Kim critical-state model, as discussed in section 2.5.2. It is also quite similar to static current profiles seen in many imaging experiments in the literature [65, 66, 148, 149]. This distribution has increasing current density from the sample centre to a peak behind the flux front, a plateau with downward gradient reaches from this peak to a shoulder near the sample edge, at which point the current quickly drops to zero.

Therefore the accepted picture of the one-dimensional current distribution predicted by the Kim model can be seen as a limiting case that holds for static current distributions with a time-stable external field.

### 4.3.5 Validity of Dynamic Current Calculation

During the analysis of dynamic currents in YBCO films, it was natural to question the validity of an application of the Biot-Savart inversion procedure to a dynamic case. The dynamic current calculation procedure analyses magneto-optical videos frame-by-frame, but each frame of MOV is not truly independent from the others. A single frame of MOV differs from a static magneto-optical image in that the magnetic field distribution seen is not generally in an equilibrium state. Therefore, the validity of application of the Biot-Savart inversion procedure to dynamic measurements must be assured. An investigation was undertaken to show that these calculations are indeed valid for the specific dynamic measurements taken.

Both the Biot-Savart law and the current continuity equation are quasi-static - they do not take into account the time-variation of electromagnetic properties. The purpose of this investigation could therefore be refined to determining whether the quasistatic approximations would still be valid in the specific dynamic case of MOV measurements of these YBCO films using the magnetic field system at ISEM. Upon investigation, it was found that the effect of time-variations on the accuracy of current calculation was small enough to be neglected. This investigation was carried out as described below:

Firstly, the accuracy of the Biot-Savart law (equation 2.11) in this case was considered, since it is only analytically accurate for quasi-static current distributions. In vector form this equation is:

\[ \nabla \times \mathbf{B} = \mu_0 \mathbf{J} \]  \hspace{1cm} (4.3)

Comparing this to Maxwell’s equations, it can be seen that this equation holds when \( \mathbf{J} \gg \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t} \), which is true if electric field \( \mathbf{E} \) varies slowly with time [133].

To test whether this assumption holds, the rate of change of electric field must be determined. Brandt found that during penetration, an electric field exists only
in penetrated regions, and that this field is proportional to the rate of change of applied magnetic field [152]. Therefore, the maximum $\frac{\partial E}{\partial t}$ can be determined by calculating the largest second derivative of the field ramping curve (figure 4.2).

In this way, the maximum value for $\epsilon_0 \frac{\partial E}{\partial t}$ during field ramping was found to be $2.55 \times 10^{-5}$ A/m$^2$, for the fastest field ramping considered (of course this quantity is zero for a static external field). This is truly negligible compared to the current density in the penetrated region, which is seen to exceed $J = 10^9$ A/m$^2$ within 2 ms in all measurements. The stated assumption therefore hold, and the Biot-Savart law is valid for the specific dynamic measurements considered in this research.

The second assumption made in the calculation procedure is that current is continuous, as expressed in equation 2.15. The more general charge continuity equation takes the form:

$$\nabla \cdot J + \frac{\partial \rho}{\partial t} = 0$$

(4.4)

as previously given in equation 2.14. Since the film is uncharged in general in the static case, the assumption is that the film does not acquire any charge over time: $\frac{\partial \rho}{\partial t} = 0$.

Investigations into the charge on superconducting films during penetration have shown that square regions near the sample corners do in fact acquire a charge proportional to the rate of external field ramping [152, 153], which may invalidate equation 2.15 in parts of the film, causing slight inaccuracy in the calculated current in these areas.

However, the area around a line through the middle of the film bisecting each edge remains uncharged throughout penetration, as discussed in [152]. The accuracy of calculations can therefore be guaranteed only for this line. The central line was therefore taken as the position of one-dimensional current profiles for all analyses, as shown in figure 4.13.

4.3.6 Vortex Velocity during Flux Penetration

The velocity of vortices during flux penetration was measured from the MOV results of section 4.3.4, and compared to a theoretical calculation based on the speed of field ramping. It was found that the results were numerically consistent with the theoretical model, although the observed changes in behaviour over time are not predicted by the model.

Flux fronts with constant velocity of the order of 100 m/s have been theoretically predicted for superconducting samples with field very quickly removed [157]. Similar speeds may also be expected for field quickly applied to a sample similar to the films investigated in this research, but the speeds actually observed are several orders of
magnitude lower. This is due to the dependence of vortex propagation speed on the finite magnetic field ramp rate [158].

The theoretical calculation of vortex velocity during field ramping was derived by Schuster et al. using the Bean model [158]. Following this derivation, the inward speed of vortices at a position $x$ is approximately given by:

$$v(x) = \frac{2\pi B_a}{\mu_0 J_c} \frac{\sqrt{x^2 - f^2}}{\ln|(1 - u)/(1 + u)|}$$

(4.5)

Where $f$ is the flux front position, $a$ is the sample half-width and $u = x\sqrt{a^2 - f^2}/a\sqrt{x^2 - f^2}$. This equation is valid throughout the penetrated region ($f \leq x \leq a$), taking $x$ positive for simplicity. All positions are measured relative to the sample centre for this calculation, as shown in figure 4.19, since this simplifies the expressions. This differs from the positions of flux front and current peak in figure 4.15, which are measured relative to the closest edge.

Equation 4.5 predicts that the inward speed of vortices during penetration should be fastest close to the flux front ($x \to f$), but quickly drops to zero for $x \geq f$ since by definition no vortices are present beyond the flux front. There is no predicted time dependence of flux front penetration speed in this simple Bean model calculation.

![Figure 4.19](image)

**Figure 4.19:** (a) Field image showing the physical position of the sample centre ($x = 0$), the flux front ($x = f$) and the sample edge ($x = a$). These quantities are used for the velocity calculation. (b) Current image showing the position of the current peak ($x = p$).

This expression was calculated using parameters relevant to the experiment:

- $B_a = 8.1 \text{T/s}$ (maximum ramp rate)
- $J_c = 10^{12} \text{A/m}^2$ (this is a typical value for high-quality films at 10K, see section 4.3.7)
- $a = 1.5 \text{mm}$ (since the sample is a 3mm square)
- $f \approx 0.95a$ (flux front penetrated to 5% depth, which is appropriate for the timing
of maximum ramp rate by comparison of figures 4.2 and 4.15) and \( x \simeq 0.951a \) (position just behind the flux front, where vortex velocities are highest)

Inputting these parameters gives a vortex velocity of approximately 0.008 mm/s. This calculation is robust to small variations of \( f \) and \( x \).

In the experimental case, the magnetic flux front is easily identified visually in magneto-optical videos as the line separating flux-filled regions of the sample near its edges and the flux-free region near the centre. This line tends to move inward from the edges during penetration, but avoid the discontinuity lines, as discussed in section 2.5.1.

For computation, the position of the magnetic flux front was defined to be the furthest point (pixel) from the sample edge with field above a threshold value \( B_t \) (chosen to be above the noise level in the flux-free region, but below 10% of the maximum field). The computational determination of flux front position ensured that there was an adjacent point (pixel) with field below \( B_t \), and the flux front position was determined independently for each time step. Generally, the calculated flux front depth increased smoothly, and closely matched what was seen in the video. It should also be noted that all samples had a low sensitivity to changes in the \( B_t \) value chosen, since field increases smoothly from zero at the flux front.

The flux front position, velocity and acceleration is shown as calculated from a typical magneto-optical video in figure 4.15. The penetration speeds observed in the ramping stage ranged from 0.01 to 0.02 mm/s as indicated in figure 4.15(b). The magnitude of this experimental value matches closely with that given by the model.

The quantitative difference between the theoretical and experimental values for vortex velocity may arise from the non-Bean-like behaviour and the transitional effects observed during penetration, as discussed in section 4.3.4. There is clearly also a qualitative difference, as the measured flux front velocity has a strong time dependence, while the theoretical model has none.

### 4.3.7 Analysis of Current-Carrying Properties by MOV

This investigation examined the differences in flux front penetration and current evolution in high quality \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) thin films compared to samples that were deliberately reduced in quality. The reduced-quality samples were damaged in such a way as to reduce critical current density without causing macroscopic damage, and as such would not have been easily identified as poor quality samples under conventional MOI.

While conventional magneto-optical imaging is useful for visual identification of large current-limiting defects such as scratches (as discussed in section 2.6.1), other
reductions in current-carrying ability are not so readily visible at the macroscopic scale. In some cases, these limitations to current flow also cannot be identified through simple measurements of the critical current density [141, 142].

High-speed magneto-optical video may be used to identify such limitations. This investigation aimed to discover new hallmarks of reduced sample quality that are visible in dynamic field and current data. To achieve this, the time-evolution of the magnetic flux and current profiles and the speed of penetration were examined in films with vastly different $J_c$ values.

**Experimental Method**

YBa$_2$Cu$_3$O$_{7-\delta}$ films were deposited by pulsed laser deposition on 5 × 5 mm substrates of SrTiO$_3$, by the method described in section 3.1.

Samples were measured with MPMS after deposition, and only high quality samples were selected for patterning as described in section 3.4.1. The samples were etched into 3 × 3 mm squares by the process described in section 3.2.

While sample patterning was usually carried out under the optimal conditions stated in section 3.3, for this investigation the quality of a group of samples was reduced in a controlled way. These samples were exposed to the ion beam for three times the average etch duration in order to allow a greater number of high-energy ions to damage the protected parts of the sample. They were then exposed to acetone for several minutes. Each of these damaging processes represents a realistic mechanism by which samples may become damaged in the lab, and which would not be detected under static MOI. Samples damaged in this manner were referred to as “over-processed”.

After patterning, the critical current was measured using MPMS, (see section 3.4.1), in order to confirm reduction of sample quality and current-carrying ability in the over-processed samples compared to the control group.

Penetration into each of the samples was then investigated using MOV. Magnetic field was applied from zero to 0.1T with a maximum ramping speed of (8.1 ± 2.3) T/s as shown in figure 4.2.

For simplicity, time-dependant current profiles over a one-dimensional slice through the sample were considered for analysis, in favour of the two-dimensional currents over the entire sample.

**Results and discussion**

The critical current density ($J_c$) was tested independently for each sample using MPMS at 10 K, as shown in figure 4.20. This initial check shows strong suppression of the critical current density in the over-processed samples by a factor of nearly $10^5$
at low fields and complete \( J_c \) suppression at applied fields \( B_a > 1 \) T.

![Figure 4.20](image)

**Figure 4.20:** Critical current density as a function of applied field at 10 K in (a) a high-quality sample, and (b) an over-processed sample, where low-field \( J_c \) is seen to have been reduced by 5 orders of magnitude.

\( T_c \) was measured to be > 90K for the high quality sample. Critical temperature could not be accurately determined for the over-processed sample due to low MPMS signal strength, but significant reduction in \( T_c \) could be assumed.

Magneto-optical video showing the dynamic behaviour of field and current in each sample was captured during and immediately after an increase of external field from zero. A selection of frames from typical magneto-optical videos of flux penetration are shown in figure 4.21 for high-quality and over-processed samples. This figure also shows the current in the samples as calculated from the field distribution at each time step. Magneto-optical videos appear visibly similar for both types of films. This similarity confirms that both films remain superconducting, without any mechanical damage to the sample surfaces. However, further investigation yields important qualitative and quantitative differences in the distribution of field and current in these films.

The first quantitative comparison between high-quality and over-processed samples involves measuring the penetration speed into each using the motion of the flux fronts, as shown in figure 4.22.

The position of the flux front (determined as described in section 4.3.6) was plotted against time for each sample in figure 4.22, using field values from magneto-optical video along the one-dimensional slices shown in figure 4.21. A horizontal slice was chosen for the high-quality sample, and a vertical slice for the reduced-quality sample in order to best avoid the defects in each.

Figure 4.22(a) shows motion of the flux front in the same high-quality sample as shown in figure 4.15. It would be possible to differentiate this curve and hence plot the velocity and acceleration of the flux fronts over time for this and other high quality samples. However, the position of the flux front in the reduced-quality
Figure 4.21: Timeline of magnetic field during penetration in high-quality and reduced-quality thin film YBCO samples, as obtained using high-speed magneto-optical video.

sample, is not differentiable, as seen in figure 4.22(b). It therefore does not produce smooth velocity and acceleration curves. This in itself is an interesting observation, and provides one simple way of identifying a lower quality sample.

It is, however, desirable to compare the velocities of flux front penetration between the high- and low-quality samples. For this purpose, a linear fit was taken over a range of approximately 50 ms in the increasing part of each penetration curve. From this linear fit, the average flux front penetration speed for the high-quality sample was found to be 12.3 ± 0.2 mm/s for the front on the left side of the sample and 14.1 ± 0.1 mm/s for the front on the right. This is consistent with the more accurate analysis taken in section 4.3.4.

From the consideration of vortex velocities in section 4.3.6, the speed of vortices near the flux front is predicted to increase as critical current in the film decreases [158].

It was therefore surprising that flux is apparently seen to penetrate more slowly into the reduced-quality sample in figure 4.22(b), with average penetration speeds being 7.0 ± 0.3 mm/s for the front at the top of the sample and 6.9 ± 0.2 mm/s for the front at the bottom. For this sample, the penetration speed does not decrease smoothly, but instead appears relatively linear (but with substantial noise) until the final penetrated position is reached. In this regard, the damaged sample more closely reflects the theoretical model [158].

Two horizontal lines are also seen in figure 4.22(b), where the flux front seems to pause before continuing to increase: below 0.8 mm for the top front, and below
Figure 4.22: Position of the flux front over time during penetration in (a) a high-quality sample, and (b) a reduced-quality sample. Flux front position was determined from magneto-optical video by finding the furthest point from the film edge with field above a threshold value for each time step. The zero position represents the edge of the measured area. The gradient of these plots gives the average penetration speed for each sample.

1.2 mm for the bottom front. These positions are associated with visible defects in the sample or indicator film.

The unexpected slower flux penetration into the reduced quality sample is explained by noting the discontinuity in the plot of figure 4.22(b) at approximately 5 ms. This discontinuity represents the time at which the field first penetrates into the sample, and hence the flux front seems to “begin” penetration around 0.6 mm for the top front, and 0.9 mm for the bottom front.

However, this position is actually some distance from the sample edge. This “initial” flux front position inside the sample in fact shows a much faster response to the field change, which is too fast to be resolved with the temporal resolution at this frame rate (and must therefore occur over a time less than ~ 1 ms). The subsequent (measured) flux speed of ~ 7 mm/s, may then be the result of reduced demagnetising fields after the initial very fast penetration. This is confirmed by analysing the magneto-optical video directly - a very fast “jump” in penetration is
observed when the field is first switched on. This fast penetration is presumably along the damaged domain boundaries where superconductivity is weakened, and then the slower penetration may start as vortices begin to enter the domains and the front is therefore pushed forward more slowly.

A second comparison between the samples was in the time-variation of current density in a one-dimensional slice for each film, as shown in figure 4.23. A difference is immediately visible in the behaviour of current density during field penetration in the reduced-quality sample.

![Figure 4.23](image)

**Figure 4.23:** Time-evolution of current in (a) the high-quality sample, and (b) the reduced-quality sample, as calculated frame-by-frame from magneto-optical video during flux penetration. Current density is plotted in arbitrary units.

Figure 4.23(a) shows the time-evolution of current in a one-dimensional slice through a high-quality square YBCO film, as detailed in section 4.3.4: A small peak forms at each sample edge within 2 ms of penetration, which increases in magnitude without significant change in position until about 20 ms. As this current-
peak increases, a long current-tail extends toward the sample centre. After this, relaxation behaviour can be seen as the current peak moves toward the centre of the sample while no longer increasing in magnitude. A current plateau forms between this peak and the sample edge, in the penetrated region, with a downward gradient toward the sample edge, which decreases over time, until the current profile settles to a distribution resembling the Kim critical-state model profile after about 200 ms. The current distribution is quite symmetric about the centre of the sample at all times.

The evolution of the current profile in the reduced-quality sample is quite different, as seen in figure 4.23(b). The peak in current forms at the sample edge as expected, and grows in magnitude during field ramping, though the shape of the current distribution has already deviated after 5 ms: there is no visible tail extending toward the centre (this may simply be because the small current tail is indistinguishable from the background noise and interpreted as zero). After the field has stabilised, the deviations become more drastic, with the peak in current not moving from the sample edge at all. The current distribution still spreads toward the sample centre as flux penetrates further inwards, but has a rounded shape with downward concavity, rather than a sharp peak at the flux front. Minimal currents are visualised in the non-penetrated region at all times for this sample. The current can still be interpreted as having a plateau in the penetrated region, but with a reversed gradient that is much steeper. After 100 ms, as flux has penetrated almost to the sample centre, the current plateau becomes much less smooth. Perturbations in the plateau appear as a series of smaller peaks that do not change position over time and hence look like long ridges in figure 4.23(b). These peaks are not symmetrically distributed about the centre, suggesting that these are caused by localised current-limiting defects that were not immediately visible in the magneto-optical video.

The final current distribution in the reduced-quality sample deviates strongly from both Bean- and Kim-like profiles described in section 2.5.2, with the current plateau having a gradient sloping toward the sample centre and peak current at the edge. The smaller peaks attributed to defects also form a major feature of the final current distribution.

For both samples, the magnitude of current in all parts of the film decreases slightly in the longer time-scale up to 500 ms due to relaxation processes, but the distribution retains approximately the same shape. This is not shown in figure 4.23 in order to emphasise the more significant features occurring in the shorter time scale. It is only at this later time that non-zero currents are seen close to the centre of the reduced-quality sample.
Conclusion

This investigation employed magneto-optical imaging in a dynamic and quantitative manner in order to characterise YBCO films with very different current-carrying abilities. A viable technique was developed for the identification of poor quality samples with defects that are not immediately apparent under MOI.

While vortex velocity was expected to increase with a reduced critical current [158], the inward speed of vortices at the flux front appeared to decrease in the damaged samples. Upon further investigation, it was shown that initial flux penetration in the damaged films was probably in fact much faster. However, this initial penetration occurs so quickly that the time resolution of imaging used in this work was too low to resolve the initial fast flux penetration in the damaged samples. Future studies may focus on analysing the high-speed penetration into similar damaged samples with a higher temporal resolution. It would be interesting to determine whether the initial ramping behaviour of the much faster penetration in damaged samples was similar to that seen in the early ramping stage for high-quality samples (as discussed in section 4.3.4).

The suppression of the macroscopic critical current density in the reduced-quality sample as measured by MPMS is seen as a major influence on the dynamic evolution of current in the damaged sample. And conversely, dynamic MOI has been shown to be useful for probing current evolution on a local scale in order to explain the dramatic reduction in overall critical current that was seen with MPMS.

4.3.8 Simulation of MOV Flux and Current Dynamics

A simulation process was carried out [159], for comparison of simulated field and current behaviour during ramping and relaxation to that which was observed experimentally, as presented in section 4.3.4.

The simulation considered the diffusive motion of vortices in relation to magnetic field, current, temperature and pinning across a one-dimensional superconducting strip. Diffusive motion of vortices occurred via vortex hopping using the Kim-Anderson model [69]. The pinning potential was given a hyperbolic field dependence in order to approximate realistic pinning as measured in previous work [70, 143, 160].

The external field was applied in a manner resembling an idealised version of the field ramping used for MOV: First, magnetic field was ramped from zero at a constant rate, to a maximum value that is reached after a simulation time of 600 µs (stated as $j = 300$, where $j$ is the 2 µs time step). For $j > 300$, the magnetic field was held at a constant maximum value, while the vortex diffusion process was allowed to continue.
Simulation Results

The simulation results are given in figure 4.24. This figure shows the variation of magnetisation in the film over time, and plots the current density profiles across the film to match the similar plots obtained from magneto-optical video in section 4.3.4.

![Simulation Results](image)

Figure 4.24: Results of the simulation: (a) Total magnetisation of the sample over time, during and after field ramping, the inset shows the same quantity over a larger domain. Current density profiles (b) during field ramping and (c) during relaxation.

Reproduced from [159], courtesy of A. V. Pan.

Note that characteristic peaks are observed on current profiles before full penetration of magnetic field is reached - in figure 4.24(b) and in (c) for $j < 500$. The peaks propagate inward together with the flux front, and then reduce in magnitude after full penetration is reached.

The latter part of the magnetization curve in figure 4.24(a), for $j > 300$, shows relaxation of magnetization. In the long time limit ($j > 500$), magnetization is seen to relax with the expected logarithmic time dependence [9]. For $300 < j < 500$, a non-logarithmic relaxation is observed.

The simulation results for current profiles during and following field ramping match very well with those found by magneto-optical video experiments, as shown in figures 4.16-4.18.
In the experimental results, a strong peak is seen in the distribution of current density at all times during penetration, most notably during the first 20 ms, when this peak dominates the current distribution. This peak value may in fact be described as an overcritical current which quickly spikes as field enters the film and then slowly drops down to a stable value over time. A very similar peak is seen in the simulation, as mentioned above, with the shape of the plateau as flux penetrates inwards being also very similar.

The simulated pinning potential giving rise to this current distribution is hyperbolic. Compare this to the hyperbolic field dependence of critical current given by the Kim critical-state model [65, 66]. The field dependences of both models are similar in form, which explains the similarity of the shape of simulated $J(x)$ curves to the predicted Kim critical-state profile.

Since this pinning potential is taken to be quite realistic as confirmed in other works [70, 143, 160], this provides a strong explanation for the shape of dynamic current and field profiles during simulation.

The field profiles obtained by simulation were also very similar to those observed by MOV (not shown here for brevity): In both cases, flux fronts penetrate inward at a position that slightly leads the current peak. One major difference is that the simulated flux fronts actually reach the sample centre, thereby achieving full penetration, where the experimentally observed flux fronts do not. This shows that the pinning in the experimental case must be stronger than that which was simulated.
Chapter 5

Scanning SQUID Microscopy of Vortex Glass

This chapter describes a multi-part experimental study of the distribution of vortices in a very-low-field glass phase. This study was carried out in collaboration with the University of Twente, where scanning SQUID microscopy (SSM) was used to examine the vortex distribution in field-cooled YBa$_2$Cu$_3$O$_{7-\delta}$ films, which had been produced by PLD at the University of Wollongong as described in section 3.1. [36, 37]

5.1 Scanning SQUID Apparatus at UTwente

Images of the magnetic field above a superconducting sample were taken by scanning Superconducting QUantum Interference Device (SQUID) microscopy.

The scanning SQUID microscope at the University of Twente consists of a $\mu$-SQUID magnetometer with a $3\ \mu m \times 5\ \mu m$ pickup loop which is raster scanned above the sample surface at a distance of approximately $5\ \mu m$, at approximately $30^\circ$ to the surface [82].

The sample and SQUID were fully immersed in liquid helium (LHe) for all measurements, using a helium bath cryostat. The sample is placed at the end of a vertical finger with the SQUID below, as shown in figure 5.1. The lower part of the finger can be inserted into the liquid helium.

Each sample to be scanned is mounted onto a small holder, which has electrical contacts for if current is to be applied to the sample. The sample holder is then mounted onto the bottom end of a long rod that extends upward through the centre of the vertical finger. Three stepper motors at the top of the finger control the $x,y$ and $z$ motion of the sample by applying force to the top of this rod. The sample position can be adjusted manually using the motion controller pictured, or
Figure 5.1: Labelled photograph of the scanning SQUID microscopy apparatus, showing the electronic control stack on the left and functional part of the apparatus on the right. The solenoid is placed over the end of the finger before insertion into the cryostat.

programmatically as during SSM data acquisition.

The SQUID sensor is mounted on the end of a cantilever that remains strong and flexible at low temperatures. This cantilever is firmly connected to the base plate at the bottom of the vertical finger, at an angle of $30^\circ$. The plane of the SQUID loop therefore makes this same angle with the plane of the sample when the cantilever is straight. The cantilever has a strain sensor attached, which is wired to the strain gauge pictured.

Field could be applied to samples using a purpose-built solenoid that can be attached to the vertical finger, surrounding the entire sample space including the sample and SQUID. This solenoid had been calibrated to produce a highly uniform field over the entire sample space. For figure 5.1, the solenoid was unmounted and placed to the side to allow the sample holder and SQUID sensor to be seen.

Once both sample and SQUID are mounted, and the solenoid is secured around them, the vertical finger is slowly lowered into the helium bath cryostat. The finger is generally lowered to such a point that the entire sample space and solenoid are immersed in LHe, but not too much lower as this would unnecessarily boil off the
The sample is lowered toward the SQUID using the $z$ stepper motor, but only once the finger has remained in its final position for a few minutes. This is to ensure that all vibrations have ceased, to protect the sample surface and the pickup loop, and to ensure that both the sample and SQUID have reached a stable temperature of 4.2 K. The strain gauge is used to determine when the SQUID comes into contact with the sample, as this causes a slight bending of the cantilever. It is important to ensure that the scan is taken at a height that is as close to the sample as possible while not being in contact.

Once the sample is at the correct $z$-height, the scanning process can begin. The sample is raster scanned using the $x$ and $y$ stepper motors, and voltage data continuously read from the SQUID. This voltage is recorded and then converted to a magnetic field value for each position of the SQUID relative to the sample surface.

### 5.1.1 Magnetic Shielding

The primary magnetic shielding for the SSM is a coil of wire wrapped around the outside of the helium bath cryostat, as visible in figure 5.1. A background field much smaller than the Earth’s field was found to persist within this shield.

Previous measurements by other members of the ICE group using a Bartington Mag-03MS three-axis magnetic field sensor found the vertical component of this background field to be approximately $2 \mu$T. The other field components were also measured and found to be negligibly small compared to the vertical component. Another measurement found the root-mean-square variation of the background field to be less than $30 \text{nT}$, using scanning SQUID microscopy of antiferromagnetic samples.

A further investigation determined the value of the background field within the shielded environment of the scanning SQUID microscope using SSM measurements of vortices in a superconducting sample. This investigation is described in section 5.3.1, and it gave the most precise value for the background field as $2.73 \mu$T in the vertical direction.

A $\mu$-metal shield with an approximate shielding factor of 25 could be placed around the SQUID and sample region, attached to the end of the vertical finger. This shield served to further reduce the background field in order to minimise its influence on the SQUID during cooling and scanning. However, the $\mu$-metal shield was not used for the measurements relating to this research, since the shield and solenoid could not be mounted at the same time.
5.1.2 Preparation of SQUID tips

The pickup loops of SQUIDs were occasionally damaged by mechanical scraping against the sample surface during scanning or positioning before a scan. Replacement SQUID devices were bulk ordered for use in the scanning SQUID microscope. Each SQUID was mounted on a silicon chip, which contained contacts for voltage, current and modulation on one half of the chip along with the SQUID loop containing two Josephson junctions. Two long thin superconducting wires extended across the chip to a $3 \, \mu m \times 5 \, \mu m$ pickup loop close to the other side. The geometry of the SQUID is given in figure 2.10(b), though this is not to scale.

The SQUIDs had been produced with the pickup loop approximately $500 \, \mu m$ from the edge of the chip, but for optimal imaging, and reduction of stray fields (as discussed in section 2.7.2), the distance from the pickup loop to the sample must be minimised. Therefore it was necessary to cut away the edge of the SQUID chip, to obtain the smallest distance possible from the loop to the edge. This would therefore ensure that the pickup loop could be scanned as close to the sample as possible without making contact. The edge of the chip was also rounded to ensure that no sharp corners could catch on the sample, and that the pickup loop would always be the closest point to the sample surface.

![Figure 5.2: Optical microscope images of the pickup loop at the end of the SQUID chip, taken (a) before cutting and (b) after cutting the chip to shape.](image)

The edge of the silicon chip was first removed roughly using a grinding disk, to reduce the distance from the loop to the edge and to round the corners. Sandpaper was then used to finely reduce the distance from the edge to the pickup loop. It was very common to damage the pickup loop during this process, and as such, the quality of cutting was checked many times under an optical microscope while grinding, and the sanding of the chip was entirely carried out under the microscope. Figure 5.2 shows a pickup loop at the end of a SQUID chip before and after the cutting process.

Any SQUID chips whose pickup loop or wiring was damaged during the cutting process were immediately discarded.
The completed SQUID chip was then glued onto the tip of the cantilever, after removal of the previous SQUID. The voltage, current and modulation contacts were then wire bonded to the appropriate contacts on the cantilever. The cantilever could then be reattached to the base of the insertion finger for scanning, and wires from the SQUID control module were soldered to the cantilever contacts in order to receive data from the SQUID.

5.1.3 Scanning SQUID Microscopy Procedure

The scanning SQUID microscopy procedure begins with the mounting of a sample onto the sample holder, which is then firmly mounted in position on the end of the mobile rod of the insertion finger, as shown in figure 5.1.

Provided that a working SQUID is in place (see above), the solenoid is placed around the end of the finger and fixed in place using three plastic screws at the top. The insertion finger is then lowered slowly into the helium bath cryostat. Full immersion of both the sample and SQUID is determined from changes in the SQUID and strain gauge signals. After waiting a few minutes for all vibrations to cease, the sample is lowered toward the SQUID using the $z$ stepper motor while carefully monitoring the strain gauge to avoid contact.

Using the SSM control software, the sample is raster scanned and a map of the voltage output from the SQUID at each point is produced. This scanning process can take several hours depending on the scan parameters chosen, and must be monitored at regular intervals to ensure that valid data is still being obtained.

Data may become invalid if the voltage suddenly changes due to the occurrence of a flux jump in the SQUID, as described in section 2.6.4. If this occurs, the scan is stopped, and it must be decided whether to use the data that has been obtained (resulting in a partial-size scan), or to discard it and repeat the scan from the beginning.

The raw SQUID data is passed to the analysis computer, and interpreted using the image acquisition program described in section 5.2.1. A quantified field image is then output.

Imaging artefacts are removed using an SPM image correction tool. This corrects for shading, line-to-line artefacts, and other common artefacts occurring in scanning probe techniques.

The current is calculated from the magnetic field data, and autocorrelation and Delaunay triangulation are then applied using the appropriate programs described in section 5.2.2.
5.2 SSM Image Analysis Software

New software was designed to acquire images using the scanning SQUID apparatus, which was much more efficient and modern than the existing SSM image acquisition program. Programs were also written to calculate current and to characterise the vortex distributions observed in SSM images.

5.2.1 Image Generation and Current Calculation Programs

The image generation program made for this research was based on a heavily modified version of the image acquisition program for MOI. This program accepted SQUID data from the SSM control software, and processed it to provide images in a usable form. A quantification step was included directly in the program, which converted raw voltage data from the SSM control program to magnetic field values for each point above the sample. This quantification step was different to that used for MOI, in that the field could be determined directly from voltage output without requiring calibration.

Finally, images were outputted in a format that could be used by the current calculation and phase characterisation programs, without requiring further conversion. This program also allowed for the determination of applied field $B_a$ from the current applied to the solenoid, and recorded this data along with other scan parameters with each image.

An external program was still required to remove artefacts from the SSM images such as line-to-line artefacts.

The current calculation program was similar to that described in section 4.2.2, but altered to suit the specific parameters of the scanning SQUID microscopy system at the University of Twente. This included the use of a smaller scan height $h$ and other apparatus-specific parameters. In addition, the option for calculation of in-plane fields was removed, as this is specific to MOI.

5.2.2 Vortex Phase Characterisation

A program was created to analyse the distribution of vortices in SSM images. More specifically, it was able to apply both autocorrelation and Delaunay triangulation to SSM images of vortex distributions, as explained below.

For magnetic microscopy techniques such as SSM with resolution on the scale of a few hundred nanometres or better, the locations of individual vortices are easily observed from the magnetic field data. On this scale, the analysis of magnetic field

---

*These artefacts were introduced in section 2.6.4.*
information is no longer focussed on the macroscopic average over many vortices, but instead on the arrangement of vortices including distances between vortices and their neighbours, and the angular separation of these neighbours. Quantitative information about the phase and order of the vortex distribution was determined using the Delaunay triangulation and autocorrelation techniques.

Autocorrelation and Delaunay triangulation techniques have been previously applied to the scanning SQUID images of vortices in a-MoGe superconducting thin films with relatively weak pinning, to quantify deviations from the vortex lattice (topological defects) around pinning sites [161].

Each of these techniques is described in detail below.

5.2.3 Autocorrelation

A spatial autocorrelation $AC(r)$ for some displacement $r$ maps the average correlation of every point in a set of data to the corresponding point separated by this displacement. When applied to magnetic field maps, the autocorrelation value for some displacement $r$ gives the probability that two points separated by $r$ have the same field value. The autocorrelation for a magnetic field map is given by:

$$AC(r) = \int B(r)B(r + r')dr'$$

(5.1)

The integral in equation 5.1 is over the image area. When such field maps show a vortex distribution, the value of the autocorrelation at $r$ is used to give an indication of the number of vortices existing with separation $r$ in a given direction.

This can be used to show the regularity of the vortex distribution, and hence to infer the phase of the vortex matter. The existence of any strong peaks in the autocorrelation implies that one (or several) vortex spacings occur more frequently than others, which shows spatial order in the lattice. The existence of any peaks at large spacings indicates that this order is long range, which is only seen in the lattice phase.

The phase characterisation program allows both one-dimensional (horizontal and vertical) and two-dimensional autocorrelations to be applied to images, in order to give an indication of the regularity of vortex spacings.

The results of application of the one-dimensional autocorrelation procedure to a model image is given in figure 5.3. A two-dimensional autocorrelation calculated from the same model image is shown in figure 5.7, where it is used for comparison with an autocorrelation taken from experimental data.
CHAPTER 5. SCANNING SQUID MICROSCOPY OF VORTEX GLASS

5.2.4 Delaunay Triangulation

Delaunay triangulation is a mathematical procedure mapping the distribution of vortices in space to a network of non-overlapping triangles. Each triangle has a vortex at each corner and has edges connecting nearest-neighbouring vortices, such that no vortex lies within the circumcircle of any triangle.

This gives a precise definition to the intuitive notion of nearest neighbours: A nearest neighbour to a vortex can be defined as any other vortex that is connected in the same triangle using the Delaunay triangulation algorithm. To put it more simply, any three vortices through which a circle can be drawn which encompasses no other vortices are defined to be nearest-neighbours. The number of nearest neighbours to a vortex can therefore be computationally determined using the Delaunay triangulation. This was important for identification of topological defect, as described in section 2.3.2, and for calculation of the hexatic order parameter, as described in the next subsection.

The internal angle of a triangle in the Delaunay triangulation is defined as the “bond angle” between the vortex at that corner and the two neighbours making up the triangle. This angle describes the angular separation of the neighbours with respect to a chosen vortex.

The bond angle can alternatively be defined as the angle of the line connecting a vortex to just one of its nearest neighbours, relative to a fixed line (such as the x-axis of the image). The use of a fixed reference line aids in the calculation of the hexatic order parameter.

For vortices at the edge of the image, both the number of nearest neighbours and the bond angle may be inaccurate. This is because some of the neighbours to these vortices may physically lie outside of the field of view. Therefore, the concept of “internal vortices” was introduced. All vortices at the edges of the measured...
area and/or at the edge of the sample were considered to be external, as were their nearest neighbours. All other vortices were considered to be internal. This is shown diagrammatically for the experimental data in figure 5.8(b).

The procedure undertaken by the program to obtain the Delaunay triangulation was as follows:

The program begins by applying a particle detection algorithm to determine the positions of vortices in SSM images, which appear as isolated bright or dark spots in the image (corresponding to the higher field in vortices). An image would then be produced of the detected particle positions. This method did not always accurately determine vortex positions, meaning that images had to be visually checked for correlation with the actual positions of vortices from the original SSM image. The parameters of the particle detection algorithm could then be varied in order to produce an optimised image that accurately determined the position of vortices. This optimisation is discussed in relation to the experiment in section 5.3.3.

The centre of each particle was taken as the vortex position, and these co-ordinates were output to give an array of spatially distributed vortex locations. A Delaunay triangulation could then be applied to the co-ordinates of vortex positions.

From this triangulation the number of nearest neighbours to each vortex was determined for all internal vortices, along with the bond angle for each of these neighbours.

As discussed in section 5.2.2, internal vortices are defined as all vortices excluding those at the edges of the measured area or at the sample edge, as well as their nearest neighbours. Vortices at the edges of the image or the sample edge could be easily identified in the program, since only these vortices had a number of nearest neighbours that was not equal to the number of triangles in which that vortex was present. The reason for this is that only vortices at the edges could have bond angles between any two of its neighbours greater than \( \pi \) radians. Whenever a bond angle was greater than \( \pi \), a triangle would not be produced on one side of that vortex. Therefore, the number of triangles for edge vortices would always be smaller than the number of triangles. Since this method only identifies vortices at the very edge, their nearest neighbours were also excluded in order to make sure the edge effects were truly removed.

Finally, from the information made available by Delaunay triangulation, the hexatic order parameter (HOP) (as defined in the following subsection) could be quickly calculated for each vortex distribution.
Hexatic Order Parameter

The hexatic order parameter is given by $|\psi_6|^2$, where $\psi_6$ is defined by equation 5.2:

$$\psi_6 = \frac{1}{N_{\text{int}}} \sum_i^{N_{\text{int}}} \frac{1}{n(i)} \sum_j^{n(i)} e^{6i\theta_{ij}}$$

(5.2)

Here $N_{\text{int}}$ is the number of internal vortices (excluding vortices at the edges, see figure 5.8) and $n(i)$ is the number of nearest neighbours to a vortex $i$. The bond angle $\theta_{ij}$ is the angle of a vector from $i$ to one of its nearest neighbours $j$, measured relative to a fixed axis.

A perfect hexagonal vortex lattice has sixfold rotational symmetry (see section 2.3.2), meaning that every bond angle is $\pi/3$ radians. The exponential function inside the sum in equation 5.2 is therefore $e^{2\pi i} = 1$ for every bond. The hexatic order parameter for the perfect lattice is then found to be $|\psi_6|^2 = 1$.

Since the range of $\theta_{ij}$ is $[0, 2\pi]$, the range of $e^{6i\theta_{ij}}$ is $[-1, 1]$. $\psi_6$ therefore takes this same range, and $|\psi_6|^2$ ranges from 0 to 1. Any deviation from the perfect lattice reduces the HOP to a value less than 1, with values close to 1 being rarely observed in practice, even for the lattice state. A value of 0 would imply a complete lack of orientational order.

Generally, very low HOP values of less than 0.01 are seen for vortices in an isotropic glass phase [36, 41], while higher values would be seen in a hexatic glass, and in the lattice phase\(^b\). While no precise threshold HOP values have been defined, the transition from an isotropic glass phase to a more orientational-ordered phase is expected to be accompanied by a sharp increase in the HOP. The field dependence of the HOP in one specific isotropic vortex glass case is investigated in section 5.3.3.

5.3 Experimental Results and Discussions

This section presents the results of two main investigations into the ordering of vortices in a particular glass phase, as published in [36] and [37]. These are given in 5.3.2 and 5.3.3, preceded by a section on background field measurement. 5.3.4 then describes the investigation of some anomalous features that were observed during the main investigations, especially the appearance of groups of closely spaced vortices.

5.3.1 Preliminary Background Field Measurement

Due to the influence of the Earth’s magnetic field, a vertical background field was present within the shielded scanning area of the scanning SQUID microscope, as

\(^b\)Vortex phases are explained in sections 2.3.2 and 2.3.3
discussed in section 5.1.1. Through preliminary scanning SQUID measurements on superconducting YBCO samples, it was found that the applied vertical field required to offset this background field was $B_a = (2.73 \pm 0.01) \mu T$. Determination of this background field was achieved through measurements of the direction of vortices under various applied field values.

The aim was to apply a field in the vertical ($z$) direction that was equal and opposite to the background field, in order to offset it. When the total external field at the sample position is directed upward, vortices in the sample are directed upward throughout the film. And when the total external field (which is the sum of background and applied magnetic field) is directed downward, vortices are directed downward throughout the film.

Therefore, a series of simple measurements were made in which the field was varied through a range of about 10 $\mu$T and the direction of a few vortices was measured for each field value. This process was quite laborious due to the necessity of heating the sample above $T_c$ and re-cooling for each new field value in order to “reset” the vortex distribution before the next measurement.

For all applied field values smaller than 2 $\mu$T, all observed vortices were seen to be directed upward, but for applied field values larger than 3 $\mu$T, the vortices were directed downward. Therefore, it was seen that the point of zero total field must lie in the range $2 \leq B_a \leq 3 \mu T$. When the field had increased past the point of zero total field, the vortices were seen to have flipped their direction.

Following this, the applied field was varied over this smaller range, and the vortex directionality measured in a similar way. This process was continued: the vortex directionality was systematically checked for increasingly precise applied field values, until the zero field point was known with sufficient precision.

The applied field value at which the vortex direction reversed was found to be 2.73 $\mu$T, with a precision of 0.01 $\mu$T. The measurements taken closest to this applied field value were also seen to have the least number of observed vortices, further confirming that this was quite close to the true zero field point. Therefore, the background field was empirically determined to be $2.73 \pm 0.01 \mu T$ in the negative direction. This value was found to correlate well with previous measurements carried out with less precision, using a Bartington Mag-03MS three-axis magnetic field sensor, which had found that $\mathbf{B} \simeq -2 \mu T \, \hat{z}$.

Following this background field test, a fixed value of 2.73 $\mu$T was subtracted from the applied field value for all subsequent SSM measurements to give the total field $B$, compensating for the background, i.e.

$$B = B_a - 2.73 \mu T. \quad (5.3)$$
All field values given in the following sections refer to the total field after background compensation, which is taken to be solely in the $z$ direction.

### 5.3.2 Analysis of Vortex Glass

The first SSM investigation involved the observation of a glass-phase vortex distribution in YBCO thin films after cooling in a magnetic field significantly smaller than the Earth’s field, along with current mapping and characterisation of the vortex phase. The results of this investigation were published in [36]. The discussion of this investigation relies heavily on the theory of vortex phases that is given in sections 2.3.2 and 2.3.3.

Autocorrelation calculations on this distribution showed a weak short-range positional order, while Delaunay triangulation showed a near-complete lack of orientational order. Using this information, the phase of the vortex matter was characterised as an isotropic vortex glass.

Most previous scanning SQUID investigations of YBCO thin films had imaged only a few individual vortices [162], for the purpose of current mapping. Therefore, this investigation aimed to observe and fully analyse vortex distributions across a larger region of a YBCO film, covering several hundred vortices.

As discussed in section 3.5, YBCO thin films deposited by PLD at ISEM generally have a high number of strong pinning sites which mainly consist of out-of-plane columnar defects [16, 136, 141, 142]. It was therefore predicted that glass-like phases would dominate the phase diagram in these films[163].

#### Experimental Details

In this work, scanning SQUID microscopy was employed to directly observe the vortex arrangements in PLD-grown YBCO thin films after field-cooling at $\mu$T fields. The ordering of the vortex phase was then examined in detail in one such film to show that a glass phase was indeed present, and to characterise the type of glass seen. Through analysis by autocorrelation and Delaunay triangulation, it was found that:

- Although most vortices had six nearest neighbours, their angular distribution did not show the orientational order of a hexatic vortex glass. Instead, the vortex arrangement was generally isotropic.

- Vortex distances were quite randomly distributed, though some short-range positional ordering was observed in the autocorrelation, which may correlate with the “grouping” behaviour described in section 5.3.4.
These features of the glass phase were replicably observed under multiple field coolings.

This investigation focussed specifically on SSM measurements of a single 200 nm YBCO film, which was repeatedly field-cooled under identical conditions, in a 6.93 $\mu$T field. The critical temperature of this film was 90.0 $\pm$ 0.5 K as measured by MPMS (see section 3.4.1).

Current in the sample was calculated from field data using the current calculation program described in section 5.2.1. The distribution of vortices was then characterised using autocorrelation and Delaunay triangulation, using the phase characterisation program described in section 5.2.2.

Results

Two typical SSM images are shown parts (a) and (b) of figure 5.4. In contrast to the magneto-optical images of magnetic field seen in the previous chapter, individual vortices are clearly resolved in these measurements. Each vortex is seen as a black circle, and these are dotted throughout the right side of each image. All scans were taken at the same position on the sample. This was ensured by choosing a position near an identifiable edge, which can be seen as the vortex-free region on the left side of each image.

It should be noted that the apparent size of vortices in these images is increased due to spread of stray fields as discussed in section 2.6.3. The vortices also appear slightly asymmetrical, with an apparent elongated ‘shadow’ on the left side of each vortex in figure 5.4(a),(b). This is due to the influence of a field component parallel to the sample surface, occurring due to the angle of the pick-up loop as shown in 2.10(a).

Since all scans were taken under identical conditions, these and other images taken during this study appear quite similar at first glance. However, figure 5.4(c) shows a superposition of the vortex positions from the two scans, revealing that none of the vortex locations are reproduced upon re-cooling. This image was created by application of the particle detection procedure, which is part of the Delaunay triangulation program described in section 5.2.2. Particle detection was applied to each of the SSM scans in order to isolate the vortices, then the resulting images of the particles (vortices) were re-coloured and overlaid. No significant overlap of vortex positions was found under any relative translation of vortex positions.

Figure 5.5 shows the circulation of current around each vortex, as calculated from the magnetic field data of figure 5.4. The brightness of each point in the image is proportional to the magnitude of current at the corresponding point in the sample. The dark spots seen throughout the sample and the bright regions around them are the current-free vortex cores and the circulating current of the vortices, respectively.
At this point it should be noted that abnormally closely spaced groups of vortices, which are statistically unlikely to occur, were observed in all SSM scans above a threshold magnetic field, as highlighted in 5.5(b). These groups are easily identified in the current map images, due to the appearance of strongly interconnected currents flowing around the perimeter of the entire group of vortices and very little current between vortices, though these features appear to be artificial, arising due to the spread of stray fields at the scan height. For a full examination of vortex groups in SSM images, see section 5.3.4.

The apparent size of vortices was determined by plotting field profiles across
CHAPTER 5. SCANNING SQUID MICROSCOPY OF VORTEX GLASS

Figure 5.5: Maps of supercurrents calculated from the field distributions of figure 5.4 respectively. Some vortex groups with apparent interaction of currents are highlighted by red circles in (b).

Figure 5.6: (a) Field profile and (b) current profile across a single isolated vortex, chosen from the SSM image in figure 5.4a. The red curves show Gaussian fits.

vortices as they appear in the SSM images. The data points are shown in figure 5.6(a). A Gaussian profile was fitted to the stray field of vortices, and the average full width half maximum (FWHM) was found to be 6 μm. This is the solid red curve in figure 5.6(a). The FWHM is significantly larger than the theoretical width of the magnetic field profile of the vortex at the film surface, which is given by $2\lambda \approx 0.5 \mu m$. The large size of vortex field profiles shows a significant spread of stray field between the film and the scan height. The apparent size may also be influenced by the spatial resolution of the scanning SQUID apparatus, which can be no smaller than the dimensions of the pickup loop.

In a similar way, interesting supercurrent profiles were obtained for the shielding currents around vortices, as shown in figure 5.6(b). A dip in current is observed at

---

See section 2.7.2 for an explanation of why this is not the true size of vortices.
the centre of each vortex as expected, and Gaussian fits were plotted as shown in the figure. The average FWHM of the current dip was found to be 3.2 \( \mu \text{m} \). This measured size was also larger than expected, due to stray current error (see section 2.7.2). Outside this region, the stray shielding currents peak sharply before gradually fading to zero at some distance from the vortex core.

**Analysis of Vortex Arrangements**

The first step in the characterisation of the distribution of vortices was an autocorrelation process, as described in section 5.2.2.

![Figure 5.7](image)

**Figure 5.7:** (a) The visualisation of the autocorrelation function from Fig. 5.4(a). The brightness of a point at some position \( \mathbf{r} \) from the centre represents the autocorrelation for the corresponding displacement. (b) Modelled autocorrelation for a finite “perfect” hexagonal lattice. The lattice in figure 2.6a was used as a model.

Figure 5.7(a) shows the calculated autocorrelation from a field map of the vortex distributions observed by scanning SQUID shown in figure 5.4(a). Figure 5.7(b) shows the visualised autocorrelation for a modelled perfect hexagonal lattice composed of a finite number of points of finite size. This represents the “perfect” defect-free distribution of vortices in the lattice state.

Compared to the “perfect” vortex lattice, the glassiness of the measured vortex distribution is evident due to the lack of regularity in this autocorrelation. Far from the central bright spot, no ordering can be seen, which implies a lack of long-range order, confirming the existence of a glass-like vortex phase.

However, the somewhat brighter ring lying very close to the central bright point indicates retention of some weak short-range order. The nature of this short-range order was not immediately clear, but when the distances between vortices were examined directly as discussed in section 5.3.4, it was seen that there was one par-
particularly prevalent vortex spacing that corresponds roughly with the radius of this ring. This ring has a fixed radius in all directions, giving some evidence for a lack of orientational order in the vortex distribution, which would be confirmed upon calculation of the hexatic order parameter.

The vertical stripe geometry of the visualisation in 5.7(a) is simply due to the rectangular shape of the vortex-containing region of figure 5.4(a) that was selected for autocorrelation. It does not imply any $x-y$ asymmetry in the vortex distribution.

To further classify this glass as hexatic or isotropic (see section 2.3.3), the orientational order must be analysed, and this is achieved by Delaunay triangulation, which effectively maps connections between nearest neighbours, as described in section 5.2.2.

![Figure 5.8](image)

**Figure 5.8:** Delaunay triangulation applied to the SSM images of vortex distribution. Each triangulation is superimposed onto the SSM image from which it is calculated (original images shown in figure 5.4). Those vortices within the red area in (b) are taken as internal.

Figure 5.8 provides a visualisation of the Delaunay triangulation applied to the vortex positions identified from SSM images. From this analysis, the distribution of the number of nearest neighbours to each internal vortex is calculated. This is plotted in figure 5.9 for three independent scans under identical field conditions.

The triangulation has been calculated based on positions of vortices found by applying particle detection to the images, as discussed in section 5.2.2. This calculation method is generally accurate, but fails when pairs of vortices are so closely spaced that the program recognizes them as a single particle. This situation can be seen in several places in figure 5.8(a), where there is more than one vortex at a single node of the triangulation. This error was seen to occur approximately once for every 40 vortices at the field value used for this investigation. It generally causes the number of nearest neighbours to be underestimated for some vortex pairs, and overestimated for others, leading to the false detection of topological defects.
section 5.2.2). This may cause a slight spread in the results of figure 5.9. Hence, the true distribution of nearest neighbours is expected to peak more sharply at 6.

Since most vortices were seen to have six nearest neighbours, the density of topological defects was lower than expected for a glass phase. It was therefore suspected that this may be a hexatic vortex glass (see section 2.3.3). The orientational order was therefore investigated using the hexatic order parameter of the vortex distribution $|\psi_6|^2$, given by equation 5.2.

The vortex distributions examined were found to have a hexagonal order parameter between 0.01 and 0.001, showing that there is very little orientational order in any scan. The phase of the vortex matter was therefore not hexatic, but an isotropic vortex glass.

**Summary**

In summary, the observed distribution of vortices in PLD YBCO thin films in this extremely diluted field-cooled vortex regime in low fields of the $\mu$T-range by scanning SQUID microscopy can be characterised as an isotropic vortex glass. The autocorrelation technique demonstrated a lack of long-range order as compared to a perfect lattice, while a fading signature of a weak short-range order has been retained, showing that the phase was glass-like. Then, employing the Delaunay triangulation technique, the glass was shown to be isotropic, although the topological defect density was lower than expected. The isotropic nature of the distribution was then confirmed through the calculation of a very low hexatic orientational order parameter.
From this evidence, it was not possible to classify the glass phase any further. It was accepted that this indeterminacy was most likely due to the highly diluted vortex regime formed by cooling in very low fields. Since the vortex density is so low in this field range, the distribution of vortices as they first nucleate (near the normal-superconducting transition) may be unlike any of the phases described in section 2.3.2, but could be a gas-like phase where any interactions between vortices are minimal \[36\]. Since cooling from the liquid/gas phase to the glass phase occurs quite slowly, thermal activations would be expected to statistically drive vortices into the nearest strong pinning positions, with minimal influence of vortex-vortex repulsion.

### 5.3.3 Field Dependence of Vortex Glass

The study of low-field vortex glass phases in a YBCO film was extended to analyse the field variation of these vortex distributions. The topological defect density and hexatic order parameter of vortex distributions were examined over a range of micro-Tesla fields using scanning SQUID microscopy. The results of this investigation were published in \[37\].

The first investigation had shown that vortices in YBCO thin films under very low field conditions existed in an isotropic vortex glass phase. The second investigation explored the field dependence of properties relating to order in this glass phase. The properties considered were the distribution of the number of neighbours to each vortex, and the hexatic order parameter. The modal number of nearest neighbours was six for all fields studied, but the proportion of vortices having six nearest neighbours was found to increase with increasing applied field (i.e. topological defect density decreased, see section 2.3.2). With a greater proportion of vortices having six nearest neighbours, the phase was loosely considered to become more “lattice-like” for higher fields.

However, the hexatic order parameter was also found to decrease with increasing field, meaning the angular distribution of vortices became less regular for higher fields in the observed field range. The distribution therefore had less orientational order at higher fields, which is actually less resemblant of an ordered lattice phase.

### Experimental Details

\(\text{YBa}_2\text{Cu}_3\text{O}_{7-x}\) films with thickness close to 200 nm were grown by pulsed laser deposition \[15, 16, 136\] on STO substrates, and lithographically patterned into 3 × 3 mm squares using the method described in section 3.2. One film was selected, with critical temperature \(T_c = 90.0 \pm 0.5\) K and the critical current density \(J_c \simeq 1.5 \times 10^{10}\) A/m² (measured at \(T = 77\) K and applied field \(B_a \to 0\) T), as measured
Figure 5.10: Scanning SQUID images of vortices near the edge of a YBCO film after field cooling at (a) the lowest field attainable (\(\sim 0.05 \mu T\)), (b) 1.47 \(\mu T\), (c) 2.10 \(\mu T\) and (d) 5.46 \(\mu T\). The brightness of each pixel represents the strength of magnetic field at the corresponding point on the sample. Shading on the left side of each vortex is due to a small tilt of the SQUID pick-up loop relative to the sample surface.

The selected YBCO film was field cooled to a stable 4.2K using the helium bath cryostat described in section 5.1. Field-cooling was repeated for several cooling field values in the low field vortex-glass region of the phase diagram (between 0.05 and 5.47 \(\mu T\)). The reason that the samples had to be field-cooled is discussed in section 2.5.1. A single high-resolution scan of the vortex distribution was taken for each field value using the scanning SQUID microscopy (SSM) technique described in the preceding sections. Current in the samples was calculated from the resulting SSM images using the current calculation program described in section 4.2.2, and the distributions characterised in terms of nearest neighbours and hexatic order parameter using the characterisation program described in section 5.2.2.
Figure 5.11: Supercurrent in YBCO film after field cooling at (a) \(~0.05\) \(\mu\)T, (b) 1.47 \(\mu\)T, (c) 2.10 \(\mu\)T and (d) 5.46 \(\mu\)T, calculated from the scanning SQUID images of figure 5.10. Brighter regions represent higher currents.

Results

Scanning SQUID images were obtained after field cooling at several field values in the specified range. Figure 5.10 gives typical magnetic field data at given field values.

The light region is the superconducting sample, with dark spots showing vortex positions. The dark area at the left is outside the sample. The vortices in this image appear larger than their actual size, and show an apparent asymmetry due to the scan height and angle of the SQUID magnetometer \[36\]. Scan (d) has a smaller area since the data for part of the scan was corrupted, while scan (a) is cropped to exclude data outside the sample. These scans were not taken in the same location on the sample due to experimental difficulties.

Supercurrent calculated in the film from similar SSM images is shown in figure 5.11, for scans taken with different field histories.

Vortices are seen here as dark spots since no current flows in the vortex core, with bright circulating currents around. Shielding currents are also seen at the
sample edge, especially in the lower-field images.

![Delaunay triangulation mapped onto scanning SQUID images](image)

**Figure 5.12:** Delaunay triangulation mapped onto scanning SQUID images at (a) \(0.05\ \mu T\), (b) \(1.47\ \mu T\), (c) \(2.10\ \mu T\) and (d) \(5.46\ \mu T\). The triangulation is represented by yellow lines connecting each detected vortex position to its neighbours.

Delaunay triangulation was applied successfully to the vortex positions from scanning SQUID data at each field, as shown in figure 5.12.

The distribution of the number of nearest neighbours to each internal vortex at a particular field is shown in figure 5.13(a). A normalised Gaussian fit is plotted for each distribution, with the total number of counted vortices (equal to the number of internal vortices, \(N_{int}\)) indicated for each. The SSM scan at \(\sim 0.05\ \mu T\) is excluded since the field value is not precise, and \(N_{int} = 0\) for this scan. The \(1.47\ \mu T\) scan is shown, but it also has a low number of internal vortices, and so is excluded from further analysis due to statistical inaccuracy.

The full width at half maximum (FWHM) of these fit curves represents the spread of the number of nearest neighbours. This width is plotted against field in part (b) of this figure. Error bars represent standard error in the Gaussian fit, which naturally decreases when a larger number of vortices are counted. It is found that the FWHM decreases linearly with applied field, but with relatively low gradient.
CHAPTER 5. SCANNING SQUID MICROSCOPY OF VORTEX GLASS

This indicates that for higher fields a greater proportion of vortices have six nearest neighbours, and hence that the number of topological defects slightly decreases with field in the range measured.

The bond angle between vortices is also determined from the Delaunay triangulation, as described in section 5.2.2. This is used to calculate the hexatic order parameter (HOP) for each field value used. The HOP is plotted against field in figure 5.14. The error ranges for HOP are calculated from an estimation of the error in distinguishing closely spaced vortices in each image, as explained in section 5.3.4.

It is seen that the HOP decreases with increasing field in an inversely proportional manner. This implies that the angles between vortices become less regular for increasing field, in the field range studied.

Discussion

As field increases, the topological defect density decreases, making the vortex distribution slightly more resemblant of a regular lattice. This was expected, since the sample is in the lowest field, lowest temperature part of a phase diagram that is similar to that shown in figure 2.5(b). It is therefore in the re-entrant part of the vortex glass region, below the re-entrant vortex lattice region. An increase in field therefore moves the sample closer to the vortex lattice region,[32] which occurs due to the increasing influence of vortex-vortex repulsion.

However, if the distribution were simply becoming more lattice-like with increasing field, the hexatic order parameter would also increase with field. Instead, HOP was found to be inversely proportional to applied field. This shows that while the number of topological defects decreases with increasing field, the angle between vor-

Figure 5.13: (a) Normalised occurrence of the number of nearest neighbours to vortices in the SSM scans at various fields. The total number of internal vortices ($N_{\text{int}}$) counted for each field is given in the legend. (b) Plot of the width of the Gaussian fit to these curves against applied field. An error-weighted linear trend line is plotted with gradient \((-5.353 \pm 0.077) \times 10^4 \text{ T}^{-1}\).
Variation of the hexatic order parameter $j_6^2$ of vortex distribution with applied field. Error bars reflect the accuracy of identification of vortex positions. The hyperbolic fit gives $1/|\psi_6|^2 = B \times (9.99 \pm 0.74) \times 10^7 \, T^{-1}$.

**Conclusion**

In this investigation, the field dependence of the ordering of vortices in a low-field vortex glass state was directly examined in a YBCO thin film, as observed by scanning SQUID microscopy of field-cooled states. The order of the vortex glass was quantified using two parameters: the spread of the number of nearest neighbours to each vortex, and the hexatic order parameter.

It was found that the distribution of nearest neighbours peaked at six for all fields studied. With increasing field, the relative number of topological defects was seen to reduce, but this did not increase the orientational order. Instead, the hexatic order parameter decreased with increasing field.

If this investigation were extended to higher fields, the vortex distribution should be seen to transition into the lattice phase at some well-defined point $[165, 166]$. The hexatic order parameter would be expected to reach a minimum before increasing sharply at the isotropic glass to lattice transition, while the number of topological defects would decrease steadily before sharply dropping at this transition point$[31]$. The HOP and nearest neighbour distribution may also vary with field in the vortex lattice regime, and may have different field dependences in the high-field vortex glass phase and in other glass-like phases such as hexatic glasses.
Direct measurements of field-dependent vortex ordering in other regions of the phase diagram may prove to be a very exciting avenue for future research. However, such studies should consider larger areas and hence larger vortex numbers for heightened statistical significance.

5.3.4 Vortex Groups Observed under SSM

A perplexing phenomenon was continually observed throughout the investigations of the low-field vortex glass phase in YBCO films: This was the appearance of “vortex groups” in many scanning SQUID images of the vortex distributions. These scans each showed several groups of two or more vortices. There were two unusual features of these groups, which will each be discussed separately in this section:

- The first was that the groups seemed to show strongly-interacting circulating currents, with an apparent lack of current throughout the region between any two closely spaced vortex cores. These current-free regions were much larger than expected due to any simple cancellation of oppositely-directed current components between the vortices. This may be an artefact, or have some unknown physical origin, as discussed in the first subsection below. An investigation of the stray field error that may have caused such an artefact was brought about as a direct result of the investigation of circulating currents, and is discussed in the second subsection.

- The second unusual feature was the distribution of vortex spacings, with one very close spacing of vortices being particularly prevalent. The origin of groups of vortices with this particular spacing is discussed in the final subsection.

Interconnected Circulating Currents

The first evidence for some abnormality in the vortex distribution was the appearance of unexpected dark spots in the current maps, occurring at the midpoints between pairs of adjacent vortices. This can be seen in the current maps calculated from SSM images in figure 5.4(c) and (d), particularly for those vortices indicated by red circles. Vortices in these images are identified by dark (current-free) spots surrounded by bright halos of circulating current. However, there were also several dark spots that appeared almost identical to vortices, though they did not correlate with the position of any vortex in the corresponding SSM image, instead being found at the midpoint of two closely-spaced vortices.

Where these dark spots appeared, groups of two or more vortices had circulating currents that were apparently continuous around the perimeter of the whole group, as identified by red circles in figure 5.4(c). This might have been expected for
vortices that were much more closely spaced, with opposing currents cancelling one another in the space between vortices, though was unexpected given the relatively large vortex spacings, as discussed in the next subsection.

The apparent connectedness of circulating currents around groups of vortices was also seen in scans taken at other field values, though it disappeared for fields below $2 \mu T$. This can be seen in figure 5.11.

One explanation is that the apparent lack of current between the vortices is merely artefact arising from stray current error (as introduced in section 2.7.1). The notion that this was an artefact is strengthened by noting that one of the poorest quality images considered, figure 5.11(d), shows the most current connectivity - with current appearing to flow around the perimeter of large groups of vortices in the centre of the image. However, this is also the image with highest vortex density, which may support a physical origin to the connectedness.

To verify that such connectivity was indeed only an artefact, the spacing between neighbouring vortices was determined in order to show that there could be no physical interaction. The distance between the midpoints of several pairs of neighbouring vortices was determined from their positions in the SSM scans taken at $B \simeq 6.93 \mu T$ (which were previously used for the investigation described in section 5.3.2). The average spacing between neighbouring vortices at this field was found to be $32 \mu m$, with a significant spread in nearest neighbour distances as expected in glassy distributions. Since both the magnetic field penetration depth ($\lambda$) [167] and the individual vortex depinning radius in these YBCO films are of the order of $0.5 \mu m$ (see section 3.5) [141, 142], the measured distance between vortices was too large to allow any significant interaction of circulating currents. Thus, the lack of current between these vortices was not found to have any identifiable physical origin.

However, the measurement of spacings between vortices in the SSM scans revealed an unexpected feature: that most of the vortices in groups were separated by approximately the same distance, while the spread of vortex separations between other neighbouring vortices was much greater. This will be investigated in the last subsection.

Since the current connectedness was found to have no physical origin, it was decided that it could with high likelihood be attributed to the stray current error, as discussed in the next subsection.

**Investigation of Stray Field Overlap**

As discussed in section 2.7.2, significant spread of the magnetic stray field occurs between the sample surface and the scan height, increasing the apparent size of vortices (see figure 2.11 in particular). This can lead to difficulty in distinguishing
vortices from their neighbours when intervortex distance is small, leading to error. For example, several pairs of vortices with apparent overlap are identified in figure 5.16. Due to these poorly resolved vortices, the current calculation procedure could not be accurately carried out for this SSM image, and error was introduced into the lattice characterisation calculations.

Figure 5.15: Field profiles across the centres of closely paired vortices showing (a) minimal stray field overlap at 13 μm separation and (b) half-height overlap at 9 μm separation, which may result in a failure to interpret these as separate vortices. (c),(d) Current profiles across the same pairs of vortices as in (a) and (b) respectively. The red lines show Gaussian fits across the central current dips, though no fit could be made for the rightmost vortex in (d).

To investigate the effects of the overlap of stray fields, linear profiles of measured field were plotted across pairs of closely spaced vortices in the 6.93 μT scans. Figure 5.15 shows the field and current profiles of pairs of vortices at 13 μm and 9 μm separation, showing that the stray fields from vortices closer than about 10 μm overlap so as to give no point of zero measured field between the vortices.

From these profiles it was therefore confirmed that this stray field error was the source of the apparent interconnectedness of circulating currents. However, a more precise analysis was impossible due to the inability to correct for stray field error (as explained in section 2.7.2). The field directly at the sample surface could not be calculated from the stray field due to the difficulty in precisely determining the
distance from sensor to sample in the scanning SQUID apparatus. If this calculation could have been carried out with a level of precision that surpasses the current equipment limitations, then any connectedness of the supercurrent profiles in the vortex groups could be examined more rigorously, and the appearance of strongly-connected circulating currents would be expected to disappear.

Another significant error also caused by the overlap of stray fields is the false reignition of two closely-spaced vortices as a single vortex by the particle detection algorithm during the Delaunay triangulation procedure. This error propagates, causing the total number of vortices to be underestimated, and further errors in the number of nearest neighbours to the closely-spaced pair and surrounding vortices. The angles between neighbouring vortices will also be miscalculated. The errors in neighbour number and angle will both affect the hexatic order parameter.

The particle detection parameters were therefore optimised to avoid such errors. This optimisation was carried out manually for each vortex distribution, and the Delaunay triangulation was examined for false vortex positions after each attempt. It was therefore possible to apply Delaunay triangulation very accurately to each measurement. The only exception was the low-quality 2.73 μT scan. The lack of accuracy in this scan is indicated by the size of its error bar in figure 5.14.

**Linear Defect**

The 2.73 μT scan was not excluded from study due to the presence of an interesting feature: a linear region of increased vortex density with length ≥ 400 μm, as shown in figure 5.16. This region still showed high linear density after the sample had been heated above $T_c$ and re-cooled (while most vortex positions were generally not reproduced). This shows that this region is a strong-pinning defect, that has linear shape and width on the single-vortex scale or smaller. This defect resembles a very small scratch, but may also be caused by a substrate twin or an extended grain boundary.

Because of the linear defect, information on the spacings of vortices in this scan was unreliable, and it was not used for the investigation of closely-spaced vortices.

Most scans following this one were taken at a fixed defect-free position near the edge of the sample, in order to avoid the influence of this defect or any other.

**Increased Prevalence of Closely-Spaced Vortices**

Just as the issue of interconnected currents was resolved, the investigation of inter-vortex spacings in the $B \simeq 6.93$ μT scans revealed another strange feature in the vortex distribution: Though the average spacing between neighbouring vortices was 32 μm, there were a disproportionately large number of vortices in the film with
nearest neighbour distances in the range of $\sim 15 \ \mu m$. This was seen in all scans, showing that the prevalence of this separation was reproduced upon re-cooling. It was subsequently determined that only these closely-spaced vortices showed the apparent lack of current over a large region between their cores, although this distance was still too large to allow any true physical interaction. It was also noted that this was the smallest intervortex spacing observed at this field, though not all pairs of neighbouring vortices were measured.

Some evidence for the increased prevalence of one particular intervortex spacing could also be seen in the autocorrelation that was carried out during the first investigation (figure 5.7), though it had initially been overlooked: A bright ring was present around the central point in the visualised autocorrelation, and the radius of this ring was approximately $15 \ \mu m$, indicating that the probability of finding a pair of vortices with a separation distance of $\sim 15 \ \mu m$ was higher than for any other distance. Another significant feature of the autocorrelation was that the area between the central bright spot and the surrounding ring was dark, indicating that there were no vortex pairs with a spacing significantly less than $15 \ \mu m$.

While stray current error explains well the apparent connectedness of currents, it does not explain the fact that significant numbers of vortices were “grouped”, having a below-average intervortex spacing with one or several neighbours. Such grouping was also observed over much of the field range considered, though no grouping behaviour could be seen for $B \leq 2 \ \mu T$. At fields below this value, the vortices were spaced very far apart and no particular vortex spacing seemed to be favoured. Some grouping may still have occurred at lower fields, though this could
not be determined with any statistical significance due to the smaller number of vortices imaged at these fields.

The reason that multiple field-coolings were undertaken for each magnetic field value in sections 5.3.2 and 5.3.3 was to examine the origin of these groups. If the vortex grouping occurred due to the existence of particular strong pinning centres, a reproducibility of the locations of the groups would be expected upon re-cooling, such as was seen for the linear defect in figure 5.16. In general, this did not occur. The positions of the groups appear randomly and independently distributed for each cooling.

It was also for this reason that the surfaces of YBCO samples were studied using atomic force microscopy, as described in section 3.4.3. The average size of domains in the AFM images of YBCO films was measured to be approximately 200 nm. This size did not correspond to the size of the vortex groups observed by SSM, or of the linear defect described in the preceding subsection. These measurements also show that there are no other significant surface features on the YBCO films whose size correlates with the magnetic features observed. The AFM images therefore confirmed that the observed grouping of vortices was not related to any structural feature on the film surface.

Hence, it was inferred that the vortex groups were not formed around any particular strong pinning centres in the films. It was therefore concluded that the existence of vortex groups must be a feature of the low-field vortex glass, arising due to the very low vortex density and possibly the sample history.

The mechanism for the formation of these groups is still under discussion, though several hypotheses have been proposed:

- The prevalence of a particular vortex spacing in the groups may have been an indicator of a straightforward mechanism such as film inhomogeneity leading to regions of heightened pinning. However, the position of the groups is not reproducible upon re-cooling and few vortices are grouped together. If there were only certain large areas of strong pinning in the films, the group positions would be reproducible; and if smaller areas of stronger pinning were distributed throughout the film, then more of the vortices should be combined into groups. Hence, inhomogeneity in the film is probably not the cause of vortex grouping.

- Another simple explanation was that a strong demagnetisation effect could lead to regions of heightened field, and hence heightened vortex density. Although thin films tend to have a strong demagnetising factor [154], these films were field-cooled at only a few μT. The demagnetisation effect should be negligible at such low fields [154], hence it is also unlikely that this is the reason for the observed grouping.
Another idea is that the existence of groups is a direct result of the sample history, as mentioned in the conclusion of section 5.3.2. Since vortices may experience a strongly diluted (gas-like) phase during cooling, thermal activations could statistically drive some vortices into pinning positions that happen to be more closely spaced than average, where they remain after the transition to the glass phase. The groups are therefore simply a feature arising from the chaotic nature of the vortex glass state. However, with this explanation it is still statistically unlikely that such grouping should occur with one pronounced intervortex spacing.

The mechanism could also be that vortices with smaller intervortex distance experience a greater vortex-vortex repulsion. Therefore, when the sample was being cooled through the liquid (or gas) to glass transition, the separation between these vortices would increase only to the point that thermal forces dominated over vortex-vortex repulsion. This vortex spacing therefore emerges in the liquid phase, and then on transition these vortices become pinned in the position observed.

A final explanation, similar to the previous, is that vortex-vortex repulsion is stronger than pinning when vortices have a smaller spacing than is observed. Any closer vortices would therefore move apart up to a certain spacing at which pinning balances vortex-vortex repulsion. This explanation might hold for any low-field vortex glass and does not rely on sample history.

In the last two cases (which seem to be the most plausible) it would be slightly misleading to think of these vortices as being “grouped”, but rather that their spacing is larger than it would be in the absence of vortex-vortex repulsion.
Chapter 6

Vortex Ratchets

This chapter describes a proposal for novel vortex ratchet devices which may allow greater rectification of supercurrent than similar previous devices.

Two novel geometries were proposed for the microstructure of vortex ratchets using 2D weak-pinning channels (the theory of vortex ratchets is explained in section 2.4.1). Ratchet patterns were produced by partial-etching of YBCO films, and initial tests were performed, through no actual diode devices have yet been produced. However, simulations have shown that under strict conditions, these novel ratchets may indeed be capable of complete vortex rectification, where vortices are only free to move in a single direction (labelled the easy-flow direction), and no motion is possible in the opposite (hard-flow) direction.

Under more realistic conditions, some motion is observed in the hard-flow direction, but this is always dampened. The rectification factor, which is the ratio of current in the forward direction to current in the reverse direction, was quite high in all cases⁴.

The realisation of practical vortex diodes is very important for production of superconducting logic circuitry, which could in turn become the basis for complex superconducting electronics [168]. Superconducting electronic devices would have very significant advantages over conventional semiconducting electronics in terms of speed, size and power consumption [168].

6.1 Ratchet Design

A novel pattern for creation of vortex ratchets was proposed, which may allow better rectification of vortex motion (and ultimately better current rectification) than has been seen using previous designs.

⁴For clarity, the terms ‘forward’ and ‘reverse’ will be used solely to refer to directions of current, while ‘easy-flow’ and ‘hard-flow’ will refer solely to directions of vortex motion.
The function of this proposed ratchet was based on the principle of asymmetric weak-pinning channels within a strong-pinning film, as demonstrated by several previous groups [60–62, 169]. The free flow of vortices along the low-pinning channels creates resistance to current directed across the channel. Ideally, the asymmetric microstructure of these channels would allow vortices to flow freely in one direction only, while not allowing any sustained vortex motion in the opposite direction. In contrast to previous ratchets based on weak-pinning channels, the new pattern is predicted to provide rectification even at zero temperature.

This ratchet could be used as an effective vortex diode: a large resistance would be created for current in the reverse direction, which causes free-flowing vortex motion (in the easy-flow direction). Conversely, there would be little to no resistance to current in the forward direction, since vortex motion in the hard-flow direction is heavily suppressed due to the shape of the channel. Further explanation of the principles of vortex diodes is given in section 2.4.3.

6.1.1 Existing Mirrored-Sawtooth Channel Geometry

Several groups have tested ratchet devices consisting of low-pinning channels with double-sawtooth shaped walls [60–62, 169]. However, this design is inherently flawed in that it cannot provide rectification at very low temperatures, as identified by Wambaugh et al. [60].

In this geometry, an asymmetric potential landscape was created within each channel by varying the width of the channel in a sawtooth manner. To realise this, both walls of the channel were patterned with a series of protrusions in the form of mirrored sawtooth shapes, creating a series of asymmetric bottlenecks, as shown in figure 6.1. It had been theorised that a vortex moving toward one such bottleneck in the easy-flow direction would encounter a shallow potential gradient which it could surmount in order to move through the bottleneck. A vortex approaching in the hard-flow direction would encounter a much steeper gradient, which was intended to restrict any passage through the bottleneck in that direction. It was expected that a sustained motion of vortices would therefore only be possible in the easy-flow direction, leading to rectification.

However, a significant flaw was discovered when this ratchet geometry was tested in simulation by Wambaugh et al.[60]:

The “Zero Temperature Flaw”

Wambaugh’s results showed that mirrored-sawtooth low-pinning channels provided good rectification at temperatures significantly higher than absolute zero, but little rectification was seen at very low temperatures. The lack of rectification as
temperature approaches zero is labelled the “zero temperature flaw”. This poses a significant problem for superconducting devices, which require the temperature to be minimised in order to maximise critical current.

The explanation for this flaw is that in the absence of thermal motion, vortices driven in either direction would tend to take the simplest path in that direction, which was along the centre of the channel in this geometry. Since the central line through the channel was not interrupted by any protrusion, the influence of the bottlenecks was minimal along this line and vortices could therefore move freely through the centre of the channel in both directions. At higher temperatures, however, vortices would wander from this central line due to thermal motion. Only then, when a vortex deviated from the centre of the channel, would it interact with the closest wall, and experience the effect of the asymmetric potential gradient.

The conclusion given by Wambaugh was that the ratchets tested could provide functional rectification, but only within a given temperature and field range.

### 6.1.2 Proposed Geometries and Their Improvements

Two novel geometries for asymmetric low-pinning channels are proposed in the following subsections.

The aim of the proposed ratchet patterns is to reduce the strong temperature dependence of vortex ratchet devices based on asymmetric low-pinning channels. Specifically, a vortex ratchet pattern similar to the mirrored sawtooth geometry was to be created with the ability to operate at low temperatures.
The inspiration for the proposed ratchet geometry came from a desire to eliminate the uninterrupted central line through the low-pinning channels, identified in figure 6.1. This would prevent vortices from moving freely along the centre of the channel at low temperature.

**Offset-Sawtooth Geometry**

To correct for the central-line flaw of the mirrored sawtooth geometry, a modification was conceived in which the sawtooth shape of one of the channel walls was offset from the other by half of its period. The length of the sawtooth protrusions from each wall could then be increased so that their tips crossed the central line.

With the sawtooth patterns offset, the bottlenecks were no longer found between the tips of two opposing sawtooth protrusions, but between the tip of one protrusion and an opposing wall. Therefore, in this geometry there were two bottlenecks per period, and these were not aligned with one another at the channel centre but alternated between one side of the channel and the other. The gap between each sawtooth protrusion was therefore increased in order to maintain a suitable separation between each bottleneck.

The protrusions from each channel wall could not extend too close to the opposite wall, otherwise the easy-flow motion of vortices would be strongly impeded. The optimal gap between the end of these protrusions and the opposite wall was to be determined experimentally, under the a priori condition that this gap must be less than half the period to avoid the zero temperature flaw.

After all of these considerations, the resulting offset-sawtooth geometry is as shown in figure 6.2.

The simplest path along an offset-sawtooth channel is not a simple line through the centre, but a serpentine path curving around each protrusion. Consequently, vortices flowing along the offset-sawtooth channel would be forced to interact with the walls without requiring any thermal activation. Rectification of vortices in these channels should therefore be possible even at temperatures approaching absolute zero.

The essential principles of vortex ratchets based on the offset-sawtooth channel geometry are as follows:

- The structure is based on a series of low-pinning channels within a high-pinning film.
- Vortices moving through the low-pinning channel experience vortex-vortex repulsion from pinned vortices in the high-pinning walls and in asymmetric protrusions from the walls.
Figure 6.2: Schematic of Offset-Sawtooth geometry for vortex ratchets using asymmetric low-pinning channels. The blue parts represent areas of high pinning, and the white areas of low pinning. The alternating locations of bottlenecks are indicated, protrusions labelled, and the central line shown to be interrupted by the protrusions. The vortex easy-flow direction is to the right.

- These protrusions cross the centre line, so that vortices moving through the channel on any simple path are forced to interact with them.

- A vortex moving through any bottleneck in the hard-flow direction will immediately encounter the edge of a protrusion at $90^\circ$, and experience a repulsion that is anti-parallel to its motion.

- A vortex moving through any bottleneck in the easy-flow direction will encounter the edge of a protrusion at an angle less than $90^\circ$, and will experience a force that tends to cause turning toward the opposite channel wall (and towards the next bottleneck).

The vortex-vortex repulsion force on vortices in the channel can also be described through an asymmetric potential field that gives rise to ratchet motion - see section 6.5.2.

**Linear Protrusion Geometry**

The offset-sawtooth geometry was refined through extensive optimisation (as described in section 6.4), with the sawtooth protrusions later being replaced by slanted linear protrusions from each of the channel walls. The linear protrusion geometry is sketched in figure 6.3.

This geometry is essentially a low-pinning channel with linear cuts protruding from the walls at some angle $\theta$ to the wall, and extending beyond the centre of the channel.
Figure 6.3: Schematic of Linear Protrusion geometry for vortex ratchets using asymmetric low-pinning channels. The blue parts represent areas of high pinning, and the white areas of low pinning. The locations of bottlenecks, protrusions and vortex traps are indicated and the interrupted central line shown. The vortex easy-flow direction is to the right.

The linear protrusion geometry functions on the same principles as listed above for the offset-sawtooth pattern, but with the addition of V-shaped ‘traps’ which are intended to further restrict vortex motion in the hard-flow direction. Rather than encountering the edge of a protrusion at 90°, a vortex moving through any bottleneck in the hard-flow direction will proceed directly into the trap created by the next protrusion in the absence of other forces.

Once a vortex is positioned within the centre of the ‘v’, there are only two paths out of the trap: either to move back against the driving force (i.e. in the easy-flow direction, though it is being driven in the hard-flow direction); or to move through the protrusion, overcoming the repulsion of the vortices pinned within the protrusion.

Since both of these options require the vortex to surmount an energy barrier, the trap functions as a potential well when the driving force is in the hard direction. Conversely, when the driving force is in the easy direction, vortices should move easily out of the traps and then through the bottlenecks in the same manner as for the offset-sawtooth geometry.

### 6.2 Ratchet Device Fabrication

The fabrication of ratchet devices involved a double-patterning of YBCO films, using two independent ion beam etches with different parameters.

High-quality YBCO films were produced by pulsed laser deposition to a nominal thickness of 200 nm, as described in section 3.1. The first etch was used to shape
these films to the overall geometry of the device, without creating the ratchet itself. The second was a partial-thickness etch, meaning that the etch parameters had been optimised to remove only part of the YBCO from exposed areas, effectively reducing the thickness of some sections of the YBCO. These thinner regions would then function as the low-pinning parts of the ratchet.

6.2.1 Initial Macroscopic Patterning

The purpose of the initial patterning was to shape the sample to an appropriate overall geometry, while not yet applying the ratchet design.

This first etch removed all of the YBCO from unwanted areas of the film, and was carried out in an identical manner to the patterning used for other YBCO samples. The patterning procedure is described in section 3.2.

Several different sample shapes were produced with the initial etch, for different purposes:

- The vortex-lensing geometry required initial shaping into $3 \times 3 \ \mu m$ squares, then ratchet patterns would be etched onto regions at the edges of the sample. Inward-directed ratchet patterns would then be placed on some parts of the sample edges and outward-directed ratchet patterns on others, in order to allow preferential penetration from some edges. An example is shown in figure 6.4 with two ratchet areas on opposite sides of the sample, with the easy direction inwards on one side, and outwards on the other.

![Figure 6.4: The vortex lensing geometry used for testing of penetration in ratchet areas. Lines on the sample represent the ratchet patterned areas, with easy direction indicated by red arrows.](image)

- The “bridge diode” geometry required initial shaping into superconducting bridge shapes. The bridge shape consists of a very thin superconducting line connecting two large superconducting regions. A bridge geometry is usually
used to directly measure critical current, since the total critical current through the bridge is given by $J_c$ multiplied by the bridge width and sample thickness. Therefore the bridge width can be reduced to reduce the supercurrent to within a measurable range. For the bridge diode, this thin region would then be patterned with vortex ratchets directed perpendicularly across it, so that critical current along the bridge in one direction would be higher than the other.

![Magneto-optical image of superconducting bridge samples of width 150 and 200 μm, at temperature 4.4K and field 43mT. The places where ratchet patterns would be applied for the bridge diode geometry are indicated by dark blue lines, with easy direction indicated by red arrows.](image)

**Figure 6.5:** Magneto-optical image of superconducting bridge samples of width 150 and 200 μm, at temperature 4.4K and field 43mT. The places where ratchet patterns would be applied for the bridge diode geometry are indicated by dark blue lines, with easy direction indicated by red arrows.

### 6.2.2 Ratchet Patterning by Partial-Thickness Etching

The ratchet pattern was applied to samples in the positions described in the previous section. This was achieved using a secondary patterning, which was a more delicate procedure for two reasons:
Firstly, it was carried out on a smaller lateral scale, with sharp features that were in some cases smaller than 1 μm. One reason for the use of two etching procedures was that the ratchets required much finer features, but in a smaller area, which necessitated an adjustment of the laser lithography parameters. Problems were encountered on these small scales since this size is close to the resolution limit of the lithography apparatus. The shape of patterns was therefore distorted by both the lithography and etching processes, as discussed in section 6.4.2.

Secondly, this was a partial-thickness etching (or partial-etch), meaning that the aim was to ablate YBCO from the sample to a well-defined depth that was less than the thickness of the sample. The YBCO would therefore be reduced in thickness in the etched areas rather than being completely removed.

The procedure for partial thickness etching was created specifically for the production of ratchet samples, since no previous work had been done by the TFT group requiring samples to be thinned in some areas. In order to successfully partially etch samples, the total etching time was reduced in an optimisation procedure that will be described in section 6.4.1.

6.2.3 Alignment

While the alignment of the sample during the lithography process (as described in section 3.2.2) only needed to be roughly accurate for singly-patterned samples, it became crucial when a second patterning was required. If the sample was not properly aligned for either the initial pattern or the secondary ratchet patterning, then the ratchet pattern might be drawn at some distance from its intended location.

There were two cases in which the dislocation of ratchet patterns were found to lead to differences in function:

- Whenever a significant part of the ratchet pattern was shifted outside the sample edge, the area of the ratchet part would be less than intended. However, such a change in the ratchet area was never found to affect the results in any significant way.

- If instead the ratchet pattern was shifted toward the sample centre, an unwanted gap may be created between the sample edge and the beginning of the ratchet area. Even a small gap between the sample edge and the ratchet area was seen to have a huge impact on vortex penetration in the vortex lensing geometry. The small strip of sample area along this edge would significantly impede vortex penetration due to the stronger edge barrier in the thicker part of the sample.

For this reason, sample alignment was ensured more thoroughly during the laser
lithography procedure for ratchet samples. The tolerance for misalignment of the $x$– and $y$–axes was therefore reduced, and corrections to alignment were often necessary prior to commencing lithography.

### 6.3 Quality Control and Testing

Several testing steps were incorporated into the ratchet fabrication procedure to ensure that the films and patterns produced were of high quality. Firstly, the quality of the superconducting film was tested using MPMS, then optical microscopy was used to ensure that films were patterned correctly, and finally profilometry was used to check the depth of partial etching.

The optical microscopy and profilometry tests were directly linked to the optimisation procedures discussed in section 6.4.

#### 6.3.1 MPMS

The first part of the quality control procedure for ratchet fabrication was testing of the superconducting properties of the un-patterned YBCO films using a Quantum Design Magnetic Property Measurement System (MPMS). This testing was carried out in the same way as was standard for every YBCO sample produced by the TFT group, as described in section 3.4.1.

#### 6.3.2 Optical Microscopy

An optical microscope in the clean room was used as the main quality check for the ratchet patterning part of the procedure.

The films were examined under the microscope at several points during the fabrication procedure:

- After the laser lithography procedure but before the initial etch, to check that the pattern had been applied correctly by the laser. This check could often be skipped as there was rarely any issue with the initial lithography.

- After the initial etch, to ensure that all unwanted YBCO was removed from the unwanted areas.

- After laser lithography but before the secondary partial etch, to check that the pattern had been applied correctly by the laser. Issues arose whenever the laser was not in focus, as discussed in section 3.3.2.

- After the secondary etch, to ensure that some YBCO remained in the etched areas (an unintentional full etch would be visible as a difference in contrast).
Figure 6.6: Microscope images of a linear protrusion ratchet pattern applied lithographically to the surface of a YBCO sample (a) before etching and (b) after an unsuccessful secondary etch - dark channels show that the sample is fully etched. The nominal width of each channel is 5µm, magnification is 50×.

Figure 6.6 shows the difference between microscopy images taken before and after an etching procedure. Part (b) of this figure shows that the sample was etched for too long. The dark areas within the channels are the visible substrate. A sample that is partially etched to the correct depth has much less contrast between the channels and the surrounding YBCO, as in figure 6.9(b).

6.3.3 Profilometry

The depth of the secondary etch was tested for each ratchet sample, using a Dektak Profilometer. This was important to determine whether the secondary etch had successfully removed only part of the thickness of the YBCO. The profilometry data for various etch times was used to determine the etching rate, as discussed in section 6.4.1.

Figure 6.7 shows profilometry results for a 200nm YBCO film etched at 400kV, 35mA for 20 minutes (these parameters are explained in section 3.2.3). This etch time was too long, and the etch depth was seen to be much greater than the sample thickness.

6.4 Optimisation of Ratchet Patterning

The majority of work carried out on these novel vortex ratchets has been devoted to the optimisation of their construction. Testing was necessary to determine the optimal etching parameters for the new partial-thickness etching procedure.

The first important parameter in the partial etching procedure was the etch depth, which was mainly controlled by varying the total etch time. Secondly, since the patterned features of the ratchet pattern were quite small, it was necessary to
 optimise the shape of the weak-pinning channels to ensure that the final etched channel matched what was desired.

6.4.1 Depth Optimisation

The production of vortex ratchet devices presented a new challenge: a partial-thickness etching had not previously been attempted. The standard ion-beam etching procedure (described in section 3.2.3) was developed to completely remove the sample and even ablate part of the substrate in exposed areas, to ensure that no trace of the superconducting material remained there. By contrast, the desired partial-etch procedure was required to be much more precise, to remove only part of the exposed YBCO. The depth of etching (defined as the reduction in thickness of sample and/or substrate material in exposed areas) therefore became a crucial parameter. An optimisation procedure was undertaken to ensure that samples could be effectively partial-etched to a desired depth.

The most important independent variable in the etching procedure was the total etch time, which naturally had a strong correlation with etch depth. In order to test this correlation, the etching procedure was applied to many YBCO samples for varying etch times. To properly emulate the conditions for ratchet etching, each of these samples had already been subject to an initial etch. One result is shown in figure 6.7, for which the etch time was clearly much too long.
The etch depth could be determined using a simple profilometry procedure, and the rate of ablation of YBCO could then be determined by plotting total etch time against etch depth. For simplicity, the STO substrate was assumed to ablate at the same rate as YBCO since their crystal structures and ablation properties are similar. This allowed the data for etch depths greater than the sample thickness to be included in the determination of ablation rate.

After a lengthy testing phase, samples were successfully etched to depths in the range of 50-100\(\mu\)m. The etch time required to achieve this depth was approximately 5 minutes. To etch for such a short duration was a significant change to the existing etching procedure (for complete etching), and therefore more precise timing of the ion beam exposure than had previously been done.

Further optimisation will be required in order to determine what etch depth (and therefore etch time) is optimal for pinning enhancement while avoiding damage to the un-etched weak-pinning channels.

### 6.4.2 Shape Optimisation

Since the pattern etched into a sample never perfectly matches the original pattern drawn in the Layout Editor program (see section 3.3.2), the specific shape of the channels in the new ratchet geometries was varied in several ways in order to optimise patterning and rectification. The parameters were first optimised to produce asymmetric channels that maintained a suitable shape after etching, and then optimised for rectification.

![Figure 6.8: Examples of the (a) offset-sawtooth and (b) linear protrusion ratchet patterns in the Layout Editor program, with design parameters indicated as defined in the text.](image)

The variable parameters in the new ratchet patterns are defined as follows:

- \(W\): Channel width. Typically 3-5\(\mu\)m.
• \( P \): Period. Typically 10-20\( \mu \)m.

• \( b \): Bottleneck width (defined as the gap between the end of a protrusion and the opposite wall). Typically 0.5-2\( \mu \)m. \( b \) must always be less than half of \( W \) to avoid the zero temperature flaw discussed in section 6.1.1.

• \( \theta \): Angle of protrusion. Typically 30-45°.

• \( s \): Size of protrusions (defined as thickness of linear protrusions, or length of the flat top on sawtooth protrusions). Typically 0-1\( \mu \)m for offset-sawtooth, or 0.5-1\( \mu \)m for linear protrusion.

Other important parameters were not independently chosen, but are uniquely defined from those given, such as the length of protrusions and the distance between the end of one protrusion and the start of the next.

The shape of the lithographically patterned channel is altered in several ways by the etching procedure when it is used to pattern very fine features. The mechanisms for this are:

• The laser spot size of \( \sim 1\mu \)m means that sharp features will be rounded in the laser lithography process. This can be seen at the ends of the protrusions in figure 6.6(a). It may also increase the size of small features, which could have been the cause of the issue seen in figure 6.9(b).

• Some photoresist at the very edges of the unexposed areas may be removed or thinned during development, which would effectively widen the channels and shrink the protrusions.

• High energy ions may ablate the edges of the protected areas during etching, especially if the photoresist has been thinned at these edges. This would also widen the channels and shrink the protrusions, as seen in figure 6.9(a) where the protrusions only just cross the central line.

All parameters were varied over many etched samples, and the final etched patterns were checked visually for alterations from the original pattern. The etched patterns often showed strong deviation from the original, and even before rectification was tested, many were eliminated since they could not function as effective ratchets. For example: the protrusions may become too thin, which would allow vortices to pass straight through; the bottlenecks may become too small and not allow vortices to move from one cell to the next; the protrusions may shorten and no longer cross the central line, allowing the zero temperature flaw (defined in section 6.1.1) to take effect.

As seen in figure 6.9, the quality of the pattern after etching is clear from visual observations under the microscope. The sample shown in (b) was etched first, with
Figure 6.9: Optical microscope images of 25µm square regions of two ratchet-patterned YBCO films that were produced for shape optimisation: (a) A successfully-etched offset-sawtooth pattern. Some rounding of sharp features is observed. (b) An unsuccessfully-etched linear protrusion pattern, where most bottlenecks have completely severed the channel. The thicker parts of the film appear darker and less smooth in both images.

\[ W = 3\mu m, \ b = 1\mu m, \ L = 11\mu m, \ \theta = 30^\circ. \] The ratio of \( b \) to \( W \) was seen to be too large, since the effective width of the bottlenecks is seen to reduce to zero after etching. The sample shown in (a) was etched later, with altered parameters \( W = 5\mu m, \ b = 2\mu m, \ L = 16\mu m, \ \theta = 30^\circ. \) This is the same sample that was shown in figure 6.6.

It can also be seen from the contrast in figure 6.9 that the sample shown in (b) is successfully etched to partial depth while the sample in (a) is not. This is because shape optimisation and depth optimisation were carried out simultaneously.

Through many tests, the shape of the asymmetric channels was successfully optimised for etching of a viable ratchet pattern.

6.4.3 Variations to Etching Method

There are many methods for producing vortex ratchets, as discussed in section 2.4.1. The two novel ratchet geometries described in this chapter have shown great potential for rectification, especially in the simulation results. These novel ratchets could be produced by many different means. In future, the partial etching procedure described here may be refined or modified, or ratchets may be produced using a completely different method.

The difference in pinning strength between the etched and un-etched regions could also be increased by depositing a two-layered superconductor using two materials with different pinning properties. This is discussed in the subsection below.
Superconducting Bilayers

The difference in pinning strength between the weak-pinning channels and strong-pinning surrounding regions could be amplified by using a superconducting “bilayer” sample [62]. An appropriate bilayer sample could be constructed by first depositing a layer of very weak-pinning superconducting material onto the substrate (using the PLD method described in section 3.1), then epitaxially depositing a layer of strong-pinning superconducting material on top of this.

This would mean that the partial-etching procedure would only be required to remove the strong-pinning layer in order to etch out weak-pinning channels. In this case, the etched regions would have weaker pinning than the un-etched regions, which is the reverse of the current situation. The difference in pinning strength between the strong- and weak-pinning regions could be increased greatly using this method.

Vortex diodes based on superconducting bilayers have previously been produced by Yu et al. [62] using the mirrored sawtooth design described in section 6.1.1. This group used NbN for the strong-pinning layer, and NbGe for the weak-pinning layer.

Energy barriers may be formed at the edges of the channel by choosing materials with a difference in penetration length for use in the bilayer [21]. The reason for this is that circulating currents are distorted when entering a superconducting region with a different penetration length [43, 170]. For vortices in the superconducting region with larger $\lambda$, this leads to a force directed away from the interface. Therefore, the weak-pinning material chosen for the channel should also ideally have a longer penetration length than the strong-pinning material, in order to increase the repulsive force from the edges of the channel and the protrusions. In this case, a repulsive force would exist on any vortices in the channel even if vortices were not present in the surrounding material.

It is therefore possible that superconducting bilayers could be used to create 2D asymmetric weak-pinning channel diodes with much more favourable properties than the current partial thickness etching method. This has not yet been undertaken, but will be a focus of future work in this area.

6.5 Results So Far

While this study into vortex diodes is still incomplete, it has shown some promising preliminary results: The novel ratchet patterns were successfully applied to YBCO samples through partial-thickness etching, (as discussed in section 6.4.2). However, magneto-optical images of the initial test samples failed to show a vortex lensing effect in a penetration test, as discussed in section 6.5.1. However, the vortex diode
The vortex diode effect was in fact tested using vortex dynamics simulations, and the feasibility of the novel ratchet patterns was confirmed [21]. A simulated linear protrusion pattern was found to give good rectification of vortex motion under specific field conditions, as discussed in section 6.5.2.

6.5.1 Magneto-Optical Imaging and Video

Ratchet-patterned samples were tested in a vortex lensing geometry using magneto-optical imaging and video. The vortex lensing geometry is explained in section 6.2.1. It was expected that vortices would penetrate faster and to a greater depth on those edges that had an inward-facing easy direction, but this lensing effect has not yet been observed.

Several different vortex lensing geometries were tested, each having ratchets patterned on different parts of the edges of the sample.

![Figure 6.10: One frame of magneto-optical video of a vortex lensing sample. The ratchet patterned areas are visible as lighter squares close to the edges, with red lines showing field profile positions and red arrows showing easy direction. No difference in final penetration depth is seen. Ratchet patterns are also present on the left and right sides of the sample, but no results could be taken from these due to a large horizontal scratch across the sample centre.](image)

From the results shown in figure 6.10 and similar measurements, the penetration depth from each edge of the sample with ratchet patterned areas was seen to be increased in comparison to adjacent unpatterned edges. This was expected due to the reduced thickness of the strong-pinning regions surrounding the channels.
However, the penetration depth was seen to be almost identical for ratchet patterned areas with inward-facing and outward-facing easy directions. The time evolution of field profiles taken along these ratchets also showed no significant difference. Rectification of vortex motion during penetration was therefore not confirmed in these MOI/MOV measurements.

A strong suggestion for the reason that rectification was not observed in these measurements is that they were zero-field-cooled. Therefore the strong-pinning parts of the ratchet pattern would have had no vortices present when field first began to penetrate, and no ratchet potential would exist for vortices in the weak-pinning channels. Since vortices most likely penetrate into the thinner (strong-pinning) parts first, no ratchet effect would be seen until the field was large enough to fill the strong pinning parts of the pattern completely. Only then would additional vortices to begin moving through the weak-pinning channel. The field dependence of rectification in the ratchet patterns is further discussed in section 6.5.2.

The non-uniformity of the vortex density in the zero-field-cooled case would also lead to uncontrolled effects, and possibly prevent the onset of rectification until still higher fields were reached. In future, similar measurements will be undertaken for field-cooled samples.

### 6.5.2 Simulation

The linear protrusion geometry was tested under simulated conditions in collaboration with Stephen Wilson [21], using the vdsim program developed by Hans Fanghor [20, 171]. The motion of vortices in a specific 2D potential was simulated, where vortices were treated as rigid massless particles moving in a uniform viscous fluid, subject to a number of forces.

The results showed that this ratchet potential did provide rectification of vortex motion, even complete rectification (where no vortex motion was seen in the hard direction) under specific circumstances.

#### Simulation Parameters

Given the initial position of each vortex, its position after a time-step $\Delta t$ was determined by calculating the resultant of a number of forces.

The forces on vortices were: pinning force, vortex-vortex repulsion, viscous drag and the Lorentz (Magnus) force. The origin and the mathematical form of each of these forces is given in section 2.3. For simplicity, simulations were performed in the absence of thermal fluctuations (at $T = 0K$). The most significant force for creating

---

bSee section 2.5.1
the ratchet potential in this simulation was vortex-vortex repulsion. The equation used for the repulsion force between pancake vortices in YBCO films was \[21]:

\[ F(r) = \frac{2\Phi_0^2}{4\pi\mu_0\lambda^2} K_1 \left( \frac{r}{\lambda} \right) \quad (6.1) \]

Where \( r \) is the separation between vortices, and \( K_1 \) is the modified Bessel function of the second kind.

The pinning potential was pre-defined for each simulation. Weak-pinning channels are often simulated using strong potential barriers at each edge of the channel to represent the effect of repulsion from pinned vortices at the channel edges [60–62]. This technique was not used in this case.

Instead, strong pinning centres in the shape of isolated dots were placed throughout each of the strong pinning regions - i.e. along the edges of the channel walls and along the protrusions. This allowed for a more realistic simulation in which the ratchet potential was created by vortex-vortex repulsion, making it a true interstitial ratchet as discussed in section 2.4.1. This also allowed for other effects such as de-pinning of the strongly-pinned vortices in the walls and protrusions, as well as “leaking” of free vortices between the walls. However, in an experimental study, the pinning potential would be high throughout a continuous region rather than in isolated dots.

![Figure 6.11](image)

**Figure 6.11:** (a) One unit cell of the pinning potential used for the ratchet simulation. The dark spots represent strong pinning, and white area has no pinning. (b) The potential energy landscape created by vortex-vortex repulsion if each pinning site is occupied by one pinned vortex. Lighter areas represent higher repulsion potential.

Figures reproduced from [21] with permission from S. Wilson.

The ratchet parameters (as defined in section 6.4.2) used for the simulation were: \( W = 0.35\mu\text{m}, \ P = 2W = 0.7\mu\text{m}, \ b = 0.4 \ W = 0.14\mu\text{m}, \) and \( \theta = 45^\circ. \) The lateral size of protrusions \( s \) was not defined in the simulation case, since only a single line of pinned vortices was used.

Each simulation had a single vortex placed in each pinning site, and a variable number \( n_f \) of “free” vortices per unit cell placed in the channel, far from the pinning sites. Simulations were run for \( n_f = 2, 4, 6, 8, \) and 16, which correspond to
applied field values $B_a = 0.072T, 0.076T, 0.080T, 0.084T$ and $0.101T$ respectively (determined from total vortex density). These field values are comparable to fields that would be applied experimentally using the MOI magnetic field system (see section 4.1.2). Each simulation was run once for current in the forward direction, and then repeated for current in the reverse direction.

Simulation Results

![Vortex Response to Applied Currents](image)

**Figure 6.12:** Vortex motion in response to applied current, in ratchet simulations for varying $n_f$. Vortex velocity is positive in the easy direction, and current is positive in the reverse direction. Figure reproduced from [21] with permission from S. Wilson.

The results are shown in figure 6.12. The vortex velocity response is asymmetric in all cases, showing rectification in vortex motion. Critical current in each direction was determined by finding the onset of vortex motion. This was determined using a linear fit to the high current part of each response curve, then taking the “Easy $J_c$” (critical current for vortex motion in the easy direction) as the x-intercept on the right side, and “Hard $J_c$” (critical current for motion in the hard direction) as the x-intercept on the left side.

For each field, the rectification factor $R$ was taken as the ratio of these two critical currents. Rectification was seen in all cases, with a peak in $R$ at 0.076 T (corresponding to $n_f = 4$). The rectification factor at this optimal field was $R = 3.5 \pm 0.3$, this can be seen through the large asymmetry in the orange curve in
CHAPTER 6. VORTEX RATCHETS

Some noise is seen in the results, and this arises from the oscillation of pinned vortices around the centres of their pinning sites. However, the pinning strength was chosen to be large enough that these vortices never left their pinning sites.

Conclusion

For all simulation results, the critical current was greater for vortex motion in the hard direction. This shows that the linear protrusion geometry can indeed be used to create a functional ratchet potential. An alternating current applied to a diode created using this ratchet geometry would therefore lead to a net vortex motion in the easy direction, experiencing a greater resistance in this direction. If the applied current were larger than \( J_c \) in the easy direction yet smaller than \( J_c \) in the hard direction, the forward resistance would be zero, but reverse resistance would be large. This is the ideal result for a vortex diode.

Expected Results Based on Simulation

Based on the simulation results, a direct measurement of rectification in similar physical ratchet devices would be expected to show a large rectification factor \( R \) over a range of applied fields.

A peak in rectification at some optimal field is expected for experimentally investigated linear protrusion ratchets, as was seen in simulation. The physical origin for the optimal field in the physical case is that lower fields would result in few to no free vortices existing in the channels (since they would fill the strong-pinning regions first), while higher fields would result in too many free vortices causing a reduction in rectification as seen in simulation.

Some deviation of experimentally measured results from the simulation results would be expected, for a number of reasons:

- Firstly, the temperature would be non-zero in the experimental case, causing thermal fluctuations to influence the behaviour of free and pinned vortices. This may lower \( J_c \) for the hard direction, since thermal activation may help the vortices through the bottlenecks.

- Free vortices in the channels would not be expected to leak through the gaps between pinned vortices as they did in simulation. This is because the protrusions do not really consist of isolated dots of high pinning strength, but continuous high-pinning regions. It is unclear whether rectification would be increased since leaking would not occur, or decreased since free vortices may
enter the protrusions and become pinned. If a free vortex did enter a pro-
trusion, the increased vortex density may then force other pinned vortices to
escape into the channel and become free.

Expanding on the last point: it would be possible to experimentally produce
ratchet devices that did have roughly the same pinning landscape as used in the
simulations. This could be achieved by the patterning of antidots in each desired
pinning location.

Therefore the peak rectification factor for the experimental case cannot be ac-
curately predicted due to the differences between simulated conditions and the real
case. Neither can the optimal field value or the experimental field dependence of
the rectification factor, but a qualitative similarity to simulation is expected.
Chapter 7

Additional Magnetic Microscopy Experiments

A variety of collaborative works were undertaken during the course of this research, some of which were not related to the major investigations. These smaller investigations also produced noteworthy results, and are therefore listed in this chapter.

Section 7.1 details the mathematical and experimental analysis of a correction procedure for MOI that was suspected to be invalid [131, 135]. Then section 7.2 discusses half-integer flux quanta in superconducting rings, and their investigation through scanning SQUID microscopy. The focus is on my contribution to these studies, which extended beyond simply taking measurements, but also involved analysis and interpretation of data, discussion of results and editing of publications.

7.1 Critical Analysis of In-Plane Field Correction

In-plane fields can have a spurious effect on magneto-optical images and therefore on the current calculated from these, as discussed in section 2.7.2. An iterative procedure proposed by Laviano et al. to correct for in-plane fields [135] was analysed mathematically and experimentally. The mathematical validity of this procedure was confirmed, though it is yet to be confirmed experimentally.

Though magnetic field is applied solely in the z-direction during MOI, in-plane magnetic fields exist at the position of the indicator film due to the bending of field around the superconducting sample by the demagnetisation effect (see section 2.3.1). To account for this phenomenon, a technique proposed by Laviano et al. was applied to current maps of the sample [135]. This technique involves computing the $x$ and $y$ components of the magnetic field from the calculated current in the film and re-calculating currents from the new field distribution in an iterative procedure.

The accuracy of such a correction technique is vital to the practice of MOI. If
accurate, it could help to avoid the calculation of incorrect and misleading current distributions, and lead to the reconstruction of higher precision images of local electrodynamic quantities, and hence more accurate determination of properties such as local current-carrying capacity [107, 108, 110, 135, 172].

An issue with the correction procedure was first noticed during an attempt to apply this procedure to MO images. At that time, it was shown through experimental results and subsequent mathematical analysis that this technique was not capable of producing physically accurate results [80]. However, the initial analysis was not mathematically rigorous, and an improved analysis was recently undertaken, as shown in [131].

This re-examination shows that the in-plane correction procedure outputs values for current that lie within a reasonable range, though it cannot analytically prove the technique’s accuracy. The procedure was also tested experimentally on several magneto-optical images of superconducting samples in order to experimentally verify the inaccuracy of this technique, and the values were seen to become un-physically large after only a few iterations.

7.1.1 Origin and Effect of In-Plane Field Components

In detecting the magnetic field around superconducting samples, the magneto-optical imaging technique measures the local z-component of the magnetic field at each point in a Faraday-active indicator film, which is placed at a finite height \( h \) above the sample [41, 63, 80, 107]. However, there also exist in-plane fields \( (B_{xy}) \), which contribute to the currents created within the sample. The indicator film is also sensitive to these field components. Such in-plane fields are caused by the bending of the imposed magnetic field around the sample due to the demagnetising effect, and due to the stray fields from each vortex bending outward as they reach the height of the indicator film.
Figure 7.1: Bending of imposed magnetic field around a superconducting sample due to the demagnetising effect and spreading of the stray field above vortices. Figure used with permission by Jack Zuber.

Figure 7.2: When there is no applied field in the $z$-direction the spontaneous magnetisation vector lies in the plane of the film. A non-zero $z$-component of the applied field perturbs the magnetisation vector by an angle $\phi$, giving it a non-zero $z$-component. Figure used with permission by Jack Zuber.

Figure 7.1 shows that in-plane fields $B_{xy}$ exist at the measurement height since the direction of the magnetic field lines are at some angle to the $z$-axis.

When a magnetic field is present at a point on the indicator film, the local magnetic moment is perturbed by an angle $\phi$ as shown in figure 7.2. In this geometry, the interaction energy, $E_{int}$, of the indicator film in presence of an applied magnetic
field with a non-zero z-component can be written in the following form [135]:

\[ E_{\text{int}} = E_A(1 - \cos \phi) + B M_s(1 - \cos(\alpha - \phi)) \]

(7.1)

where \( E_A \) is the anisotropy energy of the indicator film, \( M_s \) is the magnitude of its spontaneous magnetisation vector and \( \alpha \) is the angle between the magnetic induction \( B \) and the plane of the film.

To find an expression for the equilibrium magnetisation, an angle \( \phi \) is determined such that the interaction energy is a minimum [107]. Hence, computing the derivative of Eq. (7.1) with respect to \( \phi \) and showing it has second derivative always positive gives:

\[ \phi = \tan^{-1} \left( \frac{B_z}{B_A + B_{xy}} \right) \]

(7.2)

Here \( B_A = \frac{E_A}{M_s} \) is the magnetic anisotropy field, and \( B_{xy} = \sqrt{(B \cdot \hat{x})^2 + (B \cdot \hat{y})^2} = B \cos \alpha \).

The polarisation of light incident on the indicator film is rotated by the Faraday effect. In the MOI setup, polarised light is applied to the indicator film parallel to the film’s optical axis (in the z-direction). Therefore, the angle of rotation \( \alpha_F \) is proportional to the z-component of the magnetisation, which gives the Faraday rotation law as:

\[ \alpha_F = C M_s \sin \phi \]

(7.3)

where \( C \) is an experimental constant related to the Verdet constant but also dependent on the thickness of the indicator film’s active layer. Equation 2.9 is a simplification of this equation, which takes \( \sin \phi \approx B_z / B_A \) for negligible \( B_{xy} \).

The intensity at the analyser in the equilibrium position in a magneto-optical imaging apparatus is found by substituting eqs. (7.2) and (7.3) into (2.10), which gives a more complex dependence of light intensity on field:

\[ I = I_0 + I_{\text{max}} \cos^2 \left( \frac{C M_s B_z \sqrt{(B_A + B_{xy})^2 + B_z^2}}{\sqrt{(B_A + B_{xy})^2 + B_z^2}} + \theta \right) \]

(7.4)

In most magneto-optical experiments (including the majority of those in this thesis), \( B_{xy} \) is taken as zero in this equation for simplicity.

However, this assumption may lead to an interpretation of MO data that is quantitatively inaccurate [63, 108]. For example, magneto-optical measurements have shown unwanted current peaks near the edges of a YBCO strip, which probably arose due to in-plane fields [107].

Laviano et al. state that this causes un-physically higher electrical current (increasing with sample thickness) to be observed [135]. Therefore, the in-plane correction procedure was proposed [135] to form a relationship between the apparent
magnetic field at the detector which is obtained with the usual assumption that \( B_{xy} = 0 \) and the “actual” field at the detector including the in-plane effects.

7.1.2 Mathematical Analysis of the Correction Procedure

This section first details the mathematical derivation of the in-plane correction, then critically analyses the resulting iterative procedure to show that it converges to a physical value within a reasonable range.

Some equations in this section differ marginally from those given in [135], since insufficient information was given to follow the literature derivation precisely.

The derivation begins with equation (7.4), taking an expression for \( B_z \):

\[
\frac{B_z}{B_A + B_{xy}} = \tan^{-1} \left( \frac{\cos^{-1} \left( \frac{l/l_{max}}{CM_s} \right) - \theta}{\sin \theta} \right),
\]

(7.5)

Defining \( B_z^{(0)} \) as the value of \( B_z \) under the assumption that \( B_{xy} = 0 \) (for reasons that will become clear), this equation becomes:

\[
\frac{B_z^{(0)}}{B_A} = \tan^{-1} \left( \frac{\cos^{-1} \left( \frac{l/l_{max}}{CM_s} \right) - \theta}{\sin \theta} \right),
\]

(7.6)

Combining eqs. (7.5) and (7.6) gives:

\[
B_z = \left[ 1 + \sqrt{B_x^2 + B_y^2} \right] B_z^{(0)}
\]

(7.7)

This neatly-presented equation is used in an attempt to produce better approximations of \( B_z \) from known \( B_z^{(0)} \), \( B_x \) and \( B_y \) values while taking \( B_A \) as a constant. It was proposed [135] that an iterative procedure could be undertaken, with successive values of \( B_z \) given by:

\[
B_z^{(n+1)} = \left[ 1 + \sqrt{B_x^{(n)^2} + B_y^{(n)^2}} \right] B_z^{(0)}.
\]

(7.8)

This equation was used along with the following algorithm, in an attempt to reach convergence of calculated \( B_z \) values with the physical field:

(i) Use \( B_z^{(n)} \) to find \( J_x^{(n)}(x,y) \) and \( J_y^{(n)}(x,y) \) using the inverse Biot-Savart equations (2.19 and 2.20), beginning with \( J_x^{(0)}(x,y) \) and \( J_y^{(0)}(x,y) \) from \( B_z^{(0)} \);
(ii) Calculate \( B_x^{(n)}(x,y) \) and \( B_y^{(n)}(x,y) \) from \( J_x^{(n)}(x,y) \) and \( J_y^{(n)}(x,y) \);
(iii) Calculate \( B_z^{(n+1)} \) from \( B_z^{(n)}(x,y) \), \( B_x^{(n)}(x,y) \) and \( B_y^{(n)}(x,y) \);
(iv) Assign \( B_z^{(n+1)} = B_z^{(n)} \), then repeat from step (i), continuing until the difference
between $B_z^{(n+1)}$ and $B_z^{(n)}$ is less than some acceptable threshold.

Note that the new field values $B_z^{(n+1)}$ are always calculated from the original $B_z^{(0)}$, not from the field values at the previous iteration. A misreading of [135] may lead to replacement of $B_z^{(0)}$ with $B_z^{(n)}$ in equation (7.8), but this does not follow from (7.7) & (7.6) and the calculated $B_z$ was found to diverge with increasing iterations if this mistake was made.

Equation 7.8 is the final result given, along with the above procedure, in [135]. The resulting sequence was examined for convergence.

To complete step (ii) of the algorithm proposed above, equations for $B_x$ and $B_y$ in terms of $B_z^{(0)}$ are determined using the inversion techniques previously presented:

These are obtained by considering the 2D Biot-Savart inversion problem, but solving for $B_z$ and taking into account the finite height of the detector:

$$B_z(x, y, z, h) = \frac{\mu_0}{4\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{J_y(x', y')(h - z')dx'dy'dz'}{((x-x')^2 + (y-y')^2 + (h - z')^2)^{3/2}}.$$ (7.9)

This equation can be Fourier transformed with an application of the convolution theorem to gain an integral kernel which is very similar to that used to invert the Biot-Savart law (see equation A.18, in appendix A). Hence, the Fourier transform of (7.9) can be written as:

$$\tilde{B}_x(k_x, k_y, h) = \frac{\mu_0}{2\pi} \int_{-\infty}^{\infty} \frac{J_y(x', y')(h - z')}{h - z'} \frac{\sinh(\frac{k_d}{2})}{k} dk,$$ (7.10)

and this is integrated to find:

$$\tilde{B}_x(k_x, k_y, d, h) = \frac{\mu_0 e^{ikh}}{k}\tilde{J}_y(k_x, k_y) \sinh\left(\frac{k_d}{2}\right),$$ (7.11)

Employing a similar process for $\tilde{B}_y$:

$$\tilde{B}_y(k_x, k_y, d, h) = -\frac{\mu_0 e^{ikh}}{k}\tilde{J}_x(k_x, k_y) \sinh\left(\frac{k_d}{2}\right),$$ (7.12)

Using the inverse Biot-Savart equations (2.19 and (2.20), (7.11) and (7.12), simple expressions for the Fourier transforms of $B_x$ and $B_y$ are given in terms of $B_z$:

[135]

$$\tilde{B}_x = \frac{ik_x}{k} \tilde{B}_z^{(0)},$$ (7.13)

$$\tilde{B}_y = \frac{ik_y}{k} \tilde{B}_z^{(0)}$$ (7.14)
Since the iterative procedure is not carried out in Fourier space, these relations were converted into Cartesian space for the analysis: \[131\]

\[
B_x(x, y) = \int_{-\infty}^{\infty} \frac{ik_x}{\sqrt{k_x^2 + k_y^2}} B_z^{(0)}(k_x, k_y) e^{i(k_x x + k_y y)} dk_x dk_y
\]  
(7.15)

Applying the convolution theorem in reverse gives:

\[
B_x(x, y) = \int_{-\infty}^{\infty} B_z^{(0)}(x', y') \mathcal{F}^{-1} \left\{ \frac{ik_x}{\sqrt{k_x^2 + k_y^2}} \right\} dx' dy',
\]  
(7.16)

where \(x'\) and \(y'\) are arbitrary points on the film and the inverse transform will map \(k_x \to x - x'\) and \(k_y \to y - y'\). Considering the inverse Fourier transform shown above:

\[
\mathcal{F}^{-1} \left\{ \frac{ik_x}{\sqrt{k_x^2 + k_y^2}} \right\} = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{ik_x}{\sqrt{k_x^2 + k_y^2}} e^{i(k_x x + k_y y)} dk_x dk_y
\]  
(7.17)

Changing to a polar co-ordinate system simplifies this equation to:

\[
\mathcal{F}^{-1} \left\{ \frac{ik_x}{\sqrt{k_x^2 + k_y^2}} \right\} = \frac{1}{2\pi} \frac{\partial}{\partial x} \int_{0}^{\infty} J_0(kr) dk
\]  
(7.18)

Where \(J_0\) is the Bessel function of the first kind. By letting \(u = kr\) and noting that at all Bessel functions are normalised, this becomes:

\[
\mathcal{F}^{-1} \left\{ \frac{ik_x}{\sqrt{k_x^2 + k_y^2}} \right\} = -\frac{1}{2\pi} \frac{x}{(x^2 + y^2)^{3/2}}
\]  
(7.19)

Hence, the expressions for \(B_x\) and \(B_y\) in Cartesian space are determined from 7.13 and 7.14 to be:

\[
B_x(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} -\frac{1}{2\pi} \frac{x - x'}{((x - x')^2 + (y - y')^2)^{3/2}} B_z^{(0)}(x', y') dx' dy'
\]  

\[
B_y(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} -\frac{1}{2\pi} \frac{y - y'}{((x - x')^2 + (y - y')^2)^{3/2}} B_z^{(0)}(x', y') dx' dy'
\]  
(7.20)

These calculations were therefore verified, not showing the suspected divergence from physical values. The equations, as derived here, were then tested experimentally to determine their viability.
7.1.3 Experimental Verification

The in-plane correction technique proposed in [135] was suspected to produce inaccurate results. In order to test this experimentally, the technique was applied to Magneto-Optical images of a $3 \times 3$ mm thin YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) film that had previously been acquired using the MOI technique and apparatus described in chapter 4.

The resultant images were converted into current maps using Biot-Savart inversion (as discussed in section 2.7.1), and the Laviano procedure (using equation 7.8) was applied to give each “corrected” magnetic field profile, taking the anisotropy field to be $B_A = 80$ mT [107]. Each field profile was converted into the next iteration of the “corrected” current profile, following the algorithm given in the previous section.

However, in many cases, it was not possible to produce an accurate corrected image, due to the amplification of artefacts with each iteration.

7.1.4 Conclusion

Analysis has shown that in-plane correction procedure appears mathematically valid, but useful results have not yet been obtained experimentally.

Unfortunately, it was impossible to reproduce identically the derivation given in [135] due to insufficient detail provided in some steps. Despite this, no problem was identified with the mathematical derivation of the in-plane field correction. Issues were noted in the experimental case, however, with the amplification of artefacts.

Several alternatives to the presented in-plane correction technique may be considered, in order to avoid these issues: Firstly, the magnetic anisotropy field could be taken as non-constant across the sample due to its micro-geometry. Alternatively, the in-plane fields could be intrinsically included in the solution to the inverse problem, giving a modified inversion procedure that avoids iterative calculations [130].

7.2 SSM of Superconducting Rings with Half-Integer Flux Quanta

The flux in superconducting rings consisting of mixed s-wave and d-wave superconductors was investigated through scanning SQUID microscopy. For certain geometries, a spontaneous magnetic flux could be induced in the rings with a value that was only half of the magnetic flux quantum [124, 173]. By increasing the field in these rings, the flux was increased to higher integer and odd half-integer quantum values.
CHAPTER 7. ADDITIONAL MAGNETIC MICROSCOPY EXPERIMENTS

The superconducting rings were produced by Prof. Ariando, SSM images were acquired by myself and Dr. X. Renshaw Wang, data analysis was undertaken by Andre Timmermans and Robin Bruel, under the supervision of Prof. Hans Hilgenkamp at the University of Twente [173].

Section 7.2.1 below explains the theory behind the emergence of half-integral flux quanta, then section 7.2.2 describes their observation through SSM.

7.2.1 Theory of Half-Integer Flux Quanta

In an ordinary superconducting ring, the magnetic flux is quantised. This can be shown using the second London equation:

$$j_s = \frac{1}{c\Lambda} \left( \frac{\Phi_0}{2\pi} \nabla \theta - A \right)$$  \hspace{1cm} (7.21)

If the ring is significantly wider than the penetration depth of the superconductor, then the current will only flow in a region of width around its edge. Therefore we can find a contour within the ring for which $j_s = 0$. Integrating over such a contour gives:

$$\Phi_0 \oint \theta \cdot dl = \oint A \cdot dl \equiv \Phi$$  \hspace{1cm} (7.22)

The right hand side is equal to the magnetic flux $\Phi$ through the ring by definition of the vector potential $A$. The order parameter must be continuous around the ring (it forms a standing wave). Therefore

$$\oint \theta \cdot dl = 2\pi n$$  \hspace{1cm} (7.23)

and so from equation 7.22 it can be seen that

$$\Phi = n\Phi_0$$  \hspace{1cm} (7.24)

Thus flux is quantised in multiples of $\Phi_0$ through a regular superconducting ring. In the absence of external field, the ring is in the ground state with zero flux, and when an external field is applied, the flux through it takes only integral values of $\Phi_0$.

However, in a superconducting heterojunction, which is the interface between a conventional s-wave superconductor and a d-wave superconductor, the superconducting order parameter may undergo a $\pi$ phase change. Thus

$$\oint \theta \cdot dl = 2\pi (n + \frac{1}{2})$$  \hspace{1cm} (7.25)
and

\[ \Phi = (n + \frac{1}{2})\Phi_0 \quad (7.26) \]

In this case, the ground state of magnetic flux (for \( n=0 \)) is \( \Phi = \frac{1}{2}\Phi_0 \), and the flux through the ring never takes a zero value.

**Figure 7.3:** Spatial distribution of the order parameter in an s-wave and d-wave superconductor, showing the possible couplings at an interface. Phase change of the order parameter depends on which lobe of the d-wave superconductor is involved in coupling.

This means that in the absence of external field, a magnetic flux of \( \frac{1}{2}\Phi_0 \) is spontaneously generated in the ring, along with a corresponding spontaneous current. Application of external field will then lead to higher half-integer values of \( \Phi_0 \).

The \( \pi \) phase change leading to half flux quanta arises due to the directionality of the order parameter in d-wave superconductors. In superconductors with s-wave symmetry, the order parameter has no directional dependence and no sign change on rotation. By contrast, the order parameter in d-wave superconductors has positive and negative lobes, which are oriented 90° from one another in a flat plane. Coupling of the s-wave superconductor with a positive lobe of the d-wave superconductor causes no phase change, while coupling with a negative lobe causes a change of sign across the junction, which is equivalent to a \( \pi \) phase change. These couplings are illustrated in figure 7.3.

A composite ring composed of a s-wave and a d-wave superconducting sectors contains two such junctions, and the total phase change around the ring depends on the relative phase changes of these two junctions. If both junctions produce a zero
CHAPTER 7. ADDITIONAL MAGNETIC MICROSCOPY EXPERIMENTS

Phase change, or if both produce a $\pi$ phase change, then there will be overall zero phase change on a full revolution around the ring. These are referred to as ordinary rings or 0-rings, and behave in a similar way to non-composite superconducting rings. However, if one junction produces a $\pi$ phase change while the other produces a zero phase change, there will be an overall $\pi$ phase change in the order parameter. These are referred to as $\pi$-rings.

0-ring and $\pi$-ring samples can be produced using a high-quality single-crystalline thin-film d-wave superconductor (such as YBCO) which is cut into an annular sectors, each having a well-defined angle ranging from $15^\circ$ to $345^\circ$. An s-wave superconducting material (such as niobium) can then be deposited in such a way as to complete the annulus, with a small overlap at each end of the d-wave sector, forming the two junctions.

Since the d-wave part of the ring is a single crystal, the distribution of the order parameter is continuous throughout this sector.

It therefore follows by simple geometric consideration that if the d-wave sector has an angle of less than $45^\circ$, then two directly opposite lobes of the order parameter distribution (with the same sign) will be involved in coupling at each interface, and there will be the same phase change at each, and therefore no overall phase change.

If, however the d-wave sector has an angle around $90^\circ$, then the lobe which couples at one interface will be $90^\circ$ from the lobe that couples at the other. This means the two coupling lobes are adjacent and have opposite sign, meaning there is one junction of each type, and an overall phase change of $\pi$.

For angles around $180^\circ$, the same lobe of the d-wave distribution couples at each interface, and it produces a 0-ring. Around $270^\circ$ adjacent lobes couple, producing a $\pi$-ring, and for angles approaching $360^\circ$ it is opposite lobes again producing a 0-ring. The exact angular dependence of the overall phase change is shown in figure 7.4.

A theoretical analysis by another group of this $\pi$-loop array showed that the flux in each loop may not be independent of other loops [174]. The direction of spontaneous flux in each $\pi$-loop was shown to be determined by the flux in neighbouring loops, resulting in anti-parallel spontaneous flux in each neighbouring loop in the array [174].

7.2.2 Observation of Integral and Half-Integral Flux Quanta

Using an array of composite niobium (s-wave) and YBCO (d-wave) superconducting rings with varying angles of the d-wave sector, a number of 0-ring and $\pi$-ring samples were realised [124, 173]. By application of applied field in both the positive and negative $z$-direction, several integer and half-integer quantised flux values were measured in these rings by scanning SQUID microscopy (SSM) [173]. The SSM
apparatus and procedure are described in chapter 5.

The sample had been produced for a previous study using multiple depositions, and SSM scans had been acquired with a magnetic shield in place and with zero applied field [126]. Therefore, the spontaneous flux of $\frac{1}{2}\Phi_0$ had been observed in the $\pi$-ring samples, but higher half-integer flux quanta such as $\frac{3}{2}\Phi_0$ had not been seen, since these appear only on application of an external field. Niobium (Nb) had been chosen for the s-wave component of the rings for its relatively high critical temperature of $T_C = 9.25$ K. The Josephson junctions of the rings were made using a ramp geometry, as discussed in [173], to create the strongest connection and ensure that Cooper pairs could flow freely between the s-wave niobium and the Cu-O$_2$ planes of the YBCO.

A repetition of the measurement of spontaneous flux in six $\pi$-rings, along with the lack of spontaneous flux in two 0-rings is shown in figure 7.5. Vortices are also

Figure 7.4: Dependence of total phase change on angle of the d-wave sector in s- and d-wave composite superconducting rings. The yellow and blue regions represent the s- and d-wave sectors respectively, and the arrows at the junctions show coupling between positive (dark blue) and/or negative (red) regions of the order parameter, as in fig 7.3.
seen within the YBCO regions of the rings.

Figure 7.5: (a) Scanning SQUID micrograph of six $\pi$-rings and two 0-rings taken with an external field $\ll \frac{1}{2}\Phi_0$, showing spontaneous flux in $\pi$-rings and other labelled features. (b) Current around the rings calculated from the magnetic field data of (a).

The purpose of re-investigating these samples was to apply field to the rings and thereby observe half-integer quantised flux values other than the spontaneous $\frac{1}{2}\Phi_0$, while confirming that flux through the 0-rings took the expected integral quantised values. The zero-field scan shown in figure 7.5(a) was taken in order to determine the physical position of 0-rings and $\pi$-rings for later measurements. For all further measurements, a region of the sample containing one $\pi$-ring and one 0-ring (for comparison) was selected and scanned over a range of applied fields. The samples were cooled in zero field, then predetermined values of field were applied in each direction.

It was important to select rings for which no breakdown of the Josephson junctions was seen for further imaging. Breakdown of the junctions occurs when a current greater than $J_c$ flows through the junction, where $J_c$ varies depending on the quality of each junction. Those rings for which breakdown has occurred are most easily identified in the current mapping, figure 7.5(b), where supercurrent flows around the perimeter of the YBCO part, not crossing the junction. For some rings, very little current is seen in the Nb part of the rings, which may mean that the niobium is no longer superconducting at all for these rings. None of these rings were selected for further scans.

The flux at each point around the two selected rings was measured by SSM. A typical example for a $\pi$-ring is plotted in figure 7.6. The stray field from each
Field inside the loop integrated to give flux

Figure 7.6: 3D Plot of the spontaneous flux in and around a $\pi$-ring as measured by scanning SQUID microscopy.

Therefore the total flux through the centre of each ring was determined from the scan data by integrating the field measured over a larger area than the size of the hole in the ring, but taking care not to include vortices or any flux that is present in the junctions.

The total flux through each ring is plotted as a function of applied field in figure 7.7. This is seen to match quite well with the theoretical prediction of the field dependence of flux in each ring. Field-cooled measurements were also taken, but the flux did not match so well with the predicted values in these measurements.

Therefore, this investigation was successful in the observation of half-integer flux values other than the spontaneous flux value of $\frac{1}{2}\Phi_0$ in rings with a $\pi$ phase change upon circulation. The values of flux observed in these rings were $\pm\frac{1}{2}\Phi_0$ and $\pm\frac{3}{2}\Phi_0$. For comparison, flux values of $0$, $\pm\Phi_0$ and $2\Phi_0$ were observed in a ring with no phase change upon circulation.
Chapter 8

Conclusion

This thesis has presented magnetic microscopy results relating to the distribution and behaviour of magnetic flux vortices in thin film superconducting YBa$_2$Cu$_3$O$_{7-\delta}$ samples.

There were two primary magnetic microscopy techniques used for these investigations: The first was Magneto-Optical Imaging (MOI), which was used to examine the distribution of vortices and supercurrents on a macroscopic scale. The MOI technique was also extended to provide high-speed Magneto-Optical Video (MOV) of dynamic magnetic fields and to calculate current in superconductors as it changed over time. The second technique was Scanning SQUID Microscopy (SSM), which was used to analyse the phase of vortex matter on an individual vortex scale.

Using these techniques, several novel insights were made into the behaviour of vortices in thin film superconductors under different conditions. The most important insights and experiments were as follows:

- The new high-speed MOV technique was used to observe field and current dynamics during flux penetration. This led to the observation of several novel features including transient currents larger than the critical current density, and a transition in behaviour from increasing current density toward relaxation.

- Simulations of field and current behaviour during penetration were found to support these novel MOV results. This was done in collaboration with I. Golovchanskiy.

- Superconducting thin films of high and low quality were analysed using magneto-optical video to find differences in field and current dynamics. Features were identified which may be used to help identify low quality samples, even in the absence of visible defects.
The properties of vortices occurring in a micro-Tesla field-cooled 2D glass phase were analysed in order to characterise the order and phase of the distribution.

The field dependence of the vortex distribution in this phase was investigated. It was unexpectedly found that the orientational order decreased when field was increased.

During this investigation, an unusual bunching of vortices into closely-spaced groups was discovered. It is speculated that the origin of these groups lies in the decrease of temperature through other vortex phases during sample cooling.

A mathematical procedure that had been designed to account for the effects of in-plane fields on MOI images was analysed and shown to mathematically valid, in collaboration with J. Zuber, but the procedure did not prove viable in experiment.

Superconducting rings consisting of mixed s-wave and d-wave superconductors were analysed using scanning SQUID microscopy, in collaboration with A. Timmermans and R. Bruel. Magnetic flux was observed at the fractional quantised values of $\pm \frac{1}{2}\Phi_0$ and $\pm \frac{3}{2}\Phi_0$.

Novel vortex ratchet patterns were proposed based on the principle of asymmetric 2D weak-pinning channels. The patterns extended previous ratchet designs in order to allow rectification at temperatures approaching zero. Strong rectification was seen for one of the designs in simulation, under specific field conditions.

$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ samples were shaped using a lithographic patterning and ion beam etching procedure. This procedure was extended to allow partial-thickness etching, which is the thinning of selected parts of the superconducting film without removing all of the superconducting material in these areas. This was necessary for creation of the aforementioned novel vortex ratchet devices.

Overall, this thesis has presented several advances in the understanding of the behaviour of magnetic field vortices in superconductors. These have resulted in a number of publications, as listed on the next page. It is my hope that these insights will lead to further advances in this field, whether they come by my own hand or by another that this work may inspire. I also like to think that this work marks the beginning of a long and fruitful career in superconductivity.
Publications


Bibliography


187


(34) D. J. Bishop, P. L. Gammel, C. A. Murray, D. B. Mitzi and A Kapitulnik, “Observation of an hexatic vortex glass in flux lattices of the high-$T_c$ superconductor $\text{Bi}_{2.1}\text{Sr}_{1.9}\text{Ca}_{0.9}\text{Cu}_2\text{O}_{8+\delta}$”, Physica B, 1991, 169, 72–79.


(132) T. H. Johansen, Y. Galpern and H. Bratsberg, “Analytical Solution of the Bulk Current Inverse Problem; $B_z(x, y) \rightarrow (j_x, j_y, 0)$”, unpublished, 1997.


Appendix A

The Inverse Problem

This appendix describes a mathematical approach for inversion of the Biot-Savart law in the one- and two-dimensional cases, in order to calculate current distribution from magnetic fields measured at some distance to the current [131]. This inversion is important for calculating the current distribution in samples after the field has been imaged using magnetic microscopy, as discussed in section 2.7.1. This analysis critically examines and refines previous solutions provided by Johansen et al. [107], Jooss et al. [63] and Roth et al. [108], leading to a simpler derivation, with results that are consistent with the literature.

Since field is not measured at the same position as the current that produces it, the Biot-Savart law (equation 2.11) is used - it provides an appropriate non-local relation between these quantities. However, this relation is generally not invertible, meaning that a unique current distribution cannot be determined from a given field distribution. Therefore the “inverse problem” is the problem of finding a unique solution for the current distribution from magnetic field measurements, under appropriate physical constraints.

This problem is widely applicable to a variety of magnetic imaging techniques beyond those described in this thesis, many of which are listed in section 2.6. In particular, information about the current flowing through the human body is often required for specific applications of magnetic resonance imaging such as functional Magnetic Resonance Imaging [179] and Magnetic Resonance Elastography [180]. Also, the standard image reconstruction in magnetoencephalography and magneto-cardiography is largely based on a solution to an inverse problem closely related to that of the Biot-Savart law [91]. Each of these techniques uses a different measurement geometry and therefore imposes different constraints on the inverse problem, which in each case allow a unique solution for the current.

In this research, the inverse problem considered is for magnetic microscopy of a thin-film superconductor in a homogeneous perpendicular magnetic field. Since most magnetic microscopy techniques used for this purpose only measure the $z$-
component of magnetic field near a sample, this case is more complex than for other techniques - two current components, $J_x$ and $J_y$ must be determined from only one field component, $B_z$.

Several solutions to the inverse problem for thin film superconductors have been proposed over many years, however the conditions for which the problem has been solved have not always been the same. For example, Roth et al. solved the problem without the consideration that the magnetometer was placed at a finite height above the sample [108], whilst Johansen et al. initially only solved the problem in one dimension [107]. One of the most accurate solution for the problem was devised by Jooss et al., which takes into account the finite height of the detector and solves the problem in two dimensions [63].

A novel solution to this inverse problem is described below [131], and the final result is consistent with Jooss solution. This new technique does not require Green’s function integral identities as in [63] nor Topelitz matrices as in [130], for numerical solutions to the Biot-Savart law. Instead, computation of the integral kernel is carried out directly using a Laplace transform, and considering properties of the Bessel function of the first kind.

Note that several symbols will be defined and used in this appendix which have been left out of the list of symbols at the beginning of this thesis for simplicity. These terms are used only in the calculations here, and do not appear anywhere in the main part of the thesis.
A.1 The Inverse Problem in One Dimension

The solution to the 1D inverse problem depicted in figure A.1 was originally carried out by Johansen et al. [107]. In this section it is re-derived using a different approach and in greater detail.

The one dimensional problem considers the case of current flowing along an infinite superconducting strip oriented along the y-axis. For simplicity of this model, the current is taken to flow purely in the direction of the strip, $J_x = J_z = 0$. The remaining component, $J_y$, is allowed to vary only in the $x$-direction, i.e. along the width of the strip. The sheet thickness, $d$, is taken to be small compared to the height, $h$, of the detector above the sheet: $d \ll h$. Hence, in this geometry the sheet current is given by:

$$J_y(x) = \int_0^d j(x, z) dz,$$  \hspace{1cm} (A.1) 

where $j(x, z)$ is the local current density caused by current moving in the $y$-direction producing a magnetic field in the $xz$-plane. All quantities are invariant under translation along the $y$-axis.

An application of the Biot-Savart law (equation 2.11) in 1D gives:

$$B_z(x') = \frac{\mu_0}{2\pi} \int_{-\infty}^{\infty} \frac{x - x'}{h^2 + (x - x')^2} J(x) dx$$ \hspace{1cm} (A.2)
The integral kernel of this equation:

\[ K(x, x') = \frac{x - x'}{h^2 + (x - x')^2} = K(x - x') \quad (A.3) \]

is translational invariant in the \( x \)-direction, so an appropriate convolution can be applied. Hence, the Fourier transform of equation A.3 where \( x' \to k_x \) yields:

\[ \tilde{B}_z(k_x) = \frac{\mu_0}{2\pi} \tilde{K}(k_x) \tilde{J}(k_x) \quad (A.4) \]

Now, the inversion theorem can be used to obtaining an expression for \( J(x) \):

\[ \mu_0 J(x) = \int_{-\infty}^{\infty} \frac{\tilde{B}_z(k_x)}{\tilde{K}(k_x)} e^{ik_x x} dk_x \quad (A.5) \]

In order to compute the Fourier transform of the integral kernel it can be shown that [131]:

\[ \tilde{K}(k_x) = \int_{-\infty}^{\infty} \frac{x'}{h^2 + x'^2} e^{-ik_x x'} dx' = -i\pi \text{sgn}(k_x) e^{-h|k_x|}, \quad (A.6) \]

where \( \text{sgn}(x) \) is the sign function.

Before discretisation of the integral can take place, the functions in the integrand must be transformed into those which can be represented by an infinite series. So using equation A.6 we have:

\[ \mu_0 J(x) = \int_{-\infty}^{\infty} \frac{\tilde{B}_z(k_x)}{-i\pi \text{sgn}(k_x) e^{-h|k_x|}} e^{ik_x x} dk_x \quad (A.7) \]

However, noting that the transfer function is given by \( \frac{1}{\tilde{K}(k_x)} \), it is evident that components with high frequency will be highly amplified, since by equation A.6:

\[ \frac{1}{\tilde{K}(k_x)} \propto e^{h|k_x|} \quad (A.8) \]

Such components are to be removed. Hence, a low pass filter should be included in the analysis so that components with \( |k_x| \geq K_c \) are cut out, where \( K_c \) is a cut-off frequency. Including this cut-off frequency gives:

\[ \mu_0 J(x) = \int_{-\infty}^{\infty} \int_{-K_c}^{K_c} \frac{e^{ik_x x} B_z(x') e^{-ik_x x'}}{-i\pi \text{sgn}(k_x) e^{-h|k_x|}} dk_x dx' \quad (A.9) \]

Note that in this case, a Hanning window function was not used as in section 2.7.1.
By defining:

$$A(\xi) = \int_{-K_e}^{K_e} \frac{e^{ik_x \xi}}{-i\pi \text{sgn}(k_x)e^{-h|k_x|}} dk_x,$$  \quad (A.10)

The solution to equation A.10 is as follows:

$$A(\xi) = \int_{-K_e}^{K_e} \frac{e^{ik_x \xi}}{-i\pi \text{sgn}(k_x)e^{-h|k_x|}} dk_x$$

$$= \frac{i}{\pi} \left[ \int_{-K_e}^{0} \frac{e^{k_x(i\xi-h)}}{-1} dk_x + \int_{0}^{K_e} \frac{e^{k_x(h+i\xi)}}{1} \right]$$

$$= -\frac{i}{\pi} \left[ \frac{i\xi e^{-K_e(i\xi-h)}}{h^2 + \xi^2} - i\xi + he^{-K_e(i\xi-h)} \right]$$

$$+ \frac{i\xi e^{K_e(i\xi+h)} - i\xi - he^{K_e(i\xi+h)}}{h^2 + \xi^2}$$

$$= 2 \frac{\xi(e^{K_e h} \cos(\xi K_e) - 1) - he^{K_e h} \sin(\xi K_e)}{h^2 + \xi^2}$$

\[ \therefore A(\xi) \neq \frac{\xi(1 - e^{K_e h} \cos(K_e \xi)) + he^{K_e h} \sin(K_e \xi)}{h^2 + \xi^2} \tag{A.11} \]

Using this solution for the kernel, \(A(\xi)\), the integral in equation A.9 can be discretised since magnetic flux is measured in discrete pixel-sized units. For this process, \(\Delta = \frac{\pi}{K_e}\) was defined as the unit length, then the x-coordinates were discretised as: \(x = n\Delta, x' = n'\Delta\) and also taking \(h = t\Delta\) where \(t\) is simply the film thickness under the change of variables.

The infinite sum representing the discretised version of equation A.9 was calculated to be [131]:

$$\mu_0 J(n) = -\frac{2}{\pi} \sum_{n'} \frac{n-n'}{t^2 + (n-n')^2} \frac{1 - (-1)^{n-n'}e^{\pi t}}{\pi} 2B_z(n')$$  \quad (A.12)

Hence the 1D inverse problem has been solved. The discrete sum can be solved to give the current density given the magnetic field values. This solution differs from [107] only by a small factor of \(-\frac{2}{\pi}\).

This solution differs from a previous solution to the same 1D inversion by a factor of \(-\frac{2}{\pi}[107]\). Therefore, the current appears to flow in the opposite direction and has a slightly smaller amplitude, compared to the literature result [107]. The true current direction is shown in figure A.1 along the \(x\)-axis.
A.2 The Inverse Problem in 2D

The inverse problem in two dimensions is solved below following our approach as suggested in [131], though it is partly based on the solution given by Jooss et al. [63]. This method is however unique, since instead of using Green’s functions to evaluate the Fourier transform of the integral kernel, it is represented as a Hankel transform for which Bessel function and Laplace transform identities can then be used to compute the transform.

To derive a formula for $J_x(x,y)$ and $J_y(x,y)$ as a function of $B_z$, this solution begins with the continuity condition for the sheet current density:

$$\nabla \cdot \mathbf{J}(x,y) = 0 \quad (A.13)$$

It assumes that there is no current flowing in the $z$-direction of the sheet since it is thin.

A scalar function $g(x,y)$ is then defined by the relation:

$$\mathbf{J}(x,y) = \nabla \times \hat{z} g(x,y), \quad (A.14)$$

which is consistent with the continuity equation A.13. To alleviate the freedom of gauge choice, the Coulomb gauge condition can be applied so that $g(x,y)$ is totally defined as the local magnetisation and is the potential function for the current density.

Using this expression for $\mathbf{J}(x,y)$ in the integrand of the Biot-Savart law (equation 2.11) gives:

$$B_z(r) = \frac{\mu_0}{4\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \hat{z} \cdot \left(\nabla \times \hat{z} g(x,y)\right) \times \frac{(r-r')}{|r-r'|^3} \, dr' \quad (A.15)$$

Noting once again that only the $z$-component of the magnetic field is measured, hence the $\hat{z}$ unit vector in the integrand.

Now, it is left to compute the integrand in a form that allows the Fourier convolution theorem to be applied. It can be shown using vector calculus identities that

$$B_z(r) = -\frac{\mu_0}{4\pi} \int_{-\infty}^{\infty} \frac{2(z-z')^2 - (x-x')^2 - (z-z')^2}{((x-x')^2 + (y-y')^2 + (z-z')^2)^{3/2}} g(x',y') \, dr' \quad (A.16)$$

Taking into account the finite thickness of the sample ($d$), $z'$ is integrated over the domain $-d/2$ to $d/2$. Also, since field is measured only in a plane at distance $h$ from the sample, $z$ takes the singular value $z = h$. Equation A.17 is then obtained.
by taking the Fourier transform of the \( x' \) and \( y' \) integrals (with spatial variables \( x \) and \( y \) mapped to frequency variables \( k_x \) and \( k_y \) respectively), and applying the convolution theorem in the \( xy \)-plane.

\[
\tilde{B}_z(k_x, k_y, h, d) = -\frac{\mu_0}{4\pi} \int_{-\frac{d}{2}}^{\frac{d}{2}} \mathcal{F} \left\{ \frac{2(h - z')^2 - x^2 - y^2}{(x^2 + y^2 + (h - z')^2)^{\frac{3}{2}}} \right\} \tilde{g}(k_x, k_y) dz' \tag{A.17}
\]

The Fourier transform in this equation can be computed to give \([131]\):

\[
\mathcal{F} \left\{ \frac{2(h - z')^2 - x^2 - y^2}{(x^2 + y^2 + (h - z')^2)^{\frac{3}{2}}} \right\} = 2\pi ke^{-k(h - z')} ,
\tag{A.18}
\]

where \( k = \sqrt{k_x^2 + k_y^2} \).

Hence, the magnetic field detected becomes

\[
\tilde{B}_z(k_x, k_y, d, h) = -\frac{\mu_0}{4\pi} \int_{-\frac{d}{2}}^{\frac{d}{2}} e^{-k(h - z')} 2\pi k \tilde{g}(k_x, k_y) dz' \tag{A.19}
\]

It can be assumed that \( \tilde{g}(k_x, k_y) \) is not dependent on \( z' \) following from the assumption that current flows only in the \( xy \)-plane:

\[
\tilde{B}_z(k_x, k_y, d, h) = -\frac{\mu_0}{2} \tilde{g}(k_x, k_y)ke^{-kh} \int_{-\frac{d}{2}}^{\frac{d}{2}} e^{kd'}/d' \tag{A.20}
\]

Thus, the function \( g(x, y) \) is uniquely determined from a given field, and all that remains is to uniquely determine \( J_x(x, y) \) and \( J_y(x, y) \). This is achieved using the definition of the function \( g(x, y) \):

\[
J(x, y) = \nabla \times \mathbf{z}g(x, y) = (\partial_y g(x, y), -\partial_x g(x, y), 0) \tag{A.21}
\]

\[
\tilde{J}_x(k_x, k_y) = -ik_y \tilde{g}(k_x, k_y) \tag{A.22}
\]

\[
\tilde{J}_y(k_x, k_y) = ik_x \tilde{g}(k_x, k_y) \tag{A.23}
\]

And the continuity equation:

\[
k_x \tilde{J}_x(k_x, k_y) + k_y \tilde{J}_y(k_x, k_y) = 0 \tag{A.24}
\]

To give final expressions for both components of the current as functions of
physical variables and the magnetic field detected in the $z$-direction:

\[
\begin{align*}
\tilde{J}_x(k_x, k_y) &= \frac{ik_y}{\mu_0} e^{k h} \text{cosech} \left( \frac{kd}{2} \right) \tilde{B}_z(k_x, k_y, h, d) \\
\tilde{J}_y(k_x, k_y) &= -\frac{ik_x}{\mu_0} e^{k h} \text{cosech} \left( \frac{kd}{2} \right) \tilde{B}_z(k_x, k_y, h, d)
\end{align*}
\] (A.25) (A.26)

A simple inverse Fourier transform algorithm can be applied to calculate $J_x$ and $J_y$ and real space, and hence obtain a current map of the superconductor. The inverse problem is therefore solved, with final equations consistent with those derived by Jooss et al. [63].

Roth’s Method

Another method for solving the inverse problem for 2D currents in a thin film was historically developed by Roth et al. [108]. In contrast to the newly derived method above, this solution does not take into account the finite height of the magnetometer above the sample. Instead, it assumes detection of magnetic field directly at the sample surface. Mathematically, it differs in that the integral kernel after a Fourier transform is computed directly from the Biot-Savart law for all components of the magnetic field.

To mathematically derive the method devised by Roth et al. [108] consider the same geometry for the inverse problem, but with $h = 0$. The integral for the $x$-component of the magnetic field is therefore given by:

\[
\begin{align*}
B_x(x, y, z) &= \frac{\mu_0}{4\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathbf{\hat{x}} \cdot \mathbf{J}(r') \times \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3} \, d^3r' \\
&= \frac{\mu_0 d}{4\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{z}{[(x - x')^2 + (y - y')^2 + z^2]^{3/2}} \, J_y(x', y') \, dx' \, dy' 
\end{align*}
\] (A.27) (A.28)

Assuming again that no current flows in the $z$-direction, the current density is only a function of $x'$ and $y'$, with $z' = 0$, since current is confined to a plane located at $z = 0$. The numerator of the integrand reduces to $z J_y(x', y')$, and the integration over this thickness can be easily computed as the integrand is not a function of $z'$. This yields a Fredholm integral equation:

\[
\begin{align*}
B_x(x, y, z) &= \frac{\mu_0 d}{4\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{z}{[(x - x')^2 + (y - y')^2 + z^2]^{3/2}} \, J_y(x', y') \, dx' \, dy' 
\end{align*}
\] (A.28)

Following the same procedure as for the previous method, a Fourier transform of the above equation yields an integral kernel which must be computed:

\[
\tilde{B}_x(k_x, k_y, z) = \tilde{G}(k_x, k_y, z) \tilde{J}_y(k_x, k_y) 
\] (A.29)
Employing a convolution in the xy-plane gives:

\[ \tilde{G}(k_x, k_y, z) = \mathcal{F} \left\{ \frac{\mu_0 d}{4\pi (x^2 + y^2 + z^2)^{\frac{3}{2}}} \right\} \]  (A.30)

Now by noticing the following relation:

\[ -\frac{d}{dz} \mathcal{F} \left\{ \frac{z}{(x^2 + y^2 + z^2)^{\frac{3}{2}}} \right\} = \mathcal{F} \left\{ \frac{2z^2 - x^2 - y^2}{(x^2 + y^2 + z^2)^{\frac{5}{2}}} \right\} \]  (A.31)

Equation A.18 can be used to solve for \( \tilde{G} \):

\[ -\frac{d}{dz} \tilde{G}(k_x, k_y, z) = -\frac{\mu_0}{4\pi} \left[ 2\pi ke^{-|\vec{k}|z} \right] \]  (A.32)

Integrating both sides with respect to \( z \):

\[ \tilde{G}(k_x, k_y, z) = \frac{\mu_0 d}{2} e^{-|\vec{k}|z} \]  (A.33)

The constant of integration is omitted here - it can be shown to be 0 [108].

\[ \therefore \tilde{J}_y(k_x, k_y, d) = \frac{2}{\mu_0 d} e^{z\sqrt{k_x^2 + k_y^2}} \tilde{B}_x(k_x, k_y) \]  (A.34)

Repeating this procedure for the y and z components of the magnetic field yields:

\[ \tilde{J}_x(k_x, k_y, d) = -\frac{2}{\mu_0 d} e^{z\sqrt{k_x^2 + k_y^2}} \tilde{B}_y(k_x, k_y) \]  (A.35)

and

\[ \tilde{B}_z(k_x, k_y, z) = \frac{i\mu_0 d}{2} e^{z\sqrt{k_x^2 + k_y^2}} \left( \frac{k_y}{\sqrt{k_x^2 + k_y^2}} \tilde{J}_x(k_x, k_y) - \frac{k_x}{\sqrt{k_x^2 + k_y^2}} \tilde{J}_y(k_x, k_y) \right) \]  (A.36)

Equation A.36 is obtained by splitting the integral for \( B_z \) into two parts; one for \( J_x \) and the other for \( J_y \), then calculating a form similar to A.34 for each of these integrals. This shows that the measured \( B_z \) results from a linear combination of \( J_x \) and \( J_y \) contributions. It is then possible to use the continuity equation (A.24) to find either component individually, as described for the previous method.

Since this method does not take into account typical experimental conditions, it is considered to be less accurate. Hence, our method described above was employed for all calculations used for the current research.
A.3 Conclusion

The inverse problem for finding the current distribution in superconducting thin films from magnetic field measurements in a plane above the film has here been uniquely solved for both the one- and two-dimensional cases. A new approach was used to compute the integral kernel in the 2D case, which involves resolving the problem into a Laplace transform of a Bessel function of the first kind. The derivations are provided in great detail here for completeness, while they are heavily truncated in section 2.7.1 to aid the overall flow of the document.

For the 2D case, the solution is consistent with that previously determined in [63, 108]. However, the use of Green’s function identities were not required in this new method, while still taking into account the finite height of the magnetometer above the surface.

In the 1D case, it was found that the calculated current appears to flow in the opposite direction and has a slightly smaller amplitude by a factor of $\frac{2}{\pi}$, compared to the result in [107]. Otherwise, the solution is fully consistent with [107].