A study into the effects of fission-fragment damage on activation energies in AG/Bi2223 tapes

Damián Marinaro
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A STUDY INTO THE EFFECTS OF FISSION-
FRAGMENT DAMAGE ON ACTIVATION
ENERGIES IN AG/BI2223 TAPES

A thesis submitted in fulfilment of the
requirements for the award of the degree

Doctor of Philosophy

from the

UNIVERSITY OF WOLLONGONG

by

Damián Marinaro, BSc. (Hons.) (Adv.)

Institute for Superconducting and Electronic Materials

2003
I, Damián G. Marinaro, declare that this thesis, submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the Institute for Superconducting and Electronic Materials, University of Wollongong, is wholly my own work unless otherwise referenced or acknowledged. The document has not been submitted for qualifications at any other academic institution.

Damián G. Marinaro

20 October 2003
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ABSTRACT

Ag/Bi-2223 tapes doped with small quantities of highly enriched uranium were prepared by the powder-in-tube process and irradiated with thermal neutrons. The resulting fission fragments create randomly splayed quasi-columnar defects that provide strong flux pinning. Significant improvements in the field and angular dependence of the critical current density and in the irreversibility field were observed at 77 K, increasing with the density of fission tracks.

In tapes with equivalent fission track density, the observed increases are larger for the samples exposed to larger neutron fluences (where a lower uranium doping level requires higher neutron fluence to obtain the same fission track density). This is explained as a result of the formation of secondary uranium-containing phases physically separated from the Bi2223 phase above a solubility limit to the uranium doping. An unusual exponential dependence of $H_{irr}$ on the density of tracks was also observed, unrelated to the secondary phase formation.

The primary investigation concerns the activation energies for vortex motion in the thermally activated flux flow and flux creep regimes, probed via
resistive transition and dynamic magnetic relaxation measurements, which had not been previously explored. Both methods demonstrate the dominance of the fission tracks in the pinning landscape, particularly in fields applied parallel to the $c$-axis.

Linearity in an Arrhenius plot of the superconductor resistance against temperature over at least three orders of magnitude indicated thermally activated flux motion in tapes with and without fission damage. In fields applied parallel to the $c$-axis, the pinning energy was substantially increased by the fission tracks, $U_0$ tripling in magnitude at $\mu_0H \sim 0.3$ T, and then reconverging with the pre-irradiation $U_0$ for $\mu_0H > 2$ T. This behaviour of the activation energy is well described by an effective matching field $B_{\text{eff}} \sim 0.3$ T, identified at the maximum in the relative change of $U_0$ and also observed from changes in the behaviour of the resistively determined irreversibility field. This $B_{\text{eff}}$ is lower than the matching field $B_{\phi} \sim 2.1$ T estimated from the fission track density.

Dynamic magnetic relaxation measurements were analysed using a modified form of Maley's method [M. P. Maley et al., Phys. Rev. B 42, p2639 (1990)]. This method was modified by using an empirically determined temperature scaling to resolve the current density dependence of the effective activation energy $U_{\text{eff}}(J,H,T_0)$ in the flux creep regime from different
isothermal data points, over a wide range of temperatures and values of $J$. Correlated changes in the empirical temperature scaling form $U(T)$, an additive constant $C$, a fitting parameter $\mu$ and the divergent behaviour of $U_{\text{eff}}$ as $J \to 0$ are all consistent with a dimensional crossover from 3D elastic to 2D plastic creep at fields $\mu_0 H \approx 0.37$ T in a nonirradiated tape.

The dimensional crossover was shifted to higher fields $\mu_0 H \approx 0.65$ T after the introduction of fission-fragment damage, interpreted as evidence that the randomly splayed quasi-columnar defects promote $c$-axis vortex correlation in the Ag/Bi2223 tapes. Analysis of the relaxation data without the additional temperature scaling modification, on the other hand, produced a conflicting result, implying the complete destruction of $c$-axis vortex correlation over the entire field range explored. This observation is inconsistent with measurements of the current – voltage characteristics. Together with the unreasonable values of $C$ obtained by this analysis and in comparison to the consistency observed with the empirically determined temperature scaling modification, the results indicate that the inclusion of the empirical temperature scaling is essential to an analysis of relaxation data.

In comparison to past studies, we conclude that the efficiency of the uranium-fission method in enhancing the flux pinning is strongly dependent on the material anisotropy, through the vortex line elasticity and
dimensionality, which hence affect the vortex line accommodation to the splayed defects. This has significant consequences for the estimation of $B_{\phi}$ in materials with differing anisotropy. The present method of calculating $B_{\phi}$ directly from the fission track density multiplied by the average defect track length is an oversimplification, which does not take into account the finite elasticity of the flux lines as well as the anisotropy of the vortex system.
LIST OF FIGURES

Fig. 1.1: Crystal structure of typical anisotropic HTS materials (a) Bi₂Sr₂Ca₂Cu₃Oₓ and (b) Bi₂Sr₂CaCu₂Oₓ. The insert illustrates the crystallographic axes…………………………………6

Fig. 1.2: Structure of an isolated vortex line versus the distance from the vortex axis, r. (a) Density of the superconducting electrons; (b) magnetic flux density; (c) supercurrent density………………………………………………………………14

Fig. 1.3: (a) Uncorrelated point-like disorder; (b) Correlated disorder in the form of a columnar defect……………………………………………………………………………29

Fig. 1.4: Magnetic field profile inside a superconductor in the critical state model in (a) increasing and (b) decreasing applied fields……………………………………38

Fig. 1.5: Flux line motion by nucleation of half-loop and double-kink processes. The Lorentz force in the diagram is acting to push the lines towards the right………58

Fig. 4.1: (a) Critical current density as a function of applied magnetic field aligned parallel to the c-axis, for various combinations of uranium doping and neutron irradiation…………………………………………………………………….100

Fig. 4.1: (b) Critical current density as a function of applied magnetic field aligned parallel to the ab-plane, for various combinations of uranium doping and neutron irradiation…………………………………………………………………101

Fig. 4.2: Zero-field J_c normalised to pre-irradiation levels for various ²³⁵U doping percentages c. An approximately linear dependence on fission track density ~ cΦ_n is shown in the insert……………………………………………………….102

Fig. 4.3: Magnetic field dependence of J_c normalised to the zero-field J_c₀, for fields (a) parallel and (b) perpendicular to the c-axis………………………………………107

Fig. 4.4: H_irr as a function of cΦ_n for fields parallel to the c-axis, in (a) linear, (b) log-linear scales. The lines are a guide to the eye, representing linear or exponential behaviour of H_irr, respectively……………………………..109

Fig. 4.5: J_c normalised to the J_c at (a) μ₀H = 100 mT and (b) μ₀H = 500 mT for fields applied parallel and perpendicular to the c-axis, respectively………………112
Fig. 4.6: Angular dependence of $J_c$, normalised to the maximum current. $\theta$ is the angle of field orientation relative to the perpendicular of the broad face of the tape.

Fig. 4.7: Pinning force density $F_p$ as a function of applied fields parallel to the $c$-axis for (a) tapes with $c\Phi_n = 0$ and (b) all tapes.

Fig. 4.8: Pinning force density $F_p$ as a function of applied fields parallel to the $c$-axis for all tapes.

Fig. 4.9: Field of maximum in the pinning force density, $H_{max}$, as a function of $c\Phi_n$ for fields applied parallel to the $c$-axis in (a) linear and (b) log-linear scales. (b) demonstrates the reasonable exponential dependence.

Fig. 4.9: (c) Maximum in $F_p$ as a function of $c\Phi_n$ for fields applied parallel to the $c$-axis.

Fig. 4.10: $H_{max}$ as a function of the irreversibility field.

Fig. 4.11: Normalised pinning force density plotted against the reduced field.

Fig. 4.12: Fit to Eq. (4.2) (solid lines) of normalised pinning force density against reduced field for tapes with (a) $c\Phi_n = 0$ and (b) $c\Phi_n \neq 0$.

Fig. 4.13: (a) Example of a fit to Eq. (4.3), representative of the fits for both $c\Phi_n = 0$ and $c\Phi_n \neq 0$, as well as for both Equations (4.3) and (4.4).

Fig. 4.13: Fit to Equations (4.6) & (4.7) for (b) $c\Phi_n = 0$ and (c) $c\Phi_n \neq 0$. The solid lines represent fits to both Equations.

Fig. 5.1: Normalised resistance against temperature, at an applied field 100 mT parallel to the $c$-axis.

Fig. 5.2: Normalised resistive transitions for a virgin tape and a tape with typical doping and irradiation, in fields applied (a) parallel and (b) perpendicular to the $c$-axis. The difference between the two tapes increases further as the field is increased.

Fig. 5.3: Normalised resistive transitions for all tapes, in (a) low and (b) high applied fields, parallel to the $c$-axis.

Fig. 5.4: Normalised Arrhenius plots of the resistive transitions for all tapes.

Fig. 5.5: Field dependence of the activation energies calculated from the slopes of the Arrhenius plots, for all tapes and in fields applied parallel and perpendicular to the
c-axis. The lines are guides to the eye, representing the approximate $H^{1/2}$
dependence.................................................................150

**Fig. 5.6:** Ratio of activation energies between the irradiated and virgin tapes, calculated
from an interpolation of $U_0$ between 0 and 3 T. $U_0^{irr}$ is the activation energy of the
(0.6, 1.75) tape.......................................................................152

**Fig. 5.7:** Irreversibility line determined from the resistive transitions. The arrow indicates
the approximately location of the matching field for the (0.6, 1.75) tape........157

**Fig. 6.1:** $U_{\text{eff}}(J,H,T_0)$ for the irradiated (solid symbols) and nonirradiated (open symbols)
tapes, with a theoretically determined $U(T) = (1-t)^{3/2}$. The lines are fits to Eq.
(6.18)......................................................................................183

**Fig. 6.2:** An example of the close matching of individual isotherms using an empirical
$U(T)$, showing a section of the $U_{\text{eff}}(J)$ curve obtained for the nonirradiated tape at
an applied field of 0.65 T..............................................................185

**Fig. 6.3:** Data set of the scaling factor $G$ at each temperature, for both tapes. The errors are
approximately the size of the symbols or smaller. The lines are the fits to Eq.
(6.15)...............................................................................................185

**Fig. 6.4:** $U_{\text{eff}}(J,H,T_0)$ for the irradiated and nonirradiated tapes with the empirically
determined $U(T)$ as shown in Fig. 6.3. The lines are fits to Eq. (6.18).........190

**Fig. 6.5:** Fig. 6.4 on a linear-log scale, demonstrating the different divergent behaviours of
$U_{\text{eff}}(J,H,T_0)$ as $M_{\text{irr}} (\propto J) \rightarrow 0$.........................................................191

**Fig. 6.6:** $U_{\text{eff}}(J,H,T_0)$, from Fig. 6.4, of the (a) irradiated and (b) nonirradiated tapes, scaled
by $H^{1/2}$.....................................................................................192

**Fig. 6.7:** Field dependence of $\mu$ from a fit to Eq. (6.18) for both the theoretically and
empirically determined temperature scaling forms $U(T)$. Lines are guides to the
eye only. The arrows indicate the approximate position of the crossover field
$H_{cr}$.........................................................................................195

**Fig. 6.8:** Current – Voltage characteristics for the (a) nonirradiated and (b) irradiated tapes.
Note the change in curvature evident above and below the dashed lines.......200
# LIST OF TABLES

**TABLE 4.1:** Zero-Field $J_C$ and Irreversibility Field for All Measured Tapes… 100

**TABLE 4.2:** Fitting Parameters for a Fit of Normalised $F_p$ Against Reduced Field………………………………………………………………………………….. 132

**TABLE 5.1:** Doping and Irradiation Combinations Measured………………… 140

**TABLE 6.1:** $C$ and $\mu$ Fitting Parameters for Fit to Eq. (6.18), Obtained with $U(T) = (1-t)^{3/2}$ …………………………………………………………………………………….. 182

**TABLE 6.2:** Fitting parameters obtained from a fit of the Empirically Determined data set $G(T)$ to the form in Eq. (6.15)………………… 184

**TABLE 6.3:** Approximate Temperature Scaling Form $U(T)$ from the Fit in Fig. 6.3, and the Associated $C$ and $\mu$ Fitting Parameters………………… 187
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_0$</td>
<td>Average vortex lattice spacing</td>
</tr>
<tr>
<td>$B_\phi$</td>
<td>Matching field</td>
</tr>
<tr>
<td>$B_{\text{eff}}$</td>
<td>Effective matching field</td>
</tr>
<tr>
<td>$c$</td>
<td>Uranium-oxide doping percentage by weight</td>
</tr>
<tr>
<td>$c_{11}$</td>
<td>Uniaxial compression</td>
</tr>
<tr>
<td>$c_{44}$</td>
<td>Tilt compression</td>
</tr>
<tr>
<td>$c_{66}$</td>
<td>Shear compression</td>
</tr>
<tr>
<td>$c_L$</td>
<td>Lindemann number</td>
</tr>
<tr>
<td>$D$</td>
<td>Flux flow density</td>
</tr>
<tr>
<td>$E_{\text{core}}$</td>
<td>Core component of vortex energy per unit length</td>
</tr>
<tr>
<td>$E_J$</td>
<td>Energy of the Josephson coupling</td>
</tr>
<tr>
<td>$E_{\text{mag}}$</td>
<td>Electromagnetic component of vortex energy per unit length</td>
</tr>
<tr>
<td>$e_l$</td>
<td>Elastic line energy</td>
</tr>
<tr>
<td>$F_L$</td>
<td>Lorentz force density</td>
</tr>
<tr>
<td>$F_p$</td>
<td>Volume pinning force density</td>
</tr>
<tr>
<td>$F_{p_{\text{max}}}$</td>
<td>Maximum volume pinning force density</td>
</tr>
<tr>
<td>$f_p$</td>
<td>Elementary pinning force density</td>
</tr>
<tr>
<td>$f(T)$</td>
<td>Free energy density</td>
</tr>
<tr>
<td>$G_i$</td>
<td>Ginzburg number</td>
</tr>
<tr>
<td>$G$</td>
<td>Multiplicative scaling factor</td>
</tr>
<tr>
<td>$H^{ab}$</td>
<td>Field orientations parallel to the crystallographic $ab$-plane</td>
</tr>
<tr>
<td>$H^c$</td>
<td>Field orientations parallel to the crystallographic $c$-axis</td>
</tr>
<tr>
<td>$H_c$</td>
<td>Thermodynamic critical field</td>
</tr>
<tr>
<td>$H_{c1}$</td>
<td>Lower critical field</td>
</tr>
<tr>
<td>$H_{c2}$</td>
<td>Upper critical field</td>
</tr>
<tr>
<td>$H_g$</td>
<td>Field of vortex-glass to vortex-liquid transition</td>
</tr>
<tr>
<td>$H_{\text{irr}}$</td>
<td>Irreversibility field</td>
</tr>
<tr>
<td>$H'_{\text{irr}}$</td>
<td>Resistively determined irreversibility field</td>
</tr>
</tbody>
</table>
\( H_p: \) Field of full penetration
\( H_{max}: \) Field of the maximum in the volume pinning force density
\( I_c: \) Critical current
\( I_{Ag}: \) Current conducted through the silver sheath
\( J: \) Current density
\( J_c: \) Critical current density
\( J_{c0}: \) Critical current density in the absence of thermal fluctuations
\( J_c^{\text{inter}}: \) Inter-granular critical current density
\( J_c^{\text{intra}}: \) Intra-granular critical current density
\( k_B: \) Boltzmann constant
\( M: \) Magnetisation
\( M_{irr}: \) Irreversible magnetisation
\( M_{rem}: \) Remnant magnetisation
\( m: \) Magnetic moment
\( m_{irr}: \) Irreversible component of the magnetic moment
\( m_{ij}: \) Effective mass tensor
\( N_f: \) Number of fission events
\( N_p: \) Density of pinning centres
\( n: \) Areal density of vortices
\( R_n: \) Normal-state resistance
\( R: \) Total measured resistance
\( R_S: \) Superconductor resistance
\( R_{Ag}: \) Silver sheath resistance
\( S: \) Stopping power
\( s: \) Distance between the superconducting CuO\(_2\) planes
\( T_c: \) Critical temperature
\( T_{dc}: \) De-coupling temperature
\( T_g: \) Vortex-glass temperature
\( T_{irr}: \) Irreversibility field
\( T_{m(H)}: \) Melting temperature
\( t_0: \) Characteristic flux hop attempt time
$U_{\text{eff}}$: Effective activation energy

$U_0$: Pinning activation energy

$U_0^{\text{vir}}$: Pinning activation energy in a virgin, nonirradiated tape

$U_0^{\text{irr}}$: Pinning activation energy in a doped and irradiated tape

$V_c$: Volume of the correlated flux bundle

$V_{\text{th}}$: Thermoelectric voltage

$V_s$: Superconductor component of the measured voltage

$v_0$: Flux hop velocity

$Z$: Atomic number

$\varepsilon$: Mass anisotropy parameter

$\varepsilon_l$: Elastic line tension

$\Phi_0$: Flux quantum

$\Phi_n$: Thermal-neutron fluence

$\kappa$: Ginzburg-Landau parameter

$\lambda$: London penetration depth

$\nu$: Flux hop rate

$\nu_0$: Characteristic flux hop attempt rate

$\theta$: Angle of the applied field to the crystallographic $c$-axis

$\rho_{\text{fr}}$: Fission-fragment defect track density

$\rho_f$: Volume density of a fissionable atom

$\sigma_{\text{abs}}$: Thermal-neutron absorption cross-section

$\xi$: Coherence length

$\Psi$: Superconducting order parameter
LIST OF ABBREVIATIONS

p-Bi: Proton-induced fission of Bi nuclei
Bi2212: Bi$_2$Sr$_2$CaCu$_2$O$_x$
Bi2223: Bi$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$
BSCCO: Bi-Sr-Ca-Cu-O
CD: Columnar Defect
FLL: Flux-Line-Lattice
G-L: Ginzburg-Landau
HIFAR: High Flux Australian Reactor
HII: Heavy Ion Irradiation
HTS: High Temperature Superconductors
IL: Irreversibility Line
$I – V$: Current – Voltage
L-D: Lawrence-Doniach
LTS: Low Temperature Superconductors
MHL: Magnetic Hysteresis Loops
P-I-T: Powder-In-Tube
PPMS: Physical Properties Measurement System
SEM: Scanning Electron Microscopy
TAFF: Thermally Activated Flux Flow
T12212: TI$_2$Ba$_2$CaCu$_2$O$_x$
TEM: Tunneling Electron Microscopy
VSM: Vibrating Sample Magnetometer
XRD: X-ray Diffraction
Y123: Y$_1$Ba$_2$Cu$_3$O$_x$
LIST OF PUBLICATIONS


**Poster Presentations:**

International Conference on Materials and Mechanisms in HTSC VI, Texas, USA, February 2000, titled "The Effects of Uranium Doping and Thermal Neutron Irradiation on the Pinning Properties of Ag/Bi-2223 Tapes".


Radiation 2000 conference held by AINSE at Lucas Heights, NSW, Australia, November 2000, titled "Improvements in the Critical Currents of High Temperature Superconductors through an Uranium Doping and Thermal Neutron Irradiation Method".

CHAPTER 1:

THEORETICAL FOUNDATIONS

1.1 INTRODUCTION

Conventional, Low Temperature Superconductivity (LTS) showed tremendous potential for applications in many areas of science and technology. Persistent supercurrents were utilised in the construction of magnets capable of producing stable fields as large as 20 T. The phenomenon of the Josephson junction resulted in devices capable of measuring magnetic fields with unprecedented sensitivity. Efficient magnetic energy storage and the loss-less transmission of electrical energy over great distances without the need for dangerous high voltages were envisaged.

Their extremely low operating temperatures, however, restricted the feasibility of these applications. The cooling requirements, necessitating the use of expensive liquid helium, imposed severe technical and economic
limitations on the realisation of these visions. LTS can be found in specialist equipment, such as sensitive magnetometers and large magnet systems used for scientific and engineering studies, or employed in Magnetic Resonance Imaging devices for non-intrusive medical diagnosis.

With the discovery of High Temperature Superconductivity (HTS) in 1986, the implementation of these applications into a much wider market became one step closer to being realised. In particular, the development of materials with a critical temperature $T_c$ above the boiling point of liquid nitrogen relaxed the restrictive cooling requirements of conventional superconductors. Liquid nitrogen is a cheaper and more readily available material than the liquid helium necessary for LTS.

However, the elevated temperatures and the ceramic nature of the new superconductors led to problems that were not previously dominant in LTS materials. HTS materials primarily consist of small crystal grains, which must be well interconnected to allow the conduction of a supercurrent on a macroscopic scale. Poor connections limit the total current that may be transported. This has presented a vexing problem to the production of long lengths of HTS with a large critical current density $J_c$.

In addition to serious problems with connectivity of superconducting grains, $J_c$ is limited by the motion of magnetic flux lines that penetrate the HTS material. The higher operating temperatures, larger
material anisotropy and smaller coherence length of HTS materials means that they are considerably more susceptible to thermal fluctuations of the penetrating flux than conventional superconductors. To reduce these effects, substantial work has been done on improving the flux pinning characteristics of HTS materials.

Over the course of the last decade, much progress has been made into the problem of weak connectivity, improving the zero-field $J_c$ to a level approaching the requirements for some commercial applications. Improvements in grain connectivity, texture and phase composition are some of the areas where progress has been made. The problem of flux pinning, however, is not well resolved by these materials processing technologies for technical applications.

One method for improving flux pinning that has shown great potential is inducing the fission of $^{235}\text{U}$ atoms that have been doped into a HTS material. Work is currently underway on maximising the enhancements that can be achieved with this method.

This Thesis will focus on examining the underlying changes to the vortex dynamics caused by the fission-fragment damage. The activation energies for vortex motion, which are dependent on the strength of the flux pinning, will be directly probed. The structure of the Thesis is as follows:
• Chapter 1 will provide a brief overview of the relevant theory, providing a foundation for the subsequent studies.

• Chapter 2 is a review of the literature on the uranium-fission method in HTS. Since the defects created by the fission process are similar in structure to those created by heavy-ion irradiation, Chapter 2 will begin with a brief review of the relevant experimental issues in heavy-ion irradiation.

• Chapter 3 details the tape processing and irradiation issues.

• Chapter 4 begins the experimental examination by characterising the changes to the field dependence of $J_c$ as a result of the fission process. Technologically, $J_c$ and the irreversibility field are the most important properties of a superconductor, and the uranium-fission method is aimed at improving both. The average pinning force density is also extracted from these measurements.

• Chapter 5 probes changes to the activation energies in the thermally assisted flux flow regime through measurements of the resistive transition.

• Chapter 6 presents an examination into the activation energies in the low-temperature flux-creep regime from dynamic magnetic relaxation measurements.
1.2 ATOMIC STRUCTURE OF HTS

1.2.1 Copper-oxide HTS properties

The majority of HTS discovered to date are ceramic copper-oxide compounds, based on a layered perovskite-like crystal structure with dominant copper-oxide planes, as shown in Fig. 1.1.

Superconductivity is believed to occur in these CuO$_2$ planes, with the primary charge carrier being electron holes. The hole density is required to lie within a specific range; a maximum value of $T_c$ is obtained with 0.15-0.2 charge carriers per Cu atom in the plane. This can be adjusted through control of the oxygen stoichiometry and metallic doping. The oxide layers separating the CuO$_2$ planes are insulators.

This layering of the crystal and electronic structures results in an extreme anisotropy of the physical properties and superconducting parameters. The models proposed to describe the properties of isotropic superconductors need to account for this strong anisotropy, requiring extensions to the existing theories, or development of new theories. Anisotropic theories had been developed to account for the observations of ...
small anisotropy in some LTS materials. When high-temperature superconductors were discovered, the LTS theories were adapted to the new superconductors.

**Fig. 1.1:** Crystal structure of typical anisotropic HTS materials (a) Bi$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ and (b) Bi$_2$Sr$_2$CaCu$_2$O$_x$. The insert illustrates the crystallographic axes.
1.2.1.1 Phenomenological Descriptions of Superconductivity

Ginzburg-Landau Theory

The Ginzburg-Landau (G-L) theory was devised to provide a phenomenological description of superconductivity in an isotropic system, particularly for analysing the behaviour in external magnetic fields. Essentially, the G-L theory is composed of two coupled equations, governing the spatial variations of a superconducting order parameter $\Psi$ and the supercurrent and local magnetic flux densities. Two characteristic lengths of superconductivity were determined by the theory. The magnetic field penetration depth $\lambda$ represents the length scale determining the electromagnetic response of the superconductor, and the coherence length $\xi$ is a measure of the length scale for spatial variations of $\Psi$. The G-L parameter is defined as $\kappa = \lambda/\xi$. Type-II superconductors, of which HTS are a subset, are defined as superconductors with $\kappa > 1/\sqrt{2}$.

Anisotropic Ginzburg-Landau Theory

The G-L theory was extended to include anisotropic superconductors by introducing an effective mass tensor that reflects the conductivity along each of the principal axes, $m_i$ ($i = a, b, c$), where $(m_a m_b m_c)^{1/3} = 1$. In HTS, anisotropy is principally uniaxial. With the CuO$_2$
layers lying in the crystallographic \( ab \)-plane (see Fig. 1.1), we find that \( m_a \approx m_b \equiv m_{ab} < m_c \). The mass anisotropy parameter \( \varepsilon \) can then be defined as
\[
\varepsilon^2 = \frac{m_{ab}}{m_c} < 1. \tag{1.1}
\]

The components along each of the principal axes of \( \xi \) are then adjusted by this mass tensor such that,
\[
\xi_i = \frac{\xi}{\sqrt{m_i}}, \quad \xi = (\xi_a \xi_b \xi_c)^{1/3}.
\]

Similarly, \( \lambda \) is adjusted,
\[
\lambda_i = \lambda \sqrt{m_i}, \quad \lambda = (\lambda_a \lambda_b \lambda_c)^{1/3}. \quad \text{Thus,} \quad \varepsilon \approx \frac{\xi_c}{\xi_{ab}} = \frac{\lambda_{ab}}{\lambda_c}.
\]

The coherence lengths are several orders of magnitude smaller in HTS compounds, typically on the order of 10 nm, than those of most conventional superconductors, with coherence lengths around \( 10^{-4} \) cm. This is particularly true of \( \xi_c \) in the anisotropic G-L model as a result of the large effective mass along the crystallographic \( c \)-axis. Conversely, the penetration depths are much larger in HTS, especially \( \lambda_{ab} \).

**Lawrence-Doniach Model**

The anisotropic G-L approach is appropriate only in the limit of small anisotropy and where the coherence length is larger than the distance \( s \) between the superconducting \( \text{CuO}_2 \) planes. If \( \xi_c(T) \ll s \), \( \Psi \) in the region between the layers is very small, and hence the order parameter becomes inhomogeneous. In such cases, the discreteness of the layers becomes important. The anisotropic G-L model is then replaced by the Lawrence-
Doniach (L-D) model. In the L-D model, the superconductor is treated as an array of discrete superconducting layers, with a thickness $d$ and periodicity $s$, which are coupled by a Josephson junction.

### 1.2.2 Bi-Sr-Ca-Cu-O HTS

The focus of this work is on the properties of the cuprate high-temperature superconductor Bi$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ (Bi2223). It is one of the most technologically important HTS materials for the transport of supercurrents over large distances. Silver-sheathed Bi2223 tapes of the order of 1km in length have been manufactured, able to carry up to 20 kAcm$^{-2}$ of supercurrent.

Optimally doped Bi2223 typically has a $T_c$ close to 110 K. The crystal structure is shown in Fig. 1.1 (a). The system has three CuO$_2$ planes, with a layer of Ca separating the planes. This structure of CuO$_2$ planes is enclosed by a SrO layer on either side, which is in turn enclosed by a BiO layer at each end, to make up the unit cell. The interlayer spacing $s \approx 18 \, \text{Å}$. A second phase of the Bi-Sr-Ca-Cu-O system that also shows high-temperature superconductivity is Bi$_2$Sr$_2$CaCu$_2$O$_x$ (Bi2212). The $T_c$ of Bi2212 is lower than that of Bi2223, $T_c \approx 85$ K. As shown in Fig 1.1 (b), the crystal structure is similar to that of Bi2223. As discussed, the structural layering results in an anisotropy of the superconducting parameters. For the
BSCCO system, in zero field and at low temperatures, \( \xi_a \approx \xi_b \approx 20-40 \text{ Å} \) and \( \xi_c \approx 0.4 \text{ Å} \), resulting in an anisotropy parameter \( \varepsilon \approx 1/50 - 1/100 \).

Since \( \xi_c < s \), Bi2223 must be examined with the L-D model.

### 1.3 Superconductor Vortex Physics

The superconducting state is a true thermodynamic phase, with an associated free energy density \( f_s(T) \). The difference in the free energy densities between the normal \( f_n(T) \) and superconducting phases in zero magnetic field is

\[
 f_n(T) - f_s(T) = \frac{H_c^2(T)}{2\mu_0} \tag{1.2}
\]

where \( H_c \) is the thermodynamic critical field. This is then the condensation energy per volume gained from undergoing the superconducting phase transition. \( H_c \) is dependent on the temperature, becoming zero at \( T = T_c \).

From the definition of the condensation energy, Eq. (1.2), it becomes apparent that the superconducting phase cannot coexist with a strong magnetic field. The magnetic energy density must be balanced against the gain in condensation energy in the superconducting phase, so that a sufficiently strong magnetic field returns the system to the normal phase. For the successful application of superconductors in a working
environment that may contain strong magnetic fields, this is disastrous. However, the superconducting phase may coexist with an external magnetic field under certain conditions.

1.3.1 *Flux penetration*

1.3.1.1 *Type-I superconductors*

In type-I superconductors, a weak external applied magnetic field is expelled from the interior of the superconductor, in a process known as the *Meissner-Ochsenfeld Effect*. In the Meissner state, a screening supercurrent is created at the surface of the superconductor, penetrating over a depth $\lambda$, such that its induced magnetic field directly opposes the external applied field. It is a state of perfect diamagnetism. The flow of shielding currents induces a magnetic moment in the superconductor, and the moment per unit volume is defined as the magnetisation $M$. The magnetisation is a measure of the difference between the external and internal fields

$$M = \frac{B}{\mu_0} - H. \quad (1.3)$$

As a result of the shielding currents, the volume average of magnetic induction within the superconductor $B$ is zero, and thus $M = -H$ in the Meissner state.
The expulsion of the magnetic flux occurs once the superconducting sample is cooled below the critical temperature, $T_c$. In the normal state, $T > T_c$, magnetic flux penetrates the sample entirely. Upon cooling below $T_c$, the flux within the superconducting sample is expelled out of the interior, except in the outer layer of depth $\lambda$. A field applied after cooling cannot penetrate the superconductor deeper than $\lambda$, which is field and temperature dependent.

Flux expulsion costs magnetic energy, $H^2/2\mu_0$, which is balanced against the condensation energy gained by undergoing the phase transition into the superconducting state, Eq. (1.2). Once the external applied field reaches a value where the cost in the magnetic energy is equal to or larger than the gain in the condensation energy, superconductivity is lost and the flux fully penetrates the sample. Therefore, the magnitude of magnetic field required to destroy superconductivity is the critical field $H_c$.

### 1.3.1.2 Type-II superconductors

In type-II superconductors, of which HTS are a subset, there is a third region between the Meissner state and the normal state, in which magnetic flux can partially penetrate the superconductor. As with type-I superconductors, flux is completely expelled up to a lower critical field $H_{cl}$, except for the outer layer of thickness $\lambda$. The internal field is zero.
throughout and the magnetisation is the same as for a type-I superconductor. Above $H_{c1}$, the partial penetration of magnetic flux becomes energetically favourable and the superconductor is in the *mixed state*. In this state, superconductivity coexists with the magnetic flux confined to a limited volume within the superconducting volume. The superconductor properties are complicated by the presence of the flux, leading to the more complex physics of type-II superconductors.

A theory for the manner in which magnetic flux penetrates an isotropic type-II superconductor was developed by Abrikosov in 1957. He demonstrated that flux penetrates in the form of a regular lattice of magnetic flux lines, or *Abrikosov vortices*. Vortex penetration occurs by nucleation at the superconductor surface. The vortices are formed parallel to the applied field and each carries a quantised unit of flux

$$\Phi_0 = \frac{h}{2e} = 2.068 \times 10^{-11} \, \text{T cm}^2.$$  \hfill (1.4)

where $h$ is Planck's constant and $e$ is the electron charge. The total magnetic induction, $B$, within a superconductor in an applied field $H_a$, is then

$$B = n\Phi_0 = \frac{\Phi_0}{a_0^2} \approx \mu_0 H_a,$$ \hfill (1.5)

where $n$ is the number of vortices per cm$^2$, and $a_0$ is the average distance between the vortices in cm. The difference between the magnetic induction
and the applied field is of the order of $H_{ci}$, which is small in HTS, and so can be neglected in applied fields far beyond the lower critical field.

1.3.1.3 Vortex Structure

The structure of an isolated vortex consists of a cylindrical non-superconducting ‘normal’ core, of radius $\approx \xi$, which contains one quantum of magnetic flux, and a flow of screening supercurrents around the core that are necessary to generate the quantum of flux, as illustrated in Fig. 1.2. The term vortex is applied to the flux lines because of this supercurrent flow. In this work, both "flux line" and "vortex" will be used interchangeably to refer to the same linear object. "Vortex" may also be applied in considering a 2-dimensional structure, which will be discussed in Section 1.3.2 below.

The superconducting order parameter $\Psi$ within the vortex core is suppressed from its maximum value outside the vortex, becoming zero at the centre. The magnetic induction $B$ of a single isolated vortex is at a maximum within the core, and decays exponentially over a length-scale of the London penetration depth $\lambda$. Consequently, the screening supercurrents

![Fig. 1.2: Structure of an isolated vortex line versus the distance from the vortex axis, $r$; (a) Density of the superconducting electrons, (b) magnetic flux density, and (c) supercurrent density. [R. P. Huebener, Magnetic Flux Structures in Superconductors (Springer-Verlag, Berlin, 1979) p62.]]
are at their maximum at the edge of the vortex core. Screening currents screen the superconductor from the flux captured inside the core, with a decay length $\lambda$. An integral of $B$ over the cross-sectional area of the vortex gives the quantised flux of Eq. (1.4).

1.3.1.4 Vortex Energy

As the vortices are associated with a suppression of $\Psi$, they carry a certain amount of energy. This energy can be split into two broad components, the core and the magnetic energies. The first is related to the change in condensation energy within the vortex core. If we can consider the vortex as a cylinder with radius $\sim \xi$, then from Eq. (1.2) the energy per unit length can be given as

$$E_{core} = \frac{H_c^2}{2\mu_0} \pi \xi^2$$

(1.6)

where $H_c = \Phi_0 / (2\pi \xi \lambda \sqrt{2}) > H_{c1}$ is the thermodynamic critical field for a type-II superconductor, defined, as in Eq. (1.2), from the difference in free energy densities of the normal and superconducting phases.

The second component, the electromagnetic energy, is related to the energy of the circulating supercurrents. Taking into consideration the magnetic field energy density and the kinetic energy of the supercurrents, the electromagnetic energy per unit length of the vortex can be estimated as
\[ E_{\text{mag}} \approx \frac{H^2 c}{2\mu_0} 4\pi \xi^2 \ln \kappa. \] (1.7)

In addition, a vortex can be considered as an elastic string, with an associated elastic line tension. In an isotropic superconductor, the line tension \( \varepsilon_l \) is equal to the line energy \( e_l \). The elastic line energy is then determined by the increase in the vortex length.

1.3.1.5 Flux-line-lattice

As the applied field increases, new vortices are nucleated at the superconductor boundaries. Consequently, the number of vortices within the superconductor increases and the average separation between vortices \( a_0 \) is reduced. At vortex separations of the order of \( \lambda \), the regions of screening supercurrents of each vortex begin to overlap. Electromagnetic interactions then become significant, resulting in a repulsive force between the vortices.

In a symmetrical arrangement, the total force on an individual vortex vanishes as the forces from neighbouring vortices cancel out. An ensemble of flux lines within a superconductor tends to form a regular lattice array. In the absence of any disorder, this lattice has long-range translational order. The lattice can be described as an elastic medium, characterised by the elastic moduli for tilt \( (c_{44}) \), shear \( (c_{66}) \) and uniaxial compression \( (c_{11}) \).
Within moderate applied fields, a triangular flux-line-lattice (FLL) is the most stable arrangement, with a lattice parameter

$$a_\Delta = \left( \frac{2}{\sqrt{3}} \right)^{\frac{1}{2}} \left( \frac{\Phi_0}{B} \right)^{\frac{1}{2}} \approx \left( \frac{\Phi_0}{B} \right)^{\frac{1}{2}} = a_0.$$  \hspace{1cm} (1.8)

The vortex cores eventually overlap when the distance between vortices is reduced to \( \sim \xi \), at which point the volume of superconducting material is almost entirely eliminated. Superconductivity is then destroyed throughout the sample and the sample makes a transition to the normal state. The field at which this occurs is the upper critical field, \( H_{c2} \). At this field, the internal magnetic induction becomes equal to the external applied field and thus the magnetisation is steadily reduced from \( M = -H \) at \( H < H_{c1} \) to zero at \( H = H_{c2} \).

Throughout the rest of this chapter, we will be considering intermediate fields \( H_{c1} \ll H \ll H_{c2} \) unless stated otherwise. The first inequality insures that the vortex spacing \( a_0 \) is small compared to \( \lambda \) so that the interactions between vortex lines are strong. The second guarantees that the vortex cores will not overlap.

### 1.3.1.6 Flux-line-lattice melting

Thermal effects can destroy the order of the FLL. At finite temperatures, thermal disorder affects the vortex system by displacing the vortices from their equilibrium positions. The lattice loses long-range order
and enters a fluid state where the vortices can move past each other freely. No consistent microscopic theory of flux-line-lattice melting has been developed to date. However, a successful approach, based on the Lindemmann criterion, has been to assume that if the mean-squared displacements from thermal fluctuations exceed a fraction of the vortex spacing:

\[
\left\langle u^2(T)\right\rangle_{th}^{1/2} \approx c_L a_0,
\]

the lattice loses its long-range translational order; where \( c_L \approx 0.1 - 0.4 \) is the Lindemann number. The lattice then undergoes a melting transition into a vortex liquid state at the melting temperature, \( T_m(H) \). The shear modulus \( c_{66} \) of the vortex lattice system is also lost upon melting and the rigidity of the lattice structure vanishes.

HTS are much more susceptible to thermal fluctuations than conventional superconductors, primarily because of the larger \( T_c \). The Ginzburg number (expressed in cgs) is a measure of the strength of thermal fluctuations in a superconductor. In conventional superconductors, typically \( Gi \sim 10^{-8} \) and thermal fluctuations are weak. For HTS, however, as a result of the large \( T_c \) and small \( \varepsilon \) and \( \xi \),
Chapter 1: Theoretical Foundations

$G_i \sim 10^{-2}$. The liquid phase of the flux-line-lattice therefore occupies a large portion of the $(H, T)$ phase diagram in HTS.

1.3.2 Layering Effects

1.3.2.1 Vortices in Layered Systems

The picture given above of the vortex lattice system is complicated in HTS by the extreme anisotropy. Since superconductivity is strictly confined to the CuO$_2$ layers, the above picture of a 3-dimensional flux line is not valid. The discrete L-D model described in Section 1.2.1.1 can be applied instead to describe the superconductivity.

For simplicity, begin by considering applied fields oriented along the crystallographic $c$-axis. The L-D model then results in discrete 2D vortices restricted to individual superconducting layers, coupled across the layers by Josephson strings. The structure of a 2D vortex in the CuO$_2$ plane is similar to the cross-section of a 3D flux line, but the extent of the vortices along the field direction is limited by $\xi_c$. As $\xi_c \ll \xi_{ab}$ and the circulating supercurrents are confined to the superconducting plane over a length-scale $\lambda_{ab}$, this results in a flat structure, and hence the term 'vortex pancake' is applied.

The Josephson vortices connecting the pancakes across the layers consist of highly elliptical currents circulating about an axis that lies
parallel to the layers. The circulating currents consist of supercurrents flowing along the layers over a length-scale of $\lambda_{ab}$ and weak Josephson currents in the direction perpendicular to the layers.\(^{16}\) The energy of the Josephson coupling is given (in cgs) by\(^{17}\)

$$E_J = \frac{\Phi_0^2}{\pi(4\pi\lambda_c)^2} s.$$  \hspace{1cm} (1.11)

In the limiting case where $E_J = 0$, the vortices are instead only weakly coupled by an electromagnetic coupling term that results from the magnetic interaction of the circulating supercurrents of the 2D pancake vortices.\(^{16}\)

The magnetic field and current distributions of the 2D pancakes result in an interaction force between vortices lying both in the same CuO$_2$ plane and in neighbouring layers.\(^{16}\) As for 3D flux lines in an isotropic superconductor, the interaction between pancake vortices in the same layer (\textit{intra-layer} interaction) is repulsive, with a logarithmic dependence on the distance between them. Within different CuO$_2$ layers, individual 2D vortex-solid lattices can result from this interaction, in the same manner that the 3D flux-line-lattice is created in isotropic superconductors.

Two pancakes in different layers (\textit{inter-layer}), on the other hand, experience a weak attractive coupling force, in addition to the Josephson coupling, which favours axial alignment. The combined inter-layer coupling results in linear stacks of pancake vortices aligned along the axis.
perpendicular to the layers. If $\xi_c > s$, the pancake stacks produce magnetic field and current distributions that resemble those of the anisotropic G-L theory, even in the weak coupling limit.\(^{16}\) Thus, a string of inter-layer coupled 2D pancake vortices can approximate 3D flux lines, with the addition of the adjustments to $m$, $\xi$ and $\lambda$ as described in Section 1.2.1.1 and the behaviour of the system approaches that of the 3D flux-line-lattice solid described above. Like the 3D flux line, a coupled stack of 2D pancakes can have an elastic line energy. Unlike the isotropic case, however, the line energy is no longer equal to the line tension. The line energy has an additional dependence on the angle between the vortex and the superconducting planes.\(^{13}\)

For applied fields oriented at (non-vanishing) angles relative to the $c$-axis, the model of the string of pancake vortices is also applicable. In this case, the Josephson vortices connecting the centres of the vortex pancakes are inclined along the field direction. In applied fields parallel to the superconducting layers, flux consists entirely of Josephson vortices.

The L-D model provides a basis for a 2D description of the vortex state. However, the weak Josephson and electromagnetic coupling allow for a degree of 3D behaviour of the vortex system. When $\xi_c > s$ or the Josephson coupling is strong, the superconductivity can be described by the
anisotropic G-L theory and the weakly coupled system is said to be in a quasi-2D, or 3D line-like, state.

1.3.2.2 Dimensional Crossover

As the applied field is increased, the inter-layer Josephson coupling, Eq. (1.11), is reduced and the intra-layer interactions between the pancakes becomes dominant. The vortex system then loses its 3D line-like behaviour and begins to behave as individual 2D pancake lattices within different layers. Thus, the reduction in the inter-layer Josephson coupling as the applied field is increased leads to a field-induced dimensional crossover from 3D line-like behaviour to a pure 2D vortex pancake system. This crossover field $H_{cr}$ has been theoretically estimated as

$$H_{cr} \approx \frac{\Phi_0^2 \varepsilon^2}{s^2}.$$  \hspace{1cm} (1.12)

Above $H_{cr}$, the pancakes can still have strong intra-layer interactions despite the vanishing inter-layer coherence, resulting in the 2D vortex arrays forming independent solids within different layers. The persistence of the magnetic interaction results in a finite tilt modulus $c_{44}$ even in the decoupled state, so the individual vortex solids are weakly interacting at low temperatures.
1.3.2.3 Lattice Melting in Layered Systems

De-coupling of the 2D pancake stacks can also be thermally induced. The inter-layer coupled stacks of 2D pancakes are susceptible to thermal fluctuations in much the same manner as a 3D flux-line-lattice. Thermal energies displace pancake vortices from their equilibrium alignments; if the displacement is sufficiently strong, it will overcome the inter-layer pancake coupling. The pancake stacks are de-coupled by strong thermal fluctuations at a de-coupling temperature $T_{dc}$.\textsuperscript{16, 17} In the limit of weak Josephson coupling, where only the weak magnetic coupling is relevant, this break-up can occur at quite low temperatures.\textsuperscript{16}

As discussed in Section 1.3.1.6, the translational order of the FLL is destroyed by thermal fluctuations at the melting transition. In fields $H < H_{cr}$, melting of the vortex-lattice-solid occurs as previously discussed, at a temperature (expressed in cgs)\textsuperscript{17}

\[ T_m \approx c_L^2 L \Phi_0^2 \left( \frac{4 \pi \chi_{a,b}}{H_{cr} } \right)^2 \left[ \frac{H_{cr}}{H} \right]^{1/2}. \tag{1.13} \]

The shear modulus $c_{66} \rightarrow 0$ and long-range order is lost for $T > T_m$, but the tilt modulus $c_{44}$ remains non-zero. The pancake vortices are still coupled across the layers but the system is in a fluid state. A further increase in $T$ results in a de-coupling transition at\textsuperscript{17}
At this temperature, the tilt modulus $c_{44} \rightarrow 0$ and the system undergoes a transition into a liquid of pancake vortices. Vortex lattice melting in a layered superconductor is thus a two-stage transition.

Alternatively, if $H > H_{cr}$, the system is already in a 2D state, with weakly interacting vortex solids within different layers. Melting in this regime is very close to that found for a single superconducting layer $T_m^{2D}$, which is field independent. Both $c_{44}$ and $c_{66}$ are reduced to zero at this transition.

### 1.4 Energy Dissipation in Type-II Superconductors

The significance of the penetration of flux in a type-II superconductor, particularly in HTS, is that, under the right conditions, it allows the transport of current densities within the bulk of a superconductor. However, vortex motion results in energy dissipation in the superconducting state. To restrict the motion, flux pinning is introduced, and it is the flux pinning that is responsible for the transport of
bulk current densities. Flux penetration is therefore a necessary evil in the successful application of superconductivity.

The motion of flux within a superconductor would appear to be incompatible with superconductivity. Superconductivity is by definition the conduction of an electric current without resistance. Motion of the penetrating flux lines, however, dissipates energy, inducing an electrical resistance. To regain loss-less conductivity in the mixed state, it is therefore necessary to restrict the flux motion. In this Section, the effects of flux motion, the pinning required to restrict the motion and the effects of the pinning on the FLL properties will be discussed.

**1.4.1 Vortex Motion**

Energy dissipation in type-II superconductors arises primarily from the motion of vortices in the mixed state. From the Maxwell's equation

$$\nabla \times E = -\frac{\partial B}{\partial t},$$  \hspace{1cm} (1.15)

any change in the flux density over time will induce an electric field, $E$, in the sample. In the presence of a current density, $J$, this flux motion will dissipate energy $E \cdot J$ in an Ohmic conductor, which is equivalent to the induction of a resistivity,

$$\rho = \frac{E}{J},$$ \hspace{1cm} (1.16)

in the sample.
A physical interpretation of the dissipation process is well described for LTS, and can be extended to describe the process in HTS. A steady state current flow can be described in a superconductor as a superposition of the transport current and the screening currents around the vortex core. Motion of the vortices causes some of the transport current to be driven into the core where, because $\Psi$ is suppressed within the core, the current gains a normal component. This normal component of the current interacts with the crystal lattice and dissipates energy via viscous drag on the moving vortices, leading to the appearance of an electric field within the superconductor.

Motion of the vortex core also causes a retarded relaxation of $\Psi$. The physical properties of the crystal lattice are slightly different between the normal and superconducting states, particularly the volume $V$ and elastic constants $C$. The lattice is denser and stiffer in the normal state, $V$ and $C$ being smaller in the superconducting state, with relative changes of the order of $\Delta V/V \approx 10^{-7}$ and $\Delta C/C \approx 10^{-4}$. As the region of suppressed $\Psi$ moves, the crystal lattice relaxes, creating further dissipation as it interacts with the transport current.

The flux lines can be driven into motion by the presence of a current density. When a transport current density is applied, flux motion is driven by the action of a Lorentz force $F_L$ on the flux lines. This is the average
magnetic force of interaction between the moving charges of the supercurrent and the magnetic vortices,

\[ F_L = J \times B . \]  

(1.17)

Substituting Ampere's law

\[ J = \nabla \times H , \]  

(1.18)

into the above results in a general expression

\[ F_L = (\nabla \times H) \times B \]  

(1.19)

for both an applied current density as given by Eq. (1.17) or an applied field \( H \).

Even in the absence of an external applied magnetic field, there is dissipation within the superconductor when a driving current density is applied. From Ampere's Law, the flow of the driving current induces a magnetic field around the superconductor. If the self-field generated exceeds the \( H_{c1} \), flux penetration can occur. This self-field then dissipates energy as for any externally applied field.

The most significant effect of dissipative flux motion, particularly for practical applications, is to limit the bulk current densities that can be conducted without loss of superconductivity, and the persistent currents that can be maintained. These effects provide a severe limitation to the application of HTS in commerce and industry. Thus, it is of prime importance from a practical viewpoint to eliminate flux motion or reduce it
to the greatest extent possible. This can be achieved by improving the flux pinning properties of HTS materials.

1.4.2 Flux Pinning

To restrict flux motion from occurring, and thus reduce dissipative losses, disorder must be created from which it is energetically unfavourable for the flux lines to move. Local inhomogeneities in the superconductor properties create regions of lowered free energy within which flux lines may be localised, restricting their motion. Hence, for an ideal homogeneous superconductor any arbitrarily small current density will produce dissipative motion, as there are no points of lower free energy in which the flux lines may be accommodated. In this case, $F_L$ is opposed only by a frictional force resulting from the viscous drag of the FLL.

The sites of local depressions in the free energy of the vortex system are called flux pinning centres. Examples of typical features that affect the vortex energy and create flux pinning are:

- precipitates of second phases,
- chemical dopants,
- crystal lattice dislocations or voids,
- twinning defects,
- strain fields and

[28]
• grain boundaries.

Some of these defects can be found in virgin, as-grown materials. Others may be introduced by chemical doping during processing, or by additional working of the material after processing, for example by mechanical treatments or irradiation exposures, which will be discussed in the next Section.

The various possible kinds of pinning centres can be classified into two broad groups according to the type of disorder induced: correlated or uncorrelated disorder. A random distribution of small pinning centres with dimensions $\sim \xi$, known as point pinning centres e.g. oxygen vacancies or atomic dislocations, will induce uncorrelated disorder in the vortex lattice, see Fig. 1.3 (a). Correlated disorder, Fig 1.3 (b), on the other hand, is created by extensive defects that provide strong pinning, such as twin boundaries or tracks of non-superconducting amorphous material created by energetic heavy-ions.

![Fig. 1.3: (a) Uncorrelated point-like disorder; (b) Correlated disorder in the form of a columnar defect. [G. Blatter, *Physica C* 282-287, p19 (1997)]](image-url)
Vortex- and Bose-Glass Phases

With random, point pinning, the elastic flux lines are distorted to follow the optimal pinning configuration, leading to line wandering. The ideal regular hexagonal lattice arrangement is affected by the spatial distortion as the vortices accommodate to the pinning sites. The resulting redistribution of the flux lines is a competition between the pinning strength, the repulsive vortex interaction energy and the elastic energy of the line. The lattice is then predicted to break up into a series of domains where, within a finite correlation length, vortices keep a sense of the order of the regular FLL. This lack of true long-range order while maintaining a stable configuration is described as a glassy arrangement of vortices. Hence, the system is described as being in a vortex-glass state. Similar to the melting phase transition described in Section 1.3.1.6, the vortex-glass can undergo a phase transition into a vortex liquid along a line in the \((H, T)\) phase diagram, at \((H_g, T_g)\) the vortex-glass field and temperature.

In the case of strong correlated disorder, flux lines are localised along the length of a linear defect, providing the most efficient type of pinning. Strong correlated disorder results in the complete destruction of the original vortex lattice order. The strong localisation of the vortices on the pinning defects, however, leads to a Bose-glass phase. This Bose-glass phase is distinct from the vortex-glass phase. While both possess very
similar dynamics properties, they differ in statistical mechanics properties.\textsuperscript{13}

1.4.2.1 Pinning Mechanisms

By lowering the energy associated with the vortices, the pinning centres act as potential wells. These pinning potential wells are associated with a pinning barrier energy $U_0$ and an elementary pinning force $f_p$, equivalent to the spatial gradient of the energy, determined by the particular pinning mechanism.

As discussed in Section 1.3.1.4, the vortex energy is divided into two broad components, the core and electromagnetic energy. Pinning can therefore occur via changes in either of these two components. In addition, localised changes in the structural energy, for example due to the changes in volume and elastic properties between the normal and superconducting states discussed earlier, can provide a pinning force.

**Core Pinning Interaction**

If the core passes through a region where $\Psi$ is already zero or reduced, then all or part of the condensation energy required for the formation of the vortex core can be recovered. This region will have lower condensation energy, resulting in a lower energy requirement for the localisation of vortices at this site. Inclusions of normal state material or
voids in the crystal lattice can provide regions of $\Psi \approx 0$, whilst inclusions of secondary phases with a lower $T_c$ or $H_{c2}$ will have a reduced $\Psi$. The reduction in the core energy is dependent on the volume of suppressed $\Psi$ and on the degree of the suppression.\textsuperscript{12} Since the pinning force is the spatial gradient of the pinning energy, the maximum pinning force is limited by the size of the core $\approx \xi$ and the condensation energy, Eq. (1.6).

**Electromagnetic Interaction**

Inhomogeneities can affect the distribution of fields and supercurrents of the vortices, leading to a variation in their electromagnetic energy. In this case, the vortex can feel the pinning force even when the core is outside the pinning centre.\textsuperscript{12} For example, a vortex near the surface of the superconductor is forced to alter its distribution of circulating supercurrents so that the component of the supercurrent normal to the surface vanishes at the surface. The result is a surface potential barrier for flux entry or exit.\textsuperscript{25}

Since the electromagnetic energy varies on a length scale of $\lambda$, and $\lambda \gg \xi$ for HTS, the gradient of the electromagnetic pinning energy is much smaller than that of the core pinning energy. Thus, the electromagnetic pinning interaction is considered in most cases to be weaker than the core pinning interaction, unless the pinning centres are densely packed, in which case the electromagnetic interaction must be considered.\textsuperscript{12}
Elastic Energy Interaction

A third type of pinning interaction occurs through variations in the volume and elastic constants.\textsuperscript{26} As described in Section 1.4.1 above, the atomic lattice is contracted within the normal core. This leads to a strain field surrounding the vortex core, which interacts with lattice imperfections such as dislocations, stacking faults and voids. There is also a second-order interaction where the energy of the defects is dependent on the elastic constants, which are slightly stiffer within the vortex core. However, these contributions are negligible in comparison to the core and electromagnetic interactions.\textsuperscript{26}

1.4.2.2 Intrinsic pinning in layered HTS

The layered structure of anisotropic HTS materials leads to a strong intrinsic pinning effect. The superconducting order parameter is strongly suppressed in the region between the CuO\textsubscript{2} layers when $\xi_c$ is much smaller than the inter-layer spacing $s$. From the above discussion on the core pinning mechanism, it can be seen that this will result in a strong pinning force acting on magnetic flux in this inter-layer region. Thus, magnetic flux oriented parallel to the $ab$-plane of highly anisotropic HTS will tend to lie between the layers.\textsuperscript{13}
1.4.2.3 **Summation of pinning forces**

The total pinning force experienced by a flux-line-lattice is a summation of all of the elementary pinning forces for all of the vortices in the lattice. However, the above elementary pinning mechanisms are derived for a single isolated vortex. At higher flux densities, the elasticity of the entire flux-line-lattice must also be taken into account.

If each vortex could be individually pinned by a single pinning centre, then the total pinning force is a direct summation of each of the individual forces. This scenario is useful when considering very low densities of vortices, such that each vortex may be considered independent of the rest, or if the total FLL can be deformed plastically, so that each vortex in the lattice can be pinned at an individual position.

The rigidity of the lattice prevents the full pinning force from being applied. A certain degree of FLL elasticity is required to obtain a finite pinning force on the FLL, particularly for a random distribution of pinning sites. Elasticity allows the flux lines to accommodate to the local pinning sites, bending and distorting to occupy the most energetically favourable position in the pinning potential landscape. This results in a distortion of the lattice that maximises the pinning force along a particular direction. In the extreme situation where there is no solid lattice, such as in the case where the vortex system is fluid, effective pinning would require every
vortex to be localised within a pinning centre. The elasticity of the vortex lattice in real systems complicates the summation of the elementary pinning forces, however, beyond the simplicity of this model. When the interaction with the pinning centres distorts the FLL elastically, the direct summation is not valid.

### 1.4.2.4 Critical Current Density

Regardless of the specific method in which pinning forces are summed, for motion of a pinned FLL to occur the driving force on the vortex lattice must exceed an *average* pinning force density $F_p$. In the case of an applied current density, the resulting Lorentz force on the vortex lattice must be greater than $F_p$ before dissipative vortex motion will be induced. This requires that the current density exceed a critical value, which is determined by the magnitude of the pinning force, and is the definition of the *critical current density*, $J_c$,

$$F_L = -F_p = J_c \times B.$$  \hfill (1.20)

Throughout the rest of this work it is assumed the current density was always applied perpendicular to an applied magnetic field $H_{c1} \ll H \ll H_{c2}$, so that $H \approx B$ and

$$F_p = J_c H.$$  \hfill (1.21)
For current densities $J > J_c$ the system is in the *flux flow* regime, where flux moves freely and the resistivity approaches Ohmic behaviour for $J \gg J_c$.

By definition, the critical current density is the maximum current density that may be transported without producing dissipation. Equation (1.21) predicts an abrupt transition from dissipation-less to resistive current transport. However, in practice, the transition to resistive current transport is smoothed out by any spread in the pinning energies, thermal effects due to the high operating temperatures (see Section 1.4.3 below) and the complexities of the vortex state structure. Consequently, $J_c$ is defined in practice as the current density that produces an arbitrarily defined electric field in the superconductor, usually $1 \, \mu$V/cm. The critical current density is thus not an intrinsic property of a superconductor. Rather, it is material dependent and can be altered by processing that introduces further pinning centres (see Section 1.5).

The finite limit to the current density that may be carried without resistive dissipation results in a limit to the manner in which flux penetrates a superconductor. The pinning force opposes the propagation of flux through the superconductor by limiting the flux motion. This creates a critical gradient in the flux density, which is dependent upon $J_c$. The description of this process is known as the *Critical State Model*. 

\[\text{Critical State Model}\]
1.4.2.5 Critical State Model

Flux penetration occurs via nucleation of the vortices at the superconductor edge. As the flux is driven into the superconductor, a gradient of flux lines is induced, due to the vortex pinning. For simplicity, we will consider a 1-dimensional geometry, where the field has a component in one direction only; for example, a slab of infinite size along the $x$- and $z$-axes and finite thickness $d$ along the $y$-axis, with a magnetic field applied along $z$. The general expression given in Eq. (1.19), in the critical state where $F_L = F_p$, becomes

$$\frac{\partial H_z}{\partial y} B_z = F_p. \tag{1.22}$$

In fields well within the mixed state, $H_{c1} \ll H \ll H_{c2}$, where $H \approx B$, it can be seen that a finite flux density gradient $\frac{\partial B_z}{\partial y}$ will be maintained in the superconductor, the size of which depends on the average strength of pinning. So one can define

$$\frac{\partial B_z}{\partial y} = J_c. \tag{1.23}$$

Hence, the maximum flux gradient that can be sustained in a superconductor is limited by $J_c$. If we consider $J_c$ independent of field, then the flux gradient will be a constant equal to $J_c$. The system is then said to be in the critical state.
If a magnetic field of strength $H_a > H_{c1}$ is suddenly applied to a superconductor within which initially $B = 0$, the result of Eq. (1.23) is that the field profile is a linearly decreasing function of distance from the edge. Hence, flux penetrates only to a depth

$$\Delta \approx \frac{H_a}{J_c}. \quad (1.24)$$

Alternatively, this can be seen as the depth over which the induced shielding currents, which have a maximum current density $J_c$, are just sufficient to maintain the internal local field at zero, for $y > \Delta$. Figure 1.4 illustrates the field distributions in this critical state model. The current density thus flows with a value of $J_c$ in a surface layer of depth $\Delta$.

Increasing the applied field increases $\Delta$, until $\Delta = d/2$, at which point the field has propagated through the entire thickness of the superconductor and the currents are maintained throughout the entire bulk of the superconductor. The field of full penetration $H_p$ is then given as

$$H_p = J_c d/2, \quad (1.25)$$

![Fig. 1.4: Magnetic field profile inside a superconductor in the critical state model in (a) increasing and (b) decreasing applied fields. [V. Z. Kresin and S. A. Wolf, Fundamentals of Superconductivity (Plenum Press, New York, 1990), p150]
and for applied fields $H \geq H_p$, flux penetrates the entire volume of the superconductor, but is not homogeneous due to the vortex pinning. The entire basis for a superconducting current flow through the bulk of a mixed-state superconductor is thus the gradient of the vortex density, maintained by the pinning centres (where the field is proportional to the vortex density, since each vortex carries one flux quantum). In the Meissner state, a high current can flow without dissipation, but it is limited to a surface layer of thickness $\sim \lambda$, which is insufficient for practical applications.

Even with a homogeneous distribution of pinning centres in the sample, the vortex density will not be homogeneous. This is because $J_c$ (defined by the vortex pinning) is not high enough to cancel out the external field $H_a$ over the whole of the superconducting volume. The magnetic induction $B$ diminishes gradually from the surface of the sample, becoming zero at the depth $\Delta$, which is much larger than $\lambda$. The screening currents screen out the field from the volume inside the current loop. For a particular point $y = Q$, so that $\Delta > Q$, only the currents circulating at $y < Q$ will be contributing to the screening of external field. The greater the value of $Q$, the greater the proportion of the currents that will be contributing to
the screening and the smaller the internal field at this point. This is reflected in the gradient of the vortex density.

When the applied field is removed, the surface shielding currents reverse. The density gradient of flux exit is still equal to $J_c$. As can be seen in Fig. 1.4 (b), when $H$ is reduced to zero, flux remains trapped within the superconductor and the magnetisation remains finite. This remanent magnetisation $M_{\text{rem}}$ is a direct consequence of the pinning forces within the superconductor. If $F_p$ is increased, $J_c$ is similarly increased, with the result that both $H_p$ and $M_{\text{rem}}$ are larger.

1.4.2.6 Magnetic Hysteresis and Irreversibility

The critical state model provides an explanation for the field dependence of the sample magnetisation. In Eq. (1.3), the magnetisation was given as the difference between the average volume field of the superconductor $B$ and the external applied field $H$. In an applied field increasing monotonically from zero, the critical state model demonstrates that the magnetisation will be negative, and the internal average field is lower than the external applied field. When the field is reduced from a maximum applied field $H_a$, on the other hand, the magnetisation becomes positive due to the trapped flux, and remains finite even when the external field is reduced back to zero, resulting in the remanent magnetisation already discussed.
The magnetisation is thus dependent on the magnetic history and is said to be *irreversible*. The critical state model depends on the presence of flux pinning to introduce a critical current density. Flux pinning is therefore responsible for introducing the hysteresis into the field dependence of the magnetisation.

The hysteresis remains as long as the FLL can remain pinned and $J_c$ remains finite. In the absence of any pinning, for example, there are no barriers to the propagation of the flux within the superconductor and $J_c$ is zero. Alternatively, if the vortices are sufficiently depinned by thermal or magnetic effects (see Section 1.4.3), $J_c$ is reduced to zero. In both cases, the magnetisation is *reversible*; that is, it does not depend on the magnetic field or temperature history of the sample.

This field- and temperature-dependent change in the hysteresis of the magnetisation defines a transition line in the $(H, T)$ phase diagram. Below the line, magnetisation is irreversible due to the strong pinning of the superconductor and $J_c$ is finite. Above the line, however, pinning is negligible, $J_c$ is zero and the magnetisation is reversible. This line in the $(H, T)$ phase diagram, $(H_{irr}, T_{irr})$, is known as the *Irreversibility Line* (IL). In practice, the IL defines the limit of technical usefulness for bulk transport of a current density. In temperatures and fields above the IL, the zero $J_c$ renders the superconductor ineffective.
The critical current density can be calculated from the magnetic hysteresis in the critical state model. The difference in magnetisation between the field increasing and field decreasing branches of a magnetic hysteresis loop $\Delta M$ can be related to the strength of $J_c$. The exact dependence on $J_c$ is determined by the geometry of the superconductor and the orientation of the applied field, but is only valid in fields $H > H_p$. For example, for a plate of thickness $d$ and lateral dimensions $a$ and $b$,

$$J_c = 20 \frac{\Delta M}{a - \frac{a}{3b}}$$

(1.26)

where $a < b$.

1.4.3 Thermal de-pinning and Flux Creep

Vortex motion may also be driven by thermal excitations. At $T = 0$, the energy to de-pin a flux line from its pinning centre can only come from the Lorentz force interaction with a driving current density $J \geq J_c$ (ignoring quantum tunneling effects). At finite temperatures, however, de-pinning may be caused by thermal excitation of the flux vortices, even for $J \ll J_c$. In this case, the vortices acquire an activation energy that exceeds the pinning potential barrier energy $U_0$, leaving them free to hop to the next favourable pinning site. This process is called thermally activated flux flow (TAFF).
In conventional superconductors, flux creep is only observed at temperatures close to $T_c$, and is not of major importance. In HTS, flux creep, like lattice melting, is more dominant and is observed over a much larger section of the ($H, T$) phase diagram. This is a result of the combination of the large working temperatures and the small elementary pinning energies due to the small $\xi$.\textsuperscript{13} Therefore, in HTS, flux creep plays a much larger role in the vortex dynamics.

As seen from the critical state model, Section 1.4.2.5, pinning maintains a finite flux density gradient in the superconductor. The pinned lattice is therefore in a metastable state, far from thermodynamic equilibrium. Flux creep allows the flux density gradient of the pinned lattice to relax towards thermodynamic equilibrium through vortex rearrangement. This creates a redistribution of the current loops in the superconductor, which leads to a change in the magnetic moment over time. The flux density gradient thus relaxes towards a state where the flux is uniform throughout the superconductor. Hence, the moment is reduced, as are the screening currents.

A fundamental assumption of the critical state model, however, is that the flux density gradient is a constant, defined by the critical current density. With magnetic relaxation, this concept does not hold true, and therefore must be changed. Instead, we can consider a current density that
is determined by the time-scale of the experiment in place of the constant $J_c$, and the basic structure of the theory remains. The magnetisation of the sample is then proportional to the (time-dependent) current density $J$ rather than $J_c$.

The process of thermally activated flux creep was first pointed out by Anderson. According to the theory, creep involves bundles of flux lines hopping over the pinning barriers as a result of thermal activation and assisted by the Lorentz force. The concept of a flux bundle arises due to the vortex-vortex interaction. Even in the case where pinning is strongest for individual flux lines, the movement of a single flux line is suppressed because of the repulsive forces from the (stationary) neighbouring lines. Instead, a group of lines must move simultaneously. The size of the flux bundle depends on the screening of the vortex-vortex interaction, which is of the order of $\lambda$.

The hopping rate $\nu$ for activation of a flux bundle was given in the form of an Arrhenius relation,

$$\nu = \nu_0 \exp\left(- \frac{U_{\text{eff}}}{k_B T} \right)$$

(1.27)

where $\nu_0$ is a characteristic attempt rate and $U_{\text{eff}}$ is an effective activation energy. As pointed out above, the flux hopping process can be assisted by a driving Lorentz force, even for $J \ll J_c$. Thus, the effective thermal energy required for flux hopping is the energy barrier $U_0$ associated with the
pinning site in the absence of a driving force reduced by the Lorentz work contribution. Consequently, the effective activation energy is expected to be a decreasing function of the current density, approaching zero at $J = J_c$.

To a first approximation, it was estimated\textsuperscript{11} that the dependence would be linear in $J$, given as

$$U_{\text{eff}} = U_0 \left(1 - \frac{J}{J_{c0}}\right),$$

(1.28)

where $J_{c0}$ is the critical current density in the absence of thermal fluctuations. This approximation is known as the *Kim-Anderson* model.

Combining Equations (1.27) and (1.28), and defining the characteristic hop attempt time $t_0 = 1/\nu_0$, a basic equation of flux creep can be found,

$$J = J_{c0} \left[1 - \frac{k_B T}{U_0} \ln \left(\frac{t}{t_0}\right)\right].$$

(1.29)

Thus, flux creep in the Kim-Anderson model results in a logarithmic decay of the current density and hence the magnetisation of the sample.

The decay rate can be estimated from Eq. (1.29) as

$$\frac{1}{J_{c0}} \frac{dJ}{d \ln(t)} = -\frac{k_B T}{U_0}.$$

(1.30)

The left-hand-side of Eq. (1.30) is defined as the *normalised relaxation rate* $S(T)$. Experimentally, the decay of $J$ can be estimated using the critical state model, Section 1.4.2.5, from the time decay of the irreversible magnetisation $M_{\text{irr}} = \Delta M/2$, which is the irreversible component of the
superconductor magnetisation. Thus, measurements of the experimental
decay rate can lead to a determination of $U_0$.

The predicted logarithmic decay described well the behaviour of
conventional type-II superconductors. However, deviations from
logarithmic behaviour were observed in HTS materials, particularly at
high temperatures, even in very early research, due to the size of the creep
effect. In HTS, the Kim-Anderson model is valid only at low temperatures
and high fields. Various other models have been proposed to explain the
non-log decay, and these will be discussed in further detail in Chapter 6.

Even from this simple model, though, it is apparent that the decay is
inversely proportional to the pinning energy. Increasing the pinning energy
of a HTS material will therefore not only improve the critical current
density, but will also have the effect of reducing the decay of persistent
superconducting currents.

1.5 Pinning Improvements through Irradiation Methods

Ideally, for successful integration into commercial and industrial
applications, HTS materials must be able to conduct large amounts of
current, preferably over great distances, within any magnetic environment,
Flux pinning is responsible for the existence of large bulk superconducting currents, up to a maximum value $J_c$ that is dependent on the pinning strength. An increased pinning strength also reduces the rate of decay of persistent currents. Therefore, all of the above specifications require strong pinning of the penetrating flux lines in the mixed state.

Presently, the as-grown HTS materials have insufficient intrinsic pinning. Critical current densities are much too low for practical applications and the giant flux creep effect results in rapid decay of persistent current densities. The dominance of the melting effect also limits the useful range of temperatures and magnetic fields. It is therefore essential that flux pinning be improved by additional techniques.

1.5.1 Improving the Pinning Landscape

A number of methods have been used to address the problem of the flux pinning over the course of the last fifteen years, such as those discussed in Section 1.4.2. For example, crystal lattice dislocations or voids, as well as strain fields, can be introduced into a superconductor by mechanical treatments, such as cold-pressing. Impurities may be introduced by chemical doping before processing. This can involve doping with nano-particle inclusions of a normal metal or oxide, such as MgO or

and all with the loss-less efficiency expected of an ideal superconductor.
Ca$_2$CuO$_3$, which provides small regions of zero $\Psi$. Regions of suppressed $\Psi$ can also be created by allowing the formation of secondary phases during processing that have a different $T_c$ or $H_{c2}$. A technique that is very successful is irradiation with high-energy particles. Several different types of radiation techniques can provide possible solutions to the flux pinning dilemma. Irradiation with light particles, such as protons or neutrons, provides moderate pinning structures that have an isotropic aspect and can be created throughout the entire volume of an irradiated material. High-energy heavy-ion irradiation, on the other hand, produces much stronger pinning centres, but is limited by the short range of the incident ions within the material, as well as the directional nature of the irradiation. A compromise between the above techniques is the creation of strong pinning centres by fission-fragment damage from the fission of atoms within the superconductor matrix.

The pinning centres are created in the irradiation procedure by producing amorphous regions within the superconductor matrix that have a reduced $\Psi$. The loss of kinetic energy from the irradiating particles to the superconductor matrix results in either localised melting of the crystalline lattice or in atomic dislocations, each of which become flux pinning sites. Inevitably, a small fraction of the superconducting volume is destroyed by the irradiation defects. However, the gain in critical current density and
other features due to the improved pinning can far outweigh any small losses in volume.

Thus, not only have irradiation techniques proven to provide excellent improvements in flux pinning, but they can also be tailored to produce a variety of pinning effects. Irradiation techniques allow a great deal of control over the interaction defects created, and so are useful for the investigation of pinning and flux dynamics.

1.5.2 Energy Loss

The formation of interaction points by irradiation is determined by the energy loss of the irradiating particles. Energy loss is expressed in terms of the stopping power, \( S = \frac{dK}{dx} \), the expected value of loss of kinetic energy \( K \) per unit path length \( x \). Energy loss can occur through three separate mechanisms: Elastic or inelastic coulomb interactions with the nuclei of the target material; interaction with the atomic electrons of the material; and radiative losses, ‘bremsstrahlung’. The dominance of each mechanism of energy loss is dependent on the mass of the target atoms, and the mass and energy of the incident ions.

1.5.2.1 Radiative Energy Loss

Radiative losses result from large acceleration or deceleration of a charged particle. Radiation of the lost energy occurs in the form of gamma-
or x-rays, with an intensity that is inversely proportional to the square of the incident particle mass. This is the dominant loss for electron irradiation at low energies, but for ions the size of protons or larger radiative losses are negligible.

1.5.2.2 Nuclear Energy Loss

Nuclear energy loss dominates where the incident ion has low atomic number, \( Z \), and/or low energy. For neutron irradiation, because of their charge neutrality, there is no interaction with the atomic electrons and energy loss is completely due to nuclear collisions. If the interaction occurs elastically, momentum is conserved. The energy transferred in this process will vary with the scattering angle, from zero to a maximum energy, \( E_{\text{max}} \), corresponding to \( 180^\circ \) scattering. An inelastic collision, on the other hand, also loses a small portion of the energy to excitation of the nucleus.

The nuclear interaction, known as Rutherford scattering in the case of coulomb repulsion of ions, thus transfers energy directly to the crystalline lattice. If the energy transferred is above a material dependent threshold, the nucleus may be dislodged from its lattice position. Hence, those atoms that receive energy by direct collision with the incident radiation are known as Primary Knock-on Atoms (PKA).

For large incident energies, the transferred energy may be significant in comparison to the threshold energy required for atomic displacement.
The PKAs are then produced with a substantial kinetic energy, which considerably exceeds the threshold value for defect formation. Hence, the PKAs themselves will trigger further lattice displacements as they collide with neighbouring atoms. The end result is that cascades of displacements are created, producing a strong cluster of defects, with the extent of the cascade dependent on the initial particle mass energy and the subsequent kinematics.

**1.5.2.3 Electronic Energy Loss**

The electronic energy loss gradually becomes more predominant as $Z$ and energy are increased, until nuclear interactions become negligible. Energy is transferred, via coulomb interactions, to the atomic orbital electrons of the irradiated medium. The excess energy and momentum of the excited electrons is then transferred to the surrounding lattice atoms, resulting in a localised increase in the kinetic energy of the lattice.

The different mechanisms available for energy loss result in the creation of different types of defects. The atomic collisions involved for nuclear energy loss result in small atomic dislocations in the superconductor crystal lattice. Electronic energy loss, on the other hand, produces localised heating of the crystalline lattice, resulting in extensive regions of amorphous material, which acts as an inclusion of normal material. Different regimes of incident particle mass and energy will
therefore produce different types of pinning centres with different strengths and failures.

**1.5.3 Irradiation Techniques**

**1.5.3.1 Nucleon and Light-Ion Irradiation.**

Irradiations performed with light particles, such as protons and neutrons, of various energies, produce only small areas of lattice damage. In light ion irradiation, where the incident ions are of low Z, the dominant form of energy loss is via nuclear interactions.\(^{43}\) There is some excitation of the orbital atomic electrons and ionisation if the energy transferred is sufficient, however the primary interaction is through coulomb scattering off the atomic nuclei.\(^{44}\)

The size of the cascade of displacements produced is dependent upon the masses of the nuclei involved, the energy imparted to the primary recoil atom and the energy for displacement required, but is usually limited to a small region only several nanometres in diameter, comparable to \(\xi_c\).\(^{44}\) This region of damaged crystalline structure is surrounded by a strain field, in a nearly spherical volume,\(^{45}\) extending the size of the defect slightly.

The area of damage has a lowered \(\Psi\), or may even become completely normal, so the defect can act as a flux pinning centre through the core pinning interaction. Pinning may also occur through interaction
with the strain field of the defect. The roughly isotropic nature of the resulting damage means that there is no preferential direction for alignment between flux lines and the damage centre, so the flux pinning behaviour is isotropic. Light-ion irradiation thus produces small, isotropic point pinning centres.

The nuclear collisions involved in light ion irradiation have a very low interaction cross-section. Therefore, the incident ions have a large penetration depth, even if a supporting material, such as silver, sheaths the superconductor. This allows the damage centres to be produced approximately uniformly throughout the entire superconductor matrix.

Enhancements, however, are evident only at low temperatures and low magnetic fields, where the as-grown defects also act as pinning centres. Early experiments with light ion irradiation, using protons or neutrons of varying energies, showed moderate enhancements of $J_c$ but no improvements in the irreversibility field. Additionally, the pinning energy of the material after irradiation is not observed to be very different from that before the irradiation. These results are primarily due to the weak point pinning centres introduced. A higher concentration of point centres yields a larger pinning force, hence the improvements in $J_c$. The defect clusters, however, provide a pinning energy of only a similar size to that of the intrinsic point defects. As a result, an increase in $J_c$ is observed only at
low temperature, where the small pinning energies are relevant, and there is very little change in the irreversibility field.\textsuperscript{47} To obtain pinning centres that are effective at higher temperatures, the pinning energy of each pinning centre would have to be increased substantially.

Ideally, as the elementary pinning force is determined by the maximum gradient of the pinning potential, defects should be of the order of $\xi$ in diameter,\textsuperscript{47} which is true of the cascade defects. The strength of pinning, however, is also dependent on the length over which the vortex lines extend along the defect.\textsuperscript{48} The light-ion cascade defects act as point pinning centres only, less than several nanometres in size, so that the pinning strength is limited in this respect.

A long columnar defect (CD) track with a diameter of the order of $\xi$ should provide the strongest pinning according to this reasoning, and so is the ideal pinning centre. $J_c$ enhancement at higher temperatures and fields therefore requires irradiation with an ion mass and energy that can provide pinning centres closer in structure to the ideal pinning centre.

**1.5.3.2 Heavy-Ion Irradiation**

The atomic mass above which an ion is termed “heavy” is determined by the changes in dominance between the nuclear and electronic stopping powers. Electronic energy loss becomes progressively more important as the mass and energy of ion considered is increased.\textsuperscript{42}
Above a material dependent threshold, of the order of 2 keV/Å, the electronic energy loss component provides sufficient energy transfer to heat and melt the crystalline structure of the superconductor material through which the incident particle passes. This creates a cylinder of amorphous material along the path of the particle, within which the superconducting order parameter is reduced or eliminated.

Studies of various ion species and energies have shown that the transition from formation of small, localised defects to complete tracks of amorphous material is a smooth progression dependent on the electronic stopping power. As described above, at low masses and energies of the incident ions, damage is created primarily by nuclear interactions. As the energy or mass is increased, the electronic energy loss term is increased relative to the nuclear stopping power. At low average energy loss, the energy transfer is too small to produce localised melting of the lattice. Due to the stochastic nature of the collision processes, however, despite the low average stopping power, the instantaneous energy transferred may exceed the threshold in small isolated pockets, so that electronic induced damage can form small isolated spheres. As the electronic stopping power is further increased, the probability of the energy transferred being above threshold likewise increases, and so the damage progresses to elongated defects.
forming a discontinuous track (a *string-of-beads* effect) and eventually reaches a level where continuous tracks of amorphous material are formed.

The CD tracks of amorphous material generally have a radius of approximately 10 nm, and can be up to 20 µm in length.\(^4\) Since the strength of the pinning is dependent on the length along which the flux line is pinned, the strongest pinning occurs for flux lines oriented along the length of the column, and reduces as the angle between the two increases. Consequently, although the pinning strength is much greater than that for light ion irradiation, the pinning action is strongly oriented in the direction of incidence of the ion beam.

**Matching Field** \(B_\phi\)

The irradiation fluence expresses the average cross-sectional density of particles during the total irradiation exposure. Assuming that each particle creates one CD track, the particle fluence can be equated with the cross-sectional density of defect tracks. The density of tracks can then be related to the cross-sectional density of vortex lines \(n\), Eq. (1.5). A *matching field* \(B_\phi\) is defined as the value of applied field, at which the vortex density equals the track density,

\[
B_\phi = \Phi_0 \phi,
\]  
(1.31)
where $\phi$ is the irradiation fluence responsible for the columnar defects and $\Phi_0$ is the flux quantum. $B_\phi$ is thus proportional to the irradiation fluence, and so is commonly used to express the degree of irradiation.

From the definition of $B_\phi$, it is expected that at $B_\phi$ there is a pinning CD track available for every flux line and thus that all flux lines are pinned and all columnar pinning sites are occupied. Below $B_\phi$, since the density of tracks exceeds the total flux, there are sufficient numbers of columnar defects available for pinning and the FLL is in a Bose-glass phase, as discussed in Section 1.4.2. Vortices may hop between adjacent vacant CDs through half-loop and double-kink excitations, as illustrated in Fig. 1.5. Formation of a double-kink between two identical and parallel columns allows motion of the kinks in the direction transverse to the columns to proceed relatively unimpeded, restricted only by any residual point-defect pinning. A dispersion in the pinning energies and columnar spacing leads to variable-range hopping, which is a form of double-kink excitation where the vortex can spread out to a more energetically favourable defect that may be further than the nearest-neighbours.

Above $B_\phi$, the number of flux lines exceeds the number of pinning tracks and consequently there will be *interstitial* flux lines with only intrinsic pinning centres and the interactions with the pinned portion of the
Chapter 1: Theoretical Foundations

Fig. 1.5: Flux line motion by nucleation of half-loop and double-kink processes. The Lorentz force in the diagram is acting to push the lines towards the right. [Ref. 24].

FLL to limit their dissipative motion. These interstitial vortices will move under an applied current before the CD-trapped vortices are depinned.\textsuperscript{24}

The increased electronic stopping power of the heavy ions means that they produce much larger defect tracks than the light ions, which rely on nuclear interactions. However, this also means that the majority of the incident energy is lost quickly near the surface due to the increased interaction cross-section. The penetration of the heavy ions into the superconductor is therefore severely limited, ranging only several tens of microns on average.\textsuperscript{40} If a sheathing material surrounds the superconductor, the ions may not even reach the superconductor, or reach it with a much-reduced energy. To maximise the pinning efficiency, it would be preferable to have the defect tracks created uniformly throughout the entire
superconductor volume. This can be achieved by inducing fission of heavy elements within the superconductor matrix.

### 1.5.3.3 Fission-Fragment Damage

The third technique to be introduced was fission of a heavy element within the superconductor matrix. Fission of the $^{209}$Bi nuclei (herein referred to as $p$-Bi fission) in the Bi-Sr-Ca-Cu-O system can be induced by irradiation with 0.8 GeV protons. Doping a superconductor with $^{235}$U and then irradiating it with a thermal neutron flux produces similar results and was originally employed in some LTS materials. In the case of uranium fission, the resultant fission-fragments that produce defect pins are two high-energy heavy ions with a mean mass of 115 amu and mean energy of about 100 MeV. Conservation of atomic numbers means that the fission fragments must have $Z_1 + Z_2 = 92$, and, since the fission also involves the release of several energetic neutrons, $A_1 + A_2 \approx 233$. The exact combination of fragment masses and energies, as well as their orientation along the crystal axes, is a random process depending on the internal nuclear dynamics of the fission process.

The resulting fission-fragments create defects in a fashion similar to heavy-ion irradiation, as described in Section 1.5.3.2. Generally, the average stopping power of the fission fragments is below the threshold for track formation. As for heavy-ion irradiation (HII), though, the
instantaneous stopping power may exceed the threshold. Thus, we expect the string-of-beads effect for the formation of the fission tracks, with the extent of the break-up dependent on the exact fission-fragment mass and energy. The fission tracks therefore form quasi-columnar defects. The variation in fission fragment species and energy also results in a spread in the lengths of the columns produced.

The density of defect tracks produced is proportional to the concentration of fissionable atoms and the irradiation fluence. In the case of a $^{235}\text{U}$ doped superconductor irradiated to a thermal-neutron fluence $\Phi_n$, the probability of a single atom undergoing fission is $\sigma_{\text{abs}} \Phi_n$, where $\sigma_{\text{abs}} = 582 \times 10^{-28} \text{ m}^2$ is the thermal-neutron absorption cross-section of $^{235}\text{U}$. For $N$ atoms, the number of fission events $N_f$ is then

$$N_f = N \sigma_{\text{abs}} \Phi_n,$$  \hspace{1cm} (1.32)

provided that $\Phi_n < 1/\sigma_{\text{abs}}$. If $\Phi_n \gg 1/\sigma_{\text{abs}}$, then $N_f \approx N$. For each fission event, two energetic heavy ions are produced as fission fragments, each of which is expected to create a defect track. Thus, the number of tracks is approximately twice the number of fission events. The track density $\rho_{tr}$ is therefore approximately given by

$$\rho_{tr} \approx 2 \rho_f \sigma_{\text{abs}} \Phi_n$$  \hspace{1cm} (1.33)

where $\rho_f$ is the volume density of the fissionable atom. From the summation of elementary pinning forces discussed in Section 1.4.2.3, a
larger density of pinning centres can be expected to result in a stronger pinning force. Thus, Eq. (1.33) suggests that the pinning strength of the fission process will be proportional to both the irradiation fluence and the density of fissionable atoms.

The location of pinning tracks throughout the entire superconductor material depends on the distribution of the fissionable atoms within the superconductor crystalline matrix. In Bi2223, Transmission Electron Microscopy (TEM) studies of uranium doping effects have shown that there are no observable nonuniformities in the distribution of the uranium throughout the superconductor matrix, so that we can expect fission tracks to be uniformly distributed. Studies in Y123, on the other hand, have demonstrated that the uranium is concentrated in small inclusions, which are dispersed throughout the superconductor matrix. The uranium itself is not located within the Y123 matrix. However, the inclusions were observed to be on average approximately 300 nm in radius, so that loss of energy within the inclusion rather than the superconductor is minimal.

Fission techniques therefore provide strong, isotropic, quasi-columnar defects throughout the entire superconductor matrix, combining the strengths of both the light- and heavy-ion irradiation techniques. Combined with the easy accessibility of thermal neutrons from a nuclear reactor, such as the HIgh Flux Australian Reactor (HIFAR), the uranium-
fission method can be considered one of the best methods available for pinning enhancements in Ag-sheathed BSSCO tapes suitable for large-scale productions.

References

Chapter 1: Theoretical Foundations

As was explained in the previous chapter, the introduction of strong columnar pinning by a fission-induced process is more suitable for large-scale HTS production than the use of direct heavy-ion irradiation. The structure of the defects created by both techniques is similar and hence the interaction of individual defects with the vortices is expected to be similar. The fission technique, however, avoids the issues of the limited penetration of the heavy ions and their highly directional nature.

On the other hand, the random orientation of the fission-fragment defects complicates the dynamics of their interaction with the vortices. In comparison, HII techniques allow much greater control over the introduction of correlated disorder, as both the defect density and their orientation can be defined by the irradiation conditions. Thus, most of the work probing the effects of CDs on vortex dynamics has been performed for heavy-ion irradiation of various HTS materials. While the results cannot necessarily be directly applied to the case of fission-induced pinning, due to the random distribution of the CDs, similarities are expected because of
the almost identical nature of the individual columnar defect structure. This Section will therefore begin with a brief discussion of the relevant experimental issues in HII, and will then focus on previous findings in the field of fission-induced pinning.

2.1 **Heavy-Ion Irradiation**

While the fission process is more promising for commercial productions, the systematic degree of control over the creation of pinning centres allowed by HII means that this technique lends itself much more readily to studies on the vortex dynamics. As such, the vast majority of studies have concentrated on HII of many diverse HTS materials. The studies are too numerous to cover in detail here (Ref. 1 provides an excellent review).

In general, the majority of studies on HII have confirmed large improvements in $J_c$ and the IL, from both transport and magnetic measurements and for many different HTS systems. These enhancements are observed to increase with the level of irradiation, expressed as $B_\phi$ (Eq. 1.31), with a maximum observed in the $J_c$ enhancement at a $B_\phi$ of the order of 5 to 7 T. 

The $J_c$ and IL enhancements have been interpreted as indications of an increase in the activation energy for flux creep $U_\theta$ (defined in Section
1.4.3. Measurements of the magnetisation decay provide a more direct investigation of the activation energies, confirming large increases in $U_0$ as well as associated decreases in the normalised relaxation rate, both as defined by Eq. 1.30. The increase in $U_0$ is largely a consequence of the geometrical structure of the irradiation defects matching the linear topology of 3D flux lines. In the 3D state, there is thus a unidirectional quality to the pinning enhancements, where the increases in $J_c$ and $U_0$ are observed to be strongest when the field is aligned with the columnar defects.

Of particular interest has been the effectiveness of the columnar defects in pinning decoupled 2D pancake vortices, and their effects on the c-axis vortex pancake correlation. The crossover from 3D line-like behaviour to individual 2D pancake lattices in highly anisotropic HTS was discussed in Section 1.3.2. There is mounting evidence that parallel columnar defects created across the layers increase the inter-layer vortex coherence, promoting the 3D line-like behaviour. Under the Bose-glass framework, the strong localisation of vortices was predicted from suppression of the effects of thermal fluctuations on the pancake positions. This effect has been observed in HII studies on Bi2212 single crystals and thin films, Tl$_2$Ba$_2$CaCu$_2$O$_x$ (Tl2212) thin films and BSCCO tapes, using diverse methods such as Josephson plasma resonance, resistive dissipation and $J_c(\theta)$ scaling.
The introduction of splay (dispersion) in the columnar track directions was proposed as a method to improve pinning further. As described in Section 1.5.3.2, arrays of parallel CDs allow relaxation via double-kink formation, where a segment of flux line, localised on a columnar defect, jumps onto a neighbouring defect, as illustrated in Fig. 1.5.16. Formation of a double-kink between two identical and parallel columns allows motion of the kinks in the direction transverse to the columns to proceed relatively unimpeded, restricted only by any residual point-defect pinning. Nonparallel defects suppress the double-kink excitations by requiring an increase in the vortex length for motion along the kinks, making the process energetically unfavourable. Splay also forces vortex entanglement, which further restricts relaxation.

This effect of splay was confirmed in Y123 single crystals for a splay angle, defined as the angle of irradiation relative to the crystallographic \( c \)-axis, up to 10°. Further studies on the Y123 single crystals, however, demonstrated that sufficiently large splay angles would instead promote vortex motion and stimulate stronger vortex creep, in comparison to the parallel defect configuration. This behaviour of large splay was explained by the action of vortex cutting, whereby the \( c \)-axis coherence of the 3D vortex lines cannot be maintained. This forces the vortex line to split into independent segments, thus reducing the pinning
action to that of point defects. An optimum splay angle of 10° was established for Y123.

The large increases in $J_c$ and especially the irreversibility line seen for uniform splay in BSCCO and Hg-1212, both quasi-2D HTS with a much larger anisotropy than Y123, were difficult to reconcile with the above. Krusin-Elbaum et al. investigated the effects of randomly splayed fission-induced pinning in Hg cuprates with different degrees of anisotropy. The authors demonstrated that uniformly splayed defects provide much stronger improvement of vortex pinning in superconductors with a high anisotropy, and derived a re-scaling of the pinning distribution by the anisotropy to explain their observations.

By mapping the tracks and magnetic field into an isotropic system, the distribution of splay angles becomes focused along the $c$-axis, and the vortices gain an angular spread about the $c$-axis $\sim \varepsilon$. The overlap of the focused tracks and the angular spread of the vortices defines the matching field, also taking into account the pinning efficiency due to, for example, vortex-vortex interactions and the restricted trapping length. As such, this theory predicts that a large anisotropy leads to a focused distribution of splay angles, which effectively produces recoupling of the vortex segments.
2.2 FISSION-INDUCED PINNING

2.2.1 Uranium Fission

The technique of creating flux pinning centres through fission-fragment damage was first applied to conventional low-Tc superconductors in the 1960’s, in materials such as Nb3Al and V3Si. Fleischer et al. were the first to utilise the method in HTS, in 1989. Sintered Y123 compacts were doped with an oxide of natural uranium, UO2, to levels of c = 0, 0.08 and 0.2 wt% (0, 150 and 380 ppm U), then irradiated to thermal neutron fluences of $\Phi_n = 2 \times 10^{15}, 4 \times 10^{17}, 1.2 \times 10^{18}$ and $4 \times 10^{18}$ cm$^{-2}$. The irradiations were performed in the Brookhaven High Flux Beam Reactor, which has a ratio of neutron energies $E_n < 1$ MeV to $E_n > 1$ MeV of 1700:1. The authors examined the influence of the doping and irradiation on the magnetic hysteresis and the critical current density calculated from the loops at various temperatures (4 to 77K).

An increase in the magnetic $J_c$ was found for all uranium-doped irradiated samples, relative to both the non-doped samples and non-irradiated doped samples, even at liquid-nitrogen temperature. For example, at $c = 150$ ppm and $\Phi_n = 1.2 \times 10^{18}$ cm$^{-2}$, and in a 1T applied field, the enhancements were found to be 3.7 times at 4.5K, 20 times at 63K and 8.3 times at 77K. A maximum enhancement was observed to
occur for fluence levels somewhere between $1.2 \times 10^{18} \text{ cm}^{-2}$ and $4 \times 10^{18} \text{ cm}^{-2}$.

As reported in *J. Mater. Res.*, the previous work was extended to powdered Bi2212 and Bi2223, as well as epitaxial Y123. A 0.08 wt% doping of natural UO$_2$ was applied to the sintered Y123 and the BSCCO samples. A 60 µm natural uranium metal foil was placed over the top of the epitaxial Y123 film during irradiation to provide the fission fragment damage. The samples were irradiated in the Brookhaven medical reactor, (with a ratio of $E_n < 1 \text{ MeV}$ to $E_n > 1 \text{ MeV}$ of 33:1). The sintered Y123 sample was irradiated to $\Phi_n = 1.34 \times 10^{18} \text{ cm}^{-2}$, both the BSCCO samples were irradiated to $\Phi_n = 1.88 \times 10^{17} \text{ cm}^{-2}$, and the Y123 film was irradiated to $\Phi_n = 8.9 \times 10^{13} \text{ cm}^{-2}$. Magnetic hysteresis loops were measured in a Vibrating Sample Magnetometer (VSM) with a 3 T electromagnet, at a sweep rate of 5 mT s$^{-1}$. For the sintered Y123, both magnetic and transport measurements were made. A decrease in the transport $J_c$ was observed, despite observing increases in the magnetic $J_c$. Differences between the various samples in the magnetic $J_c$ improvements after irradiation were also reported.

The largest improvements were found for the Bi2223 sample, with improvements in $J_c$ by up to 70 times at 0.8 T and 50 K. The Bi2212 sample showed $J_c$ improvements of only up to 10 times at 0.8 T, and the
sintered Y123 showed even smaller improvements in $J_c$ of only 6 times at similar temperatures. Any improvements were negligible for the epitaxial Y123 film. These results demonstrate that the strength of pinning centres created by the fission process varies depending upon the type of superconductor. However, as the authors reported, they could find “no structural evidence to explain [the] differences”\textsuperscript{27} based on these results.

Magnetisation relaxation measurements were conducted on the 0.08 wt% doped sintered Y123 sample in a later report.\textsuperscript{28} The authors measured the time decay of the magnetisation, and observed a reduction in the field dependence of the flux creep rate $dM_{\text{irr}}/d\ln(t)$, where $M_{\text{irr}}$ is the irreversible magnetisation, at temperatures of 20 to 30K. To deduce $U_0$, the authors used the linearised analysis of Eq. (1.30), which assumes that the effective activation energy decreases linearly with increasing irreversible magnetisation. At 0.8 T, this analysis yielded a temperature dependent $U_0$ and demonstrated a slight increase in the activation energy after irradiation. This analysis, while adequately describing the flux creep behaviour of conventional LTS, is very limited in its applicability to HTS. The size of the flux creep phenomenon in HTS restricts this analysis to low temperatures.

Magnetisation relaxation experiments were also performed on powdered Bi2223 doped with 0.8wt% UO$_2$, both before and after thermal-neutron irradiation.\textsuperscript{29} Irradiations were performed in the Brookhaven
medical reactor to a fluence of $1.88 \times 10^{17}$ cm$^{-2}$. The powders consisted of flat platelet grains with a mass-weighted median (equal masses of material having diameters larger and smaller than the median) diameter of $\sim 22$ µm. The magnetic $J_c$ was calculated from hysteresis loops measured at a sweep rate of 5 mT s$^{-1}$. An increase in $J_c$ of up to 70 times at 50 K was observed.

The time decay of the magnetisation was measured at an applied field of 0.8 T for 630 s. The sample was initially swept through an entire magnetic field cycle before stopping at the measurement field, which was approached at a sweep rate of 42 mT s$^{-1}$. Several different approaches were applied to determine the effective activation energy $U_{eff}$ from the flux creep data.

From the linearised analysis of Eq. (1.30) an approximately temperature independent $U_0$ was obtained for a low temperature range. Irradiation was found to approximately double this $U_0$. As pointed out above, this analysis is very limited for HTS.

The authors also considered a nonlinear relation between $U_{eff}$ and the current density. They utilised a procedure designed by Maley et al., which begins from the rate equation for the diffusion of flux in the superconductor. The current density dependence of $U_{eff}$ is then constructed directly from the flux creep data, to within an additive parameter, assuming that $U_{eff}$ is only weakly temperature dependent in the low temperature regime. This method will be discussed in greater detail in Chapter 6. The
result revealed a strong improvement in $U_{\text{eff}}(J)$ at 0.8 T after irradiation, more than doubling $U_{\text{eff}}$ over a wide range of current densities.

Another approach taken by the authors involved determining $\partial U_{\text{eff}}/\partial J$ from the flux creep data, and then integrating to obtain $U_{\text{eff}}(J)$, assuming that $\partial U_{\text{eff}}/\partial J$ is only weakly temperature dependent in the low temperature regime. The additive integration constant was arbitrarily chosen to make $U_{\text{eff}}$ zero at the lowest measured temperature (assuming that $J \sim J_c$ at low $T$). The results were similar to those obtained by the Maley's method, above. An approximately logarithmic dependence of $U_{\text{eff}}$ on the current density was also determined.

A more extensive study on the uranium-fission method in Y123 was performed by Weinstein et al. The authors doped melt-textured Y123 with various percentages of natural uranium. Scanning Electron Microscopy (SEM) and Tunneling Electron Microscopy (TEM) studies on the microstructure of the uranium distribution demonstrated that the uranium occurs mainly in quasi-spherical deposits and not within the superconductor matrix. The inclusions were observed to have average radii of $\sim300$ nm, where the number of inclusions increased with increasing uranium doping concentration, not their size. TEM studies after irradiation showed the expected quasi-columnar tracks emanating from the uranium deposits.
In the non-irradiated state, $T_c$ and creep were unchanged for doping levels up to 1 wt-%. The trapped field was increased by the doping procedure, by up to 65% for a 0.8 wt% doping level. $J_c$ is increased significantly after neutron irradiation, by over an order of magnitude over a wide field and temperature range. $T_c$ was observed to decrease by up to 0.5K (at a fluence of $3 \times 10^{16}$ cm$^{-2}$ and 0.3 wt-% uranium doping), and the trapped field was enhanced further, with a maximum value at fluences around $6 \times 10^{16}$ cm$^{-2}$.

The studies on melt-textured Y123 doped with natural uranium were continued by Eisterer et al.\textsuperscript{32} In this study, samples of Y123 were doped with both natural and U$^{235}$ depleted uranium, and were irradiated in the Vienna TRIGA reactor in two positions: the Central Irradiation Facility, where the flux densities of fast and thermal neutrons are almost equal, and the Pneumatic Transfer System, where the fast neutron flux density is negligible. In this way, the authors were able to investigate the influence of the neutron energy. They found that cascade defects formed by fast neutrons lead to an additional enhancement in $J_c$, noticeable even when the majority of the enhancement can be attributed to the fission-fragment damage.

This work also contains an extensive theoretical discussion on fission-damage track formation and the interaction of the flux-line-lattice with the columnar tracks. Due to the random nature of the fission process,
and hence to the fission-damage track orientations, the pinning of a flux line along an entire track is a rare event. The authors discussed a simple model for the changes in pinning energy as a function of the relative angle between the flux line and the track, and the displacement length that results between the upper and lower segments of the bent flux line. They argued that the effective defect lengths, as seen by a unidirectional FLL, are distributed with a greater proportion being much shorter than the total possible damage-track length. The implication is that the average effective pinning length is only a few times larger than the size of the defects created by light ion irradiation, with only a small number of very strong pinning centres. However, the substantial pinning improvements observed in even high temperatures and fields are inconsistent with this picture. As discussed in Section 1.5.3.1, light ion irradiation results in enhancements that are evident only at low temperatures and low magnetic fields.

In 1998, Schulz et al. reported on work done with silver-sheathed Bi-2223 tapes made by the powder-in-tube (P-I-T) process (described in Chapter 3). The tapes were doped to 0.3 wt% UO$_4$. Both SEM and X-ray Diffraction (XRD) studies failed to find any evidence of separate uranium containing phases or particles, suggesting that, unlike in Y123, uranium is dispersed uniformly throughout the Bi-2223 matrix.

Doped and non-doped samples were irradiated in the Vienna TRIGA reactor to a thermal neutron fluence ($E_n < 0.55$ eV) of $4 \times 10^{20}$ m$^{-2}$, which
had a simultaneous fast neutron fluence of $5 \times 10^{20} \text{ m}^{-2}$ because of its positioning in the Central Irradiation Facility.

The resultant transport $J_c$ was characterised as a function of field and angle to the $c$-axis of symmetry $\theta$. A slight reduction in $J_c$ anisotropy was observed for the non-doped sample after irradiation (at 500 mT and 77 K), from 11 to 7, due to fast neutron cascade defects. However, the reduction in anisotropy was much more pronounced for the doped sample, being one order of magnitude at the same field and temperature. Similarly, the normalised $J_c$ showed a slight enhancement in the non-doped sample after irradiation as a result of the fast neutron defects, but not as large as the enhancement due to the fission-track creation. From the field dependence of the $J_c$, the authors estimated that the IL is also shifted to higher fields after doping and irradiation. This work illustrates clearly the improved capabilities for flux pinning of the splayed columnar fission-tracks over the spherical fast-neutron collision cascades.

Similar work was continued by Tönies et al., doping Ag/Bi2223 tapes with 0.15 and 0.025 wt% of $^{235}\text{U}$ (in the form of UO$_4$). The 0.025 wt% doped tapes, labeled type A, were irradiated in the Central Irradiation Facility of the Vienna TRIGA reactor facility, to a thermal neutron fluence of $4 \times 10^{20} \text{ m}^{-2}$, with an additional $3.2 \times 10^{20} \text{ m}^{-2}$ fast neutron ($E > 1 \text{ MeV}$) fluence. The 0.15 wt% doped tapes, type B, were irradiated to $\Phi_n = 4 \times 10^{19}$
m$^{-2}$ in the Outer Irradiation Position, which has a lower fast neutron component of 1.05 x 10$^{18}$ m$^{-2}$.

Transport $J_c$ measurements up to 6 T were measured both before and after irradiation as a function of field and $\theta$, and similar findings to those above were observed. A reduction in the low field $J_c$ (up to 0.4 T for $H||ab$ and 0.1 T for $H||c$) was observed in the type A but not the type B samples, which was attributed to damage to the inter-granular structure (see Section 3.1) from the fast-neutron fluence. The authors also claimed to find that the enhancements in absolute $J_c$ were greater for the 0.15 wt% doped samples, which have $c\Phi_n$ six tenths that of the 0.025 wt% doped samples. The IL was measured for the 0.15 wt% samples and was observed to be nearly doubled by the irradiation, for fields oriented both parallel and perpendicular to the c-axis.

### 2.2.2 Proton-Induced Fission

Both Bi and Hg have isotopes with a large capture cross-section for 0.8 GeV protons, which decay after proton capture predominantly by fission. The average mass and energy of the fission products is similar to that for $^{235}$U fission, though the former are slightly lighter due to the smaller initial masses of Bi and Hg. Thus, 0.8 GeV proton irradiation of BSCCO or Hg-cuprates can also be used to create randomly splayed, quasi-
columnar defect pinning centres that have a similar size and structure to those created by uranium fission.

The use of proton-induced Bi fission in BSCCO superconductors as an alternative method for creating splayed pinning was introduced in 1994.\textsuperscript{21} Significant increases in the magnetic hysteresis, suggesting current density improvements, as well as a considerable improvement in the IL, were observed in Ag/Bi2212 tapes. The size of the enhancements in $J$ and the IL were compared to those obtained by 1.08 GeV Au irradiation of Bi2223 tapes, and were found to be of the same order for a density of defects approximately three times larger than the effective defect density of the Bi fission, estimated from TEM studies. The authors claimed that this demonstrated the increased efficiency of the splayed columnar defects produced by fission events over the parallel CDs produced by heavy-ion irradiation.

Various subsequent studies have confirmed increases in the transport $J_c$ as well as improvements in the IL for both p-Bi\textsuperscript{63,64} as well as p-Hg\textsuperscript{22,23} fission. In moderate proton fluences, these enhancements were obtained without any losses in the self-field $J_{c0}$. Radiation damage was observed for large proton fluences, however.\textsuperscript{36} The $J_c$ for large fluences of proton irradiation, while demonstrating a weaker field dependence indicative of the enhanced flux pinning, was reduced in low fields below even the $J_c$ of a nonirradiated tape.
2.3 AREAS FOR INVESTIGATION

It has been conclusively shown that the uranium-fission method greatly enhances the field and angular dependence of $J_c$ in Y123, Bi2212 and Bi2223 samples, where the enhancement has been illustrated for both magnetic as well as transport $J_c$. Improvements in the IL have also been well documented. For probing the pinning potential energy changes more directly, though, Luborsky et al.$^{28}$ and Hart et al.$^{29}$ provide the only investigations in uranium-doped and irradiated superconductors, using magnetic relaxation methods.

The investigation by Luborsky et al.$^{28}$ was limited by the linear approximation used. The application of the linearised Kim-Anderson method is limited in HTS due to the magnitude of the flux creep. The current density dependence of the pinning energy is expected to be more complicated.$^{37}$

The nonlinear approaches taken by Hart et al.$^{29}$ are likely to reveal more information about $U_{\text{eff}}(J)$. Hart et al.$^{29}$ however, do not address the issue of the explicit temperature dependence of $U_{\text{eff}}$ instead assuming only weak temperature dependence at low temperatures. Nor do the authors examine the implications for the flux dynamics from the observed current dependence of $U_{\text{eff}}$. Theories of flux dynamics, such as collective flux creep$^{30}$ or vortex-glass theories,$^{40}$ predict the behaviour of $U_{\text{eff}}$ as a function
of current density, so that the observed current dependence can be related
to the dominant vortex dynamics.

The activation energy may also be probed from the magnetic
broadening of the resistive transition. The magnetic broadening is a
measure of the pinning strength of the material which in the limit of small
driving current density \( J \to 0 \) is dominated by a thermally activated flux
flow (TAFF) process (Section 1.4.3). The resistivity can then be written in
the form of an Arrhenius relation,

\[
\rho = \rho_0 \exp\left(-\frac{U}{k_B T}\right),
\]

(2.1)

where \( U \) is the activation energy for flux flow. The activation energy can
thus be determined from the resistive behaviour in the limit of a small
driving current.

As well as the above methods, the strength of the fission-induced
pinning can be probed by examining the magnetic field dependence of the
pinning force density \( F_p \), as given in Eq. (1.21). Before this work was
begun, none of these methods had been applied to uranium-doped HTS
tapes. Many features of the effects of the fission-fragment defect centres on
the pinning landscape may be revealed by an adequate study of \( F_p \) from
critical current measurements, as well as studies of the effective activation
energy from both resistive transition and magnetic relaxation
measurements. Therefore, the focus of this Thesis will be on an
experimental determination of the effects of fission-fragment damage on the activation energies of uranium-doped Ag/Bi2223 tapes.

References

Chapter 2: A Review of the Literature

BSCCO superconductors are an extremely important HTS material from a technological viewpoint, because a relatively high $J_c \sim 10^5 \text{Acm}^{-2}$ can be maintained in polycrystalline samples, such as silver-sheathed tapes, over very long distances, using liquid nitrogen as a coolant. The high $J_c$, however, rapidly deteriorates with an applied magnetic field, particularly at liquid-nitrogen temperatures. This is due both to a lack of intrinsic flux pinning in most field orientations and poor connectivity between the grains of superconductor material that form the tape.

### 3.1 Tape Structure

Polycrystalline tapes consist of a length of interconnected, $c$-axis oriented crystal grains of HTS material. The grains have a platelet-like structure, with a large aspect ratio; their lateral dimensions in the crystalline
Chapter 3: Sample Preparation

$ab$-plane are much larger than the $c$-axis thickness. A macroscopic transport current must traverse many such grains. However, the surface layers of the grains, referred to as grain boundaries, can impede the transport of supercurrent between the grains.

Grain boundaries may have different structural, chemical or physical properties to the interior of the grains, resulting in regions where the superconducting order parameter may be suppressed. Insulating or normal phases can occur at the grain boundaries, as well as a build up of secondary phases and impurities. As a result of the small coherence length of HTS, many of the grain boundaries will therefore prohibit inter-granular current transport, or form weak Josephson junctions that impede current transport between the grains. Thus, the superconducting connection between grains becomes of extreme importance. It is then necessary to distinguish between

- an *intra-granular* critical current density $J_{c}^{\text{intra}}$, which is the $J_{c}$ within an individual grain, determined by the flux pinning properties of the superconductor, and
- an *inter-granular* critical current density $J_{c}^{\text{inter}}$, which is the macroscopic critical current density transported across many grains. $J_{c}^{\text{inter}}$ is limited by the inter-granular links and thus will be equal or, more likely, smaller than $J_{c}^{\text{intra}}$. 

84
In low applied fields, $J_c^{\text{inter}}$ is limited by inter-granular links that are strongly dependent on applied field. For weakly Josephson coupled grains, application of quite low fields, even as low as the self-field, can have detrimental effects on the superconducting link. Hence, as an applied field is increased, the number of inter-granular links is rapidly diminished and the transport $J_c$ will decline precipitously with magnetic field. These are referred to as weak-links. The weak-link dominated region is evidenced by a power law behaviour of the $J_c$ field-dependence,

$$J_c \propto H^{-n}.$$ \hspace{1cm} (3.1)

A small fraction of grains form a strong inter-granular connection, however, allowing a macroscopic current to be conducted through a polycrystalline tape even in large applied magnetic fields. Once the majority of weak links have been eliminated and only strong inter-granular connections remain, the $J_c$ field-dependence is controlled by the flux pinning properties of the HTS material and is much weaker than in the weak-link region. This occurs because $J_c$ through strong links is even larger than the $J_c^{\text{intra}}$.

Poor alignment between the superconductor grains can also limit $J_c$. A misalignment between neighbouring grains can result in a small contact area, which acts as a bottleneck to the supercurrent transport. The issue of grain
connectivity has been well examined, and many processing procedures have been designed to maximise the strong connection between grains. One of the most technologically promising methods is the powder-in-tube method. The precursor powder, consisting of a stoichiometric mixture of the required cations, is packed into a silver tube. The tube is drawn into a long wire and then rolled to form a thin monocore tape. Tapes with multiple filaments can be made by packing several of the drawn wires into another silver tube before rolling. Sintering of the tape is performed in two stages, with an intermediate rolling stage. The grains begin to grow with the first sintering, forming small platelets. The intermediate rolling, which is done before they are fully formed, aligns these grains like a stack of cards. With the subsequent sintering, the growth of the superconductor phase, which is much faster in the ab-plane than along the c-axis, makes the platelets form well-aligned connections with each other.

The P-I-T method is only suitable for HTS with a large aspect ratio of crystalline grains and is particularly suited to the production of Bi2223 tapes. The grains produced by this method have well aligned crystalline c-axes, oriented perpendicular to the broad plane of the tape. Misalignment is typically of the order of 8°. The a- and b-axes of the grains are not well
Chapter 3: Sample Preparation

aligned, though this does not pose a problem for current transport in Bi2223.

Magnetic fields applied parallel to the broad plane of a tape are approximately aligned along the \( ab \)-plane of the superconductor. Grain misalignment, however, results in a small component of field lying along the \( c \)-axis even for perfect field alignment along the broad plane of the tape. Similarly, for fields oriented perpendicular to the broad plane of the tape, the field is generally along the crystallographic \( c \)-axis, but with a small component along the \( ab \)-plane. Therefore, when reference is made to a magnetic field being applied parallel to the \( ab \)-plane (\( c \)-axis), it should be clear that this could only be an approximation. Generally, these alignments refer to orientation with the broad face of the tape, taken as the \( ab \)-plane, and its perpendicular, the approximate crystallographic \( c \)-axis. Orientation of the field is much more critical when it is aligned along the \( ab \)-plane. This is because the projection of the field onto the crystalline \( c \)-axis has a much stronger effect than its projection onto the \( ab \)-plane. Herein, field orientations parallel and perpendicular to the crystallographic \( ab \)-plane will be denoted as \( H^{ab} \) and \( H^c \), respectively.
3.2 TAPE PREPARATION

The monocore Ag/Bi2223 tapes were fabricated at the ISEM using the P-I-T process. The precursor powders were produced with a nominal cation composition of Bi : Pb : Sr : Ca : Cu = 1.84 : 0.34 : 1.91 : 2.01 : 3.05. A series of the precursor powders were doped with an oxide of highly enriched uranium (≈98% $^{235}$U), UO$_2$.2H$_2$O, at weight percentages $c$ of 0, 0.15, 0.6 and 2.0 wt%.

The final core cross-sections were $\sim 1.4 \times 10^{-3}$ cm$^2$ for all but the 2 wt% doped tapes, which had a core cross-section of approximately $0.7 \times 10^{-3}$ cm$^2$ because a thicker silver tube was used in the tape processing. The tapes were rolled out to an overall tape width of about 4 mm. The external tape thickness was on average 0.2 mm, with a superconductor core thickness of approximately 35 µm. Individual samples with a 4 cm length were cut from the processed tapes prior to irradiation.

The zero-field critical current density $J_{c0}$ of a control tape for each doping level was measured prior to irradiation, using the method outlined in Chapter 4. The results are listed in Table 4.1, page 100. Uranium-oxide doping leads to substantial losses in $J_{c0}$, as much as 10% for even a moderate doping of 0.6 wt%. The uranium-oxide severely degrades the formation of the Bi2223 phase, resulting in the loss of $J_{c0}$.\footnote{10}
For commercial and industrial applications, it is important to identify the largest level of doping that can be added before $J_c$ is reduced below practical levels. This is required in order to minimise the irradiation-induced radioactivity of the silver sheath, while maximising the fission-induced pinning.

Silver has a moderate thermal-neutron capture cross-section and decays after neutron capture via $\beta^-$ and $\gamma$ emission. Thus, to minimise activation of the silver sheath, $\Phi_n$ must be reduced. However, the density of fission tracks, given in Section 1.5.3.3, is proportional to the product $c\Phi_n$. Thus, minimising $\Phi_n$ will minimise the activity of the silver sheath, but it will also reduce the density of fission tracks, and hence the density of flux pinning centres. To maintain equivalent pinning densities, a reduction in $\Phi_n$ must be compensated by an increase in $c$. The present method of adding uranium-oxide to the precursor powder therefore limits the effectiveness of the doping and irradiation procedure. Efforts are currently under way to reduce the initial $J_{c0}$ degradation by changing the methods and $^{235}\text{U}$ compounds that are added to the precursor powder.$^{10}$
3.3 IRRADIATION

The tapes were irradiated in a field of highly moderated thermal neutrons at the HIFAR reactor of the Australian Nuclear Science and Technology Organisation. The tapes were placed into dual titanium canisters under a natural atmosphere. The canisters were then loaded into Rig X7 of the reactor via a pneumatic transfer system. Rig X7 is located in the graphite moderator region, within which epithermal and fast neutron components of the neutron flux are minimal. The temperature is estimated at 180°C in the regions surrounding the rig.

The tapes were irradiated with a neutron flux during exposure of approximately $1 \times 10^{21} \text{ m}^{-2} \text{ s}^{-1}$, up to a total fluence of $\Phi_n \text{ m}^{-2}$. The titanium canisters are relatively transparent to thermal neutrons, having no major activation channels, resulting in minimal attenuation of the neutron flux. The absorption of thermal neutrons by the silver sheath must be taken into consideration, though. This reduces the effective neutron fluence reaching the superconductor matrix, and hence the density of fission events. However, the silver sheath is sufficiently thin in comparison to the mean penetration depth of neutrons that the attenuation can be disregarded.
The particular combinations of doping and irradiation that were successfully produced are listed in Table 4.1. Herein, the doping and irradiation combinations will be expressed as \((c, \Phi_n)\), where \(c\) is in wt\% and \(\Phi_n\) is in units of \(10^{19}\) m\(^{-2}\). For example, a 0.6 wt\% doped tape exposed to a fluence \(\Phi_n = 2.25 \times 10^{19}\) m\(^{-2}\) is expressed as \((0.6, 2.25)\). The \(T_c\)s in self-field of a select few tapes were determined from ac susceptibility measurements.\(^9\) In the virgin nondoped, nonirradiated tape, \(T_c = 108\) K. \(T_c\) was not altered substantially after 0.6 wt\% doping, and after a doping and irradiation treatment, \(T_c\) is only reduced by approximately 0.5 K.

Unfortunately, many tapes were physically damaged during some stage in the irradiation process. It is thought that this may have occurred when loading the canisters into the irradiation rig, due to the particular pneumatic transfer system used. The canisters are shot by compressed gas over several tens of metres through a transfer tube that contains at least one sharp bend.\(^{12}\) The tapes were carefully packed into the canisters, filling empty spaces with rolled aluminium foil to avoid excessive movement of the tapes. However, sharp jolts may still have cracked the fragile core. The physical damage was not discovered until after all of the irradiations had been completed, and in many cases, it was not obvious until measurements were begun. The tapes reported here do not appear to have sustained
physical damage, with the possible exception of the (2, 0.05) tape, as will be discussed in the next chapter, where transport measurements of $J_c$ are presented.

References

The critical current density is one of the basic parameters that define the suitability of a superconductor material to commercial and technological applications, giving the value of the maximum dissipation-less current. An experimental examination of the uranium-fission induced pinning should therefore begin with a characterisation of the changes in the field dependence of the critical current density, particularly at the technologically relevant liquid nitrogen temperatures $T = 77$ K. Measurements of $J_c$ also allow the changes to the average pinning force density $F_p$ experienced by the flux line lattice to be determined. These measurements will establish a firm basis to explore other experimental issues later.
4.1 **Experimental Methods**

The theoretical $J_c$ as defined by Eq. (1.21) predicts an abrupt increase in voltage from a theoretical zero limit at $J = J_c$. However, experimentally, the voltage is observed to rise smoothly with $J$. This results from the transition to dissipative behaviour being broadened as a result of thermal fluctuations, as well as a possible spread in pinning energies and the complexities of the vortex state structure. Thus, the use of a voltage criterion to define a practical $J_c$ is necessary. In practice, the critical current density is identified as the limiting current density that may be sustained before a measurable electric field is induced in the sample. Generally, in transport measurements, and as in this work, this criterion is selected as 1 $\mu$V/cm to define the transport $J_c$.

There are several methods available for measuring energy dissipation in HTS. These include techniques such as transport measurements of induced voltage, magnetic measurements of hysteresis loops and magnetic relaxation, and calorimetric measurements of the heat produced by the superconductor. Of the methods available, transport measurements are perhaps the most straightforward.

The measurements were performed using a standard four-probe layout. A dc transport current is applied through contacts at either end of the tape under study. The contacts for the voltage measurement are connected to the
tape between the current contacts, a distance $\Delta x$ cm apart. Using an electric field criterion of $\Delta E = 1 \mu V/cm$, the voltage defining $J_c$ is then $\Delta x \cdot \Delta E$.

To ensure that a homogeneous current flows between the voltage contacts, where possible the distance between the voltage and current contacts is kept large, generally twice the sample width. The quality of the contacts is also an important issue. The resistance of the contacts should be as small as possible, to minimise energy dissipation in the contacts themselves. Energy dissipation in the contacts can lead to heating at the contact point and then to a localised change in the superconductor temperature. As a general rule, dissipation in the contacts should be kept below 0.1 mW, as this level can be dissipated into the sample holder without significantly heating the sample. For Ag-sheathed Bi2223 tapes, careful soldering using an appropriate solder is sufficient.

Thermoelectric voltages $V_{th}$ may also be produced, through both contact heating and heating of the measurement wires. These thermal voltages are only small, but since the voltages of interest are also minimal, $V_{th}$ can mask the measurements of the superconductor voltages. Averaging the difference in voltage measurements obtained under opposite current polarity can eliminate the thermal voltages. Since $V_{th}$ is proportional to the power dissipation, it is independent of the sign of the applied transport current, whereas the
superconductor voltage is dependent upon the sign. Taking the difference in voltages between a positive and a negative applied current therefore yields a result that is twice the superconductor voltage,

\[ V_s = \frac{1}{2} \left[ V(I^+) - V(I^-) \right] \] (4.1)

where \( V_s \) is the superconductor voltage and \( V(I^\pm) \) is the measured voltage at an applied current \( I \) with \( \pm \) polarity.

Fluctuations in the contact or wire temperatures produce fluctuations in \( V_{th} \), which lead to a component of noise in the measurement. This noise cannot be entirely eliminated due to its non-periodic nature, thus causing a drift in the voltage measurement. The temperature fluctuations responsible for the noise can be caused, for example, if the sample is in direct contact with the heat exchange gas, from inhomogeneities in the gas temperature, or even by bubbles in a cryogenic liquid bath that can result from inadequate cryostat thermal insulation. It is therefore important to insulate the sample from the cryostat environment and have it in good thermal contact with a portion of the sample holder, whose temperature may be remotely controlled through the use of a small heater element. The sample holder used here has a copper block as the sample base, with a heater element and temperature sensor embedded within. The heater is controlled in a feedback loop with the temperature sensor.
As described in Section 3.2, the samples for irradiation were typically cut to 4 cm in length. Both contacts for the voltage measurements were typically placed a distance $\Delta x \sim 1$ cm, in the centre of the broad plane of the tape. Voltage measurements were performed with a Keithley 1902 nanovolt pre-amplifier coupled to a Keithley 2001 Digital Multimeter (DMM), which can achieve sensitivity down to 5 nV. The functions of the Keithley DMM allow a voltage measurement to be defined as a zero point, so that subsequent measurements display the relative voltage difference. With the voltage zeroed under an applied driving current, the voltage displayed upon reversal of the polarity of the driving current is the relative difference, which, from Eq. (4.1) above, is twice the superconducting voltage. Therefore, to determine $J_c$ with a 1 $\mu$V/cm voltage criterion, the magnitude of the applied current is adjusted until the measured voltage difference is $2 \Delta x \Delta E = 2 \mu$V.

Any drift in the voltage due to thermal effects will affect the voltage zero point, introducing an unavoidable systematic error. A useful double-check to ascertain the severity of the voltage drift is to return the current polarity to the initial setting, where it is expected that the relative voltage display should return to zero in the absence of drift. A maximum drift in the zero point of $\pm 30$ nV was considered acceptable.
The transport measurements of the field and angle dependence of \( J_c \), \( J_c(H) \) and \( J_c(\theta) \) respectively (where \( \theta \) is the angle between the field and the axis perpendicular to the broad plane of the tape), were conducted on a 1.2 T electromagnet. High field measurements of \( J_c(H) \) in fields parallel to the tape \( ab \)-plane were performed in a 12 T Oxford Instruments superconducting magnet. Due to the limited availability of this magnet system, only few tapes were measured in this field range. All measurements were performed with the samples immersed in liquid nitrogen, \( T = 77 \) K.

The measurements performed determine the critical current \( I_c \). To convert this to a current density, it is assumed that the entire superconductor cross-section \( A \) is involved in the current transport and \( J_c \) is then simply given as \( I_c/A \). However, the effects of grain connectivity and misalignment result in a distorted current path (see Section 3.1), so that the total superconductor cross-section does not accurately represent the cross-sectional area of current transport. Despite this, for a polycrystalline system \( I_c/A \) is physically meaningful, and is particularly useful for comparison purposes between tapes of different sizes.

The measured \( I_c \) must be corrected for parallel silver conduction. Within the Ag sheathed tapes, there is parallel conduction of an applied current through both the superconductor and the Ag sheath. At very low
current densities and applied fields, where there is very little dissipative flux motion induced, the current flows almost entirely through the superconductor. At $I_c$, a small proportion of the applied current is conducted through the Ag sheath. The proportion of Ag current $I_{Ag}$ can be found by measuring in very high fields $H \gg H_{irr}$ the current that induces a voltage equal to the voltage criterion $\Delta E$. The resistivity of HTS is typically greater than that of Ag at these high fields, thus it can be assumed that the current is conducted almost entirely through the Ag sheath. $I_{Ag}$ is then subtracted from the measured $I_c$. In the following, $I_{Ag}$ was always measured in applied fields $\mu_0 H = 1 \text{T}$, except where $H_{irr}$ approached 1 T, in which case $I_{Ag}$ was measured for the highest field available. The $J_c$ reported in the following Sections have been adjusted for parallel conduction in this manner.

### 4.2 Results

#### 4.2.1 $J_c$ Field Dependence

The field dependence of the critical current was measured for the various combinations of $(c, \Phi_n)$ listed in Table 4.1. Figure 4.1 shows the field dependence of $J_c$ in field orientations (a) parallel and (b) perpendicular to the
crystallographic $c$-axis, up to the misorientation angle of the crystal grains in the tape (Section 3.2).

**TABLE 4.1**

**ZERO-FIELD $J_c$ AND IRREVERSIBILITY FIELD FOR ALL MEASURED TAPES**

<table>
<thead>
<tr>
<th>Doping Percentage $c$ (wt%)</th>
<th>Thermal Neutron Fluence $\Phi_n$ ($10^{19}$ m$^{-2}$)</th>
<th>Fission Track Density $c\Phi_n$ ($10^{19}$ wt% m$^{-2}$)</th>
<th>Zero-Field $J_{c0}$ ($10^3$ Acm$^{-2}$)</th>
<th>Irreversibility Field $\mu_0 H_{err}$ (mT)</th>
</tr>
</thead>
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<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>24.4</td>
<td>700</td>
</tr>
<tr>
<td>0.15</td>
<td>5</td>
<td>0.75</td>
<td>23.6</td>
<td>665</td>
</tr>
<tr>
<td>0.15</td>
<td>6</td>
<td>0.9</td>
<td>19.6</td>
<td>1120</td>
</tr>
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<td>0.15</td>
<td>1.25</td>
<td>0.75</td>
<td>18.6</td>
<td>10645</td>
</tr>
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<td>1.35</td>
<td>18.1</td>
<td>1200</td>
</tr>
<tr>
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<td>3</td>
<td>1.8</td>
<td>16.6</td>
<td>12405</td>
</tr>
<tr>
<td>0.6</td>
<td>4</td>
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<td>14.5</td>
<td>12715</td>
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<td>0.1</td>
<td>13.0</td>
<td>495</td>
</tr>
<tr>
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<td>0.4</td>
<td>13.0</td>
<td>770</td>
</tr>
<tr>
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<td>10.0</td>
<td>1620</td>
</tr>
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<td>(0.6, 2.25)</td>
<td>(0.6, 3)</td>
</tr>
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<td>(0.15, 6)</td>
<td>(2, 0.05)</td>
<td>(2, 0.2)</td>
</tr>
<tr>
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<td>(2, 1)</td>
<td>(2, 1)</td>
<td>(2, 1)</td>
<td>(2, 1)</td>
</tr>
</tbody>
</table>

**Fig. 4.1:** (a) Critical current density as a function of applied magnetic field aligned parallel to the $c$-axis, for various combinations of uranium doping and neutron irradiation.
4.2.1.1 Zero-Field $J_c$

As discussed in Section 3.2, the doping process reduces the zero-field $J_c$ of the tapes before irradiation. The reduction is most significant for the 2 wt% doping level, where $J_{c0}$ is almost half the virgin value. The values of $J_{c0}$ for both non-irradiated and irradiated tapes are given in Table 4.1. It can be seen that the irradiation procedure also results in a drop in $J_{c0}$. The quasi-columnar defects destroy a small fraction of the superconducting volume, as mentioned in Section 1.5.1, and thus a reduction in $J_{c0}$ is a likely result. As seen in Fig. 4.2, which plots $J_{c0}$, normalised to the pre-irradiation $J_{c0}$, as a function of $c\Phi_n$, 

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig4.1.png}
\caption{(b) Critical current density as a function of applied magnetic field aligned parallel to the $ab$-plane, for various combinations of uranium doping and neutron irradiation.}
\end{figure}
the zero-field critical current density falls approximately linearly with $c\Phi_n$ for a constant doping level. Thus, the loss of $J_{c0}$ after irradiation is a cumulative effect of the fission track density. Measurements of $J_{c0}$ were also made on tapes with a combination (0.6, 10) and (0.6, 20) (Fig. 4.2). The very low $J_{c0}$ for these tapes confirmed the increasing damage with $c\Phi_n$, although the linearity breaks down, possibly due to defect track overlapping at these high track density levels.

Fig. 4.2: Zero-field $J_c$ normalised to pre-irradiation levels for various $^{235}$U doping percentages $c$. An approximately linear dependence on fission track density $\sim c\Phi_n$ is shown in the insert.
The $J_{c0}$ reduction is also related to the magnitude of irradiation. At equivalent $c\Phi_n$ the reduction in $J_{c0}/J_{c0\text{ (nonirradiated)}}$ is greater for a lower doping level, because of the greater $\Phi_n$. Irradiation by neutrons may damage the inter-granular link structure, which also leads to a reduction in $J_{c0}$. The fact that these effects are observed as a result of the neutron fluence as well as the fission damage suggests that the neutron irradiation itself generates defect points in addition to the fission induced defects, see Section 4.2.4. A similar effect has been observed in fast-neutron irradiated Ag/Bi2223 tapes, where the low-field $J_c$ was degraded by the irradiation process, but ultimately exceeded the non-irradiated $J_c$ in an increasing field. This behaviour was attributed to damage of the weak-link inter-granular connections by the fast-neutron fluence.4

**4.2.1.2 Weak-Link Dominated $J_c$**

The field dependence of $J_c$ demonstrates a weak-link dominated regime, as evidenced by a power law behaviour of $J_c(H)$, Eq. (3.1), that is observed in all of the tapes in low fields $\mu_0 H^F < 80$ mT and $\mu_0 H^{ab} < 300$ mT. In this field region, $J_c$ drops off rapidly with a small increase of the applied field, due to the quenching of superconductivity at the weakly-linked grain boundaries as explained in Section 3.1. The $J_c$ of all the doped tapes remains lower than that
of the virgin tape within the weak-link region. This is particularly pronounced in the 2 wt% doped tapes and the (0.6, 4) tape, which show a significantly smaller $J_c$ than all the rest of the tapes.

In applied fields parallel to the $c$-axis, $n = -0.46$ for all the tapes with $c\Phi_n = 0$, where $n$ is the power law exponent of Eq. (3.1). The power law exponent is slightly reduced to a common value of $n = -0.38$ for all the fission-damaged tapes. A similar universality is also observed in fields parallel to the $ab$-plane, where the exponents are only slightly larger than those in the $c$-direction, $n = -0.57$ for all $c\Phi_n = 0$ and $n = -0.47$ for all $c\Phi_n \neq 0$ tapes.

4.2.1.3 High Field $J_c$

In applied fields above this weak-link dominated region, the magnetic field dependence of $J_c$ is significantly lessened after irradiation. In comparison, the $J_c$ of both the virgin and the doped, non-irradiated tape continue their rapid decline and show little flux pinning strength. Within this field region, current transport is limited principally by the flux pinning within the grains (Section 3.1). The lowered field dependence after doping and irradiation is thus a signature of the increased flux pinning that results from the fission damage tracks.
Improvements in the field dependence are even observed for the 2 wt% doped tapes, which have the worst $J_{c0}$, except at the lowest irradiation level. The high-field $J_c$ of the 2 wt% doped tapes is improved with increasing irradiation. These tapes, however, must be irradiated to a sufficiently high fluence before $J_c$ returns approximately to pre-doping levels. From Fig. 4.1, this can be seen to occur for a fluence $\Phi_n \approx 0.2 \times 10^{19} \text{ m}^{-2}$ in fields $\mu_0 H^c > 300 \text{ mT}$ and $\mu_0 H^{ab} > 1 \text{ T}$. Above this fluence level, $J_c$ of the 2 wt% doped tapes exceeds the $J_c$ of the virgin tapes in fields above the weak-link dominated region. The $J_c$ then begins to approach a magnitude comparable to the other $(c, \Phi_n)$ combinations, but still with a much lower $\Phi_n$. Thus for a suitable increase in $^{235}\text{U}$, the irradiation level can be substantially reduced while maintaining similar improvements in flux pinning, but at the expense of the low-field $J_c$.

In applied fields parallel to the $c$-axis, the best tape performance with regards to the magnitude of $J_c$ depends on the range of magnetic field examined. In low applied fields, $\mu_0 H^c < 450 \text{ mT}$, the highest $J_c$ was recorded for the $(0.15, 5)$ tape. This is largely because the 0.15 wt% doping does not lower the zero-field $J_c$ significantly, unlike the larger doping levels. At around 300 mT, $J_c$ of the $(0.15, 5)$ tape begins to decrease faster with applied field than the $J_c$ of the $(0.6, 3)$ tape, and the two intersect at $\mu_0 H^c \sim 450 \text{ mT}$. 
Above 450 mT, the (0.6, 3) tape exhibits the strongest $J_c$. However, the (0.6, 4) tape has the weakest field dependence of all measured tapes in applied fields above the weak-link regime, so that at $\mu_0 H_c \sim 950$ mT the $J_c$ of the (0.6, 4) tape exceeds that of the (0.6, 3) tape. Thus, while the strongest pinning (characterised by the weakest field dependence) is observed for $c = 0.6$ wt% and $\Phi_n = 4 \times 10^{19}$ m$^{-2}$, the largest $J_c$ can be achieved;

1. in moderate fields; with a low irradiation fluence and a moderate doping level, resulting in lower induced radioactivity, or

2. in low fields; with a smaller doping levels and higher irradiation fluence, resulting in less zero-field critical current density degradation.

**4.2.1.4 Irreversibility Field $H_{irr}$**

Irradiation increases dramatically the resistively determined irreversibility field $H_{irr}$, defined in Section 1.4.2.6 as the field at which the transport $J_c$ is reduced to zero. Experimentally, $H_{irr}$ is defined at a field where $J_c$ becomes immeasurably low, so that $H_{irr}$ can be (arbitrarily) defined as the field at which $J_c / J_{c0} \sim 10^{-3}$. Thus, to better compare the irreversible behaviour of $J_c$, we normalised the critical current densities to their zero-field values, as illustrated in Fig. 4.3. The results for $H_{irr}$ are included in Table 4.1.
Fig. 4.3: Magnetic field dependence of $J_c$ normalised to the zero-field $J_{c0}$, for fields (a) parallel and (b) perpendicular to the $c$-axis.
The doping process appears to lower $H_{irr}$, as does moderate irradiation of a nondoped tape. As was explained in Section 3.2, uranium-oxide doping degrades the formation of the Bi2223 phase during processing. This is the cause of the decrease in $J_{c0}$. Neutron irradiation, on the other hand, destroys a small fraction of the inter-granular links (see Section 4.2.1.1). This has a similar effect on $J_{c0}$, which is more obvious in the nondoped tapes because of a lack of introduced pinning. Since an arbitrarily small level of $J_c$ defines the irreversibility field, the destruction of the inter-granular links can also result in the reduction of $H_{irr}$. From this, it is clear that oxide doping has a detrimental effect on not only the zero-field critical current but also the irreversibility field, adding to the importance for finding a suitable uranium-doping compound that will lower $H_{irr}$ and $J_{c0}$ less than the uranium oxide.

Fission track damage clearly increases $H_{irr}$, as is expected if the damage tracks provide strong pinning. Surprisingly, though, the irreversibility field appears to scale exponentially rather than linearly with $c\Phi_n$ for fields parallel to the $c$-axis, as illustrated in Fig. 4.4. While the exponential dependence is most obvious for the 0.6 wt% doped tapes, the line of $H_{irr}$ as a function of $c\Phi_n$ is common to both the 0.6 wt% and 2 wt% doped tapes studied.
Fig. 4.4: $H_{irr}$ as a function of $c\Phi_n$ for fields parallel to the $c$-axis, in (a) linear, (b) log-linear scales. The lines are a guide to the eye, representing linear or exponential behaviour of $H_{irr}$, respectively.

Chapter 4: Critical Current Density
An exception is the irreversibility field of the 2 wt% doped tape with negligible irradiation, (2, 0.05) (Fig. 4.4). The irreversibility field for this tape is extremely low, especially in comparison to the virgin tape, and lies well away from the common line. However, we cannot discount the possibility of mechanical damage (see Section 3.3) to the (2, 0.05) tape, which resulted in these modest values of \( J_c \) and \( H_{irr} \). As will be discussed below, the extreme reduction in the \( J_c \) field behaviour of this tape suggests that the tape has suffered some mechanical damage, resulting in the loss of supercurrent transport in a section of the tape.

In contrast, the (0.15, 5) tape has a \( H_{irr} \) that lies above the universal line (Fig. 4.4). The deviation of approximately 10% is greater than would be expected from any scattering of the data due to experimental uncertainties. This increase could be accounted for by an additional \( \Phi_n \)-dependent component to the increase in \( H_{irr} \). This will be discussed further in Section 4.2.4.

It is not likely that the observed exponential dependence of \( H_{irr} \) on \( c\Phi_n \) can be explained by the additional \( \Phi_n \) dependence. The exponential behaviour is common to both the 0.6 wt% and 2 wt% doped tapes, with points for both doping percentages lying on a common line (with the exception of the damaged (2, 0.05) tape). This is despite the 2 wt% doped tapes having a much
lower thermal-neutron fluence compared to the 0.6 wt% doped tapes. Hence, a \( \Phi_n \) component to the increase in \( H_{irr} \) does not seem to account for the observed exponential dependence, for fluences up to \( \Phi_n = 4 \times 10^{19} \text{ m}^{-2} \).

**4.2.1.5 Improvements of \( J_c \) normalised at high fields**

In normalising \( J_c \) to \( J_{c0} \), the effects of the doping and irradiation on the weak-link behaviour still confuse the analysis of the high-field behaviour. To isolate the flux pinning improvements, observable only in high fields, \( J_c \) can instead be normalised to the \( J_c \) at an arbitrary field that lies just above the weak-link region. This allows the \( J_c \) field dependence to be more clearly compared between tapes. \( J_c \) was normalised to the values \( \mu_0 H_c = 100 \text{ mT} \) and \( \mu_0 H_{ab} = 500 \text{ mT} \), at which the tapes are expected to lie outside the weak-link dominated region (see Section 4.2.1.2). The results are plotted in Figure 4.5. The excellent flux pinning demonstrated by the (0.6, 4) tape in both field directions, despite the poor low-field \( J_c \), is now more apparent.

Of special note in Fig. 4.5 (a) is that the (0.6, 3) and (2, 1) tapes, which have comparable fission track densities (\( c\Phi_n \sim 1.8 \) and 2, respectively), have a very similar field dependence in fields parallel to the \( c \)-axis. The rest are also approximately arranged in order of \( c\Phi_n \), confirming that the flux pinning force is directly related to the density of columnar defects.
Fig. 4.5: $J_c$ normalised to the $J_c$ at (a) $\mu_0H = 100$ mT and (b) $\mu_0H = 500$ mT for fields applied parallel and perpendicular to the $c$-axis, respectively.
The (0.15, 5) and (2, 0.05) tapes are again two notable exceptions. The worsened $J_c$ field dependence of the (2, 0.05) tape may be an indication of mechanical damage. The $J_c$ of this tape is extremely field dependent. Assuming that the density of fission tracks is negligible due to the very low $\Phi_n$, it could be surmised that this is a result of the doping affecting strong inter-granular links. However, mechanical damage cannot be ruled out.

The (0.15, 5) tape, on the other hand, shows a greater pinning improvement than expected from the $c\Phi_n$ dependence of the other tapes. It shows a comparable field dependence to the (0.6, 2.25) tape, despite having different track densities, $c\Phi_n = 0.75$ compared to 1.35. As for $H_{irr}$ (Section 4.2.1.4), this behaviour suggests that there is a component of flux pinning resulting directly from the neutron irradiation, which only becomes evident at high fluence levels. The $c\Phi_n$ dependence in fields oriented along the $ab$-plane is similar to that in fields parallel to the $c$-axis.

**4.2.1.6 $J_c$ Anisotropy**

The differences in the magnitude and field dependence of $J_c$ and $H_{irr}$ observed between the two different field orientations are explained by the layered structure of BSCCO. As explained in Section 1.4.2.2, the layered structure results in strong confinement of the vortices between the CuO$_2$ layers.
for a magnetic field applied parallel to the $ab$-plane. Thus, there is a strong intrinsic inter-layer pinning for fields oriented in this direction, which is lacking for fields applied perpendicular to the layers. As noted in Section 3.1, the crystal grains in the tapes are misaligned (typically about $8^\circ$). Thus, as a result of the inter-layer pinning being much stronger than the intra-layer pinning, $J_c$ for $H$ parallel to the tape plane is actually defined by the projection of $H$ on the $c$-axis of the crystals, and therefore by the misalignment angle.

This difference between the intrinsic and intra-layer pinning leads to an anisotropy in the critical current that is evident as both a much weaker field dependence of $J_c$ and a much larger irreversibility field for fields $H^{ab}$ compared to that for $H^c$. In the $J_c$ measurements above, this anisotropy is clear. For magnetic fields applied parallel to the crystallographic $c$-axis, $J_c$ drops under the limit of measurement in fields below 2 T, even in the tapes with the strongest pinning. This is in contrast with $J_c$ in fields applied parallel to the tape plane, where $J_c$ does not drop below the limit of measurement until well above the 12 T limit of measurements in the tapes with strongest pinning.

Figure 4.6 shows the angular dependence of $J_c$ in a 500 mT applied field and at temperature $T = 77K$, where $\theta$ is the angle of the field orientation relative to the axis perpendicular to the tape plane. At this value of the field, transport is entirely through strongly-linked grains. The observed effect of the
fission-track damage is to reduce the anisotropy in $J_c$. The anisotropy is reduced by up to 46 times for a 0.6-wt% doping and $\Phi_n = 3 \times 10^{19} \text{ m}^{-2}$ at an applied field of 500 mT.

![Diagram](image)

**Fig. 4.6:** Angular dependence of $J_c$, normalised to the maximum current. $\theta$ is the angle of field orientation relative to the perpendicular of the broad face of the tape.

The observed reduction in anisotropy as a result of the induced columnar defects cannot necessarily be associated with a reduction in the anisotropy of the superconductor parameters or a change in the vortex structure from 2D to 3D. The likely reason for the change is the larger effect of the fission-track pinning at 500 mT on fields that are aligned with the $c$-axis.
as compared with fields aligned along the \( ab \)-plane. This is because of the much stronger intrinsic pinning along the \( ab \)-plane of the superconductor. The pinning potential trapping the flux between the planes is quite large, especially in low applied fields, and may mask any additional pinning that is introduced. There is no such strong intrinsic pinning along the \( c \)-axis, however, so that any additional pinning has an immediate effect on \( J_c \).

### 4.2.2 Implications for Applications

Overall, the highest critical current density observed in the examined tapes, over a wide applied field range both parallel and perpendicular to the \( c \)-axis, was for the (0.6, 3) tape. In this case, the measured \( J_c \) was 25 times larger than that of the non-irradiated tape at 77 K, in an applied field \( \mu_0 H^{ab} = 5 \) T, and 15 times larger at \( \mu_0 H^c = 500 \) mT. Thus, in terms of absolute \( J_c \) performance, \( c = 0.6 \)-wt\%, \( \Phi_n = 3 \times 10^{19} \) m\(^{-2} \) appears to be an optimum combination before degradation occurs.

The level that is considered as optimum for an industrial application, however, depends entirely on any restrictions or requirements placed upon the tape by the particular application. If total available critical current over a wide range of applied fields is important, then a moderate uranium doping, followed by a moderate thermal neutron irradiation (for example, 0.6-wt\% and
3 x 10^{19} \text{ m}^{-2} \) provides significant improvements in \( J_c - H \) performance. The level of silver radioactivity produced is quite high, though, which may cause problems for applications that are used in public areas or workspaces.

Some applications, on the other hand, place an importance on the minimisation of the silver radioactivity and require only improvements in the irreversibility field, and thus the flux pinning. Then a higher percentage of \(^{235}\text{U}\) and a low (but still sufficient) \( \Phi_n \) are best (such as 2 wt%, 1 x 10^{19} \text{ m}^{-2} \) if moderate \( J_c \) degradation is acceptable. If changes to the processing procedure can reduce the degradation of \( J_{c0} \) caused by the addition of \(^{235}\text{U}\) compounds, then this combination would be optimal for nearly all applications when the silver radioactivity is taken into consideration. This is also the optimal case if a simple reduction in the anisotropic behaviour of the tapes is required in low applied fields.

### 4.2.3 Pinning Force Density

\( J_c \) is by definition (Section 1.4.2.4) the current density at which the Lorentz force density \( F_L \) on the FLL equals an average volume pinning force density \( F_p \). The volume pinning force density can thus be approximated from experimental measurements of the critical current density as given in Eq.
(1.21), \( F_p = J_c H \). Using the measured \( J_c \), the resulting pinning force densities are shown in Fig. 4.7 for fields \( H^c \) and in Fig. 4.8 for fields \( H^{ab} \).

Fig. 4.7 (a) presents the pinning force densities for the tapes without fission-fragment damage, in applied fields oriented parallel to the crystallographic \( c \)-axis. Thermal-neutron irradiation of a non-doped tape reduces the maximum pinning force density \( F_{p\text{max}} \) to approximately 82% of \( F_{p\text{max}} \) of the virgin tape, but without any noticeable shift in the field \( H_{\text{max}} \) at which the maximum in \( F_p \) occurs. Doping, on the other hand, leads to a larger reduction in \( F_{p\text{max}} \) (down to \( \sim 60\% \) of the virgin \( F_{p\text{max}} \) for a 0.6 wt\% doping level) as well as a shift to slightly lower \( H_{\text{max}} \).

Pinning force densities for all of the doped and irradiated tapes are shown in Fig. 4.7 (b) along with the virgin tape for comparison. The initial rise of \( F_p \) with applied field is the same for almost all of the tapes, with the exception of the (0.6, 4) tape and all 2 wt\% doped tapes. The similar behaviour is indicative of common mechanisms defining \( J_c \) in low applied fields \( H < H_{\text{max}} \), despite the presence or absence of columnar defects. The exceptions to the common behaviour are from those tapes that show a significantly reduced \( J_c \) in the weak-link dominated regime (Section 4.2.1.2). For the rest, however, the common behaviour observed extends beyond the weak-link region.
Fig. 4.7: Pinning force density $F_p$ as a function of applied fields parallel to the $c$-axis for (a) tapes with $c\Phi_n = 0$ and (b) all tapes.
Both $F_{p_{\text{max}}}$ and $H_{\text{max}}$ are increased after introduction of fission-fragment defects, shown in Fig. 4.9 as a function of the fission-track density $c\Phi_n$. It should be noted that the broad maxima of $F_p(H)$ makes it difficult to determine the exact values of $H_{\text{max}}$ and $F_{p_{\text{max}}}$, which adds a scatter to the data. For several tapes, there are an insufficient number of points in the region of the peak, making it further difficult to determine the maximum accurately. The error bars shown in Fig. 4.9 represent an estimated deviation in the possible location of $(H_{\text{max}}, F_{p_{\text{max}}})$, and are larger for those tapes that have an ill-defined peak due to the lack of data points.
Fig. 4.9: Field of maximum in the pinning force density, $H_{\text{max}}$, as a function of $c\Phi_n$ for fields applied parallel to the $c$-axis in (a) linear and (b) log-linear scales. (b) demonstrates the reasonable exponential dependence. The lines are a guide to the eye.
Fig. 4.9: (c) Maximum in $F_p$ as a function of $c\Phi_n$ for fields applied parallel to the $c$-axis.

Despite the data scattering, Fig. 4.9 (b) shows an approximately exponential increase of $H_{max}$ with $c\Phi_n$, similar to that observed for the $c\Phi_n$-dependence of $H_{irr}$, Section 4.2.1.4. However, Fig. 4.9 (a), which shows the same data on a linear scale, demonstrates that a linear relation might also be fit to the data. For the available data, both the exponential and linear relation describe the data equally well. The (0.15, 5) tape again appears to have a much larger increase than expected from the $c\Phi_n$ dependence, up to 1.4 times greater, as observed for $H_{irr}$. This additional increase may be related to the additional $\Phi_n$-dependence of $H_{irr}$ described above, which was accounted for by
an increase in pinning due to the effects of neutron irradiation damage. Plotting $H_{\text{max}}$ against the irreversibility field determined for each tape, Fig. 4.10, an approximate linear correlation between the two can indeed be observed, $H_{\text{max}} \sim 0.4H_{\text{irr}}$. Both $H_{\text{irr}}$ and $H_{\text{max}}$ reflect the strength of the flux pinning, hence the correlation between them. A linear or power law fit to Fig. 4.10 does not describe well the relation between $H_{\text{max}}$ and $H_{\text{irr}}$. Thus, it is likely that the exponential dependence of $H_{\text{max}}$ on $c\Phi_n$, Fig. 4.9(a), is correct.

![Graph](image)

**Fig. 4.10:** $H_{\text{max}}$ as a function of the irreversibility field. The line is a guide to the eye only.
Unlike $H_{\text{max}}$, $F_{p_{\text{max}}}$ does not follow any simple relation with $c\Phi_n$, as illustrated in Fig. 4.9 (c). There is an ill-defined linear increase in $F_{p_{\text{max}}}$ with $c\Phi_n$ noticeable for the 0.6 wt% doped tapes in neutron fluences up to $3 \times 10^{19}$ m$^{-2}$, indicative of the overall increase in pinning strength with increasing fission-fragment density. The (0.6, 4) tape, however, has a much reduced $F_{p_{\text{max}}}$, as do all the 2 wt% doped tapes. A large uranium-oxide doping percentage or a large density of fission fragments evidently diminishes the maximum pinning force density. This is correlated with the reduced $J_{c0}$ and low field $J_c$ of these tapes. For the (0.6, 4) tape, however, $F_p$ is extended to higher applied fields, resulting in the observed $J_c$ improvements in high applied fields.

Despite the different behaviours, scaling the applied magnetic field to the irreversibility field and normalising $F_p$ to $F_{p_{\text{max}}}$, as shown in Fig. 4.11, reveals two distinct forms for tapes with fission-fragment defects ($c\Phi_n \neq 0$) and tapes without ($c\Phi_n = 0$). The tapes with $c\Phi_n = 0$ demonstrate a common functional form over the entire field range, with a peak in $F_p/F_{p_{\text{max}}}$ at $H_{\text{max}} \sim 0.22H_{irr}$. After the introduction of fission-fragment damage, the functional form is distinct to that of the tapes with $c\Phi_n = 0$, with the peak shifted to a common value of $H_{\text{max}} \sim 0.33H_{irr}$. There is a slight difference between the
curves in each of the two groups, but this difference is much smaller than the difference between the groups.

The 2 wt% doped tapes are again an exception. The peak in $F_p/F_{pmax}$ for the $(2, 0.05)$ tape occurs at the same value of $H/H_{irr}$ as for those tapes without fission-fragment damage and the functional form is also similar. The small fission-fragment density for this tape thus produces a negligible amount of pinning. For the $(2, 0.1)$ tape, on the other hand, $H_{max}$ is similar to that of the tapes with columnar defect damage. The form of the $F_p/F_{pmax}$ curve, however, approaches that of the $c\Phi_n = 0$ tapes in fields higher than the peak field.

![Graph showing normalised pinning force density plotted against the reduced field.](image)

**Fig. 4.11:** Normalised pinning force density plotted against the reduced field.
An elastic pinning theory was developed to explain the observed scaling of $F_p$ in conventional type-II superconductors, predicting that the peak occurs when pinning is overcome by FLL shear. The model predicts that at low reduced fields $h = H/H_{c2}$ in conventional superconductors, the pinning force density is dominated by the pinning force exerted by defects against the Lorentz force. This is an increasing function of $h$, as the Lorentz force increases with magnetic field. At high values of $h$, the FLL is proposed to shear plastically around the pinning centres. This shear process in the range of high $h$ dominates the pinning force density, which is dependent on the shear modulus of the FLL, a decreasing function of $h$ near $H_{c2}$. The competition between these two processes produces a maximum in $F_p$ at a reduced field where the two effects are approximately equal in strength. In the case of HTS, as the pinning force density is reduced to zero at the irreversibility field rather than $H_{c2}$, $H_{irr}$ can be substituted to define the reduced field instead.

This elastic pinning theory was developed for a large distribution of weak pinning centres, and only few strong pins, so that shear is only weakly dependent on the pinning strength. As a consequence, an increase in the pinning strength is predicted to shift the peak in $F_p$ to lower reduced fields. It was suggested that this elastic pinning model describes well the $F_p$ scaling in Ag/Bi2223 tapes.
However, the observations of the shift in the peak field $H_{\text{max}}$ as pinning is increased in the uranium-doped Bi2223 tapes show the opposite. The increasing $J_c$ in high applied fields as the density of quasi-columnar pinning centres is increased is testament to the strong induced pinning forces. As mentioned above, the linear relation observed between $H_{\text{max}}$ and $H_{\text{irr}}$ suggests that $H_{\text{max}}$ also reflects the strength of the pinning.

The more extended defects introduced, the weaker is the plastic shear deformation. The strong alignment of the vortices along the columns effectively increases the shear modulus of the pinned vortices. The assumption of shear that is independent of pinning is therefore not valid for our strongly pinned, uranium-doped and irradiated, Ag/Bi2223 tapes. Hence, the elastic pinning model as it is described in Ref. 7 cannot successfully describe the flux dynamics in these tapes.

Another phenomenological model, the Dew-Hughes model, was developed to account for the experimentally observed scaling of the pinning force densities by considering direct summation of the elementary pinning forces. This model successfully described the $F_p$ scaling using the form

$$F_p / F_{p\text{max}} = A h^p (1 - h)^q$$  \hspace{1cm} (4.2)

where $h = H/H_{c2}$, $A$ is a numerical constant and $p$, $q$ are fitting parameters. The model considers various types of elementary pinning mechanisms, as
discussed in Section 1.4.2.1, and distinguishes between point, surface and volume pinning as well as pinning by normal or superconducting centres. Within the model, the actual pinning mechanism is described by particular values for the parameters $p$ and $q$.

As for the elastic pinning model above, in the case of various HTS materials $F_p$ exhibits scaling behaviour when $H$ is instead normalised by the irreversibility field. The model of Dew-Hughes is empirically adjusted so that, with $h = H/H_{irr}$, Eq. (4.2) adequately describes the scaling of $F_p$ in HTS.\textsuperscript{8, 11, 12}

The fits to Eq. (4.2) for the nondoped tapes are shown in Fig. 4.12 (a). The functional form can be well described by Eq. (4.2), with parameters $p = 0.79 \pm 0.01$ and $q = 3.00 \pm 0.10$. The resulting parameters are similar to those found previously for Ag/Bi$_{2223}$ tapes prepared using various processing techniques.\textsuperscript{8} They are not, however, indicative of any particular pinning mechanism according to the Dew-Hughes model.

For the doped and irradiated tapes, a fitting procedure was performed for the (0.6, 1.25) and (0.15, 5) tapes, representative of the irradiated tapes, shown in Fig. 4.12 (b). They show a limited fit to Eq. 4.2, with $p = 1.04 \pm 0.03$ and $q = 2.48 \pm 0.05$, but it does not adequately follow the curve at $H_{max}$. As the majority of tapes show a functional form close to that of the (0.6, 125) tape, it
is apparent that Eq. 4.2 does not adequately describe the tapes after introduction of fission-fragment defect pinning centres.

Fig. 4.12: Fit to Eq. (4.2) (solid lines) of normalised pinning force density against reduced field for tapes with (a) $c\Phi_n = 0$ and (b) $c\Phi_n \neq 0$. 
Since the Dew-Hughes model was developed for LTS, it does not take into account flux creep, which is a more significant effect in HTS, as described in Section 1.4.3. In Ref. 11, the authors extend this model to include thermally activated flux creep. They begin by assuming the Kim-Anderson model, Eq. (1.28), and that $U_0$ is proportional to the volume $V$ of flux line that is coherently activated. Two possibilities for this activation volume are considered, $V = a_0^3 \propto H^{-3/2}$, or $V = a_0^2 \xi \propto H^1$. Substituting Eq. (1.28) for $J_c$ in Eq. (1.21) and solving for $F_p = 0$ and $H = H_{irr}$ leads to an expression for $H_{c2}$ as a function of $H_{irr}$, where the irreversibility field is defined at the limit of zero pinning force density. Replacing $H_{c2}$ in Eq. (4.2) with the resultant expression gives, for $V = a_0^3$,

$$F_p = R_1 h^p \left(1 - h^{3/2-p}\right), \quad (4.3)$$

and for $V = a_0^2 \xi$,

$$F_p = R_2 h^p \left(1 - h^{1-p}\right), \quad (4.4)$$

where $R_1$ and $R_2$ are constant factors, $h = H/H_{irr}$, and $p$ is the fitting parameter as given in Eq. (4.2).

The Kim-Anderson model is strictly valid only for $J$ close to $J_c$. The authors also take into account a more general expression for $J_c$, taken from observed $E$-$J$ characteristics.\(^{11}\)
Chapter 4: Critical Current Density

\[ J_c = J_0 \left( \frac{E_c}{E_0} \right)^{\frac{kT}{U_0}} \]  

(4.5)

where \( E_c \) is the electric field criterion used to define \( J_c \) and \( E_0 \) is the minimum measurable electric field. Using the same procedure, the authors find expressions for \( F_p \) of

\[ F_p = R_3 hC^{h^{3/2}p} \]  

(4.6)

for \( V = a_0^3 \), and

\[ F_p = R_3 hC^{h^{1-p}} \]  

(4.7)

for \( V = a_0^2 \xi \), where \( R_3 \) and \( C \) are constant factors.

The above expressions for \( F_p \), Equations (4.3) to (4.7), were fit to the normalised \( F_p \) curves of Fig. 4.11, some of which are shown in Fig. 4.13. Eq. (4.3) inadequately describes the form for the virgin (0, 0) tape, where Fig. 4.13 (a) shows the best possible fit found. A poor fit was also obtained for Eq. (4.4), and the doped, irradiated tapes fared no better. An adjustment for flux creep using the Kim-Anderson model thus fails to adequately describe the scaling behaviour of \( F_p \) for these Ag/Bi2223 tapes.

Fitting is more successful for the generalised \( J_c \) adjustment, as shown in Fig. 4.13 (b), (c). However, for both the (0, 0) and (0.6, 1.25) tapes the fits with Equations (4.6) and (4.7) are almost identical except for the fitting
parameters, presented in Table 4.2. From the identical behaviour of the fits, it is impossible to distinguish between the two approximations for the activation volume. In addition, the fitting parameters do not suggest any particular pinning mechanism according to the values given by the Dew-Hughes model.

### TABLE 4.2
**FITTING PARAMETERS FOR A FIT OF NORMALISED $F_p$ AGAINST REDUCED FIELD**

<table>
<thead>
<tr>
<th>Doping percentage $c$ (wt%)</th>
<th>Thermal Neutron Fluence $\Phi_n$ ($10^{19}$ m$^{-2}$)</th>
<th>$R_1$</th>
<th>$C$</th>
<th>$p$ Eq. 4.6</th>
<th>$p$ Eq. 4.7</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>12.0</td>
<td>2.8</td>
<td>0.80</td>
<td>-0.20</td>
</tr>
<tr>
<td>0.6</td>
<td>1.25</td>
<td>5.5</td>
<td>4.8</td>
<td>0.75</td>
<td>-1.0</td>
</tr>
</tbody>
</table>

**Fig. 4.13:** (a) Example of a fit to Eq. (4.3), representative of the fits for both $c\Phi_n = 0$ and $c\Phi_n \neq 0$, as well as for both Equations (4.3) and (4.4).
**Fig. 4.13:** Fit to Equations (4.6) & (4.7) for (b) $c\Phi_n = 0$ and (c) $c\Phi_n \neq 0$. The solid lines represent fits to both Equations.
**In-plane field**

Very similar results were found in fields parallel to the tape $ab$-plane, with a larger magnitude of $F_{p_{\text{max}}}$ and $H_{\text{max}}$. As for $H^c$, $H_{\text{max}}$ and $F_{p_{\text{max}}}$ are increased by the fission-fragment damage, with the exception of the (0.6, 4) tape which has a lower than expected $F_{p_{\text{max}}}$. The peak in $F_p$ occurs at a lower reduced field, however, with $H_{\text{max}} \sim 0.13 H_{\text{irr}}$ for the tapes without fission and $H_{\text{max}} \sim 0.20 H_{\text{irr}}$ for fission-damaged tapes. Scaling the normalised $F_p$ to the reduced field demonstrates the same distinction in functional form between the fission-damaged and pristine tapes. The fitting analyses conducted above for $H^c$ also behave similarly. The fitting parameters are different, but still do not indicate any particular pinning mechanisms.

**4.2.4 $\Phi_n$ Dependence**

Despite the small decrease in $J_{c0}$ (Section 4.2.1.1), irradiation with a high thermal-neutron fluence results in increases of $J_c$ in a magnetic field (Section 4.2.1.3) as well as $H_{\text{irr}}$ (Section 4.2.1.4) and $H_{\text{max}}$ (4.2.3). However, between tapes with equivalent $c\Phi_n$ these increases are larger for the tapes that were exposed to larger neutron fluences.

There are a couple of possible explanations. Firstly, a similar effect has been observed in Y123, where the uranium is unevenly distributed within the
superconductor matrix. If the uranium reaches a solubility limit, excess uranium will be deposited in phases physically separated from the superconductor, which then has minimal effect on the flux pinning. In Bi2223, the uranium was also observed in secondary phases. These phases were typically 1-20 µm in size, with a typical composition of U : Sr : Ca of about 1 : 1.5 : 1.5. In addition, the region around the particles usually had depressed Sr and Ca content (or higher Bi and Cu content) than the rest of the Bi-2223 matrix. The size of the phase is significant in comparison to the ~ 6 µm range of a single fission fragment.

Another possibility is that pinning defects are created by a component of the irradiation flux. The energy of the thermal-neutrons ~ 0.025 eV is too small to create further defect structures other than through uranium fission. Thermal-neutron capture and gamma emission may occur, creating minor displacements, but the cross-sections for these interactions are negligible.

A fast-neutron component of the flux, on the other hand, could produce extra point pinning centres. The irradiations were performed in the graphite region of the reactor, which highly thermalises the neutron flux, so that any fast neutron component is expected to be < 1% of the total flux. At the maximum fluence of $6 \times 10^{19} \text{ m}^{-2}$, the fast-neutron component would not be
expected to be greater than \( \sim 1 \times 10^{18} \text{ m}^{-2} \). However, this may prove to be sufficient to produce the observed increases in pinning.

### 4.3 Conclusion

Significant improvements in both the field and angular dependence of \( J_c \) as well as in the irreversibility field were observed at liquid nitrogen temperatures in the uranium doped and irradiated Ag/Bi2223 tapes. For commercial, industrial or technological applications, the optimum doping and irradiation combination depends entirely on the conditions or restrictions placed upon the tape characteristics. If large improvements in \( J_c \) over a wide range of applied fields are sought, moderate doping and \( \Phi_n \) levels provide excellent performance. Much lower radioactivity levels, on the other hand, can be achieved with a high \(^{235}\text{U} \) content and low \( \Phi_n \) while still retaining the improved flux pinning performance, but the \( J_c \) levels are much lower as well.

The pinning force densities calculated from the \( J_c \) field dependence confirmed the increase in pinning strength with fission-fragment defect density. None of the current models were successful in identifying the relevant pinning mechanisms, though.
References

In zero applied field, the transition from normal resistance $R_n$ to $R = 0$ occurs at $T \approx T_c$. The critical temperature is conventionally defined (arbitrarily) as the temperature at the midpoint of the transition, where $R = \frac{1}{2} R_n$. The magnetic broadening of the resistive transition occurs as a result of the dissipation due to thermally activated flux motion. As shown in Section 1.4.1, the energy dissipation resulting from flux motion induces a sample resistivity, which, from Eq. (1.15), is proportional to the magnitude of flux in motion. Thus, in an applied magnetic field, as the temperature is lowered through $T_c$, the transition to zero resistivity is widened. With a small driving current, the Lorentz force on the flux is small and flux motion is dominated by thermally
activated depinning. Therefore the resistive transition is also dominated by thermal activation.

An increase in flux pinning reduces the thermal activation, and thus affects the behaviour of the resistive transition by acting to reduce the magnetic broadening. The action of the flux pinning centres in reducing the motion of the flux lines lowers the energy dissipated in the superconductor, and hence the resistivity of the sample. The magnetic broadening of the resistive transition can therefore be used as a basis for comparing the pinning strengths of various materials, where the stronger pinning is indicated by a reduced broadening.

In the limit of small driving currents $J \ll J_c$, the resistance is the result of a thermally activated process, and can be expressed in terms of an activation energy $U_0$,\[ R = R_0 \exp\left(-\frac{U_0}{k_BT}\right). \quad (5.1) \]

The vortex system is then in the TAFF regime (Section 1.4.3). Defining an Arrhenius plot of the resistance against temperature, $\ln R$ vs $T^{-1}$, the slope of the linear portion corresponds to the height of the pinning potential well $U_0/k_B$, extrapolated to $T = 0$. The linearity is expected to break down close to $1/T_c$, where fluctuations of the superconducting order parameter set in.
Chapter 5: Activation Energies from Resistive Measurements

5.1 EXPERIMENTAL TECHNIQUES

Four different combinations of \((c, \Phi_n)\) were measured, listed in Table 5.1 below. Two separate samples with a \((0.6, 1.75)\) combination were measured, both tapes having been cut from the same pre-irradiation parent tape and irradiated simultaneously. Initial measurements on the tapes were performed in fields below 1 T. When extending the measurements out to higher applied fields, a second \((0.6, 1.75)\) tape was used; three common fields were re-measured. The data from both \((0.6, 1.75)\) tapes, for both the resistive transition and the derived activation energies described in Section 5.2, below, conformed to within experimental uncertainty, \(\pm 2\%\). As a result, in the following sections the results will not be distinguished.

A low frequency ac method was used to perform the resistance measurements, with \(I_{rms} = 1\ mA\) and an applied frequency \(f = 18.4\ Hz\), in the standard four-probe arrangement described in Section 4.1. The measurements

<table>
<thead>
<tr>
<th>Doping Percentage</th>
<th>Thermal Neutron Fluence</th>
<th>Fission Track Density</th>
<th>Irreversibility Field</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c) (wt%)</td>
<td>(\Phi_n) ((10^{19}\ m^{-2}))</td>
<td>(c\Phi_n) ((10^{19}\ wt%\ m^{-2}))</td>
<td>(\mu_0H_{irr}</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>700</td>
</tr>
<tr>
<td>0.15</td>
<td>5</td>
<td>0.75</td>
<td>1120</td>
</tr>
<tr>
<td>0.6</td>
<td>1.25</td>
<td>0.75</td>
<td>960</td>
</tr>
<tr>
<td>0.6</td>
<td>1.75</td>
<td>1.25</td>
<td>1070</td>
</tr>
</tbody>
</table>
were performed in a temperature range 40 – 120 K and in magnetic fields $\mu_0H \leq 3$ T both parallel and perpendicular to the $c$-axis.

The samples were placed within a continuous flow cryostat, using liquid He as coolant. The gas flow is regulated through a needle valve, which can be controlled either manually or automatically using a PID (Proportional-Integral-Differential) temperature controller. An evaporator is located at the base of the sample chamber. It houses a heater, which is also regulated by the temperature controller. A temperature sensor in the evaporator monitors the temperature of the gas atmosphere. A second temperature sensor is mounted on the sample holder to monitor the stability of the temperature at the sample location.

To avoid fluctuations in the temperature the gas flow was kept constant, with the needle valve kept open at 35%, and the temperature was controlled entirely through the evaporator. Beginning at a stable temperature $T = 120$ K, heating from the evaporator was ramped down so that cooling occurs at a rate of 0.1 K/min. This minimised the lag between the temperature of the gas atmosphere and that of the sample.

A lock-in amplifier (SR830) was used as an ac current source, connected to the tape sample with a 1 k$\Omega$ resistor in series. The resistor ensures a constant current level through the tape, even when the
superconductor resistance drops to zero. With the 1 kΩ resistor in series, a 1 V\text{rms} output from the lock-in provides 1 A\text{rms}. The resistance of the sample and connecting wires was negligible, compared to the resistor.

The same lock-in amplifier was used to measure the voltage after differential pre-amplification by a transformer amplifier. The pre-amplifier provides amplification of 100 times, decouples the low-temperature set-up from the lock-in amplifier and differentiates the input signals. The output from the pre-amplifier was measured using the internal reference signal of the lock-in. A 20-µV range was sufficient to provide satisfactory accuracy over the entire temperature range, with a voltage resolution of approximately 1 nV.

5.2 RESULTS AND DISCUSSION

5.2.1 Resistive Transitions and Magnetic Broadening

The resistive transitions of all tapes are displayed in Fig. 5.1 at an applied field of 100 mT. Relative to the virgin (0, 0) tape, the resistive behaviour of the irradiated tapes is very similar. Hence, to simplify the
Chapter 5: Activation Energies from Resistive Measurements

Fig. 5.1: Normalised resistance against temperature, at an applied field 100 mT parallel to the c-axis.

discussion, when comparing between irradiated and non-irradiated tapes we will discuss the features of the (0.6, 1.75) tape as being typical of a U/n treated tape.

The shapes of the resistive transitions are consistent with parallel conduction between the superconductor core and the silver sheath, as described in Section 4.1. Parallel conduction results in the total measured resistance, $R$, being a combination of the superconductor and silver sheath resistances, $R_s$ and $R_{Ag}$, respectively. At low temperatures, where thermally induced dissipation is negligible and $R_s \ll R_{Ag}$, the current flows only through
the superconductor and our measurements are not affected by the silver sheath. Once the dissipation increases and \( R_s > R_{Ag} \), the current flows primarily through the sheath material and \( R_{Ag} \) dominates the resistive behaviour.

In a previous study on the magnetoresistance of silver-sheathed Bi2223 tapes\(^4\), it was ascertained that \( R_s \) coincides with \( R \) within a few tenths of a degree K above \( R = 0 \), in a region where \( R_s \) is typically less than 10% of \( R_n \), the normal state resistance at a temperature \( T \) just above \( T_c \). We employ this result to define the resistive behaviour below 10% \( R_n \) as being indicative of solely the superconductor resistance.

### 5.2.1.1 Comparison of Irradiated and Nonirradiated Tapes

Fig. 5.2 compares the resistive transitions of the virgin tape and the typical treated tape, (0.6, 1.75), at various applied fields. Figures 5.2 (a) and (b) show the results for the magnetic fields applied parallel and perpendicular to the \( c \)-axis of the tapes, respectively. The zero-field superconducting transition temperature, \( T_{c0}(R = \frac{1}{2}R_n) \), was not significantly affected by the doping and irradiation treatment. A reduction in \( T_{c0} \) of less than 0.5 K is found after irradiation, as was confirmed by ac susceptibility measurements of \( T_c \), Section 3.3.
Fig. 5.2: Normalised resistive transitions for a virgin tape and a tape with typical doping and irradiation, in fields applied (a) parallel and (b) perpendicular to the $c$-axis. The difference between the two tapes increases further as the field is increased.
An appreciable decrease in the resistive transition broadening by magnetic field was observed after irradiation for fields $\mu_0 H > 10$ mT. As explained in Section 5.1 above, this reduction in the transition broadening is an indication of the improved flux pinning in the irradiated tapes. There was no observed reduction in the broadening with the field oriented in the $ab$-plane, however, until larger applied fields. This implies that the strong intrinsic pinning for flux oriented along the $ab$-plane is dominant over the introduced fission-damage pinning. This will be discussed further in Section 5.2.2.3.

5.2.1.2 Comparison of $c\Phi_n$ combination effects

Fig. 5.3 highlights the effect of the different doping and irradiation levels on the broadening of the resistive transitions at various fields applied parallel to the $c$-axis. While the behaviour of the treated tapes may appear similar in relation to the virgin tape, there are some apparent changes that can be observed between the different combinations of $c$ and $\Phi_n$. As discussed in Chapter 4, observations on the effects of doping and irradiation on $J_c$ and the pinning force density $F_p$ revealed that the overall pinning strength is proportional to the density of fission tracks $\sim c\Phi_n$, but that there are also indications that the pinning may have an additional weak dependence on $\Phi_n$. 
Fig. 5.3: Normalised resistive transitions for all tapes, in (a) low and (b) high applied fields, parallel to the c-axis.
The effect of the fission-track density on pinning strength is confirmed by observations on the tapes with equivalent doping. The (0.6, 1.75) tape, which has the highest $c\Phi_n = 1.05 \times 10^{19}$ m$^{-2}$ of the measured tapes, shows a smaller broadening than the (0.6, 1.25) tape in fields $\mu_0 H_c > 100$ mT, as would be expected for a tape with a higher pinning density. The difference in $\Phi_n$ is sufficiently small that we can associate the change in broadening solely to changes in the fission-track density.

However, notwithstanding the approximately equal fission-track densities, $c\Phi_n = 0.75 \times 10^{19}$ wt% m$^{-2}$, of the (0.6, 1.25) and (0.15, 5) tapes, the resistive transitions are markedly different. At an applied field of $\mu_0 H = 400$ mT, the resistive transitions are almost identical for the (0.15, 5) and (0.6, 1.75) tapes. As the field is increased, the broadening becomes substantially smaller for the (0.15, 5) tape compared to both of the 0.6-wt% doped tapes, even, despite the slightly higher $c\Phi_n$ of the (0.6, 1.75) tape. This would seem to indicate that the changes in resistive transition are due not only to the changes in fission track density, but also depend on the thermal neutron fluence, $\Phi_n$. Taken together, these findings indicate that both $c\Phi_n$ and $\Phi_n$ participate in the enhancement of the vortex pinning in uranium-oxide doped Ag/Bi2223 tapes, as was concluded in Chapter 4.
5.2.2 Activation Energies

All of the Arrhenius plots, lnR vs $T^{-1}$, Fig. 5.4, exhibit linear behaviour over approximately three orders of magnitude at $R < 10\%$ of $R_n$, where we define $R_n = R(112 \text{ K})$. As explained above, the resistive behaviour at $R < 10\% R_n$ can be attributed to the properties of the superconductor core, independent of the sheath conduction. The linearity of the lnR vs $T^{-1}$ plots is expected for thermally activated flux motion with an activation energy $U_0$ as given in Eq. (5.1) above.

Fig. 5.4: Normalised Arrhenius plots of the resistive transitions for all tapes
The activation energies determined in this manner are displayed in Fig. 5.5 as a function of applied field. The intrinsic activation energy of the virgin tape exhibits a field dependence $U_0 \propto H^{-0.55}$ in fields both parallel and perpendicular to the $c$-axis. This is consistent with previous magnetoresistance measurements on Ag/Bi2223 tapes, as well as Bi2212 thin films, which give $U_0 \propto 1/\sqrt{H}$. The square-root field dependence of the activation energy is associated with thermally activated plastic shear deformations of a 3D flux lattice or 3D viscous flux liquid.

**Fig. 5.5:** Field dependence of the activation energies calculated from the slopes of the Arrhenius plots, for all tapes and in fields applied parallel and perpendicular to the $c$-axis. The lines are guides to the eye, representing the approximate $H^{-1/2}$ dependence.
5.2.2.1 *Comparison of Irradiated and Non-irradiated Tapes*

As with the resistive transition broadening, the major changes after the doping and irradiation treatment occur for fields oriented along the $c$-axis. Unlike the field dependence of the intrinsic $U_0$, however, the field dependence of $U_0$ after irradiation, in fields parallel to the $c$-axis, is not indicative of any simple flux flow mechanism. Rather, a smooth variation in $U_0$ is observed, suggesting that different sets of pinning sites become involved at different field values, which may be an indication of matching field effects.

At an applied field $\mu_0H^c \sim 10$ mT, $U_0$ for the virgin and all of the irradiated tapes, $U_0^{\text{vir}}$ and $U_0^{\text{irr}}$ respectively, are almost equal. In applied fields above this, $U_0$ is increased by the fission-track defect pinning and the post-irradiation activation energy $U_0^{\text{irr}}$ deviates from the intrinsic activation energy, $U_0^{\text{irr}} > U_0^{\text{vir}}$. In this region, the magnetic field dependence of $U_0^{\text{irr}}$ is initially much weaker than that of $U_0^{\text{vir}}$ (Fig. 5.5). The magnitude of the deviation steadily increases with applied field, reaching maximum deviation at an applied field of approximately 300 mT. The ratio of $U_0^{\text{irr}}/U_0^{\text{vir}}$ is demonstrated in Fig. 5.6, where $U_0^{\text{irr}}$ is taken from the measurements of the (0.6, 1.75) tape and $U_0^{\text{vir}}$ is extrapolated out to 3 T using Fig. 5.5, assuming that it follows the same $H^{-0.55}$ field dependence out to high fields. An interpolation at fixed field
Chapter 5: Activation Energies from Resistive Measurements

Fig. 5.6: Ratio of activation energies between the irradiated and virgin tapes, calculated from an interpolation of $U_0$ between 0 and 3 T. $U_0^{irr}$ is the activation energy of the (0.6, 1.75) tape. 

values was used to calculate the ratio demonstrated in Fig. 5.6. The small fluctuations in the data points are an artifact of the interpolation. The relevant feature is the dominant peak in $U_0^{irr}/U_0^{vir}$ at $\mu_0 H \approx 300$ mT, where $U_0^{irr} \sim 2.6U_0^{vir}$.

These results corroborate the conclusions from the $J_c(H)$ measurements in Chapter 4. The substantial reductions in $J_c$ field dependence, the increase in $H_{irr}$ and the shift of $F_p$ to higher applied fields were all features associated with a strong improvement in the flux pinning as a function of $c\Phi_n$. The correlation between these independent measurements thus confirms the observation that increases in $J_c$ of uranium-doped and neutron-irradiated
Ag/Bi2223 tapes can be directly attributed to increases in the pinning potential barrier due to the introduction of the splayed columnar pinning centres.

Above 300 mT, the difference in $U_0$ decreases slowly. In Fig. 5.5, a broad change in slope is observed, beginning at approximately 300 mT as the magnetic field dependence of $U_0^{irr}$ becomes stronger. The result is that $U_0^{irr}$ appears to re-converge with the intrinsic activation energy at $\mu_0H_F \sim 2$ T.

The decline in the pinning efficiency of the fission-track defects above 300 mT is related to the matching effect mentioned in Section 1.5.3.2. Below the matching field $B_\phi$, the density of pinning centres exceeds the density of flux lines, resulting in more than sufficient numbers of columnar defects (CDs) available for pinning. The flux lines are strongly localised within individual fission-track defects and the system is in the Bose-glass phase, resulting in the weak field dependence of $U_0^{irr}$ below $B_\phi$.

Above $B_\phi$, however, the number of flux lines is greater than the total available pinning centres. Consequently, a number of flux lines lie between the quasi-columnar defect sites, with only intrinsic pinning centres and interactions with the pinned portion of the FLL limiting their dissipative motion. The activation energy is therefore limited in fields greater than $B_\phi$ by the action of the interstitial flux lines and thus approaches $U_0$ of the virgin sample.
The concept of a matching field was originally defined for a parallel track configuration. It can, however, be extended to isotropic defects. The volume density of fission events can be calculated from Eq. (1.33) as was discussed previously. Multiplying this by the average length of a fission track has been suggested to give the maximum density of fission tracks within the superconducting plane. This figure can then be employed to calculate the matching field as for unidirectional CDs, given by Eq. (1.31). From TEM examination of the p-Bi fission damage in Ref. 10, an estimation of the density of tracks viewed along a direction perpendicular to the tape plane yielded a value very close to the $B_\phi$ calculated from the irradiation details using this procedure. Using the combination of $c = 0.6 \text{ wt}\% \approx 1 \times 10^{20} \text{ } ^{235}\text{U} \text{ cm}^{-3}$, $\Phi_n = 1.75 \times 10^{19} \text{ m}^{-2}$ and an average track length of 5 $\mu$m, the density of fission fragments is $\sim 1 \times 10^{11} \text{ cm}^{-2}$, indicating a $B_\phi \approx 2.1 \text{ T}$. 

This matching field effect has been recently observed in Bi2212 single crystals after both heavy ion irradiation (HII) and proton-induced Bi fission, as well as in heavy ion irradiated Ta-Ge films. For the highly directional CDs produced by HII, a sharp difference in the field dependence of $U_0$ is observed to occur at approximately $B_\phi$. A weak field dependence is observed below $B_\phi$, while $U_0$ decreases rapidly at magnetic fields $\sim B_\phi$ and approaches that of the nonirradiated crystals for fields far above the matching field.
The isotropic fission tracks created by p-Bi fission,\textsuperscript{11} however, demonstrated a broad change in $U_0$ rather than a sharp transition. The smoother change in field dependence observed for the isotropic defects may be an indication of a broad distribution of activation energies.\textsuperscript{11} The varying angular orientation of the fission tracks results in an irregular defect landscape, in comparison to the regular defect structure of unidirectional HII. There is an energetic cost in tilting a flux line to accommodate to the angled defects.\textsuperscript{11} The finite elastic energy of the flux lines limits the angle over which the lines can accommodate to a track inclined with respect to the field direction. Tracks with a larger inclination are ineffective at trapping the vortices. Flux will, therefore, rarely accommodate along the entire length of a CD,\textsuperscript{13} and the length over which a flux line is pinned varies. Additionally, there are suggestions that the activation energy is affected by the projection of the columnar defect in the superconducting plane.\textsuperscript{14} The random orientation of the CDs results in a wide range of ellipsoidal cross-sections in the superconducting plane, which could result in a range of pinning strengths. These factors all combine to create a pinning landscape with a broad variation in activation energies that has been suggested to explain the broad transition in field dependence of $U_0$.\textsuperscript{11}
The broad distribution in pinning also leads to a lower observed $B_\phi$ than calculated from fission track densities. The matching fields of the Bi2212 single crystals in Ref. 11 were determined from the scaling of the field-dependence of $T(R = 0)$. The authors equated this line on the $H$-$T$ plane with the irreversibility line. The samples with unidirectional defects demonstrated a sharp transition in the temperature dependence of $H_{irr}$ near $B_\phi$, and a universal behaviour, between samples with different irradiation fluences, when scaled to the matching field. The isotropic defects, on the other hand, displayed a similar crossover in the temperature dependence but at a magnetic field lower than $B_\phi$. The effective matching field $B_{eff}$, which scaled the behaviour onto a universal line nearly identical to that of the unidirectional samples, was found to be $\sim B_\phi/2.6$.

Figure 5.7 shows the $T(R = 0)$ line in the $H$-$T$ plane, which can be defined as a transport irreversibility field $H_{irr}^t$, calculated from the measured resistive transitions, Figures 5.2 and 5.3. The broad transition in the temperature dependence of $H_{irr}^t$ is clearly observed for the (0.6, 1.75) tape at a $B_{eff} \sim 300$ mT identical to that found from the field dependence of the activation energy. Compared to $B_\phi \sim 2.1$ T calculated above, $B_{eff} \sim B_\phi/7$. The $B_{eff} < B_\phi$ is an indication of the lowered pinning efficiency of a fraction of the isotropic defects, arising from properties such as the finite elastic
energy, which limits the maximum tilt angle for flux line accommodation, and the action of vortex-vortex interactions. This effect is consistent with the proposed theory of Krusin-Elbaum et al. as discussed in Section 2.1, where the effects of anisotropy on the effective matching field are considered.

The reduced effective matching field in comparison to $B_\phi$ also indicates that the present method for calculating $B_\phi$, by multiplying the total fission track volume-density with the average fission track length, is an oversimplification of the isotropic pinning landscape. It confirms that the real situation is complicated by the material anisotropy. $B_\phi$ thus depends on the track pinning efficiency and the elastic line energy, as well as on the
material anisotropy, which can effectively rescale the angular distribution of the columnar tracks.\(^{17}\)

### 5.2.2.2 Comparison of \(c\Phi_n\) combination effects

The magnitude and field dependence of \(U_0^{\text{irr}}\), Fig. 5.5, is quite similar for the three irradiated tapes. \(U_0\) corresponds to the average activation energy of a single pinning centre. If the quasi-columnar fission-fragment defects dominate the distribution of activation energies, in comparison to any pre-existing pinning centres, then \(U_0\) remains the same regardless of the density of fission tracks, as observed. The field dependence of \(H_t^{\text{irr}}\), Fig. 5.7, is also very similar for the three irradiated tapes. Unlike \(U_0\), \(H_t^{\text{irr}}\) is expected to be dependent on \(c\Phi_n\), as shown in Section 4.2.1.4. Thus, the similarities in \(H_t^{\text{irr}}\) are consistent with the rather similar \(c\Phi_n\) values of the three tapes (Table 5.1).

It is difficult to distinguish a separate \(B_{\text{eff}}\) for the lower \(c\Phi_n\) tapes (0.6, 1.25) and (0.15, 5) from either Fig. 5.7 or Fig. 5.5. There are insufficient data points in Fig. 5.5 at the crucial fields between 100 and 400 mT for both these tapes to allow an accurate interpolation of the field dependence as this effect was not anticipated within this field range. Regardless, it is not expected that \(B_{\text{eff}}\) would be very different. The relative difference in \(c\Phi_n\) between the tapes suggests that \(B_{\text{eff}}\) is expected at approximately 215 mT. However, it is possible
that the difference in $B_{\text{eff}}$ would be even smaller when the effects of the limited flux line accommodation discussed above are taken into consideration.

5.2.2.3 Comparison of $U_0$ for $H^{ab}$

With $H$ perpendicular to the $c$-axis, the doping and irradiation treatment does not appear to substantially alter $U_0$. The field dependence of both $U_0^{\text{vir}}$ and $U_0^{\text{irr}}$ is indicative of thermally activated plastic shear deformations, as was discussed in Section 5.2.2 above. The similar field dependence of $U_0$ in fields parallel and perpendicular to the $ab$-plane is difficult to account for.

Above $\mu_0 H^{ab} = 200\text{mT}$, $U_0^{\text{irr}}$ appears to slightly exceed $U_0^{\text{vir}}$, but the change in $U_0$ is minor. This can be understood if the magnitude of the pinning potential energy of the introduced columnar fission-damage tracks is not larger than the pre-existing pinning energy. The thermally activated flux flow process will then be dominated by the energy of the pre-existing pins. In the case of a field applied along the $ab$-plane of a Bi2223 grain, the pinning potential trapping the flux in the insulating layers between the Cu-O$_2$ planes, where the order parameter is highly suppressed, is quite large, especially in small fields, as discussed in Section 1.4.2.2. The larger values of $U_0$ in fields $H^{ab}$ compared with those in fields $H^c$ are consistent with the stronger intrinsic pinning.
From the lack of change in $U_0$ observed after irradiation, it appears that the intrinsic inter-layer pinning remains dominant over the induced columnar defect pinning at low applied fields. There is also little improvement observed in normalised $J_c$ for $\mu_0H^{ab} < 1$ T, as seen in Fig. 4.3 (b). However, significant improvements are observed in fields greater than 1 T, implying an improvement in the pinning force. The correlation between the increases in $U_0$ and the $J_c$ field dependence observed in the preceding Sections for $H$ parallel to the $c$-axis suggests that resistivity measurements in fields perpendicular to the $c$-axis greater than 1 T will show increases in $U_0$ after doping and irradiation.

### 5.3 Conclusion

The effects of the uranium doping and neutron irradiation treatment on the broadening of the resistive transitions demonstrate the strong pinning of the induced fission-fragment damage. Linearity in an Arrhenius plot of the superconductor resistance against temperature over at least three orders of magnitude indicated thermally activated flux motion. With the fields applied parallel to the $c$-axis, the pinning energy is substantially increased, $U_0$ tripling in magnitude at $\mu_0H \sim 300$ mT, and then re-converging with the pre-
irradiation $U_0$ at $\sim 2$ T. This behaviour of the activation energy is well described by the effect of an effective matching field $B_{\text{eff}} \sim 300$ mT.

The effective matching field is also suggested by the transition in field-dependence of $T(R = 0)$, identified as a transport irreversibility field $H_{\text{irr}}^t$, at an identical value of $B_\phi$. This effect had not been previously observed in the uranium-doped Ag/Bi2223 tapes, but could reasonably be expected from observations of similar effects in Bi2212 single crystals after both heavy-ion irradiation\textsuperscript{11, 12} and p-Bi fission.\textsuperscript{11}

The importance lies in the fact that the observed value for $B_{\text{eff}}$ is lower than the matching field $B_\phi$ calculated from the density of fission tracks times the average track length. This method of calculating $B_\phi$ is therefore an oversimplification, which does not take into account the random nature of the fission process as well as the finite elasticity of the flux lines.

In magnetic fields $\mu_0 H^{\alpha} \leq 1$ T, the pinning energy of the induced columnar defects does not, however, dominate over the intrinsic pinning, thus providing only a limited increase in the activation energies observed.
Chapter 5: Activation Energies from Resistive Measurements

References

In Section 1.4.3 the concept of an effective activation energy $U_{\text{eff}}$ was introduced as the thermal energy required to allow flux vortex hopping between pinning centres. The effective activation energy was defined as the pinning potential energy barrier $U_0$ reduced by the work contribution of a driving Lorentz force. Consequently, the effective activation energy is expected to be a decreasing function of the current density, approaching zero at $J = J_c$.

Since the activation energy determines the rate of flux creep, and hence the decay of the supercurrent density, measurements of the relaxation of the superconductor magnetic moment provide a useful means of investigating the activation energies. The $U_{\text{eff}}$ can be examined over a wide range of current densities by analysing the relaxation at various temperatures and applied fields. Theories of flux dynamics, such as collective flux creep or vortex-glass theories, predict the behaviour of $U_{\text{eff}}$. 
as a function of current density, so that the observed current dependence
can then be related to the dominant vortex dynamics.

The flux creep probed by the magnetic relaxation corresponds to a
different vortex system than the one probed by the resistive transition
measurements.\footnote{In these relaxation experiments, the system is close to the
critical state, so that $J \sim J_c$. Since the measurements are performed below
the irreversibility line, the system is assumed to be in the vortex-glass
state,\footnote{although, as will be seen, this is not necessarily always valid.}
Proper analysis of the magnetic relaxation data is crucial to the
correct interpretation of the results. Hence, the analysis theory will be
examined in detail, followed by a discussion of the experimental methods
and lastly the results and their interpretation.

\section{Analysis Theory}

There are a number of different methods available for analysis of
relaxation data.\footnote{One of the most widely cited in the literature is Maley’s
method.} Unlike the Kim-Anderson model, Eq. (1.28), this method avoids
making any assumptions about the form of the current dependence of $U_{\text{eff}}$,
but, rather, determines the form directly from measurements of the
magnetic relaxation.
6.1.1 Maley’s Method

Assuming the Arrhenius relation given in Eq. (1.27) for the hopping rate of a thermally activated flux bundle,

\[ v = v_0 \exp \left( - \frac{U_{\text{eff}}}{k_B T} \right) \]  

(1.27)

Beasley et al. derived a form for the flux flow density,

\[ D = - \left( \frac{\nabla B}{|\nabla B|} \right) B a v_0 \exp \left( - \frac{U_{\text{eff}}}{k_B T} \right), \]  

(6.1)

where \( a \) is an average hop distance of the activated flux bundles. The flux-flow density, \( D \), is defined as the amount of flux, per unit length and time, that crosses a line perpendicular to both the flux density \( B \) and the flux density gradient \( \nabla B \). In a one-dimensional approximation \( \nabla B = |\nabla B|n \), where \( n \) is a unit vector in the direction of the flux density gradient, such that Eq. (6.1) becomes

\[ D = -n B a v_0 \exp \left( - \frac{U_{\text{eff}}}{k_B T} \right). \]  

(6.2)

Conservation of flux requires that \( dB/dt = -\nabla \cdot D \), and thus a one-dimensional form of the flux density rate equation can be expressed as

\[ \frac{dB}{dt} = \nabla \cdot \left[ n B a v_0 \exp \left( - \frac{U_{\text{eff}}}{k_B T} \right) \right]. \]  

(6.3)

This expression is integrated over the sample volume to express the rate equation for the average flux density \( \langle B \rangle \). For simplicity, an infinite slab
geometry, with a finite thickness $d$, is assumed so that, using the divergence theorem, the integration results in the expression

$$\frac{d\langle B \rangle}{dt} = \mu_0 \left( \frac{dM}{dt} + \frac{dH}{dt} \right) = \frac{2\mu_0 H_a \nu_0}{d} \exp \left(-\frac{U_{\text{eff}}(J,H,T)}{k_B T} \right),$$

(6.4)

where the flux density has been expanded in terms of the magnetisation $M$ and the magnetic field $H$, as given in Eq. (1.3), and it is assumed that $B \approx \mu_0 H$ for fields much higher than $H_{c1}$ (Eq. (1.5)).

Maley et al. considered the case of static magnetic relaxation, where the time decay of the superconductor magnetisation is measured under a constant applied field. Thus, $dH/dt = 0$ and Eq. (6.4) is rearranged to obtain an expression for the effective activation energy,

$$U_{\text{eff}}(J,H,T) = -k_B T \left[ \ln \left( \frac{dM}{dt} \right) - \ln \left( \frac{2H_a \nu_0}{d} \right) \right].$$

(6.5)

Measurements of the sweep rate dependence of the magnetisation, referred to as dynamic magnetic relaxation, can also be used in combination with Eq. (6.4) to find $U_{\text{eff}}$. In this case, $d\langle B \rangle/dt$ is expressed as

$$\frac{1}{\mu_0} \frac{d\langle B \rangle}{dt} = \frac{\partial M}{\partial H} \frac{\partial H}{\partial t} + \frac{dH}{dt} \approx \frac{dH}{dt},$$

(6.6)

where $\partial M/\partial H$ can be neglected in comparison to $dH/dt$. Equation (6.4) can then be rearranged to give
\[ U_{\text{eff}}(J,H,T) = k_B T \left[ \ln \left( \frac{H}{H_0} \right) + \ln \left( \frac{2a\nu_0}{d} \right) \right], \] (6.7)

where \( \dot{H} = \frac{dH}{dt} \) is the applied magnetic field sweep rate.

It has previously been shown that both of the above methods are equivalent.\(^7\)\(^8\) Thus, both Equations (6.5) and (6.7) allow the experimental determination of the current dependence of the effective activation energy, without the need for any prior assumption about the form of the dependence. This analysis technique is known as Maley's method.\(^5\)

Hereafter, discussion will be restricted to dynamic magnetic relaxation measurements. This method was considered to be the most time effective, as it does not require the temperature and field to be held constant over long periods of time. The drawback is that the available ranges of sweep rate are limited by the large inductance of the superconducting magnet used, limiting the effective relaxation rates that may be probed.

The current density is also derived from the measured magnetic hysteresis loops (MHL) of the superconductor sample. The measured magnetic moment \( m \) contains parasitic components, for example a background signal from the sample holder. Relaxation occurs only for the irreversible component of the superconductor moment \( m_{irr} \), however, which can be calculated as

\[ m_{irr} = \frac{m^+ - m^-}{2}, \] (6.8)
where \( m^+(-) \) is the magnetic moment of the field increasing(decreasing) branch of the hysteresis loop. This expression is not valid for edge and geometrical barrier pinning, however their contributions are negligible for the samples measured. From the critical state model, the irreversible magnetisation \( M_{irr} \) of the sample can be directly related to the (time-dependent) average circulating current density of the superconductor, where the magnetisation is the volume average of the magnetic moment, \( M = m/V \). As was discussed in Section 1.4.3, this is not \( J_c \) because of the strong flux creep in HTS. At a fixed temperature, the current density that may be probed is limited. To extend the range of \( J \), the MHL are measured over a range of temperatures.

Plotting the \( U_{eff} \) calculated from Eq. (6.7) against \( M_{irr} \), the resulting figure presents the approximate current density dependence of the effective activation energy. Experimentally, the plot consists of a series of disjointed isothermal curves. The second logarithmic term on the right-hand-side of Eq. (6.7),

\[
C \equiv \ln \left( \frac{2a
\nu_0}{d} \right), \tag{6.9}
\]

is not accessible from the experimental relaxation measurements, but is rather an additive constant to the isothermal curves. It is therefore taken as a fitting parameter, which can be adjusted to provide a smooth, continuous \( U_{eff}(J) \) curve. \( C \) is assumed to be temperature independent at \( T \ll T_c \) (or
only weakly temperature dependent at higher T)\(^5\). This assumption stems from the reasoning that \(C\) does not have a strong dependence on \(a\) or \(\nu_0\), both temperature dependent terms, as they are only logarithmic arguments within the constant.\(^{11}\) A similar argument was also applied to the field dependence of \(C\).\(^{11}\)

### 6.1.2 Temperature Scaling

McHenry et al.\(^{11}\) concluded that the magnitude and field dependence of the additive constant \(C\) in Eq. (6.5), required for a smooth fit to their data, was physically unreasonable. To overcome this, the introduction of a temperature scaling term was suggested to account for a change in the pinning potential barrier height at high temperatures, thus separating the explicit temperature dependence of \(U_{\text{eff}}\) from its current dependence. This modified form of Maley’s method can be verified by considering \(U_{\text{eff}}(J,H,T)\) as a separable function\(^{12}\) of applied field, temperature and current density. Then \(U_{\text{eff}}(J,H,T)\) can be expressed as

\[
U_{\text{eff}}(J,H,T) = U_0 U(T) U(H) U(J),
\]

(6.10)

where \(U_0\) is a characteristic pinning energy, and \(U(T)\), \(U(H)\) and \(U(J)\) describe the temperature, field and current dependencies of \(U_{\text{eff}}(J,H,T)\), respectively. It follows that
Chapter 6: Magnetic Relaxation

\[ U_{\text{eff}}(J, H, T_0) = U_0 U(H) U(J) = \frac{U_{\text{eff}}(J, H, T)}{U(T)} \]  \hspace{1cm} (6.11)

is the effective activation energy scaled to a characteristic temperature, \( T_0 \).

\( U(H) \) and \( U(J) \) then describe the field and current dependencies at \( T_0 \). To determine \( U(T) \), two different approaches may be considered.

6.1.2.1 Theoretically Determined \( U(T) \)

The first, as followed by McHenry et al.,\(^{11}\) takes known temperature scaling forms determined theoretically and applies them directly to the data. \( U_{\text{eff}}(J, H, T) \) is divided by the predetermined scaling term and values for the additive constant \( C \) are then chosen to provide the best continuity of \( U_{\text{eff}}(J, H, T_0) \) over the entire temperature range. The scaling term \( U(T) \) can be changed or adjusted until an additive constant is found that provides a smooth continuous fit.

Tinkham,\(^{13}\) by working the temperature dependencies of \( H_c \) and \( \lambda \) in the Ginzburg-Landau relations, found a temperature scaling factor of the form

\[ U(T) = 1 - t^2 \left( 1 - t^4 \right)^{1/2} / t ; t = \frac{T}{T_c}, \]  \hspace{1cm} (6.12)

which can be approximated to

\[ U(T) \approx (1 - t)^{3/2}, \]  \hspace{1cm} (6.13)
valid over a wide range of temperatures above \( t \approx \frac{1}{2} \). More specifically, this \( U(T) \) reflects the temperature dependence of the depairing current density, \( J_{c0} \), which is \( J_c \) at \( H = 0 \) and without any reductions by thermal fluctuations. \( U(T) \) is thus, by definition, not reduced to zero above \( T_{\text{irr}} \).\(^{13}\)

The form of Eq. (6.12) also predicts a temperature dependence of the form

\[
U(T) \approx \left(1 - t^2\right) 
\]

(6.14)

at low temperatures, \( t \ll \frac{1}{2} \). Temperature scaling functions of the form (6.13) and (6.14) can be found in the literature to obtain a smooth fit for \( U(J) \) under the modified form of Maley’s method.\(^{11, 14}\)

Other forms have been proposed without theoretical argument, for example a combination of Equations (6.13) and (6.14), \((1-t^2)^{3/2}\).\(^{13}\)

**6.1.2.2 Empirically Determined \( U(T) \)**

The second approach involves determining \( U(T) \) empirically from the experimental data.\(^{14}\) Since it can be reasonably expected that any dependence on temperature of \( C \) will be least at very low temperatures, the value of the additive constant \( C \) is first chosen to provide continuity of \( U_{\text{eff}} \) at the lowest two isotherm data sets (highest \( J \)). After adding this constant value of \( C \) to all isotherms, successive isotherms are adjusted by a multiplicative scaling factor \( G \) that produces a continuous fit to the adjacent lower isotherms. Once all isotherms have been adjusted, the resulting curve should be a continuous \( U_{\text{eff}}(J,H,T_0) \) curve. By curve fitting
the data set of \( G \) against \( T \) thus obtained, the functional form can then be determined, generally in the form\(^\text{14} \)

\[
G(T) = \left(1 - \frac{T^m}{a}\right)^n = U(T)^{-1}, \tag{6.15}
\]

where \( m, n \) and \( a \) are fitting parameters.

### 6.1.3 Theoretical \( U(J) \) Models

As explained in Section 1.4.3, the linear Kim-Anderson model fails to explain the non-logarithmic time decay of magnetisation in many HTS materials. Beasley \textit{et al.}\(^6\) predicted the need for a more realistic non-linear current density dependence of the activation energies and thus derived Eq. (6.3) to explain the resulting magnetic relaxation. Various subsequent theoretical descriptions of the vortex state as well as empirical models have been developed that predict the form of \( U_{\text{eff}}(J) \). Some of these are discussed below.

#### 6.1.3.1 Collective Creep Theory

The collective creep theory developed by Feigel'man \textit{et al.}\(^1\) considers thermal activation of flux bundles similar to the concept proposed by Anderson\(^\text{16} \) (Section 1.4.3). Collective creep treats an elastic flux line lattice, see Section 1.3.1.5, pinned by a system of weak, randomly distributed pinning centres. Thus, local displacements of the flux lines are
small, but the long-range order of the lattice is destroyed by the collective action of a large number of pinning centres.

Unlike the original flux bundle concept,\textsuperscript{16} however, the volume of the correlated flux bundle $V_c$ is dependent on the current density, becoming infinitely large as $J$ approaches zero. Consequently, collective creep theory predicts a diverging barrier as $J \to 0$. The theory predicts a power-law current density dependence of the activation energy for $J < J_c$,

$$U(J) = U_0 \left( \frac{J_c}{J} \right)^\mu,$$

(6.16)

where the exponent $\mu$ is dependent on the dynamics of the flux creep, such as the bundle volume and vortex dimensionality. For example, in the case of 3D flux lines three regimes were identified:\textsuperscript{17}

- For low fields and temperatures, and thus large $J < J_c$, relaxation is dominated by thermally activated hopping of individual flux lines, and $\mu = 1/7$ is predicted;
- In intermediate fields, relaxation occurs via jumps of small flux bundles, with the bundle size smaller than the London penetration depth $\lambda$, predicting $\mu = 3/2$;
- For higher fields, the bundle size becomes much larger and $\mu = 7/9$.

Collective relaxation of a 2D vortex system was considered by Vinokur \textit{et al.}\textsuperscript{18} with similar results to the above. The power-law exponents derived
become $\mu = 7/4, 13/16$ and $1/2$ for small, medium and large bundle regimes, respectively.

### 6.1.3.2 Vortex-Glass Theory

The concept of a vortex-glass was introduced in Section 1.4.2, resulting from the disordering of a FLL by a randomly distributed system of pinning centres. A model for the flux dynamics was developed with this vortex-glass concept as a starting point.\[^{19}\] Similar expressions for magnetic relaxation and the current dependence of the activation barrier were obtained as in the collective pinning model.

### 6.1.3.3 Empirical Models

An interpolation formula was developed, which can account for the majority of the above current dependencies,\[^{20}\]

$$U_{\text{eff}}(J,H,T) = \frac{U_c(H)}{\mu} \left[ \left( \frac{J_c(T)}{J} \right)^\mu - 1 \right], \quad (6.17)$$

where $U_c(H), J_c(T)$ and $\mu$ are fitting parameters, and $U_c(H)$ incorporates the field dependence of $U_{\text{eff}}$. With the inclusion of temperature scaling as described above and $U_{\text{eff}}(J,H,T_0)$ as defined in Eq. (6.11), Eq. (6.17) becomes
Chapter 6: Magnetic Relaxation

\[ U_{\text{eff}}(J, H, T_0) = \frac{U_c(H)}{\mu} \left[ \left( \frac{J_c(T_0)}{J} \right)^\mu - 1 \right]. \quad (6.18) \]

For \( J_c(T) \gg J \), Eq. (6.17) reduces to the form predicted by the vortex-glass and collective creep theories, Eq. (6.16). In the limit of \( \mu \to 0 \), Eq. (6.17) can be reduced to a logarithmic \( J \) dependence of \( U \) as proposed by Zeldov et al.\textsuperscript{21} and even a value of \( \mu = -1 \) describes the Kim-Anderson model, Eq. (1.28). All of the models described above demonstrate that the \( \mu \) parameter is a good indicator of the dominant vortex dynamics.

6.1.4 Current – Voltage Analysis

Measurement of the current-voltage characteristics over a range of applied fields at constant temperature can also yield information on \( \mu \). In dynamic relaxation measurements, the applied field sweep rate defines an electric field \( E \) within the sample, according to Eq. (1.15), so that \( dH/dt \propto E \). A normalised dynamic relaxation rate can then be defined as\textsuperscript{9}

\[ Q(T) = \frac{d \ln J}{d \ln E}. \quad (6.19) \]

Substituting for the sweep rate in Eq. (6.7) and combining with Eq. (6.17), the following expression can be derived for \( \mu \),\textsuperscript{22}

\[ \mu = -Q(T) \left[ \frac{d^2(\ln E)}{(d \ln J)^2} \right]. \quad (6.20) \]
Thus, the parameter $\mu$ may be related to the curvature of the $\ln E$ vs $\ln J$ curves.

6.2 EXPERIMENTAL METHODS

Dynamic magnetic relaxation measurements were performed on two 0.6 wt% doped tapes, a control tape with no thermal-neutron irradiation and one with $\Phi_n = 2.25 \times 10^{19}$ m$^{-2}$. For the magnetic measurements, pieces with dimensions of approximately 4 x 3 x 0.2 mm$^3$ were cut from the original lengths of tape. The transport current – voltage ($I - V$) measurements were then conducted on the remaining length of tape.

The magnetic measurements were performed in an Oxford Instruments Vibrating Sample Magnetometer (VSM). The system can obtain magnetic hysteresis loops with a maximum field sweep rate of 1.2 T/min and at a sensitivity of $2 \times 10^{-6}$ emu. The samples were measured within the cryostat previously described in Section 5.1, which has a lower temperature limit of 4.2 K. As in the resistive transition measurements, to avoid fluctuations of the temperature, the gas flow was controlled manually and the temperature was controlled automatically via the evaporator. Despite this, the stability of the temperature was not good at $T < 15$ K, but above 15 K stability better than ±0.1 K was achieved. Thus,
MHL were limited to $T > 15$ K. The samples were zero-field cooled to the measurement temperature and given time to reach thermal equilibrium before measurements were begun.

Field inhomogeneities can be a problem for magnetic moment measurements. Movement of a superconductor sample through a non-uniform magnetic field can cause the sample to undergo a minor hysteresis loop. Many such loops can cycle the magnetic state to its reversible limit. However, in the VSM this effect is not of major concern. Measurements of the magnetic moment are performed by vibrating the superconductor sample between a set of pick-up coils, where the moment is calculated from the voltage induced in the coils. The amplitude of the sample movement was 1.5 mm at a frequency of 45 Hz. Homogeneity of the field was better than $1 \times 10^{-4}$ of its maximum value within a 1 cm diameter at the measurement point. As the tape dimensions and the amplitude of the vibrations were smaller than this, the field can be assumed to be homogeneous throughout the sample even when accounting for the vibrations.

Measurements were originally begun on a Physical Properties Measurement System (PPMS). Problems were encountered, though, with an inconstant field sweep rate, which was not an issue on the VSM. Further, the sample in the PPMS moves over several centimetres upon a
measurement, experiencing a field inhomogeneity that is likely to affect the relaxation measurements. Thus, those early data sets have been subsequently discarded, and only the VSM data is considered.

The total moment measured by the VSM is a sum over all the microscopic circulating current paths within the superconductor sample, and thus the calculated $J$ is a volume average. Different relaxation rates in separate regions will affect the averaged behaviour, but it is not possible to distinguish between the regions using this method. This is particularly true for HTS tape samples such as the Ag/Bi2223 tapes considered here, which are composed of a multitude of interconnected grains. Thus, it may not be possible to gain any direct insight into the precise flux dynamics as a result of relaxation measurements, but a direct comparison between virgin and fission-damaged tapes can still elucidate any changes in the underlying processes.

As explained above, the current density is estimated from the irreversible magnetisation $M_{irr}$ calculated from the MHL. The measurements of the current density are therefore limited to fields below the irreversibility line, above which $M_{irr}$ is immeasurably small. In the non-irradiated sample, the irreversibility line lies close to 1 T at $T = 60$ K. Therefore, analysis for measurements performed in applied fields close to 1 T would be restricted to $T \leq 55$ K, which would impose a severe limit on
the range of \( J \) available for the \( U_{\text{eff}}(J) \) analysis. Time constraints on the availability of the equipment, see Chapter 7, also forced the measurement length time to be kept low. Thus, the hysteresis loops were only measured to a maximum field of \( \pm 1 \) T, to enable data analysis above 55 K and a larger range of \( J \).

The hysteresis loops were recorded in magnetic fields applied parallel to the crystallographic \( c \)-axis. The hysteresis loops were repeated at magnetic field sweep rates of 0.05, 0.1, 0.25, 0.5, 0.75 and 1.0 T/min and within a temperature range 15 – 70 K.

The transport \( I – V \) characteristics were measured using the standard four-probe dc method and equipment discussed in Chapter 4. Magnetic fields up to 0.6 T were applied, also parallel to the crystallographic \( c \)-axis. The measured \( I – V \) were corrected for parallel silver conduction\(^{24}\) as explained in Chapter 4.

### 6.3 Experimental Results and Discussion

Similar to the limitation of measurements to fields below \( H_{\text{irr}} \), the analysis of the relaxation data is limited to fields well above the field of full penetration \( H_p \).\(^{11}\) The rate equation of Eq. (6.3) is derived assuming complete flux penetration. \( H_p \) can be estimated from the field of minimum
magnetisation $H_m$ of the virgin hysteresis curve, the first leg of a hysteresis loop begun at $M = 0, H = 0$. The field of full flux penetration is approximately given by $1.5H_m$.\(^{10}\)

In the irradiated sample, $H_p$ lies above 0.15 T for $T < 30$ K. Thus no analysis is performed for fields below 0.15 T where the available temperature range for analysis is severely limited. Therefore, including the restrictions from $H_{irr}$ discussed above, analysis of the dynamic relaxation data is restricted to fields $0.15$ T $\leq \mu_0 H < 1$ T. The analysis was hence conducted at applied fields of 0.15, 0.3, 0.5 and 0.65 T, with extra analysis at 0.8 T for the irradiated tape. In addition, the data for temperatures below 30 K are not included in the analysis at $\mu_0 H = 0.15$ T for the irradiated tape.

**6.3.1 Standard Maley's Method**

In applying Maley's method, Eq. (6.7), to the relaxation data it was difficult to find a single value of additive constant $C$, Eq. (6.9), which could provide smooth continuity of $U_{eff}$ over the entire temperature range. To line up isotherms in the low $T$ range, values of $C \sim 15$ were required. However, this left the higher temperature isotherms extremely misaligned. To compromise in the quality of fit between the high and low $T$ ranges, values of $C \sim 40$ were needed at low applied magnetic fields, and $C \sim 30$ in higher fields.
It becomes apparent that these values for $C$ are too large when Eq. (6.9) is rearranged to give

$$\frac{v_0}{d} = \frac{1}{2} e^C,$$  \hfill (6.21)

where $v_0 = a v_0$ is the flux hop velocity. From the literature, values of $C$ between 10 and 20 and $v_0 \approx 10 \text{ ms}^{-1}$ are expected. A value of $C = 15$ would result in an estimate of $a v_0/d = 1.6 \times 10^6 \text{ s}^{-1}$. In comparison, the values of $C = 30$ and 40 observed here result in $a v_0/d = 5.3 \times 10^{12}$ and $1.2 \times 10^{17} \text{ s}^{-1}$, respectively, which are both many orders of magnitude larger than physically viable.

Hence, the above experimentally derived values for $C$ of 30 and 40 are unacceptably large. These would also result in an unexpectedly strong field dependence of $C$. Consequently, we consider the standard Maley's method to yield physically unreasonable values for $C$, in agreement with McHenry et al.

### 6.3.2 Temperature Scaling

The results obtained after analysis with the modified form of Maley’s method as outlined in Section 6.1.2 above are shown in Tables 6.1 and 6.3.
6.3.2.1 Theoretically Predetermined $U(T)$

Table 6.1 contains the results found from an application of the first scaling approach, as outlined in Section 6.1.2.1, using the temperature scaling form given in Eq. (6.13). Since the continuity fit for each isotherm was determined by eye, values of $C$ are only accurate to ±1.

A continuous $U_{\text{eff}}(J,H,T_0)$ was found at each applied field in both samples, as illustrated in Fig. 6.1. However, the isotherms did not always fit in a smooth manner, and in some cases, the slopes of neighbouring isotherms were slightly offset. To compensate for the misalignment in one set of isotherms, $C$ could be altered, but usually with the effect of bringing others out of line. Changing the form of $U(T)$ was also tried, but with similar problems. Using the form in Eq. (6.14) and again altering $C$, it was just as difficult to distinguish a good fit and in most cases, especially in low fields, the results were even more disjointed. The result shown is the best compromise that could be determined by eye.

<table>
<thead>
<tr>
<th>TABLE 6.1</th>
<th>$C$ AND $\mu$ FITTING PARAMETERS FOR FIT TO EQ. (6.18), OBTAINED WITH $U(T) = (1-t)^{3/2}$.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.6-wt% UO$_2$, nonirradiated</td>
</tr>
<tr>
<td>$H$ (T)</td>
<td>0.15</td>
</tr>
<tr>
<td>$C$</td>
<td>12</td>
</tr>
</tbody>
</table>
When comparing changes in $U_{\text{eff}}$, it is important to restrict comparisons to those at equivalent $J$. The current density is directly affected by changes in the relaxation rate, which is determined by $U_{\text{eff}}$. However, the effective activation energy is itself a function of the current density. Thus, comparing $U_{\text{eff}}$ at the same current density, the effect of $J$ on $U_{\text{eff}}$ can be disregarded. Compared at a fixed applied field and an equivalent $J$, the effective activation energies after introduction of fission-fragment damage are approximately 50% larger than those of the nonirradiated tapes, over the entire field and current density range.
6.3.2.2 Empirically Determined $U(T)$

The slopes of the isotherms in $U(J)$ are much better aligned if a discrete temperature scaling factor $G$ is chosen for each isotherm, as discussed in Section 6.1.2.2. With this method, it was possible to obtain a very smooth $U(J)$ curve, with the slopes of each of the neighbouring isothermal $U(J)$ matching closely, see Fig 6.2. Such smooth transitions between the isothermal $U(J)$ were not obtainable with a predetermined form of $U(T)$ (Fig. 6.1). Consequently, we will focus our discussion on the data obtained by the empirical scaling approach.

The data set of $G$ determined at each temperature in this fitting procedure can be plotted as a function of temperature, where an empirical expression for the functional form of $U(T)$ can be fit to the data set. Figure 6.3 displays the empirically determined temperature scaling data set $G(T)$, along with a curve fit to Eq. (6.15). The resulting fitting parameters are shown in Table 6.2, accurate to approximately ±3%.

<table>
<thead>
<tr>
<th>TABLE 6.2: FITTING PARAMETERS OBTAINED FROM A FIT OF THE EMPIRICALLY DETERMINED DATA SET $G(T)$ TO THE FORM IN EQ. (6.15).</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H$ (T)</td>
</tr>
<tr>
<td>m</td>
</tr>
<tr>
<td>n</td>
</tr>
<tr>
<td>a</td>
</tr>
</tbody>
</table>
Chapter 6: Magnetic Relaxation

**Fig. 6.2:** An example of the close matching of individual isotherms using an empirical \( U(T) \), showing a section of the \( U_{\text{eff}}(J) \) curve obtained for the nonirradiated tape at an applied field of 0.65 T.

**Fig. 6.3:** Data set of the scaling factor \( G \) at each temperature, for both tapes. The errors are approximately the size of the symbols or smaller. The lines are the fits to Eq. (6.15).
The empirically determined temperature scaling forms for both tapes can be seen to fall into two broad groups. At low applied fields, the scaling forms are very close to the theoretically determined form \( U(T) \approx (1-t)^{3/2} \) (Eq. (6.13)) within the specified uncertainty. Only the form of \( U(T) \) at \( \mu_0 H = 0.3 \) T for the irradiated tape is difficult to account for, with \( n = -1.70 \pm 0.05 \) being larger than the expected \( n = -1.50 \).

There is a significant change in the functional form at applied fields \( \mu_0 H \geq 0.5 \) T in the nonirradiated tape and similarly in fields \( \mu_0 H \geq 0.65 \) T in the irradiated tape. At these fields, \( U(T) \) shows a remarkably different behaviour than at low fields, following a form similar to that of Eq. (6.14). Changes in \( U(T) \) are thus more obvious when the temperature scaling term is derived directly from the data, rather than applying a term determined \textit{a priori}.

It is generally found in the literature that a predetermined \( U(T) \) is often chosen in the form of either Eq. (6.13) or (6.14).\(^{11,14}\) The empirically determined \( U(T) \) for both the irradiated and nonirradiated tapes can also be successfully fitted with these two equations. However, it is necessary to stress that the nature of the temperature scaling function obtained from magnetic relaxation is determined by the type of vortex dynamics. That is, \( U(T) \) may depend not only on \( \lambda \) and \( H_c \), but also on the dimensionality of
the vortices, type of pinning centers, interlayer vortex coupling and many other factors. Therefore, Equations (6.13) and (6.14), which are derived by taking into account only the temperature dependence of $\lambda$ and $H_c$ are not necessarily indicative of a fundamental description of any temperature dependencies of the magnetic relaxation data. The successful fit of Equations (6.13) and (6.14) to the empirically obtained $U(T)$ might only result from the fitting procedure being performed over a range of temperatures that is too narrow to discern reliably the accuracy of the fitting. Consequently, the apparently good fit of $G(T)$ with these equations is merely coincidental and no conclusions based on their derivation are applied to the data.

Table 6.3 summarises the results for $U_{\text{eff}}$ obtained from Eq. (6.7) and the empirical temperature scaling approach. For the irradiated tape, the additive constant $C$ is not significantly different to that determined using the a priori temperature scaling, as listed in Table 6.1, for applied fields

<table>
<thead>
<tr>
<th>TABLE 6.3: APPROXIMATE TEMPERATURE SCALING FORM $U(T)$ FROM THE FIT IN FIG. 6.3, AND THE ASSOCIATED C AND $\mu$ FITTING PARAMETERS.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>H (T)</strong></td>
</tr>
<tr>
<td><strong>C</strong></td>
</tr>
<tr>
<td><strong>U(T)</strong></td>
</tr>
<tr>
<td><strong>$\mu$</strong></td>
</tr>
</tbody>
</table>
\[ \mu_0 H \leq 0.5 \text{ T}. \] C is increased by only slightly more than the margin of error (±1) for the nonirradiated tape in applied fields \( \mu_0 H = 0.15 \) and \( 0.3 \) T.

The values of additive constant \( C \) determined by the empirical scaling approach are very similar at the lowest applied fields between both the irradiated and nonirradiated tapes. The minor difference may be due to the problem of determining \( C \) by eye. This similarity is an indication that \( 2a\nu_0/d \) is very similar in both tapes at these applied fields. Since both tapes were constructed using the same processing, with the exception of the uranium doping, it can be reasonably expected that the characteristic sample dimension \( d \) will be very similar for both tapes. Thus, the flux hop velocity \( \nu_0 \approx a\nu_0 \) is very similar in both the irradiated and nonirradiated tapes at low applied fields \( \mu_0 H \leq 0.3 \) T. As was discussed above, values of \( C \approx 15 \) as observed here result in an estimate of \( a\nu_0/d \approx 1.6 \times 10^6 \text{ s}^{-1} \). Assuming \( d = 35 \) µm (average core thickness for the tapes), this result gives a reasonable estimate for the flux hop velocity: \( \nu_0 \approx 55 \text{ ms}^{-1} \), in reasonable agreement with expectations from the literature.\(^7\),\(^26\)

In applied fields \( \mu_0 H \geq 0.5 \) T in the nonirradiated tape, and \( \mu_0 H \geq 0.65 \) T in the irradiated tape, however, \( C \) is significantly larger than in lower applied fields, and also larger in comparison to the value resulting from the theoretically determined temperature scaling approach. This observed increase in \( C \), which is correlated with the change in the form of
\( U(T) \), represents a large change in the flux hop velocity as a function of applied field. The increase to \( C = 19 \) results in \( a\nu_0/d \approx 8.9 \times 10^7 \text{ s}^{-1} \), less than two orders of magnitude larger than the low field estimate, which is still an acceptable estimate, especially in comparison to the results from the standard Maley's method, above.

Figure 6.4 displays the resulting \( U_{\text{eff}}(J,H,T_0) \) curves obtained using the empiric temperature scaling, in a log-log plot. A comparison at both extremes of the applied field analysed (0.65 and 0.15 T) reveals that, after introduction of fission-fragment damage, the effective activation energies are increased to approximately twice those of the control tape without fission-induced defects. This difference is at a maximum of 2.3 times for \( J \) close to zero, and reduces as \( J \) approaches \( J_c \), with a minimum change in \( U_{\text{eff}} \) of approximately 1.5 times at the upper limit of \( J \) measured.

The difference in \( U_{\text{eff}} \) is much more pronounced at an applied field of 0.5 T, reaching almost 15 times the nonirradiated \( U_{\text{eff}} \) as \( J \to 0 \). This is a result of the different limits of \( U_{\text{eff}}(J,H,T_0) \) as \( J \) approaches zero that are observed in different applied fields and which can be better perceived by re-plotting Fig. 6.4 in a linear-log format, Fig. 6.5. In low applied fields, \( U_{\text{eff}} \) diverges as \( J \to 0 \), as predicted by the collective creep and vortex-glass theories for an elastic FLL, Section 6.1.3. This is distinguished from the
Fig. 6.4: $U_{\text{eff}}(J,H,T_0)$ for the irradiated and nonirradiated tapes with the empirically determined $U(T)$ as shown in Fig. 6.3. The lines are fits to Eq. (6.18).

high field behaviour, where $U_{\text{eff}}$ can be seen to approach a constant value $U_0$ as $J \to 0$, characteristic of creep by plastic shear deformations.

The crossover from elastic to plastic creep is in fact correlated with the empirically determined form of $U(T)$. A diverging $U_{\text{eff}}$ is found for all applied fields where $U(T)$ is approximately described by $(1-t)^{3/2}$, whereas a form of $(1-t^2)$ defines a $U_{\text{eff}}(J,H,T_0)$ curve that approaches a constant value for $J \to 0$. In the nonirradiated tape, the crossover from divergent to non-divergent behaviour is found to occur at $\mu_0 H = 0.5$ T, but it occurs at a higher applied field, $\mu_0 H = 0.65$ T, in the irradiated tape. Therefore, at an applied field of 0.5 T, the current dependencies of the two tapes differ, and
hence the diverging activation energy of the irradiated tape deviates to a large degree from the activation energy of the nonirradiated tape as $J \to 0$.

In an intermediate field $\mu_0 H = 0.3$ T, the difference is larger than the average increase (excluding the unique situation at 0.5 T discussed above), with $U_{\text{eff}}$ of the irradiated tape approaching 6 times that of the nonirradiated tape at $J \sim 0$. The behaviour of $U_{\text{eff}}$ with current density for the irradiated tape at this applied field is also observed to be quite different to the rest. As $J$ approaches zero, $U_{\text{eff}}$ at 0.3 T reaches, and appears to surpass, the effective activation energy in the lower applied field $\mu_0 H = 0.15$ T.
**Fig. 6.6:** $U_{\text{eff}}(J,H,T)$, from Fig. 6.4, of the (a) irradiated and (b) nonirradiated tapes, scaled by $H^{1/2}$
The difference in current density dependence is further highlighted when considering the scaling of the effective activation energies with applied field. $U_{\text{eff}}(J,H,T_0)$ was found to scale with $H^{-1/2}$ for both the irradiated and nonirradiated tapes over all applied fields, as shown in Fig. 6.6. The individual $U_{\text{eff}}(J,H,T_0)$ curves scale onto two common curves, again divided according to the form of the empirically determined temperature scaling term. As mentioned in Section 5.3.2, a square-root field dependence of the activation energy is associated with thermally activated \textit{plastic} shear deformations of a 3D flux lattice or 3D viscous flux liquid.27 This is in contradiction to the evidence from the divergence of $U_{\text{eff}}$, above, which suggests a change from elastic to plastic pinning as a function of applied field.

The only exception to the field scaling was for an applied field of 0.3 T in the irradiated tape, which could not be scaled in line with any of the other curves. The dependence of $U_{\text{eff}}$ on $J$ at this field is distinct to the $J$ dependence at any other applied field that was analysed. This may be construed as evidence of a matching field $B_{\text{eff}}$ close to 0.3 T.

Since $B_{\text{eff}} \propto B_\Phi$ is simply a relation between the vortex density and the columnar pin density, it can be reasonably expected that it is independent of temperature. Thus, assuming that $B_{\text{eff}}$ is directly proportional to $c\Phi_n$, and with $B_{\text{eff}} \sim 0.3$ T for a (0.6, 1.75) tape, $B_{\text{eff}}$ can be
estimated as \( \sim 0.39 \) T for a (0.6, 2.25) tape. Considering the broadened transition in the field dependence of \( U_0 \) observed close to \( B_\phi \), this allows for matching field effects to be observed in fields close to 0.3 T. Pinning is expected to be much more efficient at \( B_{eff} \), hence the peak in \( U_0^{irr}/U_0^{vir} \) in Section 5.2.2.1. Therefore, it can be reasonably expected that, at fields close to the matching field, \( U_{eff}(J,H,T_0) \) will be enhanced in comparison to lower applied fields, especially as \( J \rightarrow 0 \), thus explaining the observed behaviour at 0.3 T.

The current density dependence can be better characterised by fitting the experimental \( U_{eff}(J,H,T_0) \) curves with the functional form of Eq. (6.18) and examining the resulting values of the fitting parameter \( \mu \).

### 6.3.3 Field Dependence of \( \mu \)

The values of \( \mu \) obtained after curve fitting the experimentally determined \( U_{eff}(J,H,T_0) \) with the form in Eq. (6.18) are given in Tables 6.1 and 6.3 for both approaches to the temperature scaling, and are plotted against applied field in Fig. 6.7. It can be seen that the resulting \( \mu \) values are a decreasing function of applied field, with an apparent crossover from positive to negative \( \mu \) values at a field \( H_{cr} \). Using the empirically determined \( U(T) \), this crossover occurs at \( \mu_0H_{cr} \approx 0.37 \) T for the nonirradiated tape and approximately 0.65 T after irradiation. \( H_{cr} \) is
slightly higher when scaling is performed with a predetermined $U(T)$, but the transition of $\mu$ is evident regardless of the particular technique used for determining $U(T)$.

As noted above, changes in the divergence of $U_{\text{eff}}$, and hence the vortex dynamics, are more obvious when the temperature scaling term is derived directly from the data, rather than by applying a term a priori. The crossover to negative $\mu$ is then observed to occur simultaneously with the changes in the divergence of $U_{\text{eff}}$, as well as in the form of $U(T)$ and the value of $C$. These features all appear to be correlated. It is possible that the changes in $C$ and $\mu$ are artifacts of the empirical scaling procedure, as they

![Graph](image)

**Fig. 6.7:** Field dependence of $\mu$ from a fit to Eq. (6.18) for both the theoretically and empirically determined temperature scaling forms $U(T)$. Lines are guides to the eye only. The arrows indicate the approximate position of the crossover field $H_{cr}$.  

195
both are determined by the scaled behaviour of $U_{\text{eff}}(J)$. However, the empirically determined temperature scaling term relies directly on the measured relaxation data, thus it is expected to reflect a true physical behaviour. Additionally, this is the only way to obtain a smooth $U_{\text{eff}}(J)$ curve over a wide enough range of $J$ to make the data analysis meaningful. The changes in $U(T)$, $C$ and $\mu$ are expected to occur simultaneously for all three parameters because this change reflects a transition between two different vortex states. All of this points to the superiority of this approach.

### 6.3.4 3D - 2D Transition

The shift from positive to negative $\mu$ has been proposed to be an indication of a transition from 3D elastic creep to a 2D plastic creep regime\(^{29}\). As discussed in Section 1.3.2.2, in anisotropic HTS, such as Bi\(_{2223}\), it has been established that there is a dimensional crossover in the vortex structure when either magnetic fields or thermal fluctuations destroy the Josephson coupling between vortices in adjacent layers.\(^{30}\) At low temperatures, the theoretical criterion for this field induced 3D to 2D transition is given by Eq. (1.12),\(^{30}\)

$$\mu_0 H_{cr} \propto \frac{\Phi_0 \epsilon^2}{s^2}. \quad (1.12)$$

It has been proposed\(^{29}\) that the field at which $\mu$ changes sign can be used as an estimate of $H_{cr}$. Studies of the dynamic magnetic relaxation in
Tl$_2$Ba$_2$CaCu$_2$O$_8$ thin films revealed negative values of $\mu$, defined as in Eq. (6.18), that were associated with a vortex-glass temperature $T_g = 0$. The $T_g(H)$ defines the temperature at which thermal fluctuations force the vortex-glass phase to undergo a transition into a vortex liquid phase. A characteristic feature of a 2D system is that the vortex-glass cannot exist at a finite $T$. The barriers to plastic deformation in the 2D state remain finite as $J \to 0$, and hence so does the resistivity, limiting the glassy behaviour in 2D. Therefore, the association of $T_g = 0$ with a negative $\mu$ is taken as an indicator of a 2D state.

The change in sign of $\mu$ observed thus suggests a 3D-2D transition in the system at $\mu_0H_{cr} \sim 0.37$ T in the nonirradiated tape and approximately $0.65$ T after irradiation. Due to a lack of exact data on the anisotropy ratio in Ag/Bi2223 samples, the theoretical crossover field can only be estimated. Blatter et al. estimate $\mu_0H_{cr} = 0.36$ T. The value of $\mu_0H_{cr} \approx 0.37$ T observed here from the relaxation measurements on the nonirradiated tape is remarkably close to this estimate.

The substantial changes observed in $U(T)$ at $\mu_0H_{cr}$ may be consistent with a de-coupling of the pancake vortices in neighbouring layers. Clem demonstrated that the inter-layer magnetic coupling between pancake vortices, in the limit of zero Josephson coupling, could be destroyed by thermal excitations even at quite low temperatures. The dimensional
Chapter 6: Magnetic Relaxation

crossover requires a large reduction in the Josephson coupling, so that in
the 2D state, above $\mu_0 H_{cr}$, the limit of zero Josephson coupling may apply.
Below $\mu_0 H_{cr}$, where the Josephson coupling is strong, the dependence on
thermal excitations is not as great at such low temperatures as for the 2D
state. Thus, at low temperatures, the dependence of the vortex dynamics on
thermal excitations will be greater in the 2D regime than in the 3D regime.

Similarly, the change in $C$ observed at $\mu_0 H_{cr}$ may also be consistent
with a pancake de-coupling transition. The viscosity of vortex motion
depends on the pinning and interaction of the flux vortices. In a crossover
to 2D pancakes, the effectiveness of pinning centres on vortices in
neighbouring layers is reduced. The weak interlayer vortex interaction
also allows vortices in one layer to slip past pinned vortices in adjacent
layers. This leads to a reduction in the viscosity of the vortex pancake
motion. The reduced viscosity allows a faster hop velocity, which will lead
to a commensurate increase in $C$.

The change in the divergent behaviour of $U_{eff}$ as $J \rightarrow 0$ may also be
explained from a dimensional crossover. In the 2D state, the diverging
activation energy barriers predicted for the 3D elastic FLL are not
expected. Above $H_{cr}$, rather, creep is dominated by the plastic shear of
dislocations, which have a finite barrier against creep even as $J \rightarrow$
Thus the behaviour of $U_{\text{eff}}(J,H,T_0)$ as $J \to 0$ above and below the crossover field, Fig. 6.6, is consistent with a dimensional crossover.

The shift in $\mu_0H_{cr}$ to higher applied fields after irradiation signifies that the 3D vortex-glass regime is extended to higher applied fields by the fission-fragment damage centres. Restoring the 3D state by significantly increasing the inter-layer vortex correlation can restore the finite $T_g$. The glassy phase is restored, and will revert to the liquid phase at a higher $H_{cr}$. Hence, it is apparent from these relaxation measurements, that splayed defect pinning promotes the $c$-axis vortex correlation of Ag/Bi2223 tapes.

### 6.3.5 Current – Voltage Results

An independent measurement of the field dependence of $\mu$ was sought to confirm the behaviour observed from the relaxation measurements. The field dependence of $\mu$ is related to the curvature of $\ln E - \ln J$ curves through Eq. (6.20). Figure 6.8 (a) and (b) shows the resulting $\ln E - \ln J$ curves at 77 K for the nonirradiated and irradiated tapes, respectively. A crossover from negative to positive curvature can be identified in both tape samples as the applied field is increased, at values of 0.08 T and 0.44 T in the nonirradiated and irradiated tapes, respectively.

The crossover in curvature is generally identified as the vortex-glass to liquid transition at $(H_g, T_g)$, with $T_g = 77$ K in these measurements. It
Fig. 6.8: Current – Voltage characteristics for the (a) nonirradiated and (b) irradiated tapes. Note the change in curvature evident above and below the dashed lines.
cannot be immediately interpreted as a dimensional crossover into a 2D state, as it is possible to undergo a transition into the liquid phase while still maintaining \( c \)-axis vortex coherence; that is, if \( H_g < H_{cr} \), in a situation similar to that described in Section 1.3.2.3. However, in the situation where the vortex-glass to liquid transition would occur at fields \( H_g > H_{cr} \), the transition is limited by the crossover to the 2D state, which has \( T_g = 0 \). Thus, the observed increase in the transition is only possible if \( H_{cr} \) is also increased.

The transition in the curvature of \( \ln E - \ln J \) is consistent with the dimensional crossover, despite the values of \( H_{cr} \) derived from the magnetisation measurements being higher than the \( H_g \) derived from the \( \ln E - \ln J \) curves. From fluctuation theory,\(^{35}\) the temperature dependence of the crossover field, due to thermal fluctuations of the pancake vortices, is approximately given by

\[
\mu_0 H_{cr} \propto 1/T. \tag{6.22}
\]

The dynamic relaxation measurements are scaled to a temperature \( T_0 \approx 20\)K, determined by the lowest temperature used for the scaling analysis. The \( I - V \) curves, on the other hand, are measured at \( T = 77 \) K. We thus expect a difference of \( \sim 1/4 \) if both measurements represent the field of dimensional crossover. The relative difference between the two measurements on the nonirradiated samples is \( \sim 0.22 \), in good agreement
with these expectations. As a result of the strong columnar pinning, the fluctuation theory is not entirely applicable to the irradiated tape samples without alteration, where the difference is observed to be $\sim 0.6$. However, this limits the dimensional crossover to $H_{cr} \geq 0.44 \, \text{T}$ at $77 \, \text{K}$.

Consequently, the crossover in the $\ln E - \ln J$ curvature is equated with a change in the sign of $\mu$. The field at which the crossover occurs at $77 \, \text{K}$ is thus shifted from $\mu_0 H_{cr} \approx 0.08 \, \text{T}$ in the nonirradiated tape to $\mu_0 H_{cr} \approx 0.44 \, \text{T}$ after irradiation. The $I - V$ measurements therefore also indicate a shift in $H_{cr}$ to higher applied fields as a consequence of the fission-induced damage, confirming that the effect of the randomly splayed, quasi-columnar defect pinning is to drive the applied field at which the $3D - 2D$ transition occurs to higher fields.

### 6.3.6 Dimensionality of an Irradiated Tape under the Standard Maley's Method

To further highlight the consistency found with the empirical temperature scaling method, these results can be compared to those obtained in the absence of temperature scaling. The sign of $\mu$, and thus the deduced dimensionality of the system, is found to be altered radically without the application of a temperature scaling procedure. Over the entire
range of applied fields, even at $\mu_0 H = 0.15$ T, a fit to Eq. (6.18) results in $\mu < 0$ for the irradiated tape.

The results obtained with the standard Maley's method therefore suggest that the vortex system in the irradiated tape is in a 2D plastic creep regime over all applied magnetic fields. This implies the complete destruction of the $c$-axis vortex correlation by the introduction of randomly splayed columnar pins, hence the drastic reduction in $\mu_0 H_{cr}$. The loss of vortex correlation could possibly be explained as a result of vortex cutting\textsuperscript{13d} by the highly splayed columnar pins. This would, however, reduce the effects of the pinning to that of random point disorder,\textsuperscript{13b} inconsistent with the observed improvements in $J_c$ and its field dependence as well as the increases in the resistively determined $U_0$, and the magnetically determined $U_{eff}(J)$ (see Ref.\textsuperscript{38} and the preceding Chapters). The increase in $T_g$ obtained after irradiation from $I-V$ measurements would also contradict a loss of vortex correlation. Otherwise, it would imply that $T_g$ is finite in the 2D vortex state, when compared to the lower $T_g$ in the nonirradiated tape.

As was discussed in Chapter 2, in less anisotropic Y123 heavy-ion irradiation with large splay angles was observed to result in the loss of $c$-axis vortex coherence.\textsuperscript{13} Work with Hg cuprates demonstrated, however, that uniformly splayed defects provide much stronger improvement of
vortex pinning in superconductors with a higher anisotropy.\textsuperscript{[40]} In addition, the theory of anisotropic rescaling of the pinning landscape presented in Ref. 40 predicts the recoupling of vortex segments, and thus an increase in the dimensional crossover field, for large splay angles in a strongly anisotropic material. These factors serve to strengthen confidence in the modified form of Maley's method with an empirically determined $U(T)$.

Further, comparisons of the uranium-fission method on Y123, Bi2212 and Bi2223 samples revealed that pinning enhancements were greater in the more highly anisotropic BSCCO superconductors, particularly Bi2223.\textsuperscript{[41]} At the time, the authors were unable to provide any structural evidence to explain the differences. Therefore, based on the observations obtained in this Thesis of the increase in the dimensional crossover, and hence the $c$-axis vortex correlation, in fission-damaged Bi2223, it becomes clear that the difference lies in the lower anisotropy of the Y123.

### 6.3.7 Related Issues

The observation from the divergent behaviour of $U_{\text{eff}}(J,H,T_0)$ as $J \to 0$ of the extension of elastic creep to higher applied fields with fission induced pinning centres is in agreement with the behaviour of $F_p$ determined from the $J_c$ measurements (Section 4.2.3). It may also indicate
that the basic idea of the elastic pinning theory, the competition between elastic and plastic creep with \((H_{\text{max}}, F_{p\text{max}})\) defined at the balance point, is still essentially correct for strong columnar pinning. The difference in the divergent behaviour of \(U_{\text{eff}}\) between the irradiated and nonirradiated tapes reveals that the elastic creep behaviour is extended to higher fields by the fission-fragment damage. The increase in \(H_{\text{max}}\) to higher fields can likewise be interpreted as a shift in the balance between elastic and plastic vortex creep to higher applied fields from the elastic pinning theory.\(^{42}\)

As measurements are taken over a wide temperature range, it might be expected that a thermally induced crossover in the \(U(J)\) dynamics at a fixed applied field would be evident, characterised by a change in the empirical temperature scaling and \(\mu\) at high and low \(T\) isotherms. This is not observed within these results, however.

One possible reason is that the procedure for finding the functional form of \(U(T)\) may have been performed over an inadequate temperature range to discern a change. Perhaps there are an insufficient number of data points to distinguish the changing behaviour, and further isotherms are required. The functional form obtained by a fit of \(G(T)\) describes the overall temperature dependence of the data set of \(G\), and so reflects the scaling form that is dominant over a majority of the temperature range. With a larger number of data points, a change in scaling behaviour may be
distinguishable before the fitting procedure is undertaken, allowing the fitting form to be altered for high and low $T$ ranges.

6.4 Conclusion

Dynamic magnetisation relaxation measurements were conducted on a 0.6 wt% uranium-oxide doped, nonirradiated sample and a 0.6 wt% doped sample irradiated to $\Phi_n = 2.25 \times 10^{19} \text{ m}^{-2}$. The data were analysed using a modified form of Maley's method with an empirically determined temperature scaling, which revealed a field-induced crossover in the vortex dimensionality. This dimensional crossover was inferred from the correlated changes in:

- the empirical temperature scaling form $U(T)$;
- the additive constant $C$; and
- the fitting parameter $\mu$.

Additionally, the divergent behaviour of $U_{\text{eff}}$ as $J \rightarrow 0$ is consistent with the dimensional crossover.

The applied field $H_{cr}$ at which the crossover occurs is increased by the introduction of randomly splayed quasi-columnar fission-fragment defects. Analysing the data using the standard Maley's method without any temperature scaling, however, produced a conflicting result, where the
dimensionality remains in a 2D state over the entire field range even after introduction of the strongly pinning fission-induced defects.

The independent \( \ln E - \ln J \) measurements demonstrate a strong agreement with the results obtained using the modified form of Maley's Method. Together with the consistency observed with the empirically determined \( U(T) \), as well as the unreasonable values of \( C \) obtained from analysis without the temperature scaling, the results indicate that the inclusion of the empirical temperature scaling is an essential part of the relaxation analysis. Further, the standard Maley’s method cannot provide a smooth \( U_{\text{eff}}(J) \) over the entire temperature range measured.

These observations therefore support the conclusion that the 3D–2D dimensional crossover is shifted to higher applied fields after the introduction of fission-fragment damage. Hence, the randomly splayed quasi-columnar pins created by the uranium-fission process promote the \( c \)-axis vortex correlation in the Ag/Bi2223 tapes.

References

Chapter 6: Magnetic Relaxation

7.1 Conclusion

The uranium-fission method has demonstrated substantial improvements in both $J_c$ and $H_{irr}$ at liquid nitrogen temperature. These improvements are observed to increase with the density of fission fragments, but are offset by a loss in the zero-field $J_c$ caused by the uranium-oxide doping process, the fission-fragment damage and even high levels of neutron fluence. Combined with the thermal-neutron induced Ag radioactivity, the application of this method to commercial materials requires careful consideration of the balance between the levels of uranium doping and thermal-neutron fluence.

In Chapter 2, the effect of fission-fragment damage on the activation energies determined from resistive transition and magnetic relaxation measurements was identified as an area that had not been adequately explored
in uranium-doped HTS, particularly Ag/Bi2223. In this Thesis, both the resistive transition and magnetic relaxation measurements confirm the significant increases in the activation energies of the pinning defects introduced by fission-fragment damage. While the two methods probe different flux creep regimes, both demonstrate the dominance of the randomly-splayed quasi-columnar defect structures in the pinning landscape, particularly in fields applied parallel to the \(c\)-axis.

One of the most important results from the resistive transition measurements in Chapter 5 was the identification of an effective matching field effect. \(B_{\text{eff}}\) was observed below the matching field \(B_{\phi}\) as it is generally calculated in the literature: as the density of fission tracks multiplied by the average defect track length. This method of calculating \(B_{\phi}\) was concluded to be an oversimplification, which does not take into account the finite elasticity of the flux lines or the anisotropy of the vortex system.

A field-induced crossover in dimensionality was observed in a virgin tape at a field \(\mu_0H \approx 0.37\) T, which is shifted to \(\mu_0H \approx 0.65\) T after the introduction of fission-fragment damage with \(c\Phi_n = 1.35\) wt% m\(^2\). The splayed quasi-columnar defects produced by the uranium-fission process thus promote \(c\)-axis vortex correlation in the Ag/Bi2223 tapes. Based on these
observations, the reported differences in the efficiency of the fission method between Y123 and Bi2223 are ascribed to the differences in anisotropy.

7.2 FURTHER WORK

An examination of various \((c, \Phi_n)\) tapes via dynamic magnetic relaxation studies was originally intended for this Thesis. Time constraints and a lack of access to the required equipment as a result of repeated severe equipment failures restricted the investigations possible. Thus, the effects of \(c\Phi_n\) on the dimensionality crossover probed through magnetic relaxation measurements on tapes with various combinations \((c, \Phi_n)\) remains as a possible avenue of research. Probing the changes with \(c\Phi_n\) will establish the dependence of \(H_{cr}\) on the defect density, which may lead to an empirical method for estimating \(H_{cr}\) from randomly splayed defect densities in Ag/Bi2223.