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Study on newly discovered iron-based superconductors

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Study on Newly Discovered Iron-based Superconductors

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ABSTRACT

Discovered in 2008, iron pnictides are the latest high temperature superconductors which have aroused enormous attention in the scientific community. The discovery of iron based superconductors (FBSs) marked the foundation of a new era in the field of superconductivity by replacing the Copper Age by the Iron Age. This discovery has given scientists the chance to study the superconducting and magnetic properties in a different family of high temperature superconductors, as understanding the nature of superconductivity in unconventional superconductor is crucial for designing new materials with higher critical temperature ($T_c$). These materials would be good candidates for use in electricity generators, cheaper medical imaging scanners, and extremely fast levitating trains because superconducting materials with higher $T_c$ would not require expensive coolants to reach the superconducting transition temperature. Therefore, the discovery of FBSs was a significant achievement in the condensed matter community.

The main focus and novelty of this work is twofold: firstly, the pinning potential, thermally activated flux flow behaviour and superconducting properties of iron based superconductors, mostly hole doped BaFe$_2$As$_2$ pnictides and arsenic free FeSe$_{1-x}$Te$_x$ chalcogenides was investigated in details. Secondly, the magnetic and transport properties of parent compound BaFe$_2$As$_2$ and non superconducting Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ was studied using magnetic, magnetoresistance and neutron diffraction measurements.

Understanding the vortex pinning mechanism in FBSs is crucial for practical applications and fundamental study due to the relatively high critical temperature,
high upper critical field ($B_{c2}$), high critical current density, very high intrinsic pinning potential, and nearly isotropic superconductivity of these compounds, and also due to the possibilities for the fabrication of superconducting wire. In order to understand the pinning mechanisms in these systems, scaling analysis of the normalized pinning force as a function of reduced field was performed. Analysis using the Dew-Hughes model has suggested that point pins alone cannot explain the observed field variation of the pinning force density. According to the collective flux pinning model, the field dependence of the magnetization shows that the flux pinning in $\text{Ba(Fe}_{1-x}\text{Ni}_x\text{)As}_2$ is dominated by the spatial variation in the charge carrier mean free path.

Irradiation has been employed in order to increase the pinning potential, and as a result, the critical current density in $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ single crystal. The $\text{C}^{4+}$ irradiation cause little change in the superconducting critical temperature, but it can enhance in-field critical current density ($J_c$) by a factor of up to 1.5, with enhanced flux jumping at 2 K. Also, the magnetic optical imaging results confirm the enhancement of $J_c$ in the irradiated samples. These results suggest that light $\text{C}^{4+}$ ion irradiation is an effective method for the enhancement of $J_c$ in FBSs compared to heavy ion irradiation and neutron irradiation.

In addition, The angular dependence of the upper critical field and the pinning potential of underdoped $\text{BaFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ single crystals have been investigated by measuring magneto-transport at different magnetic fields and angles. Furthermore, by scaling the angular dependence of the resistance, based on the anisotropic Ginzburg-Landau (GL) theory, an anisotropy value of less than 2.1 was determined for different temperatures below the superconducting transition temperatures. Based on these results, the pinning potential is strongly angle dependent for $\theta \leq 45^\circ$ and
almost angle independent for $\theta \geq 45^\circ$, while $B_{c2}$ increase monotonically with increasing angle.

Also, the thermally activated flux flow (TAFF) behaviour of arsenic free Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystals were analysed using the conventional Arrhenius relation and modified TAFF model. It was found that the Arrhenius curve slopes are directly related to, but not equal to, the activation energies of Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystals. Therefore, the use of a modified TAFF model, $\rho(T, B) = \rho_{0f} \exp(-U/T)$, is suggested, where the temperature dependence of the prefactor $\rho_{0f} = 2\rho_c U/T$ and the nonlinear relation of the thermal activation energy are considered.

Furthermore, a detailed investigation was carried out to understand the magnetic and magnetoresistance behaviour of non-superconducting Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$. It should be noted that understanding the antiferromagnetic order of iron ions itself is also important for both fundamental study and practical application. It is very interesting to design new magnetic device based on spin dependent transport properties of pnictide materials. Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ is the first doped compound with no superconductivity phase and magnetic phases is the only competitor as Cr concentration increases. Transport and magnetic measurements show an interesting two fold symmetry in non superconducting Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x = 0.303$) compound which depend on temperature and magnetic field. In order to understand the temperature and magnetic field response of iron pnictide at atomic level, neutron diffraction studies were performed for Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x=0.303$).
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CHAPTER 1
INTRODUCTION

This work was inspired by the discovery of superconductivity in iron-based superconductors, the second family of high temperature superconductors after copper-based high temperature superconductors. Finding superconductivity in compounds containing iron, a magnetic element, after two decades of intensive research on the high temperature cuprates was a surprise to the scientists in this field, as they thought that the magnetic nature of iron would disrupt the pairing of electrons in the superconducting state. Iron based superconductors and copper based superconductors reveal some similarities and differences: both have layered structure and show an unconventional paring mechanism. Also, they both have very high upper critical fields and high critical current densities. Iron based superconductors, however, possess several advantages compared to copper based superconductors: Firstly, the parent compound of iron based superconductors is semi-metallic, as opposed to the Mott insulator parent compound of copper based superconductors. Secondly, iron based superconductors exhibit low anisotropy compared to the very high anisotropic properties for copper based superconductors.

The major contributions and novelty of the current work can be summarised as follow:

I. An extensive literature review has been carried out for the development of Fe-based superconductors with focus on the supercurrent carrying ability, models of flux pinning, upper critical field and thermally activated flux flow models in chapter 2.
II. Superconducting properties in electron doped BaFe$_{2-x}$Ni$_x$As$_2$ single crystal are reported in Chapter 4. Also the flux pinning mechanism of this compound was investigated using the Dew-Hughes model and the collective flux pinning model. Analysis using the Dew-Hughes model has suggested that point pins alone cannot explain the observed variation of the pinning force density. Also, based on the collective flux pinning model, flux pinning is dominated by the spatial variation in the charge carrier mean free path.

III. In Chapter 5, the possibility of enhancement of the critical current density was investigated in Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ single crystals. It was found that C$^{4+}$ ion irradiation is an effective method to enhance the critical current density in Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ single crystals by a factor of up to 1.5.

IV. In Chapter 6, the angular dependences of the pinning potential and the upper critical field are investigated for under doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals. The determination of the angular dependence of these properties is important for understanding how the critical current changes with both angle and field. Also, the anisotropy value was determined by scaling the angular dependence of the resistivity based on the Ginzburg-Landau theory, which is more reliable compared to estimating the anisotropy using the ratio of the upper critical field in the $ab$-plane to that along the $c$-axis.

V. In Chapters 7 and 8, the flux pinning mechanism, and electrical and magnetic anisotropy in Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ single crystal are investigated. According to the Dew-Hughes model, the spatial variation in the charge carrier mean free path is responsible for the pinning mechanism in this compound. Also, the thermally activated flux flow behaviour of Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ single crystal is investigated using the conventional Arrhenius relation and the modified
thermally activated flux flow model. It is shown that the Arrhenius curve slopes are directly related to, but not equal to, the activation energies of Fe_{1.06}Te_{0.6}Se_{0.4} single crystals. Therefore, the use of a modified thermally activated flux flow model, \( \rho(T, B) = \rho_0 f \exp(-U/T) \), is suggested, where the temperature dependence of the prefactor \( \rho_0 f = 2 \rho_c U/T \) and the nonlinear relation of the thermal activation energy are considered. The modified thermally activated flux flow model results are in good agreement with the very high value of the critical current density of this compound from experimental data.

VI. Structural, transport and magnetic properties of the parent compound BaFe\(_2\)As\(_2\) and the non superconducting Ba(Fe\(_{1-x}\)Cr\(_x\))\(_2\)As\(_2\) compounds are presented and discussed in chapter 9. The angular dependence of resistivity manifested a twofold symmetry for the parent compound BaFe\(_2\)As\(_2\) and all the studied concentration of Cr; however, hysteresis in the twofold symmetry is observed with increasing Cr concentration. Especially, a very sharp and wide hysteresis twofold symmetry observed for Ba (Fe\(_{1-x}\)Cr\(_x\)) \(_2\)As\(_2\) (x=0.303) compound which can be interesting for practical application. The Laue data at 100 K and 4 K reveals that the systematic broadening of families of the \((hh0)\) family beneath 50 K is consistent with a subtle orthorhombic phase transition, however, it is clear that other peaks including those in the perpendicular \((h-hk)\) lines are affected, suggesting that crystal strain in the perpendicular direction is an important consideration. The moderate increase in intensity at the index nominally assigned (-101) is consistent with G-type AF transition reported for this material.

VII. Finally a summary is given in chapter 10.
2.1 Fundamental properties of superconductivity

Superconductivity can be described as a phenomenon of zero electrical resistivity or infinite electrical conductivity below a certain temperature. A material becomes superconducting at a certain temperature, called the superconducting transition temperature, $T_c$, which varies from very small values of a few millikelvin to values above 100 K [1].

![Figure 2-1](Image)

Figure 2-1 Experimental data obtained on mercury by Kamerling Onnes showing the superconducting transition for the first time [1]

For the first time, in 1911 Heike Kamerlingh Onnes, discovered that the electrical resistance of mercury suddenly drops to zero at a temperature of 4.2 K. [2] (Figure 2-1). His further study on other materials showed that the transition from the
normal to the superconducting state occurs at different temperatures for different materials.

Figure 2-2 Periodic table showing all superconducting elements and their $T_c$ values.

<table>
<thead>
<tr>
<th>Periodic Table</th>
<th>Superconductor</th>
<th>Superconductor under pressure</th>
<th>Special form is a superconductor</th>
<th>Not a superconductor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elements</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 2-2 Shows the periodic table, with identification of all known elemental superconductors and their $T_c$. Elements that become superconducting at atmospheric pressure are indicated the by dark pink colour. The $T_c$ of these elements varies from 9.3 K for niobium to 0.0003 K for rhodium. The orange coloured cells indicate elements that become superconductors under high pressure. The light pink cells are elements that are superconductors in specific forms. For example, chromium in the form of thin films, palladium after irradiation with alpha particles, platinum as a compact powder, and carbon in the form of nanotubes.
The Meissner effect was discovered by W. Meissner and R. Oschsenfeld in 1933. When a superconducting material is cooled below its $T_c$, magnetic fields are excluded from the material and the material act as a perfect diamagnet, as shown in Figure 2-3 [3]. The field is excluded, however, only if it is below a certain critical field which depends on the materials, temperature, and geometry of the specimen. Superconductivity disappears above this critical field. Zero resistance and the Meissner effect are the defining characteristic of superconductors that make them very useful for practical applications. Zero resistance means zero energy loss when superconductor materials are used to carry electrical current.

![Figure 2-3: Meissner effect, the expulsion of external magnetic field from inside the superconductor in the superconducting state by creating surface current [4]. The field is applied at (a) $T > T_c$ and (b) $T < T_c$.](image)

The Meissner effect has implications for making powerful superconducting magnets for magnetic resonance imaging (MRI) and magnetic-levitation (maglev) trains which allow safe and high speed transportation.
In 1950, the Ginzburg-Landau theory of superconductivity was developed by Landau and Ginzburg. This theory had great success in explaining the macroscopic properties of superconductors. The microscopic theory of superconductivity was proposed in 1957 by Bardeen, Cooper, and Schrieffer (BCS). It is known as BCS theory, and Bardeen, Cooper, and Schrieffer were awarded the Noble Prize for it.

According to the BCS theory, the electrons can pair together, or Cooper pairs can form, because of lattice distortion. The Cooper pair acts like a boson and therefore, is able to move easily through the lattice without any electrical resistance. Cooper pairs cause a reduction in the Fermi energy, resulting in an energy gap or superconducting gap. In order to break the Cooper pairs, there needs to be enough energy to overcome the energy gap [6]. BCS theory can only explain the superconductivity in conventional superconductors, but cannot account for the pairing mechanism in high temperature superconductors.

Figure 2-4: (a) Magnetization curve for type I superconductor. (b) Magnetization curve for type II superconductor.

In 1958, Abrikosov divided superconducting materials into two groups by their behaviour in magnetic fields: type I and type II superconductors. In type-II
superconductors, the coherence length, $\xi$, is shorter than the penetration depth, $\lambda$. The magnetization of type I and type II superconductors is shown in Figure 2-4.

For the elemental superconductors and other type I superconductors, the superconductivity is quenched in relatively low magnetic field. In contrast, type II superconductors have two critical field strengths for a given temperature, a lower critical field ($B_{c1}$) and an upper critical field ($B_{c2}$). Below $B_{c1}$, these materials act exactly like type I superconductors and magnetic field cannot penetrate inside the material, and above $B_{c2}$, they act like normal materials at low temperature. Between $B_{c1}$ and $B_{c2}$, however, the superconductors have a unique property. They have a resistance of zero, but allow a certain amount of flux penetration in the form of vortices. The area between $B_{c1}$ and $B_{c2}$ is known as the vortex state or mixed state; where the superconducting state and the normal state coexist. Each vortex can be described as a long cylinder with its axis parallel to the external magnetic field. Inside the cylinder, the superconducting order parameter is zero. These vortices are surrounded by a superconducting region. The radius of the cylinder is of the order of Ginzburg-Landau coherence length, $\xi_{GL}$ [3]. The supercurrent circulates around the vortices within an area with radius $\approx \lambda$, and it forms a regular triangular lattice under ideal conditions, as shown in Figure 2-5(b).

The ideal triangular vortex lattice, however, can only occur in a homogeneous superconductor. In fact, the material structure has a great influence on the vortex pattern [3]. For example, a vortex can be pinned or trapped by defects or impurities in the material. This phenomenon is called flux pinning. Flux pinning is only possible when there are defects in the crystalline structure of the superconductor, such as grain boundaries, impurity particles, and or crystal imperfections. This
phenomenon is used in high temperature superconductors in order to prevent flux motion or creep, which can create resistance and decrease the critical current density and upper critical field. The magnetic field at which the pinning effect is not effective, is called the irreversibility field, as shown in Figure 2-5(a).

Figure 2-5: (a) Magnetic phase diagram of high temperature superconductor. (b) Vortices in the mixed state of type II superconductor. The grey area is the normal state at the centre of vortices. The magnetic field and shielding current are schematically drawn for one flux line [7].

In 1962, Brian Josephson demonstrated the existence of a superconducting current through a tunnelling device made from two superconductors separated by a thin insulating layer. The effect is known as the Josephson Effect and has had practical applications in making various sensitive measurements, including the determination of fundamental physical constants and measurements of magnetic fields that are a billion times weaker than the Earth’s field. Josephson was awarded Nobel Prize for this discovery in 1973.

Figure 2-6 illustrates the $T_c$ of some known superconductors versus the date of their discovery. The modern era of superconductivity started in 1986 with the discovery of superconductivity in lanthanum barium copper oxide with $T_c = 35$ K. In
February of 1987, a perovskite ceramic material was found to superconduct above liquid nitrogen temperature at 90 K. Discovered in 2000, MgB$_2$ with $T_c$ of 40 K, is a conventional superconductor because its superconducting properties can be explained by BCS theory.

In February 2008 an iron-based family of high temperature superconductors was discovered. Hideo Hosono of the Tokyo Institute of Technology discovered that lanthanum oxygen fluorine iron arsenide becomes a superconductor at 26 K. This discovery was followed by the revelation of an even higher $T_c$ of 43 for the same compound under applied pressure [8], which inspired researchers to use chemical pressure. Following research by other groups suggested that replacing the lanthanum by other rare earth elements, such as cerium, samarium, neodymium, and praseodymium, lead to superconductivity at critical temperatures up to 56 K [9].

Figure 2-6  Superconducting critical temperatures of several superconductors as a function of year of discovery. The inset is an enlargement of the discoveries since 2006.
The discovery of iron based superconductors (FBSs) represents the foundation of a new era in the field of superconductivity replacing Copper era by Iron era. This discovery gives scientists a great opportunity to study about superconducting and magnetic properties in a different family of high temperature superconductors, also to understand the nature of superconductivity in unconventional superconductors is the crucial aspect for designing new superconductors with higher $T_c$. The FBSs materials would be good candidates for use in electricity generators, cheaper medical imaging scanners, and extremely fast levitating trains, because superconducting materials with higher $T_c$ would not require expensive coolants to reach the superconducting transition temperature. Therefore, the discovery of FBSs was a significant milestone in the condensed matter community.

### 2.2 MgB$_2$, Cuprates and Iron-based Superconductors

Cuprate superconductors were the only known superconductors that worked far above liquid-helium temperatures for more than 20 years. In 2008, the discovery of a very novel class of superconductors based on iron was a surprise to the scientific community. Hideo Hosono and his colleagues announced the first non-cuprate high-temperature superconductors in the form of LaFeAsO$_{1-x}$F$_x$ [10]. Superconductivity in the iron-based compound amazed scientists, as they thought that magnetic nature of iron would disrupt the pairing of electrons. Soon after only one month, the critical temperature of these compounds was doubled by replacing La by other rare earth elements with smaller atomic radius.

Table 2-1 summarizes the features of the electronic structures and physical properties of FBSs in comparison with cuprates and MgB$_2$. All of these compounds have layered structures. The FBSs contain FePn (Pn = As or Se) layers similar to the CuO$_2$
layers in cuprates, which are considered to be necessary for the occurrence of superconductivity. The parent compound of FBSs is an antiferromagnetic metal, whereas the parent compound of cuprates is a Mott insulator. According to experimental studies such as angle-resolved photoemission spectroscopy (ARPES) [11] and band calculation studies [12], FBSs are multiband superconductors, as each of the five Fe 3d bands crosses the Fermi level and several disconnected electron and hole sheets of the Fermi surface are formed by the hybridized d-orbital of Fe [11, 12]. On the other hand, in cuprates, superconductivity occurs when hole or electron carriers are doped into the CuO$_2$ layer. The Fermi level exists in the Cu 3$d_{x^2-y^2}$-O 2$p_o$ degenerate band, and as a result, a single electronic band is involved in superconductivity. MgB$_2$ is a two-band superconductor, in which the B 2$p_o$ and 2$p_z$ bands are involved in the Fermi level. In cuprates, perfection of the CuO$_2$ layer is essential for obtaining superconductivity, but in FBSs, the FeAs layer is less sensitive. In fact, doping into the iron sites also can induce superconductivity [13, 14]. This is due to the highly delocalized nature of the iron 3d-electrons in FBSs [15]. The electronic anisotropy of FBSs is generally smaller than for cuprate superconductors [16].

The pairing mechanisms for FBSs are still under debate. It is likely that the spin fluctuation mechanism is responsible for pairing in these compounds. Studies by Mazin et al. suggested that the Fermi surface of these compounds consists of two electron cylinders around the tetragonal M point and two electron cylinders plus a heavy three dimensional hole pocket around the Γ point. They proposed that the order parameters have s-wave symmetry, but different signs on the electron pockets and hole pockets (± symmetry) [17]. This is different from the $d_{x^2-y^2}$ and s-wave
symmetry for cuprates and MgB$_2$ superconductors, respectively. Kontani et al., predicted a different pairing mechanism based on orbital fluctuation for these materials [18]. They suggested that the pairing mechanism is based on s-wave order parameters without sign reversal, $s^{±}$ symmetry.

Table 2-1 Comparison of MgB$_2$ [19, 20], cuprates [21, 22, 23, 24, 25], and iron-based superconductors [15].

<table>
<thead>
<tr>
<th></th>
<th>MgB$_2$</th>
<th>Cuprates</th>
<th>FBSs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parent compound</td>
<td>Metal</td>
<td>Antiferromagnetic Mott Insulator</td>
<td>Antiferromagnetic metal (semi-metal)</td>
</tr>
<tr>
<td>Maximum $T_c$</td>
<td>40 K</td>
<td>~140 K</td>
<td>57 K</td>
</tr>
<tr>
<td>Impurity effect</td>
<td>Sensitive</td>
<td>Sensitive</td>
<td>Robust</td>
</tr>
<tr>
<td>Superconducting symmetry gap</td>
<td>S-wave</td>
<td>d-wave</td>
<td>Extended s wave</td>
</tr>
<tr>
<td>Fermi level</td>
<td>2-band</td>
<td>3d single band</td>
<td>3d 5 band</td>
</tr>
<tr>
<td>$\mu_0B_c$ (T)</td>
<td>~30T</td>
<td>~100 T</td>
<td>70 T</td>
</tr>
<tr>
<td>Coherence length($\xi_{ab}$)(nm)</td>
<td>10</td>
<td>2.2 (YBCO) [26]</td>
<td>1.5-3</td>
</tr>
<tr>
<td>Coherence length($\xi_c$)(nm)</td>
<td>2</td>
<td>0.4 (YBCO)</td>
<td>0.6-1.5</td>
</tr>
<tr>
<td>$J_c$(A/cm$^2$) at T=5 K</td>
<td>$10^5$ [27]</td>
<td>$3\times10^6$ [28]</td>
<td>$2\times10^6$ [29]</td>
</tr>
<tr>
<td>Anisotropy($\gamma_H$)</td>
<td>3-5</td>
<td>4-14 for YBCO</td>
<td>2-5</td>
</tr>
<tr>
<td>Penetration depth($\lambda_{ab}$)(nm)</td>
<td>50</td>
<td>120 (YBCO)</td>
<td>200-500</td>
</tr>
<tr>
<td>Ginzburg number, $G_1$</td>
<td>$10^{-5}$</td>
<td>$5\times10^{-4}$</td>
<td>$1.5\times10^{-5}$-1$\times10^{-3}$</td>
</tr>
<tr>
<td>Pairing Mechanism Structure</td>
<td>Unconventional BSC</td>
<td>Unconventional Layered</td>
<td>Unconventional Layered</td>
</tr>
<tr>
<td>Pairing Symmetry</td>
<td>s [30]</td>
<td>$d_{x^2-y^2}$</td>
<td>$s_T$, $s_{++}$</td>
</tr>
<tr>
<td>Proposed pairing mechanism</td>
<td>Phonon [30]</td>
<td>Spin fluctuation</td>
<td>Spin fluctuation</td>
</tr>
<tr>
<td>Band involved in superconductivity</td>
<td>B 2$p_\sigma$, 2$p_x$</td>
<td>Cu 3$d_{x^2-y^2}$-O 2$p_\sigma$</td>
<td>Fe 3d</td>
</tr>
<tr>
<td></td>
<td>2 band [30]</td>
<td>single band</td>
<td>5 bands</td>
</tr>
</tbody>
</table>

FBSs show several advantages over cuprates. Firstly, the parent compound of FBSs is semi-metallic, in contrast to the insulator parent compound for cuprates. Secondly, FBSs exhibit low anisotropy which is not strongly dependent on the level.
of doping, and impurity does not significantly affect $T_c$ [31]. This is different from cuprates, where their anisotropic and nearly two-dimensional nature results in weak pinning and significant thermal fluctuation in these compounds.

### 2.2.1 Crystal structures and families of iron based superconductors (FBS)

In the five years since the discovery of FBSs, several compounds belonging to different families have been discovered. Figure 2-7 illustrates the crystal structure of the different families of FBSs discovered so far. Even though they have different structures and compositions, they all share a common blocking layer of iron-pnictogen (P, As) or iron chalcogen planes (Se, Te). These blocking layers provide a quasi-two-dimensional character to the crystal structure, as they have ionic bonds to the FeAs layer, while the FeAs layer itself possess a combination of covalent bonding for Fe-As and metallic bonding for Fe-Fe [32]. In a similar way to the cuprates, where the copper oxide layer is responsible for high temperature superconductivity, it is widely believed that the interaction that leads to superconductivity is initiated from the iron layers. In all FBSs, Fe and As (Se) atoms form FeAs$_4$ or FeSe$_4$ tetrahedral, where $T_c$ of FBSs depends on the angle between FeAs or FeSe bonds and the height of tetrahedral [33, 34].

The first FBS, LaFePO with $T_c = 4$ K, was discovered in 2006 [35]. This material has ZrCuSiAs type crystal structure with space group of $P4/nmm$. In 2008, superconductivity was also found in a LaFeAsO$_{1-x}$F$_x$ compound with $T_c$ of 26 K [36]. This discovery opened up a new window of opportunity in the field of superconductivity, as this material represents a new family of unconventional superconductors having $T_c$ relatively higher than conventional superconductors.
Soon after this discovery, the highest $T_c$ of this family of materials increased to 55 K by substitution of samarium [8, 9, 37] and neodymium [38] for lanthanum. These materials are described as 1111-FeAs compounds. The crystal structure of 1111-FeAs compounds consists of negatively charged FePn layers, where Fe atoms form a planar square lattice, and positively charged REO layers, with $RE$ a rare earth element. Although 1111-FeAs family exhibits the highest $T_c$ among all iron-based superconductors, it is difficult to study about its properties in detail, because it is challenging to grow a big single crystal and the currently available single crystals are too small. After the discovery of 1111-FeAs, superconductivity was found in hole doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ with $T_c = 38$ K [39] and electron doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [13] compounds with $T_c = 22$ K (the so-called 122-FeAs compounds), followed by LiFeAs (111-FeAs) [40] with $T_c = 18$ K. In 1111-FeAs and 122-FeAs, superconductivity results from electron or hole doping or can be induced by pressure or by isovalent doping. In 111-FeAs, however, superconductivity occurs at zero doping. The 122-FeAs and 111-FeAs compounds have simpler structure compared to 1111-FeAs compounds. Although all three families share the same FePn layers, the
blocking layer, which separates them, is different for each family: rare earth oxide for 1111-FeAs, alkaline earth metals for the 122-FeAs family, and alkali metals for the 111-FeAs.

**Table 2-2 Possible doping for the 122-FeAs family.**

<table>
<thead>
<tr>
<th></th>
<th>$T_c$ (K)</th>
<th>$a$ (Å)</th>
<th>$c$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KFe$_2$As$_2$ [41]</td>
<td>3.8</td>
<td>3.8414</td>
<td>13.837</td>
</tr>
<tr>
<td>CsFe$_2$As$_2$ [41]</td>
<td>2.6</td>
<td>3.9261</td>
<td>12.376</td>
</tr>
<tr>
<td>SrFe$_2$As$_2$ [41]</td>
<td>0</td>
<td>3.8894</td>
<td>15.066</td>
</tr>
<tr>
<td>K$<em>{0.4}$Sr$</em>{0.6}$Fe$_2$As$_2$ [41]</td>
<td>36</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cs$<em>{0.4}$Sr$</em>{0.6}$Fe$_2$As$_2$</td>
<td>37</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ba$<em>{0.89}$Rb$</em>{0.05}$Sn$_{0.06}$Fe$_2$As$_2$ [42]</td>
<td>23</td>
<td>3.925</td>
<td>13.2096</td>
</tr>
<tr>
<td>CaFe$_2$As$_2$ [43]</td>
<td></td>
<td>3.872</td>
<td>11.73</td>
</tr>
<tr>
<td>Ca$<em>{0.5}$Na$</em>{0.5}$Fe$_2$As$_2$ [43]</td>
<td>20</td>
<td>3.829</td>
<td>11.862</td>
</tr>
<tr>
<td>Eu$<em>{0.5}$K$</em>{0.5}$Fe$_2$As$_2$ [43]</td>
<td>32</td>
<td>3.8671</td>
<td>13.091</td>
</tr>
<tr>
<td>SrFe$<em>{1.85}$Ni$</em>{0.15}$As$_2$ [44]</td>
<td>9.8</td>
<td>3.947</td>
<td>12.16</td>
</tr>
<tr>
<td>CaFe$<em>{1.94}$Co$</em>{0.06}$As$_2$ [45]</td>
<td>17</td>
<td>3.887</td>
<td>11.687</td>
</tr>
<tr>
<td>BaFe$<em>{1.8}$Co$</em>{0.2}$Fe$_2$As$_2$ [46]</td>
<td>22</td>
<td>3.9639</td>
<td>12.98</td>
</tr>
<tr>
<td>Ba(Fe$<em>{1.4}$Ni$</em>{0.6}$)$_2$Fe$_2$As$_2$ [47]</td>
<td>20</td>
<td>3.98+</td>
<td>13.03</td>
</tr>
<tr>
<td>Ba(Fe$<em>{1.4}$Rh$</em>{0.6}$)$_2$As$_2$ (x=0.057) [48]</td>
<td>24</td>
<td>3.957</td>
<td>12.939</td>
</tr>
<tr>
<td>Ba(Fe$<em>{1.4}$Pd$</em>{0.6}$)$_2$As$_2$ (x=0.053) [48]</td>
<td>19</td>
<td>3.946</td>
<td>12.953</td>
</tr>
<tr>
<td>SrFe$<em>{1.75}$Rh$</em>{0.25}$As$_2$ [49]</td>
<td>22</td>
<td>3.945</td>
<td>12.25</td>
</tr>
<tr>
<td>SrFe$<em>{1.5}$Ir$</em>{0.5}$As$_2$ [49]</td>
<td>22</td>
<td>3.955</td>
<td>12.24</td>
</tr>
<tr>
<td>SrFe$<em>{1.85}$Pd$</em>{0.15}$As$_2$ [49]</td>
<td>9</td>
<td>3.95</td>
<td>12.28</td>
</tr>
<tr>
<td>BaFe$<em>{1.25}$Ru$</em>{0.75}$As$_2$ [50]</td>
<td>21</td>
<td>4.01</td>
<td>12.81</td>
</tr>
<tr>
<td>Sr(Fe$<em>{1.3}$Ru$</em>{0.7}$)$_2$As$_2$ [51]</td>
<td>13.5</td>
<td>3.97</td>
<td>12.1</td>
</tr>
</tbody>
</table>

In addition, BaFe$_2$As$_2$ and LiFeAs compounds do not contain oxygen. It is suggested that FeAs layer is the key ingredient for superconductivity in these materials, and superconductivity is not uniquely related to oxide materials, as in the case for cuprates [52]. It is very tricky to study 111-FeAs compounds, as they are highly reactive with air. 122-FeAs family has several doping possibilities, as shown
This family has ThCr$_2$Si$_2$ structure with $I4/mmm$ space group. The most studied compounds are hole doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ and electron doped Ba (Fe$_{1-x}$Co$_x$)$_2$As$_2$, as it is easy to grow a big, good quality single crystal. Both share the same parent compound, BaFe$_2$As$_2$, which is an antiferromagnetic metal and goes into a magnetically ordered phase transition below 140 K. Extremely over-doped KFe$_2$As$_2$, which is non-magnetic, shows superconductivity at $T_c = 3$ K. The first arsenic-free family was discovered in the form of $\alpha$-FeSe with $T_c = 8$ K at ambient pressure by Hsu et al [53]. The so-called 11-FeCh family, where Ch is chalcogen, has the simple $\alpha$-PbO structure with space group of $p4/nmm$ and contains a stack of FeCh$_4$ tetrahedral layers. The $T_c$ value of $\alpha$-FeSe increased to 37.6 K under pressure of 8.9 GPa [54]. This family also includes FeTe$_{1-x}$Se$_x$ and FeTe$_{1-x}$S$_x$, with $T_c = 14$ K [55] and 7 K [56], respectively. A new 122 compound (122-FeCh) with the pnictogen replaced by a chalcogen was discovered in defect structured $A_x$Fe$_2$Se$_2$ ($A = K$, Rb, Cs, and Tl) with $T_c = 32$ K [57]. Superconductivity has been reported in FBSs with more complicated blocking layers for Sr$_2$VFeAsO$_3$ (21113-FeAs) [58] and Sr$_3$Sc$_2$Fe$_2$As$_2$O$_5$ (32225-FeAs) [59], with $T_c = 37$ K and 41 K, respectively. Table 2-3 summarizes the structures, space groups, compositions and $T_c$ values of different FBSs.

Table 2-3 Structures, compositions, dopants, and $T_c$ of different iron-based superconductors. $T_s$ and $T_{SDW}$ are the structural and spin density wave transition temperatures.

<table>
<thead>
<tr>
<th>Composition</th>
<th>Structure</th>
<th>Space group ((T=300\ \text{K}))</th>
<th>Dopant</th>
<th>$T_s$ (K)</th>
<th>$T_{SDW}$ (K)</th>
<th>$T_c$ (K)</th>
<th>Low temperature ((\mu_B/\text{Fe atom}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>LnFePO ((Ln=\text{La, Sm, Gd}))</td>
<td>1111</td>
<td>$P4/nmm$</td>
<td>F(O)</td>
<td>3-7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LaFeAsO [60]</td>
<td>1111</td>
<td>$P4/nmm$</td>
<td>F(O)</td>
<td>158</td>
<td>134</td>
<td>26</td>
<td>0.36 [61]</td>
</tr>
<tr>
<td>PrFeAsO [62]</td>
<td>1111</td>
<td>$P4/nmm$</td>
<td>F(O)</td>
<td>154</td>
<td>135</td>
<td>47</td>
<td>0.35 [63]</td>
</tr>
<tr>
<td>Compound</td>
<td>Structure</td>
<td>Symmetry</td>
<td>F(O)</td>
<td>141</td>
<td>41</td>
<td>0.8 [63]</td>
<td></td>
</tr>
<tr>
<td>------------------</td>
<td>-----------</td>
<td>--------------</td>
<td>-------</td>
<td>-----</td>
<td>----</td>
<td>----------</td>
<td></td>
</tr>
<tr>
<td>CeFeAsO</td>
<td>1111</td>
<td>P4/nmm</td>
<td>F(O)</td>
<td>155</td>
<td>140</td>
<td>0.8 [63]</td>
<td></td>
</tr>
<tr>
<td>NdFeAsO</td>
<td>1111</td>
<td>P4/nmm</td>
<td>F(O)</td>
<td>150</td>
<td>141</td>
<td>0.25 [63]</td>
<td></td>
</tr>
<tr>
<td>SmFeAsO</td>
<td>1111</td>
<td>P4/nmm</td>
<td>F(O)</td>
<td>175</td>
<td>135</td>
<td>0.00 [63]</td>
<td></td>
</tr>
<tr>
<td>[66, 67] GdFeAsO</td>
<td>1111</td>
<td>P4/nmm</td>
<td>F(O)</td>
<td>135</td>
<td>141</td>
<td>~56</td>
<td></td>
</tr>
<tr>
<td>LnFeAsO (Ln=La,Sm,Gd)</td>
<td>1111</td>
<td>P4/nmm</td>
<td>Vacancy (O)</td>
<td>28- 55</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CaFeAsF</td>
<td>1111</td>
<td>P4/nmm</td>
<td>Co(Fe)</td>
<td>134</td>
<td>114</td>
<td>0.49 [68]</td>
<td></td>
</tr>
<tr>
<td>CaFe2As2</td>
<td>122</td>
<td>I4/mmm</td>
<td>Co(Fe)</td>
<td>171</td>
<td>171</td>
<td>0.8 [63]</td>
<td></td>
</tr>
<tr>
<td>BaFe2As2 [69]</td>
<td>122</td>
<td>I4/mmm</td>
<td>K(Ba)</td>
<td>142</td>
<td>142</td>
<td>0.87 [70]</td>
<td></td>
</tr>
<tr>
<td>BaFe2As2 [71]</td>
<td>122</td>
<td>I4/mmm</td>
<td>Co,Ni(Fe)</td>
<td>142</td>
<td>142</td>
<td>18-22</td>
<td></td>
</tr>
<tr>
<td>SrFe2As2</td>
<td>122</td>
<td>I4/mmm</td>
<td>Co(Fe)</td>
<td>205</td>
<td>205</td>
<td>0.94 [63]</td>
<td></td>
</tr>
<tr>
<td>[72, 73] EuFe2As2 [74]</td>
<td>122</td>
<td>I4/mmm</td>
<td>K(Eu)</td>
<td>190</td>
<td>190</td>
<td>32</td>
<td></td>
</tr>
<tr>
<td>BaFe2As2 [75]</td>
<td>122</td>
<td>I4/mmm</td>
<td>Cr,Mo(Fe)</td>
<td>56- 140</td>
<td>No SC</td>
<td></td>
<td></td>
</tr>
<tr>
<td>LiFeAs [76]</td>
<td>111</td>
<td>P4/nmm</td>
<td>Co(Fe)</td>
<td>50</td>
<td>40</td>
<td>0.09 [63]</td>
<td></td>
</tr>
<tr>
<td>NaFeAs [77]</td>
<td>111</td>
<td>P4/nmm</td>
<td>Co(Fe)</td>
<td>50</td>
<td>40</td>
<td>0.09 [63]</td>
<td></td>
</tr>
<tr>
<td>FeTe</td>
<td>11</td>
<td>P4/nmm</td>
<td>Co(Fe)</td>
<td>72</td>
<td>72</td>
<td>2.25 [79]</td>
<td></td>
</tr>
<tr>
<td>FeSe1-xTex</td>
<td>11</td>
<td>P4/nmm</td>
<td></td>
<td></td>
<td>14</td>
<td></td>
<td></td>
</tr>
<tr>
<td>K0.8Fe2-ySe2</td>
<td>122</td>
<td>I4/mmm</td>
<td></td>
<td></td>
<td>551-540</td>
<td>3.31 [81]</td>
<td></td>
</tr>
<tr>
<td>Rb0.8Fe2-ySe2</td>
<td>122</td>
<td>I4/mmm</td>
<td></td>
<td></td>
<td>540-534</td>
<td>3.11 [81]</td>
<td></td>
</tr>
<tr>
<td>Cs0.8Fe2-ySe2</td>
<td>122</td>
<td>I4/mmm</td>
<td></td>
<td></td>
<td>525-504</td>
<td>~27</td>
<td></td>
</tr>
<tr>
<td>Tl0.4K0.3Fe2-ySe2</td>
<td>122</td>
<td>I4/mmm</td>
<td></td>
<td></td>
<td>515-496</td>
<td>~25</td>
<td></td>
</tr>
<tr>
<td>Tl0.4Rb0.4Fe2-ySe2</td>
<td>122</td>
<td>I4/mmm</td>
<td></td>
<td></td>
<td>512-500</td>
<td>~31</td>
<td></td>
</tr>
<tr>
<td>Sr2ScFePO3</td>
<td>21113</td>
<td></td>
<td></td>
<td></td>
<td>155-37</td>
<td>0.1 [82]</td>
<td></td>
</tr>
<tr>
<td>Sr2VFeAsO4</td>
<td>21113</td>
<td>No structural transition</td>
<td>155</td>
<td>37</td>
<td>0.1 [82]</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
2.2.2 Phase diagram

Figure 2-8 presents electronic phase diagrams for different families of FBSs with distinct areas for the antiferromagnetically ordered spin density wave (SDW) and superconducting phases. Undoped parent compounds, such as LaFeAsO, BaFe$_2$As$_2$, and FeTe, are semi-metals with antiferromagnetic or spin wave magnetic ordering, and they show the structural transition from tetragonal to orthorhombic at temperatures close to the magnetic transition temperature. Superconductivity arises from electron or hole doping, or can be induced by pressure [83] or by isovalent doping in the 1111-FeAs and 122-FeAs families [60].

For LaFeAsO$_{1-x}$F$_x$, the antiferromagnetic (AFM) and superconducting phases are totally separated as a function of doping and do not coexist. It is reported, however, that AFM and superconducting phases overlap for SmFeAsO$_{1-x}$F$_x$ [84], in a similar way to 122-FeAs phase diagram [85].

![Phase diagrams of different families of iron-based superconductors](image-url)
A very similar phase diagram has been reported for compounds with electron, hole and isovalent substitution in 122-FeAs system [Figure 2-8 (c)]. Superconductivity in 122-FeAs arises from several types of d-metal substitution into Fe sites, including from Fe [51], Co [46], and Ni [47, 91] columns. Cr [92], Mn [93], and Cu [52] doping, however, results in the suppression of AFM ordering without inducing superconductivity. It is suggested that, absence of superconductivity in Ba(Fe1-xCrx)2As2 can be explained by the presence of long range magnetic order for all concentrations of these compounds, and G-type AFM order occurs as the spin density wave (SDW) AFM order is suppressed [94]. This is different from what happens in all other electron doped 122-FeAs compounds. It should be noted that undoped compounds in different families of FBSs show different behaviour. For example, LaFePO, LiFeAs, and FeSe are non-magnetic and exhibit superconductivity even without doping. In contrast, undoped LaFeAsO and BaFe2As2 are non-superconducting antiferromagnetic metals, in which superconductivity can be induced after suppression of magnetic order by electron or hole doping.

2.2.3 **Transport properties**

Figure 2-9 shows the temperature dependence of electrical resistivity for LaFeAsO1-xFx [95], Ba1-xKxFe2As2 [96], BaFe1.8Co0.2As2, LiFeAs [97], FeSe1-xTe x [98], and KxFe2-ySe2 [99] (122*) compounds. Substitution of fluorine for oxygen site results to superconductivity in LaFeAsO0.89F0.11 with $T_c = 26$ K. With applied pressure of 4 GPa to LaFeAsO0.89F0.11, $T_c$ increases, reaching 43 K. In 122-FeAs, superconductivity results from electron or hole doping, or can be induced by pressure or by isovalent doping.
Superconductivity is reported in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ [39], Sr$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ [100], Sr$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ [101], Ca$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ [100], and BaFe$_{1.8}$Co$_{0.2}$Fe$_2$As$_2$ [46], with $T_c$ of 38 K, 32 K, 26 K, 21 K and 22 K, respectively. The LiFeAs compound reveals superconductivity with $T_c = 18$ K [97]. It should be noted that superconductivity occurs without doping in this compound. Superconductivity was reported in FeSe system with $T_c = 8$ K, as the first arsenic free compound. The $T_c$ of this compound increases to 36.7 K with applied pressure of 8.9 GPa [102]. In addition, superconductivity was reported in FeTe$_{0.5}$Se$_{0.5}$ with $T_c = 14$ K [103] and K$_{0.82}$Fe$_{1.63}$Se$_2$ with $T_c = 31.5$ K [99].

![Figure 2-9](image)

Figure 2-9 Temperature dependence of electrical resistivity for (a) LaFeAsO$_{1-x}$F$_x$ [95], (b) Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [96], (c) BaFe$_{1.8}$Co$_{0.2}$As$_2$, (d) LiFeAs [97], (e) FeSe$_{1-x}$Te$_x$ [98], and (f) K$_x$Fe$_{2-y}$Se$_2$ (122') [99] compounds.
The temperature dependence of resistance for all 1111-FeAs, 122-FeAs, 111-FeAs, and 11-FeAs compounds demonstrates metallic behaviour ($d\rho/dT > 0$), and the resistivity decreases with decreasing temperature. The 122* family reveals a different behaviour depending on composition. For example, for $K_{0.84}Fe_{1.59}Se_2$, the temperature dependence of resistivity shows an anomaly at 150 K, while superconductivity appears at 33 K, while for $A_{0.8}Fe_{1.6}Se_2$, resistivity decreases monotonically with decreasing temperature. Also, $FeSe_{1-x}Te_x$ with $x > 0.25$ does not show metallic behaviour, and resistivity increases with decreasing temperature [98]. In all cases, the resistivity values at room temperature are as high as $\rho \approx 1 \text{ m}\Omega \cdot \text{cm}$, while for a good metal like copper, $\rho \approx 1 \mu\Omega \cdot \text{cm}$. According to the band structure calculations, FBSs are classified as submetallic compounds [104].

### 2.2.4 Magnetoresistance in FBSs

Figure 2-10 presents magnetoresistance measurements for different single crystal samples: $NdFeAsO_{0.7}F_{0.3}$ (Nd-1111) [105], $Ba(Fe_{0.9}Co_{0.1})_2As_2$ (Ba-122), $FeSe_{0.5}Te_{0.5}$ (Fe-11), and $Tl_{0.58}Rb_{0.42}Fe_{1.72}Se_2$ (122*) [106] single crystals for $B//c$. Similar to the cuprates, for FBSs, thermal fluctuations of vortices are unavoidable, resulting in thermally activated flux flow, as the resistance curve $R(T,B)$ shifts to lower temperature and also broadens as the field increases. For example, Nd-1111 shows similar transition broadening to $YBa_2Cu_3O_{7-x}$ (YBCO) with increasing field (Figure 2-10). On the other hand, thermal fluctuations are negligible in Ba-122 compounds as the resistive transition curves $R(T,B)$ only shift to lower temperature but do not broaden as the field is enhanced. The broadening is intermediate for Fe-11 and 122*.
2.2.5 Upper critical field

One of the key parameters for superconducting materials is upper critical field, which provides crucial information about fundamental superconducting properties such as the anisotropy, coherence length, dimensionality of the superconductivity, and pair-breaking mechanism. $B_{c2}$ is an intrinsic property of type II superconductors, which can be measured from the resistive transition curves, $R(T, B)$. Based on the conventional BCS theory, $B_{c2}$ is linear in $T$ near $T_c$ and saturates at 0 K.

The $B_{c2}$ value can be estimated using the one band Werthamer- Helfand- Hohenberg (WHH) formula,

$$B_{c2} = -0.693T_c \left[ dB_{c2}/dT \right]_{T_c}$$

2-1
Where \([dB_{c2}/dT]_T\) is obtained from the slope of the \(B_{c2}\) vs. \(T\) curve near \(T_c\). However, the estimated value of \(B_{c2}\) based on the WHH formula is not valid for the low temperature range, as the \(B_{c2}\) values are extrapolated from measurements at low magnetic field and high temperature using the WHH formula.

Table 2-4 \(B_{c2}(T)\) and related parameters of different FBSs. \(B_{c2}^{ab}(T)\), \(B_{c2}^{c}(T)\), \(B_{c2}^{c}(T)\) exp., \(B_{c2}^{c}(T)\) exp., \(r, \gamma_H, \epsilon_{ab}, \epsilon_{c}, \Delta, \) and \(G_i\) represent the upper critical field for \(B//ab\) and for \(B//c\), Pauli limit for \(B_{c2}\), experimental value of the upper critical field for \(B//ab\) and for \(B//c\), Maki parameter, upper critical field anisotropy, coherence length for \(B//ab\), coherence length for \(B//c\), superconducting energy gap, and the Ginsburg number, respectively.

<table>
<thead>
<tr>
<th>(B_{c2}(T)) and related parameters of different FBSs. (B_{c2}^{ab}(T)), (B_{c2}^{c}(T)), (B_{c2}^{c}(T)) exp., (B_{c2}^{c}(T)) exp., (r, \gamma_H, \epsilon_{ab}, \epsilon_{c}, \Delta, ) and (G_i)</th>
<th>(NdFeAs(O,F))</th>
<th>(Ba,K)Fe_2As_2</th>
<th>Ba(Fe,Co)_2As_2</th>
<th>LiFeAs</th>
<th>Fe(Se,Te)</th>
<th>KxFe2Se2</th>
<th>Tl0.58Rb0.42Fe1.72Se2</th>
</tr>
</thead>
<tbody>
<tr>
<td>(T_c) (K)</td>
<td>47</td>
<td>37-38</td>
<td>22</td>
<td>18</td>
<td>14</td>
<td>32</td>
<td>33</td>
</tr>
<tr>
<td>(\mu_0(dB_{c2}^{ab}/dT)) (T/K)</td>
<td>9</td>
<td>5.4</td>
<td>6</td>
<td>3.3</td>
<td>8.9</td>
<td>1.4</td>
<td>12</td>
</tr>
<tr>
<td>(\mu_0(dB_{c2}/dT)) (T/K)</td>
<td>1.85</td>
<td>2.9</td>
<td>2</td>
<td>1.2</td>
<td>3.6</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>(\mu_0B_{c2}^{ab}(0)) (T)</td>
<td>304</td>
<td>104</td>
<td>103.5</td>
<td>39.8</td>
<td>86</td>
<td>60</td>
<td>273</td>
</tr>
<tr>
<td>(\mu_0B_{c2}^{c}(0)) (T)</td>
<td>62-70</td>
<td>56</td>
<td>34.5</td>
<td>14.5</td>
<td>36.9</td>
<td>45</td>
<td></td>
</tr>
<tr>
<td>(\mu_0B_{c2}^{c}(0)) (T) exp.</td>
<td>87.4</td>
<td>70.1</td>
<td>41</td>
<td>33.5</td>
<td>26</td>
<td>59.5</td>
<td>61.4</td>
</tr>
<tr>
<td>(\mu_0B_{c2}^{c}(0)) (T) exp.</td>
<td>57(34 K)</td>
<td>57</td>
<td>53(6 K)</td>
<td>24.2</td>
<td>47</td>
<td>54(18 K)</td>
<td></td>
</tr>
<tr>
<td>(\mu_0B_{c2}^{c}(0)) (T) exp.</td>
<td>43(18 K)</td>
<td>55</td>
<td>40(4.2 K)</td>
<td>15</td>
<td>47</td>
<td>52(4 K)</td>
<td></td>
</tr>
<tr>
<td>(\alpha (B//ab))</td>
<td>3.5</td>
<td>1.9-2.2</td>
<td>2.03</td>
<td>1.74</td>
<td>2.3</td>
<td>5.6</td>
<td></td>
</tr>
<tr>
<td>(\gamma_H(T_c))</td>
<td>6</td>
<td>2</td>
<td>2.7</td>
<td>2.5</td>
<td>2</td>
<td>2</td>
<td>8.1</td>
</tr>
<tr>
<td>(\epsilon_{ab}(nm))</td>
<td>2.3</td>
<td>2.17</td>
<td>2.45</td>
<td>4.8</td>
<td>2.56</td>
<td>2.3</td>
<td></td>
</tr>
<tr>
<td>(\epsilon_{c}(nm))</td>
<td>0.26</td>
<td>2.17</td>
<td>1.48</td>
<td>1.7</td>
<td>2.56</td>
<td>1.4≤ (\epsilon_{c}) ≤2.3</td>
<td></td>
</tr>
<tr>
<td>(\Delta(meV))</td>
<td>4-7, 10-18</td>
<td>1.8-4.6, 9-11</td>
<td>1.9-4.4, 5-7.4</td>
<td>2.3</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(G_i [113])</td>
<td>(10^{-2}\cdot10^{-4})</td>
<td>(1.7\times10^{-4})</td>
<td>(1.3\times10^{-3})</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
FBSs reveal very high upper critical fields that are promising for practical applications. For example, the temperature dependence of $\mu_0B_{c2}$ of single crystalline Nd-1111 shows a weak upward curvature with a large initial slope of 9 T/K [107]. Using WHH formula, a very high value of $\mu_0B_{c2}(0) = 304$ is obtained, however, this value is overestimated for his compound. This is confirmed by determining $\mu_0B_{c2}$ at low temperature and high magnetic field. High magnetic field resistive measurement of LaFeAsO$_{0.89}$F$_{0.11}$ show a remarkable enhancement of $B_{c2}$ at low temperature compared to the values expected from the $dB_{c2}/dT$ slope near $T_c$. Two band superconductivity is suggested for this compound, as similar behaviour was observed for MgB$_2$ films, and the $B_{c2}$ enhancement can be described in the framework of two gap scenario [114]. The multiband superconductivity of FBSs is confirmed by ARPES experiments [115].

2.2.5.1 Pair breaking mechanism

There are two mechanisms that contribute to the pair breaking of Cooper pairs and the suppression of superconductivity due to external applied magnetic field [116]: (I) orbital pair-breaking of Cooper pairs in the superconducting state due to the Lorentz force acting on Cooper pair and the destruction of superconductivity when kinetic energy goes above condensation energy of the Cooper pair; (II) Pauli paramagnetic pair breaking because of Zeeman effect, which aligns the spins of two electrons with the applied field. Figure 2-11 schematically illustrates pair breaking mechanism of a singlet Cooper pair by external magnetic field.
Werthamer, Helfand, and Hohenberg studied the temperature and impurity dependence of $B_{c2}$ for type II superconductors [117]. In single band $s$-wave, weakly coupled type II superconductors, the effect of Pauli spin paramagnetic and spin-orbital scattering have been accounted in WHH model through Maki parameter, $\alpha$, and spin-orbital scattering, $\lambda_{so}$ [117]:

$$\ln \frac{\gamma}{\tau} = \left( \frac{1}{2} + \frac{i\lambda_{so}}{4\gamma} \right) \psi \left( \frac{1}{2} + \frac{\hbar + \frac{\lambda_{so}}{2} + i\gamma}{2t} \right) + \left( \frac{1}{2} - \frac{i\lambda_{so}}{4\gamma} \right) \psi \left( \frac{1}{2} + \frac{\hbar + \frac{\lambda_{so}}{2} - i\gamma}{2t} \right) - \psi \left( \frac{1}{2} \right) \quad 2-2$$

Where $\psi(x)$ is the digamma function, and

$$\gamma \equiv \left[ (\alpha h)^2 - \left( \frac{\lambda_{so}}{2} \right)^2 \right]^{\frac{1}{2}} \quad 2-3$$

$$\left( h = \frac{4\mu_0 \beta_{c2}(T)}{\pi^2 T_c \left( \frac{d\mu_0 \beta_{c2}(T)}{dT} \right)_{T=T_c}} \right) \quad 2-4$$

Here, $\lambda_{so}$ is the strength of spin orbital scattering and $\alpha$ is Maki parameter. If the orbital effect is dominant ($\alpha = 0$) and $\lambda_{so} = 0$, equation 2-2 can be simplified as
\[ \ln \left( \frac{1}{T} \right) = \psi \left( \frac{1}{2} + \frac{\hbar}{2T} \right) - \psi \left( \frac{1}{2} \right) \]  

At \( T = 0 \) K, the orbital limit of upper critical field can be obtained from

\[ \mu_0 B^c_2(0) = -0.693 T_c \left( \frac{d \mu_0 B^c_2(T)}{dT} \right)_{T=T_c} \]  

If only the spin paramagnetic effect is taken into account, however, the Pauli limiting field at \( T = 0 \) K is

\[ \mu_0 B_p(0)[T] = \frac{\Delta}{\sqrt{\mu_B}} = 1.86 T_c \]  

Where \( \Delta \) is the \( s \)-wave superconducting gap.

The effect of paramagnetic-limited effect should be considered for determining the actual \( B_{c2} \) when Pauli limit of \( \mu_0 B^p_{c2}(0) \) and orbital limit of \( \mu_0 B^{orb}_{c2}(0) \) are comparable. Therefore,

\[ \mu_0 B^p_{c2}(0) = \mu_0 B^{orb}_{c2}(0) / \sqrt{1 + \alpha^2} \]  

Where the Maki parameter is expressed as

\[ \alpha = \sqrt{2} B^{orb}_{c2}(0) / B^p(0) \]  

Generally in conventional superconductors, \( \mu_0 B^p(0) \) is higher than \( \mu_0 B^{orb}_{c2}(0) \), and therefore, \( B_{c2} \) is mostly limited by orbital pair breaking mechanism. In unconventional superconductors, however, such as FBSs, the influence of paramagnetic effects on the temperature dependence of \( \mu_0 B_{c2} \) and \( \gamma_H \) need to be taken into account [118]. Several reports indicate that paramagnetic effect plays an important role in determining \( \mu_0 B_{c2} \) [119]. FBSs show a large Maki parameter, \( \alpha \), and
there are several disconnected electron and hole sheets at the Fermi surface that result from the hybridized \(d\)-orbitals of iron. \cite{115} Therefore, the multiband effect on \(\mu_0 B_{c2}\) should also be considered. Figure 2-13 shows the temperature dependence of \(\mu_0 B_{c2}\) for different families of FBSs. The temperature dependence of \(\mu_0 B^c_{c2}\) for 1111-FeAs reveals a pronounced upturn in the curvature at low temperature. \(\mu_0 B^\text{ab}_{c2}\), however, shows a downturn in the curvature with decreasing temperature \cite[Figure 2-12(a)]{105, 114}. It is possible to describe both types of behaviour using two-band theory with the high diffusivity ratio of the electron and hole bands. In addition, the spin paramagnetic effect also needs to be taken into account, especially for \(B//ab\).

For hole doped 122-FeAs system as shown in Figure 2-12(b), \(B^\text{c}_{c2}(T)\) follows an almost linear temperature dependence down to 10 K, while in contrast, \(H^\text{ab}_{c2}(T)\) exhibits a convex shape \cite{96}, which tends to saturate with decreasing temperature. As with 1111-FeAs, this also can be explained within the two-band theory \cite{120, 121}. It is likely that spin paramagnetic effect is essential for \(B//ab\) \cite{120}. On the other hand, it is possible that \(B^\text{c}_{c2}(T)\) trend is related to its multiband electronic structure \cite{120}.

It should be noted that, \(B_{c2}\) of (Ba,K)Fe\(_2\)As\(_2\) extrapolates to a similar value of 70 T for both \(B//ab\) and \(B//c\) as the temperature goes to zero. This is different from what occurs in cuprates and organic superconductors, where in-plane critical field is much higher than when the field is applied perpendicular to the planes \cite{122}. Also, electron doped 122-FeAs shows similar behaviour to hole doped 122-FeAs system, and \(B^\text{ab}_{c2}(T)\) and \(B^\text{c}_{c2}(T)\) converge at a similar value at zero temperature \cite{110, 120}.

For 111-FeAs system, the temperature dependence of \(\mu_0 B_{c2}\) for \(B//ab\) and \(B//c\) show trend towards saturation at low temperature. It is suggested that temperature
dependence of $\mu_0B_{c2}$ is mainly determined by the orbital-limited field and that $\mu_0B_{ab}^{c2}(T)$ can be described by the spin paramagnetic effect [97, 123].

Figure 2-12 Temperature dependence of $\mu_0B_{c2}$ for (a) LaFeAsO$_{0.89}$F$_{0.11}$ [114], (b) (Ba,K)Fe$_2$As$_2$ [96], (c) LiFeAs [97], and (d) Fe$_{1.06}$Te$_{0.89}$Se$_{0.11}$ [123] single crystals. Insets: The anisotropy of $\mu_0B_{c2}$.

In 11-FeCh system (Figure 2-12(d)) both spin paramagnetic and multiband effects contribute to $\mu_0B_{c2}(T)$. Compared to iron pnictides, however, the multiband effect is much weaker and spin paramagnetic effect is dominant [124]. Both $\mu_0B_{ab}^{c2}(T)$ and $\mu_0B_{c}^{c2}(T)$ can be explained using WHH theory with the spin paramagnetic effect when neglecting spin orbital scattering. It was reported that Fe$_{1.11}$Fe$_{0.6}$Se$_{0.4}$ with relatively low $T_c = 14$ K had a very high $\mu_0B_{ab}^{c2} = 50$ T [125]. It is likely that the disorder induced by Te(Se) substitution and excess of Fe in Fe-11
system results in dominance of spin paramagnetic effect [116, 119]. Similar to the case of 122-FeAs, $\mu_0B_{c2}$ shows isotropic behaviour at low temperature. High magnetic field measurements show that if the zero temperature value of $\mu_0B_{c2}$ is much lower than the corresponding orbital limit, then the paramagnetic limit may play a role in pair breaking for both $B\parallel ab$ and $B\parallel c$ [123]. LiFeAs reveals a relatively low value of $\mu_0B_{c2}$ compared to other FBSs [Figure 2-12 (c)]. The estimated value of $\mu_0B_{c2}^e = 14.5$ T using equation 2-6 is in good agreement with the experimental value of 15 T [97]. On the other hand, the spin paramagnetic effect should be considered for $\mu_0B_{ab}^{c2}$. The $B_{c2}(T)$ and the related parameters of some FBSs are summarized in Table 2-4, where $B_{c2}^{ab}(T)$, $B_{c2}^{c}(T)$, $B_0^{c2}(T)$, $B_{c2}^{ab}(T)$ exp., $H_{c2}^{c}(T)$ exp., $\alpha$, $\gamma_h$, $\varepsilon_{ab}$, $\varepsilon_{c}\Delta$, and $G_i$ represent the upper critical field in the orbital limit for $B\parallel ab$ and for $B\parallel c$, the Pauli limit for $B_{c2}$, the experimental value of the upper critical field for $B\parallel ab$ and for $B\parallel c$, the Maki parameter, the upper critical field anisotropy, the coherence length for $B\parallel ab$, the coherence length for $B\parallel c$, the superconducting energy gap, and the Ginsburg number, respectively.

Figure 2-13 shows the normalize $B_{c2}$ as a function of normalized temperature for different families of FBSs for $B\parallel ab$ and $B\parallel c$. The normalized temperature dependence of $B_{c2}$ presents a linear increase with decreasing temperature for $B\parallel c$, and the results nearly collapse onto the same curve for different families of FBSs. Superconductivity for $B\parallel c$ is mainly destroyed by the orbital effect as the $B_{c2}$ values approach or fall slightly below the corresponding orbital limited field. The normalized temperature dependence of $B_{c2}$ reveals different behaviour for $B\parallel ab$, and it is suppressed far below the corresponding orbital limited field. In the case of $B\parallel ab$, $B_{c2}^{ab}(T)$ can be described using the WHH model and equation 2-2. It should be noted that the paramagnetic-limited effect plays a key role for $B\parallel ab$. Also, the curvature is
affected by the Maki parameter value. For example, LiFeAs with the lowest $\alpha$ shows lower curvature, and Tl$_{0.58}$Rb$_{0.42}$Fe$_{1.72}$Se$_2$ with the highest $\alpha$ reveals a greater curvature (Figure 2-13).

![Figure 2-13 Normalized $B_{c2}$ as a function of normalized $T$ for different families of FBSs for (a) $B//c$ and (b) $B//ab$. The dashed lines in (b) represent the fitting based on the WHH model [118].](image)

For all FBSs except 11-FeSe, the spin paramagnetic effect is important in pair breaking and suppression of superconductivity for $B//ab$, as $B_{c2}^{ab}$ ($T$) shows a concave shape at low temperature. The enhancement of $B_{c2}^c(T)$ at low temperature may be correlated to the multiband effect, however.

### 2.2.5.2 Anisotropy of upper critical field

The anisotropy parameter of $\mu_0B_{c2}$ ($T$), $\gamma_H$ ($T$), is defined as $\mu_0B_{c2}^{ab}$ ($T$)/$\mu_0B_{c2}^c(T)$. $\gamma_H$ ($T$) near $T_c$ is moderate for 1111-FeAs ($\gamma_H = 5-8$) and small for other FBSs ($\gamma_H = 2-3$). The $B_{c2}$ anisotropy, $\gamma_H$, is mostly affected by the temperature dependence in two directions. For example, Nd-1111 compound shows strong temperature dependence in its anisotropy. The value of $\gamma_H$ is 5 at low temperature and increases to 10 at $T =$
Also, for Ba-122 and Fe-11, it decreases with decreasing temperature [113], and finally, for 122*, the $\gamma_{H}$ is about 2 close to $T_c$, but shows a maximum of $\gamma_{H} \approx 3.6$ around 27 K [112]. It is interesting to note that the anisotropy for the K and Co doped BaFe$_2$As$_2$, FeTe$_{0.6}$Se$_{0.4}$, and K$_{0.8}$Fe$_{1.76}$Se$_2$ systems is as large as 3 close to $T_c$, but it drops toward 1 as the temperature is decreased to 0 K [112]. Nearly isotropic superconductivity in FBSs is a remarkable physical phenomenon, which is not completely understood yet. This phenomenon is different from what occurs in other layered superconductors. It is likely that the three dimensional electronic structure of FBSs compared to other layered superconductors contributes to nearly isotropic superconductivity in these compounds. Also, the coherence length of FBSs is comparable to or even bigger than the distance between neighbouring FeAs layers. Therefore, it is likely that interlayer coupling occurs. As mentioned above, the spin paramagnetic effect is dominant for $B//ab$ and orbital pair breaking effect for $B//c$. Generally, orbital pair breaking effect is more effective near $T_c$, where the limiting effect can be due to Zeeman splitting with increasing magnetic field. Therefore, Pauli paramagnetic effect may become strong enough to overcome the orbital pair breaking mechanism, resulting in close value for $\mu_0B_{c2}^{ab}$ and $\mu_0B_{c2}^{c}$. In addition, the temperature dependence of $\mu_0B_{c2}$ and its anisotropy is more complicated due to multiband superconductivity in FBSs [118]. It should be noted that $\gamma(T)$ decreases with decreasing temperature for all FBSs. It is likely that multiband effect is responsible for this behaviour.

2.2.6 Thermal fluctuation in FBSs

FBSs are type-II superconductors with very high $B_{c2}$ and a large amount of magnetic field penetration in the mixed state of $\mu_0B-T$ phase diagram. Therefore,
very interesting vortex physics is expected in these materials. In the mixed state, thermal fluctuation effects on the vortex behaviour can be estimated from the Ginsburg number, \( G_i \),

\[
G_i = \frac{1}{2} \left( \frac{k_B T_c}{\mu_0 B_c(0) \xi_{ab}(0) \xi_c(0)} \right)^2
\]

which is the squared ratio of thermal energy \( k_B T_c \) to condensation energy in the volume occupied by a Cooper pair [126]. Here, \( k_B \) is the Boltzmann constant, \( \mu_0 \) is the vacuum permeability, \( \mu_0 B_c(0) \) is the thermodynamic critical field \( (\varphi_0) \) is the magnetic flux quantum, \( \lambda_{ab} \) is the London penetration depth in the \( ab \)-plane, \( \xi_{ab}(0) \) and \( \xi_c(0) \) are the coherence lengths in the \( ab \)-plane and \( c \)-axis at \( T = 0 \) K, respectively. The \( G_i \) can be rewritten as follows [108, 124]:

\[
G_i \propto \gamma_m^{4/3} m^4 T_c^4 n^{-8/3}
\]

where \( \gamma_m \) is the mass anisotropy, \( m \) is the effective mass, and \( n \) is the carrier density. Therefore, \( G_i \) can be enhanced if \( T_c \), \( m \), or \( \gamma_m \) is increased, or if the carrier density decreases. In conventional superconductors, vortex thermal fluctuation is negligible due to small \( G_i \) on the order of \( 10^{-10} - 10^{-6} \). On the other hand, static and dynamic properties of vortex structure can be affected strongly by thermal fluctuation in cuprates with high \( G_i \approx 10^{-3} \). Thermal fluctuations also influence vortex properties in FBSs with \( G_i \) values of \( 10^{-4} - 10^{-2} \). Different phases of vortex matter from conventional Abrikosov lattices to the vortex glass/liquid phases exist in these materials [108]. Many FBSs also reveal strong vortex fluctuations similar to those in low anisotropy cuprates, even though their \( T_c \) is 2-3 times lower than for cuprates. It is likely that the low carrier density and large effective mass of FBSs result in a
relatively large thermal fluctuation effect in these materials. Although 11-FeCh compounds have smaller $T_c$ values than 122-FeAs materials (Table 2-4), their $G_i$ is larger, which could be related to the lower carrier density and significant mass enhancement of 11-FeCh compounds.

2.2.7 Thermally activated flux flow

Thermally activated flux flow, TAFF, behaviour is the consequence of the thermal fluctuation in FBSs, where the vortex bundles hop between neighbouring pinning centres. According to the TAFF model, the resistivity in the TAFF can be described as [126, 127]

$$\rho = \left(\frac{2\nu_0 LB}{J}\right) \exp \left(-\frac{Jc_0 BVL}{T}\right) \sinh \left(\frac{JBVL}{T}\right)$$  \hspace{1cm} 2-12

where $\nu_0$ is an attempt frequency for a flux bundle hop, $L$ is the hopping distance, $B$ is the magnetic field, $J$ is the applied current density, $Jc_0$ is the critical current density in the absence of flux creep, $V$ is the bundle volume, and $T$ is the temperature. For very small $J$ and $JBVL \ll 1$, equation 2-12 can be summarized as

$$\rho = \left(\frac{2\rho_c U}{T}\right) \exp \left(-\frac{U}{T}\right) = \rho_{0f} \exp \left(-\frac{U}{T}\right)$$  \hspace{1cm} 2-13

Where $U = Jc_0 BVL$ is the thermal activation energy and $\rho_c = \nu_0 LB/Jc_0$, which is usually considered to be temperature independent. If $\rho_{0f}$ is a constant, then

$$\ln \rho(T, \mu_0 B) = \ln \rho_{0f} - U(T, \mu_0 B)/T$$  \hspace{1cm} 2-14

Furthermore, based on the condensation model [127]

$$U(T, \mu_0 B)/T = (\mu_0 B_c(t))^2 \varepsilon^n(t)$$  \hspace{1cm} 2-15
Here, $\mu_0B_c$ is the thermodynamic critical field, $\varepsilon$ is the coherence length, $t = T/T_c$ and $n$ depends on the dimensionality of the vortex system with a range of 0 - 3.

At $T$ close to $T_c$, $\mu_0B_c \propto 1 - t$ and $\varepsilon \propto (1-T)^{1/2}$, so

$$ U(T, \mu_0B) = u_0(\mu_0B)(1 - t)^q $$

where $q = 2-n/2$.

For $n = 2$, we have $U(T, \mu_0B) = u_0(\mu_0B)(1 - t)$, and then we can write the Arrhenius relation for $\ln \rho - 1/T$:

$$ \ln\rho(T, \mu_0B) = \ln\rho_0(\mu_0B) - U_0(\mu_0B)/T $$

and finally,

$$ U_0(\mu_0B) = -\partial \ln\rho(T, \mu_0B)/\partial T^{-1} $$

According to equation 2-18 if we plot $\ln\rho$ vs. $1/T$, the linear part should appear in the TAFF region, and the slope of the linear part corresponds to the $U_0(\mu_0B)$. $\ln\rho_0 (\mu_0B)$ can be obtained using the y-intercept of the plot.

Figure 2-14 shows that the experimental data can be fitted very well using the Arrhenius relation for NdFeAsO$_{0.7}$F$_{0.3}$ [105], (b) Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$ [128], (c) Fe$_{1.03}$Te$_{0.55}$Se$_{0.45}$ [129], and (d) Tl$_{0.58}$Rb$_{0.42}$Fe$_{1.72}$Se$_2$ [106] single crystals. The good linear behaviour demonstrates that temperature dependence of $U(T, \mu_0B)$ is almost linear. The log$\rho(T, \mu_0B)$ values for NdFeAsO$_{0.7}$F$_{0.3}$ and Fe$_{1.03}$Te$_{0.55}$Se$_{0.45}$ extrapolate to same temperature, $\sim T_c$. This is not the case, however, for Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$ and Tl$_{0.58}$Rb$_{0.42}$Fe$_{1.72}$Se$_2$ single crystals.
Figure 2-14 Arrhenius plots for the resistivity in various fields (B//c) for (a) NdFeAsO$_{0.7}$F$_{0.3}$ [105], (b) Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$ [128], (c) Fe$_{1.03}$Te$_{0.55}$Se$_{0.45}$ [129], and (d) Tl$_{0.58}$Rb$_{0.42}$Fe$_{1.72}$Se$_2$ [106] single crystals.

As shown in Figure 2-15 the field dependence of $U(T, \mu_0B)$ is a power law dependence ($U(T, \mu_0B) \propto \mu_0B^{-\alpha}$) on magnetic field for NdFeAsO$_{0.7}$F$_{0.3}$ and Fe$_{1.03}$Te$_{0.55}$Se$_{0.45}$ single crystal. The fitted $\alpha$ value of 0.5 for Fe$_{1.03}$Te$_{0.55}$Se$_{0.45}$ indicates that vortices are mainly pinned by collective point defects in high-field region. In the case of NdFeAsO$_{0.7}$F$_{0.3}$, field dependence of $U(T, \mu_0B)$ for B//c and B//ab are considerably different. $U(T, \mu_0B)$ for B//ab reveals a weak power law decrease $U(T, \mu_0B) \propto B^{0.17}$ over the entire field. The magnetic field dependence of pinning potential show a different behaviour for B//c, however, and it exhibits a conventional field dependence characteristic of TAFF. The value of $U(T, \mu_0B)$ is constant at low
field $B < 3$ T, where single vortex pinning dominates, followed by a power-law decrease $U(T, \mu_0 B) \propto B^{-1.1}$, representing the existence of collective pinning for $B > 3$ T [105]. On the other hand, situation is remarkably different for $\text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2$ single crystal, and $U(T, \mu_0 B)$ drops very slowly with field as $B^{-0.09}$ and $B^{-0.13}$ for $B//ab$ and $B//c$, respectively [128]. $\text{Tl}_{0.58}\text{Rb}_{0.42}\text{Fe}_{1.72}\text{Se}_2$ single crystal exhibits similar behaviour, but $U(T, \mu_0 B)$ drops faster with field as $B^{-0.6}$ and $B^{-0.7}$ for $B//ab$ and $B//c$, respectively. The value of $U(T, \mu_0 B)$ for $\text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2$ is three times larger than for $\text{NdFeAsO}_{0.7}\text{F}_{0.3}$ crystal and is almost field independent [128].

Figure 2-15 Field dependence of $U(T, \mu_0 B)$ for $\text{NdFeAsO}_{0.7}\text{F}_{0.3}$ (red squares) [105], (b) $\text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2$ (black squares) [128], (c) $\text{Fe}_{1.03}\text{Te}_{0.55}\text{Se}_{0.45}$ (green squares) [129], and (d) $\text{Tl}_{0.58}\text{Rb}_{0.42}\text{Fe}_{1.72}\text{Se}_2$ (blue squares) [106] single crystals. The closed and open symbols represent $B//ab$ and $B//c$, respectively.

2.2.8 Critical current density

Figure 2-16 presents magnetization hysteresis loops (MHL) of (a) $\text{SmFeAs}_{1-x}\text{F}_x$ [130], (b) $\text{(Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ [131], (c) $\text{Ba(Fe}_{1-x}\text{Co}_{x})_2\text{As}_2$ [132], (d) $\text{LiFeAs}$ [133], $\text{FeSe}_{0.7}\text{Te}_{0.3}$ [134], and (f) $\text{K}_{x}\text{Fe}_{2-x}\text{Se}_2$ [135] samples at different temperatures for...
$B//c$. The symmetric curves indicate that bulk current dominates in all the samples. The occurrence of flux jumping in an FBS was reported by our group in $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ for the first time [128]. Flux jumping can be detected in type II superconductors in large samples with high $J_c$ and small specific heat, if ramp rate of magnetic field is sufficiently fast. Figure 2-16 (b) and (d) shows the flux jumping in $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ [131] and LiFeAs [133] single crystals.

The critical current density, $J_c$, is calculated from the width of MHL using Bean model [136]. According to this model, for a rectangular shaped crystal with dimensions $c < a < b$ and $B//c$, the $J_c$ is obtained by

$$J_c=20\Delta M/ [a (1-a/3b)] \quad (2.19)$$

where $a$ and $b$ are the sample dimensions and $\Delta M$ is the difference between the magnetization values for decreasing and increasing field at a particular applied field.

FBSs reveal high critical current density, $J_c$, which is almost field independent at low temperature. This is in good agreement with strong pinning associated with atomic scale defects resulting from chemical doping, nanoscale coherence lengths, and high value of $B_{c2}$ in iron-based superconductors. The weak field dependence of $J_c$ suggests that FBSs have superior $J_c$ behaviour, which is favourable for potential application in high magnetic fields. Figure 2-16 shows the magnetic field dependence of $J_c$ at different temperatures for $B//c$ for (a) SmFeAs$_{1-x}$F$_x$ [137], (b) $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ [131], (c) Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [132], (d) LiFeAs [133], (e) FeSe$_{0.5}$Te$_{0.5}$, and (f) K$_{0.58}$Fe$_{1.58}$Se$_2$ [135] single crystals.
Figure 2-16 Magnetization hysteresis loops at different temperatures for $B//c$ for (a) SmFeAsO$_{1-x}$F$_x$ [130], (b) (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ [131], (c) Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [132], (d) LiFeAs [133], (e) FeSe$_{0.7}$Te$_{0.3}$ [134], and (f) K$_x$Fe$_{2-y}$Se$_2$ [135] samples.

Zhigadlo et al. reported $J_c$ of $2 \times 10^6$ A/cm$^2$ at $T = 5$ K for SmFeAsO$_{1-x}$F$_x$ single crystal [137], which was almost field independent up to 13 T. A fishtail effect and very high $J_c$ of $5 \times 10^6$ A/cm$^2$ at $T = 4.2$ K has been reported for Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ single crystal [138]. Prozorove et al. revealed $J_c$ of $2.6 \times 10^6$ A/cm$^2$ at $T = 5$ K for
electron doped Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ single crystal [139]. Their study demonstrated the fishtail effect as well as a very large magnetic relaxation rate, which were analysed using the collective pinning and creep model. Yamamoto et al. reported the lower $J_c$ of $4 \times 10^5$ A/cm$^2$ at $T = 4.2$ K for BaFe$_{1.8}$Co$_{0.2}$As$_2$ single crystal and a fishtail effect for the same compound [132].

Figure 2-17 Magnetic field dependence of $J_c$ at different temperatures for $B//c$ for (a) SmFeAs$_{1-x}$F$_x$ [137], (b) (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ [131], (c) Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [132], (d) LiFeAs [133], (e) FeSe$_{0.5}$Te$_{0.5}$, and (f) K$_{0.58}$Fe$_{1.58}$Se$_2$ [135] samples.
Also, the fishtail effect has been observed in LiFeAs single crystal for $B//c$. No fishtail effect was detected, however, for the same sample for $B//ab$ [140]. In the case of 11-FeCh system, Tean et al. reported that $J_c$ was $\sim 1 \times 10^5$ A/cm$^2$ at low temperature for FeTe$_{0.61}$Se$_{0.39}$ single crystal [55]. Li et al. reported $J_c$ of $10^{-3}$ A/cm$^3$ for K$_x$Fe$_{2-y}$Se$_2$ single crystal [141]. The $J_c$ of K$_x$Fe$_{2-y}$Se$_2$ was enhanced to $7.4 \times 10^4$ at $T = 1.8$K by post annealing and quenching, which is 50 times higher than that of as-grown single crystal. The value of $J_c$ for post annealed K$_x$Fe$_{2-y}$Se$_2$ single crystal is still smaller than for FeTe$_{0.61}$Se$_{0.39}$ and other iron pnictides [130, 142].

The high values of $J_c$ reveal highly effective pinning in FBSs and give a reason to expect that it can be further increased by suitable material design and treatment. Pinning can be further increased by introducing artificial defects through heavy ion or neutron irradiation [143, 144, 145, 146, 147]. It was demonstrated that thermal neutron irradiation leads to $J_c$ improvement by a factor of 1.5-3 [144], while heavy ion irradiation using Au [145], Pb [148, 149], or Ta [143] ions increases $J_c$ by a factor of 3-10 as a result of columnar defects [55, 143, 145].

2.2.9 Peak effect and second magnetization peak effect

For some superconductors, $J_c$ obtained from MHLs increases with magnetic field after the first peak of penetration field. This is the so-called second magnetization peak (SMP) or fishtail effect. In low temperature superconductors, e. g. MgB$_2$, Nb$_3$Sn, etc., the SMP corresponds to a hump feature in $J_c(B)$ far below the $B_{c2}$, while the peak effect (PE) occurs near $B_{c2}(T)$ [150]. It is suggested that PE is associated with the rapid softening of the flux line lattice (FLL), and it occurs in the same part of the ($B$-$T$) phase diagram where phase transitions in the FLL are expected to take place [151]. The peak effect in unconventional superconductors such as YBCO
occurs just below the vortex liquid phase transition, far below $B_{c2}$. It is suggested that the occurrence of PE is associated with increased pinning by twin boundaries. It is likely that softening of the shear modulus near the melting transition allows vortex lattice to properly adjust to the pinning centres, thereby increasing the pinning force [152]. The occurrence of an SMP has been reported in SmFeAsO$_{0.9}$F$_{0.1}$ [153], where the authors claimed weak and collective pinning for the system. The SMP was observed only for the samples with near optimal doping for NdFeAsO$_{0.85}$ [143] and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [139, 154, 155, 156], and weak and collective pinning are concluded for most studies. The SMP has also been observed in optimally doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [138, 154]. It is worth noting, however, that most forms of inhomogeneity, such as $T_c$ variation, impurity phase, doping variation, etc., might prevent the occurrence of SMP [157]. For example, under-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ systems with $T_c$ below 28 K do not show the SMP. Also, SMP has been reported for Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ [154, 158], LiFeAs [140], FeTe$_{1-x}$Se$_x$ [134], and PrFeAsO$_{0.6}$F$_{0.1}$ [159].

The SMP has been detected for electron and hole doped Ba-122 single crystals for $B//c$ [154]. The SMP disappeared for $B//ab$, indicating an anisotropic effect in the flux pinning for these compounds [154]. The peak moves to a higher field with decreasing temperature [138]. Similar behaviour was observed for $RE$Ba$_2$Cu$_3$O$_{7-\delta}$ [160]. It is likely that the SMP has same mechanism in both compounds [138]. Sun et al. suggest that SMP originates from the small-size normal core pinning and it is the result of crossover from elastic collective creep to the plastic vortex creep [139].
Table 2-5 SMP in 122-FeAs and 11 single crystals.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$T_c$(K)</th>
<th>Measurement</th>
<th>Results</th>
<th>Mechanism of SMP</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Ba}<em>0.6\text{K}</em>{0.4}\text{Fe}_2\text{As}_2$ [138]</td>
<td>36.2</td>
<td>MHL</td>
<td>Peak position dropping to lower field with increasing temperature</td>
<td>Same mechanism as $\text{REBa}_2\text{Cu}<em>3\text{O}</em>{7-\delta}$</td>
</tr>
<tr>
<td>$\text{Ba}<em>{0.68}\text{K}</em>{0.32}\text{Fe}_2\text{As}_2$ [154]</td>
<td>38.5</td>
<td>MHL</td>
<td>SMP observed for $B//c$, No SMP for $B//ab$</td>
<td>SMP originates from small size normal core pinning effect and crossover from elastic collective creep to the plastic vortex creep</td>
</tr>
<tr>
<td>$\text{Ba}<em>{0.72}\text{K}</em>{0.28}\text{Fe}_2\text{As}_2$ [161]</td>
<td>32.7</td>
<td>Magnetic relaxation</td>
<td>SMP in MHL Magnetic relaxation measurements</td>
<td>Crossover in the pinning mechanism from collective to plastic pinning as the field increases</td>
</tr>
<tr>
<td>$\text{BaFe}<em>{1.91}\text{Ni}</em>{0.09}\text{As}_2$ [154]</td>
<td>18.5</td>
<td>MHL</td>
<td>SMP observed for $B//c$, No SMP for $B//ab$</td>
<td></td>
</tr>
<tr>
<td>$\text{BaFe}<em>{1.82}\text{Ni}</em>{0.18}\text{As}_2$ [162]</td>
<td>8</td>
<td>Measuring flux creep over the SMP for $B//c$ and $B//ab$</td>
<td>SMP observed for $B//ab$ and $B//c$</td>
<td>SMP not associated with a softening in vortex pinning before melting, nor a change of pinning regime within the collective pinning model.</td>
</tr>
<tr>
<td>$\text{BaFe}<em>{0.6}\text{Co}</em>{0.1}\text{As}_2$ [142]</td>
<td>22</td>
<td>MHL</td>
<td>Normalized pinning force as a function of reduced field, independent of $T$ around SMP</td>
<td>Dominant dense vortex pinning mechanism</td>
</tr>
<tr>
<td>$\text{BaFe}<em>{0.925}\text{Co}</em>{0.075}\text{As}_2$</td>
<td>25</td>
<td>MHL, Magnetic relaxation</td>
<td>SMP in MHL Magnetic relaxation measurements show a minimum in the relaxation rate placed in between the SMP onset and the peak field.</td>
<td>SMP is associated with a vortex structural phase transition from rhombic to square lattice that occurs at field and temperatures corresponding to the minimum point of the relaxation rate.</td>
</tr>
<tr>
<td>$\text{Ba(Fe}<em>{0.93}\text{Co}</em>{0.07})_2\text{As}_2$ [163]</td>
<td>22</td>
<td>Transport, MHL, Magnetic relaxation</td>
<td>SMP in MHL, very large magnetic relaxation rate</td>
<td>Crossover from collective to plastic creep regime in fields higher than the field at which SMP is maximum.</td>
</tr>
<tr>
<td>$\text{BaFe}<em>{1.85}\text{Co}</em>{0.15}\text{As}_2$ [154]</td>
<td>24.5</td>
<td>MHL</td>
<td>SMP observed for $B//c$, No SMP for $B//ab$</td>
<td>Crossover from collective to plastic creep regime in fields higher than the field at which SMP is maximum.</td>
</tr>
</tbody>
</table>
for under doped and over-doped samples.

<table>
<thead>
<tr>
<th>Material</th>
<th>Temperature</th>
<th>Relaxation</th>
<th>SMP Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeTe$<em>{0.7}$Se$</em>{0.3}$ [134]</td>
<td>10.9</td>
<td>MHL, Magnetic relaxation</td>
<td>SMP in MHL for B//c and position shifts with decreasing temperature and relaxation time. Vortex configuration that determines the SMP at specific B value does not depend on the temperature in the range of 0.4T$_c$-0.75T$_c$, and temperature effect on vortex-vortex and vortex-defect interaction near SMP is negligible.</td>
</tr>
<tr>
<td>FeSe$<em>{0.5}$Te$</em>{0.5}$ [165]</td>
<td>14.3</td>
<td>MHL, Magnetic relaxation</td>
<td>SMP in MHL for B//c. Possibility of an order-disorder transformation across SMP.</td>
</tr>
</tbody>
</table>

### 2.2.10 Pinning mechanisms

In order to understand the nature of pinning in FBSs, it is useful to study the temperature and field dependencies of vortex pinning force, $F_p = \mu_0 B J_{c}$. According to Dew-Hughes theory, if a dominant vortex pinning mechanism exists, then the normalized pinning force at different temperatures should collapse into one curve, and a scaling law of $F_p \approx h^p (1-h)^q$ will be observed. Here, $p$ and $q$ are two parameters whose values depend on the origin of pinning mechanism, and $h$ is the reduced field, $h = B/B_{irr}$, where $B_{irr}$ is the magnetic field where $J_c(B,T)$ extrapolates to zero. In type–II superconductors, there are two main categories which contribute to the flux pinning: first, spatial variations in the charge carrier mean free path ($\delta l$) and secondly, pinning arising because of spatial fluctuations in $T_c$ across the sample ($\delta T_c$) [126, 166]. Based on the Dew-Hughes model, different values of $p$ and $q$ are expected, depending on the special pinning mechanism involved, and crucial information can be obtained by analysing the scaled $F_p(h)$ curves. For example, in the case of $\delta l$ pinning, $p = 1$ and $q = 2$ with maximum $h = 0.33$ are expected for point pins, while $p = 0.5$ and $q = 2$ with $h_{\text{max}} = 0.2$ are related to surface pinning such by as grain boundaries. There is no maximum for $h_{\text{max}}$ in the case of volume $\delta l$ pinning with $p = 0$ and $q = 2$. For $\delta T_c$ pinning, the maximum pinning force is anticipated at
higher $h_{\text{max}}$ values. For example for point pins with $p = 2$ and $q = 1$, the maximum $h$ occurs at $h_{\text{max}} = 0.67$. For surface pins, $h_{\text{max}} = 0.6$ with $p = 1.5$ and $q = 1$, and finally, for volume pins, $h_{\text{max}} = 0.5$ with $p = 1$ and $q = 1$. Figure 2-18 presents the normalized pinning force as a function of reduced magnetic field in different families of FBSs at different temperatures below $T_c$ of each compound. For all cases, the reduced magnetic field is scaled as $B/B_{\text{irr}}$. Different values of $p$ and $q$ were obtained for different FBSs, as summarised in Table 2-6.

Table 2-6 Summary of $p$, $q$, and $h_{\text{max}}$ values for different families of FBS.

<table>
<thead>
<tr>
<th>Material</th>
<th>$p$</th>
<th>$q$</th>
<th>$h_{\text{max}} = p/(p+q)$</th>
<th>Peak position</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba$<em>{0.6}$K$</em>{0.4}$Fe$_2$As$_2$</td>
<td>1</td>
<td>2</td>
<td>0.33</td>
<td>0.33</td>
</tr>
<tr>
<td>Ba$<em>{0.68}$K$</em>{0.32}$Fe$_2$As$_2$</td>
<td></td>
<td></td>
<td></td>
<td>0.43</td>
</tr>
<tr>
<td>BaFe$<em>{1.8}$Co$</em>{0.2}$As$_2$</td>
<td>1.67</td>
<td>2</td>
<td>0.45</td>
<td>0.45</td>
</tr>
<tr>
<td>BaFe$<em>{1.85}$Co$</em>{0.15}$As$_2$</td>
<td></td>
<td></td>
<td></td>
<td>0.37</td>
</tr>
<tr>
<td>BaFe$<em>{1.29}$Ru$</em>{0.71}$As$_2$</td>
<td>1.95</td>
<td>2.5</td>
<td>0.44</td>
<td>0.45</td>
</tr>
<tr>
<td>BaFe$<em>{1.91}$Ni$</em>{0.09}$As$_2$</td>
<td></td>
<td></td>
<td></td>
<td>0.32</td>
</tr>
<tr>
<td>FeTe$<em>{0.7}$Se$</em>{0.3}$</td>
<td></td>
<td></td>
<td></td>
<td>0.27</td>
</tr>
<tr>
<td>FeTe$<em>{0.6}$Se$</em>{0.4}$</td>
<td>1.54</td>
<td>3.8</td>
<td>0.28</td>
<td>0.28</td>
</tr>
<tr>
<td>K$<em>x$Fe$</em>{2-y}$Se$_2$</td>
<td>0.86</td>
<td>1.83</td>
<td>0.32</td>
<td>0.33</td>
</tr>
</tbody>
</table>

In the case of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ single crystal [138], it is suggested that the observed value of $h_{\text{max}} = 0.33$ is likely to be related to small-size normal cores, such as with arsenic deficiency. Yamamoto et al. obtained the values of $p = 1.67$ and $q = 2$ with $h_{\text{max}} = 0.45$ for BaFe$_{1.8}$Co$_{0.2}$As$_2$ single crystal [142]. They suggest that the observed temperature independent and symmetric $F_p(h)$ curves with $h_{\text{max}} = 0.45$ are correlated to a dense vortex pinning nanostructure, likely due to the inhomogeneous distribution of cobalt ions. The peak at $h_{\text{max}} = 0.45$ in BaFe$_{1.29}$Ru$_{0.71}$As$_2$ single crystal indicates a
dense vortex pinning nanostructure which results from inhomogeneous distribution of the Ru dopant [167]. On the other hand, Sun et al. noticed that the arsenic deficiency or inhomogeneous distribution of dopant could not be responsible for strong pinning in FBSs, and they claimed that the strong vortex pinning could have resulted from fluctuating magnetic/structural domains [154]. In their study, they obtained the values of $h_{\text{max}} = 0.43$, 0.37, and 0.32 for Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$, BaFe$_{1.85}$Co$_{0.15}$As$_2$, and BaFe$_{1.91}$Ni$_{0.09}$As$_2$ single crystals, respectively. According to their results, $B_{c2}$ and $B_{\text{sp}}$ decrease faster with decreasing temperature for BaFe$_{1.91}$Ni$_{0.09}$As$_2$ than for Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ and BaFe$_{1.85}$Co$_{0.15}$As$_2$ single crystals. Therefore BaFe$_{1.91}$Ni$_{0.09}$As$_2$ single crystal with larger $\Delta T_c$ compared to Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ or BaFe$_{1.85}$Co$_{0.15}$As$_2$ seems to be inhomogeneous. In addition, the potassium doped 122 reveals the highest pinning among the three different single crystals because this compound has the most fluctuating domains. Therefore, they conclude that the As deficiency or inhomogeneous distribution of dopant could not play an important role in determining the strong pinning in FBSs [154]. The obtained value of $p = 1.54$ and $q = 3.8$ with $h_{\text{max}} = 0.28$ for FeTe$_{0.6}$Se$_{0.4}$ indicate that vortex pinning is by a mixture of the surface and the point core pinning of the normal centres [168]. Das et al. reported $h_{\text{max}} = 0.36$ for FeTe$_{0.5}$Te$_{0.5}$, with $p = 1.65$ and $q = 2.95$. They suggested that point pinning alone cannot explain the pinning mechanism of this compound. The variation in the values of $h_{\text{max}}$ for FeTe$_{1-x}$Se$_x$ could be a result of change in the pinning force due to change in the Se and Te concentration. Here, it is worthwhile to notice that different groups use different criteria to normalize the field. For example, Das et al. employed $B_{c2}$ for field normalization, while Yadav et al. used $B_{\text{irr}}$ to normalize the field. Therefore, the differences arising from the
different criteria used for estimation of $h_{\text{max}}$ must be taken into an account, as $B_{\text{irr}}$ and $B_{c2}$ could have different temperature dependencies [165].

Figure 2-18 Reduced field dependence of normalized flux pinning force at various temperatures for (a) Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ [138], (b) Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [142], (c) BaFe$_{1.29}$Ru$_{0.71}$As$_2$ [167], (d) BaFe$_{1.91}$Ni$_{0.09}$As$_2$ [154], (e) FeTe$_{0.6}$Se$_{0.4}$ [168], and (f) K$_x$Fe$_{2-y}$Se$_2$ [135].

Lie et al. [135] reported the temperature independent scaling law of $F_p(h)$ with $p = 0.86$, $q = 1.83$, and $h_{\text{max}} = 0.33$ for K$_x$Fe$_{2-y}$Se$_2$ single crystal, which is close to the expected values for core normal point pinning. The scaling law was observed for all
temperatures above 10 K for this single crystal. It is likely that the normal point pinning mechanism is dominant above 10 K. They suggest that the point pinning centres could come from the random distribution of Fe vacancies after quenching [135]. It is likely that complex domain structures due to orthorhombic distortion could be responsible for the intrinsic pinning in FBSs. The existence of a complex domain structure was reported in the parent compound of AFe$_2$As$_2$ based on transmission electron microscopy [169], polarized light microscopy, and high energy synchrotron [170]. Moreover, it was reported that intertwined orthorhombic magnetic/structural domains could play a key role in the strong intrinsic pinning in under-doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals [171]. There is no evidence, however, for the presence of disordered structural domains in the optimally doped compound.

### 2.2.11 Application of FBSs

FBSs are favourable for practical applications due to their high $B_{c2}$ and high isotropic $J_c$. They are attractive for electrical power and magnetic applications, while the coexistence of magnetism and superconductivity makes them interesting for spintronic applications. Shortly after the discovery of FBSs, fabrication of La-1111 and Sm-1111 wires using the in situ powder-in-tube (PIT) method was reported by Gao et al. (Figure 2-19) [172, 173]. The PIT wires reveal a self-field $J_c = 4000$ A/cm$^2$ at 5 K and weak field dependence of $J_c$, which was promising for the practical applications. Later, the transport $J_c = 2700$ A/cm$^2$ was obtained for Sm-1111 wires using ex-situ PIT and annealing at low temperature. Also, the fabrication of (Sr,K)Fe$_2$As$_2$ wires using PIT methods with transport $J_c = 1200$ A/cm$^2$ at $T = 4.2$ K was reported by the same group [174, 175]. They employed ex-situ PIT method for the same compounds and obtained transport $J_c = 3750$ A/cm$^2$ at $T = 4.2$K [176].
Tagano et al. reported a self-field $J_c = 1104$ A/cm$^2$ at $T = 4.2$ K for Ag- sheathed (Ba,K)Fe$_2$As$_2$ wires using the ex-situ PIT method [177]. Also, the fabrication of wire for a compound in the FeCh-11 family with $J_c = 1000$ A/cm$^2$ was reported [178]. Further improvement of the transport $J_c$ is crucial, however, for practical application of FBS wires. Also, the texturing of grain boundaries in FBS wires needs to be considered to overcome the weak-link behaviour of high angle grain boundaries.

![Figure 2-19 Cross-section of SmFeAsO$_{0.7}$F$_{0.3}$ wire [173].](image)

![Figure 2-20 (a) Magnetic field dependence of $J_c$ for Ba(Fe,Co)$_2$As$_2$ epitaxial thin film with magnetic field applied perpendicular to the plane of the film, with a magnetic optical image. (b) Cross-sectional TEM micrographs of films on STO/LSAT [179], with inset showing the corresponding selected area electron diffraction pattern.](image)

The first epitaxial films of Sr(Fe,Co)$_2$As$_2$ were fabricated on (La,Sr)(Al,Ta)O$_3$ (LSAT) single crystal substrate [180]. The thin films reveal nearly isotropic
superconductivity at low temperature, although the transport $J_c$ was only 20 kA/cm$^2$ at 4.2 K. The Sr(Fe,Co)$_2$As$_2$ thin films were extremely sensitive to humidity, and amusingly, the un-doped SrFe$_2$As$_2$ shows superconductivity below 25 K after exposure to humid air [181]. Ba(Fe,Co)$_2$As$_2$ epitaxial thin films show much higher stability against water vapour compared to the Sr(Fe,Co)$_2$As$_2$ thin films [182]. High quality Ba(Fe,Co)$_2$As$_2$ epitaxial thin films have been synthesized on SrTiO$_3$ (STO) or BiTiO$_3$ substrates by using a buffer layer technique, as shown in Figure 2-20 [179].

The Ba(Fe,Co)$_2$As$_2$ films reveal very high value of $J_c = 1$ MA·cm$^{-2}$ at $T = 4.2$ K, which is even higher than for bulk single crystals. The interesting point is that field dependence of $J_c$ is weak, even at $B = 14$ T for $B//c$, representing strong flux pinning of this thin film. Based on the microstructure analysis using transmission electron microscopy [Figure 2-20(b)], strong flux pinning is related to the line defects aligned along the c-axis. The magneto-optical images [Figure 2-20(a) inset] confirm that epitaxial thin films of Ba(Fe,Co)$_2$As$_2$ on STO/LSAT are uniform without any weak links [179]. The fabrication of (Ba,K)Fe$_2$As$_2$ thin film is complicated due to reactive nature of potassium. For the first time, (Ba,K)Fe$_2$As$_2$ thin films have been synthesized by an ex-situ pulsed laser deposition (PLD) method, where the un-doped BaFe$_2$As$_2$ films were deposited at room temperature and then post annealed with potassium shots in a silica tube [183]. Also, molecular beam epitaxy (MBE) was used for situ growth of (Ba,K)Fe$_2$As$_2$ thin film [184]. It is necessary to coat the (Ba,K)Fe$_2$As$_2$ thin films with polystyrene resin, however, as they degenerate quickly in air [30]. It should be noted that the fabrication of thin film for 1111-FeAs [185, 186] and 11-FeCh [187, 188] has been reported.
CHAPTER 3
EXPERIMENTAL TECHNIQUES

3.1 Sample Preparation

3.1.1 Ba(Fe_{1-x}M_x)_2As_2 Single crystals

Single crystals of Ba(Fe_{1-x})_2M_xAs_2 were grown using the high temperature self-flux method where M is a transition metal (Co, Ni). First FeAs and MAs were prepared by placing a mixture of As powder and Fe/M powder in a quartz tube. These were reacted at 600°C for 10 hours. A mixture of FeAs/MAs and Ba pieces was then placed in an alumina crucible. The whole assembly was sealed in a large quartz tube and heated to 1180°C for 15 hours, which was followed by a reaction at 1180°C for 10 hours [189]. The as-grown single crystal was cleaved and cut into a rectangular shape for measurements. The 122-FeAs samples were prepared at Seoul National University.

3.1.2 Fe_{1.06}Te_{0.6}Se_{0.4} Single crystals

The Fe_{1.06}Te_{0.6}Se_{0.4} single crystals were grown using the self-flux method [190]. The high purity Fe Pieces, Te lumps, and Se shot were weighed, mixed, and placed in an alumina crucible. The starting materials with composition of Fe_{1.06}Te_{0.6}Se_{0.4} under argon atmosphere from 200-800 mbar were packed into evacuated ampoules. The ampoules were heated at 970-1000°C for 12 hours and then slowly cooled down to 400°C at the cooling rate of 2°C/h. The 11-FeSe sample was made in the Max-Planck-Institut für Festkörperforschung, Heisenbergstr.
3.2 Transport and magnetotransport measurements

Transport properties were measured over a wide range of temperatures, $2 \text{ K} < T < 300 \text{ K}$, and magnetic fields up to 14 T using a Quantum Design Physical Properties Measurement System (PPMS).

3.2.1 Resistivity measurements

The four-probe technique was employed for resistivity measurements of samples. In this method, four copper wires are attached to the surface of the sample using silver paste, where the two middle contacts are for the voltage and the two outer contacts are for the current. As the current passes through the end of the sample, it generates a voltage that is proportional to the resistivity. Figure 3-1 shows the DC resistivity puck with three samples. The resistivity versus temperature was measured in magnetic field up to 13 T at the current of 5 mA.

![Figure 3-1 DC resistivity puck with three samples for resistivity measurements.](image)

Upper critical field, $B_{c2}$, was defined by the 90 % drop of resistance from transition temperature, $T_c$, of RT curves under different magnetic fields for both $B//ab$ and $B//c$ directions.
3.2.2 **Horizontal rotator for angular dependent transport measurements**

The horizontal rotator were employed to study the angular dependence of resistivity under different magnetic fields. Single crystalline samples are mounted on the removable sample holder. A thermometer is placed in direct contact with the sample holder. The sample holder rotates about the horizontal axis over a range of \(-10^\circ < \theta < 370^\circ\).

![Horizontal rotator and its sample holder for measuring the angular dependence of resistivity.

3.3 **Magnetic measurements**

3.3.1 **Vibrating Sample Magnetometer (VSM)**

The measurement is carried out by oscillating the sample near a detecting coil. The VSM option contains a linear motor transport for vibrating the sample, a coilset puck for detection, and the MultiVu software application.

The sample is attached to the end of a sample rod. The magnetic measurement can be accomplished for \(1.9 \: K < T < 400 \: K\) and for magnetic field up to 14 T. The magnetic
hysteresis loops are collected at different temperatures under different magnetic fields. For a long sample with rectangular cross-section with the dimensions of $l \times w$ perpendicular to the magnetic field direction, critical current density can be calculated using the extended Bean model:

$$J_c = 20\Delta M \left(l \left(1 - \frac{l}{3w}\right) \right) \quad l < w$$

Where $\Delta M$ is the width of the magnetization loop and can be calculated using the relation $\Delta M = M^r - M^r$, and $M^r$ and $M^r$ are negative and positive branches of the hysteresis loop, respectively. $J_c$ is in A/m$^2$, and $\Delta M$ is in emu/m$^3$.

Figure 3-3 Schematic drawing of VSM motor and its sample puck [191].
3.3.2 Magnetic measurement using superconducting quantum interference device (SQUID) 5 T magnetic property measurement system (MPMS)

Some of the magnetic measurements were conducted using the 5 T MPMS. These measurements included zero-field-cooling (ZFC) and field-cooling (FC) measurements, and magnetic hysteresis loops at several temperatures. The $J_c$ was calculated using the extended Bean model.
CHAPTER 4

MAGNETORESISTANCE, CRITICAL CURRENT DENSITY, AND MAGNETIC FLUX PINNING MECHANISM IN NICKEL DOPED BAFE₂AS₂ SINGLE CRYSTALS

4.1 Introduction

The discovery of the first iron based superconductors in fluorine doped LaOFeAs [192] aroused great interest due to the high upper critical field, high critical current density, and very high intrinsic pinning potential compared with MgB₂ and other conventional superconductors, and even cuprate superconductors. Later, superconductivity was discovered in potassium doped BaFe₂As₂ with maximum \( T_c = 38 \text{ K} \).

A second peak effect has been observed in Ba(Fe\(_{1-x}\)Co\(_x\))₂As₂ [139, 154, 193, 194], Ba(Fe\(_{1-x}\)Ni\(_x\))₂As₂ [154], and Ba\(_{1-x}\)K\(_x\)Fe₂As₂ single crystals [154, 195]. The second peak effect can result a crossover from the elastic collective creep to the plastic vortex creep¹³ regime, or it is associated with the structural phase transition from a rhombic lattice at low field to a square lattice above a transition field [194]. The absence of second magnetisation peak effect in CaFe₂₋xCoₓAs₂ indicates that the vortex dynamics in this compound is consistent with plastic creep rather than the collective creep model [196].

There are two main pinning mechanisms in type II superconductors: (I) \( \delta l \) pinning from spatial variation in the charge carrier mean free path, \( l \), and (II) \( \delta T_c \) pinning due to randomly distributed spatial variation in the transition temperature, \( T_c \). It has been reported that strong pinning centres in PrFeAsO₀.₉ and NdFeAsO₀.₉F₀.₁
arise from oxygen deficiency and dopant atoms, which results in pinning by local variations in the mean-free path [197]. Analysis of temperature and field dependencies of magnetic relaxation suggests a crossover from collective to the plastic creep regime near the peak position of second magnetization peak (SMP) [139] in Ba(Fe_{0.93}Co_{0.07})_{2}As_{2} single crystal. Strong intrinsic pinning due to structural domains in the superconducting orthorhombic phase [171] of Ba(Fe_{1-x}Co_{x})_{2}As_{2} is also observed. Similar results were found for BaFe_{1.8}Co_{0.2}As_{2}, where the temperature and field dependence of $J_c$ were attributed to the inhomogeneous distribution of Co atoms [132]. Furthermore, it has been suggested that the very large $J_c$ and fishtail effect at high temperature below $T_c$ in Ba_{0.6}K_{0.4}Fe_{2}As_{2} originates from the small-size normal core pinning centres [198].

Flux jumping has been observed in Ba_{1-x}K_{x}Fe_{2}As_{2} single crystal\textsuperscript{20}. Flux jumping, however, has not been reported in electron doped Ba(Fe_{1-x}Ni_{x})_{2}As_{2} so far. Also, there have been no reports on the pinning potential behaviour of Ni doped BaFe_{2}As_{2} single crystal. In this Chapter, the pinning potential behaviour in optimal electron doped BaFe_{1.9}Ni_{0.1}As_{2} single crystal is presented. Also a systematic study of the flux pinning mechanism of optimally doped BaFe_{1.0}Ni_{0.1}As_{2} single crystals point to the presence of a SMP or fishtail effect in magnetization hysteresis loops,(MHL) In order to understand the pinning mechanisms in this system, scaling analysis of the normalized pinning force as a function of reduced field was performed. Analysis using the Dew-Hughes model has suggested that point pins alone cannot explain the observed field variation of the pinning force density. According to the collective flux pinning model, field dependence of magnetization shows that flux pinning in the sample is dominated by the $\delta l$ pinning.
4.2 Experiment

Single crystal with the nominal composition BaFe$_{1.9}$Ni$_{0.1}$As$_2$ was prepared by a self-flux method. The as-grown single crystal was cleaved and cut into a rectangular shape for transport and magnetic measurement. The microstructure of the as-obtained samples and the morphology were studied using field emission scanning electron microscopy (FE-SEM, JEOL7500), operated at 50 kV. The transport properties were measured over a wide range of temperature and magnetic fields up to 13 T. with applied current of 5 mA using a physical properties measurement system (PPMS, Quantum Design). The temperature dependence of magnetization and MHL were measured using a magnetic properties measurement system (MPMS, Quantum Design). The critical current density was calculated using Bean model.

4.3 Results and discussions

Figure 4-1 shows a field emission scanning electron microscope (FE-SEM) image of the cleaved edge of a BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal. The FE-SEM image shows that BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal has a typical two-dimensional (2D) crystal structure.

Figure 4-1 FE-SEM image of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal.
The temperature dependence of resistivity of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal at zero magnetic field is shown in Figure 4-2. The resistivity decreases with decreasing temperature from 300 K to 19.4 K, supporting metallic behaviour of this compound. The resistance drops to zero at the onset $T_c$, $T_{c_{on}} = 17.6$ K, which is lower than that reported in Ref. [14].

![Figure 4-2 Temperature dependence of resistivity for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal at zero field.](image)

The superconducting transition width, $\Delta T_c = 0.8$ K, was calculated using the temperature difference between 90% and 10% values in the drop-off of the resistivity at zero magnetic field, which indicates a sharp superconducting transition temperature and high quality single crystal.

Figure 4-3 show the temperature dependence of the resistivity under different magnetic fields up to 13 T. The onset of superconductivity slowly shifts to lower temperature with increasing magnetic field, which is related to the nearly isotropic superconductivity in 122 family [199].
Figure 4-3 Temperature dependence of resistivity under different magnetic fields up to 13 T.

The upper critical field, $B_{c2}$, is extracted from the $\rho T$ curves under different magnetic field for $B//c$ and $B//ab$ and it is characterized as the field at which the resistance becomes 90% of the normal state resistivity, $\rho_n$. Also the resistivity onset field $B_{10}(T)$ and $B_{0.5}(T)$ defined by $\rho(T, B) = 0.1\rho_n(T,B)$ and $0.05\rho_n(T,B)$, respectively. The $B_{0.5}(T)$ are close to the irreversibility field, $B_{irr}$, which qualifies the onset of vortex critical state [200]. Figure 4-4 shows the temperature dependence of all these fields for $B//ab$ and $B//c$. The estimated slopes for $B_{c2}$ and $B_{irr}$ are -6.09 and -4.22 TK$^{-1}$ for $B//c$. Different slopes, $dB_{c2}/dT$, have been reported for Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ single crystal, ranging from 5.42 TK$^{-1}$ for $x = 0.09$ [154] to the high value of 9.9 TK$^{-1}$ for $x = 0.1$ [201]. The upper critical field was estimated by using the conventional one-band Werthamer–Helfand–Hohenberg (WHH) theory: $B_{c2}(0) = -0.69 T_c \left(\frac{dB_{c2}}{dT}\right)$, assuming that the upper critical field is limited by orbital pair breaking effect. The $B_{c2}$ values were estimated to be 46.6 and 81.5 T along the $ab$-plane and $c$-axis, respectively. The estimated $B_{irr}$ values are 29.3 and 56.5 T for $B//ab$ and $B//c$ at zero temperature, respectively. The estimated irreversibility field is close to the estimated upper critical field, which is related to the strong vortex pinning or weak thermal fluctuation in this compound.
There are two mechanisms for destroying superconductivity by applying magnetic field. The first one is orbital effect, which is related to the pair breaking of Cooper pairs by the Lorentz force via charge and opposite momenta on the Cooper pairs. The other one is paramagnetic effect, where a single pair is broken by the Zeeman Effect. The estimated upper critical field calculated from the WHH theory is higher than BCS paramagnetic limit, \( B_p^{BCS} \), in the weak coupling area. By using the weak coupling BCS formula, \( B_p^{BCS} = \frac{1.84}{T_c} \). We obtain \( B_p^{BCS} = 35.7 \) T. The estimated \( B_{c2} \) from WHH formula is 2.3 times this limit along the \( c \)-axis, showing that Zeeman paramagnetic pair breaking possibly is essential for \( B_{c2}^{ab} \). Also, it reveals the unconventional superconducting mechanism in this family. A similar result was reported for Co doped 122 family [132]. Using the value of \( B_{c2}^{ab} \), we calculated the Ginzburg-Landau coherence length, \( \xi_{GL} = \left( \frac{\phi_0}{2nB_{c2}} \right)^{1/2} \), where \( \phi_0 = 2.07 \times 10^{-7} \) Oe.cm\(^2\). The obtained coherence length is 2.7 and 2.01 nm for \( B/ab \) and \( B/c \) at \( T = 0 \) K, respectively. According to our data, the estimated anisotropy for
BaFe$_{1.9}$Ni$_{0.1}$As$_2$ is $\gamma = B_{c2}^{ab}/B_{c2}^c = 1.7$, for the temperature range of $12 < T < 18$ K, which indicates nearly isotropic superconductivity in this compound.

Figure 4-5 Arrhenius plot of the electrical resistivity at different magnetic fields for $B/ab$ and $B/c$.

Thermally activated flux flow is responsible for the broadening of resistivity transition and can be described by the following equation: $\rho(T, B) = \rho_n \exp \left(-U_0 (T, B)/k_B T\right)$, where $\rho_n$ is the normal state resistivity and $k_B$ is the Boltzmann constant. In Figure 4-5, we plot $\log \rho$ vs $T^{-1}$ at different magnetic fields. The linearity of $\log \rho$ versus $1/T$ indicates the thermally activated energy behaviour of resistivity. Slope of the curves is the pinning potential, $U_0$.

The best fit to the experimental data yields a value of the pinning potential of 5300 K for $B//c$ and $B/ab$ at the low magnetic field of 0.1 T. The pinning potential value for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal is 5 times greater than that of Bi-2212 crystal [202]. This value is lower than the reported value of 9100 K for Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$ single crystal for $B/ab$ [203], probably due to the different dopants. The magnetic field dependence of the pinning potential is shown in Figure 4-6. The activation energy drops very slowly with increasing applied magnetic field for $B < 1$ T, scaled
as $B^{-0.11}$, and then decreases as $B^{-0.56}$ for $B > 1$ T for $B//c$. This means that the pinning potential is almost field independent for $B < 1$ T.

Figure 4-6 Magnetic field dependence of the activation energy for $B//ab$ and $B//c$.

4.4 Evidence for fluctuation in mean free path induced pinning in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystals

The inset of Figure 4-7 shows the temperature dependence of magnetization measured after zero-field-cooling (ZFC) and field-cooling (FC) of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal, with a field of 200 Oe applied parallel to the crystallographic $c$-axis. The $T_c$ of 17.7 K was determined from the onset of transition.
The magnetization hysteresis (MH) loops of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal at various temperatures between 3 and 15 K for $B//c$, are shown in the main panel of Figure 4-7. The almost perfect symmetry of the MH loops with respect to the x-axis indicates that bulk pinning is dominant [158]. The minimum in the magnetization, which is located slightly above zero field in a given MH loop, characterizes the onset of the second magnetization peak (SMP) or fishtail effect, FE. At this field, the applied magnetic field penetrates completely into the bulk sample after zero field cooling [165]. The fishtail effect can be observed at all temperature below 15 K, similar to the behaviour of (Ba,K)Fe$_2$As$_2$ [195] and BaFe$_{1-x}$Co$_x$As$_2$ crystals [132]. Arrows indicate the onset ($B_{SMP}^{onset}$) and peak ($B_{SMP}^{Peak}$) positions of the SMP in Figure 4-7 for $T = 3$ K. The
onset and peak positions of the SMP move to lower magnetic field as the temperature is raised from 3 to 15 K.

In some of the conventional superconductors, such as MgB$_2$ [204] and Nb$_3$Sn [205], the peak effect occurs at a field close to the upper critical field. It is believed that the peak effect is associated with the metastability of an underlying first-order vortex melting transition, where softening of the vortex pinning due to thermal fluctuation leads to a better accommodation of the pinning centres by the vortex lattice [205]. This explanation of the peak effect in Nb$_3$Sn does not appear to be applicable to the fishtail effect observed in cuprate and pnictide superconductors, in which the peak effect occurs far from the normal phase boundary. In the case of cuprates, it has been suggested that a first-order disorder driven transition is responsible for the second magnetization peak in this system [206]. Salem- Sugui et al. [162] studied the vortex dynamics of an over-doped BaFe$_{1.82}$Ni$_{0.18}$As$_2$ single crystal by measuring flux creep over the fishtail peak and suggested that the fishtail peak could not have arisen due to the softening in the vortex pinning prior to melting nor from a change in the pinning regime within a collective model. Also, their study of an optimally doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal did not show any evidence of a pinning crossover occurring near the SMP of $MH$ loops [158]. Magnetic studies by Prozorov et al. [139] and Shen et al. [193] interpreted the SMP as signifying a crossover from elastic to plastic vortex creep. The same result was obtained by Kopeliansky et al., who suggested that the SMP is associated with a vortex structural phase transition from a rhombic to a square lattice [194].

The $J_c$ values were extracted from $MH$ loops, using Bean’s model [136], where
\[ J_c(B) = 20 \times \frac{\Delta M(B)}{l(1-l/3w)} \], with \( l \) and \( w \) being the sample dimensions perpendicular to the applied magnetic field, \( l < w \), and \( \Delta M \) the width of hysteresis loop, where \( \Delta M(B) = (M(B_{\downarrow}) - M(B_{\uparrow})) \), with \( M(B_{\uparrow}) \) and \( M(B_{\downarrow}) \) representing the magnetization in increasing and decreasing directions of the magnetic field, respectively.

Figure 4-8 shows the field dependence of \( J_c \) at different temperatures for \( B//c \). The obtained value of \( J_c = 0.14 \times 10^6 \text{ A/cm}^2 \) at zero field and \( T = 10 \text{ K} \) is comparable with the reported value of \( J_c = 0.23 \times 10^6 \) for an optimally doped sample [154]. The inset of Figure 4-8 presents normalized \( J_c \) versus \( B \) plots corresponding to the magnetic hysteresis loops of Figure 4-7 at selected temperatures. The onset position, \( B_{\text{onset}}^{\text{SMP}} \), and peak position, \( B_{\text{peak}}^{\text{SMP}} \), of the SMP are marked for \( T = 4 \text{ K} \).
position of the second peak shifts toward lower field monotonically with increasing temperature, for example, $B_{NMP}^{peak}$ decreases considerably from 6.8 T at 4 K to 0.7 T at 15 K, but $B_{NMP}^{onset}$ drops slowly from 1.7 T at 4 K to 0.2 T at 15 K, respectively. Similar behaviour was observed for YBa$_2$Cu$_3$O$_{7-\delta}$ superconductors. It is likely that the SMP has the same mechanism in both families.

Figure 4-9 Vortex phase diagram of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal determined from magnetic measurements for $B//c$. The dashed line represents the fitting curve using $B = A (1-T/T_c)^n$. The inset shows the temperature dependence of $B_{c2}$ and $B_{irr}$ obtained from the $\rho - T$ curves for $B//c$.

Figure 4-9 presents a vortex phase diagram of the BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal. Three characteristic fields, the irreversibility field, $B_{irr}$, $B_{NMP}^{peak}$, and $B_{NMP}^{onset}$, were determined from magnetic measurements, as shown by the solid symbols in Figure 4-9. It is clear that the $B_{irr} - T$, $B_{NMP}^{peak} - T$, and $B_{NMP}^{onset} - T$ are temperature dependent. The large area between $B_{irr} - T$ and $B_{NMP}^{peak} - T$ suggests that the vortex dissipation is through plastic motion in this area, as proposed by Shen et al. for optimally Co doped BaFe$_2$As$_2$ [193]. The dashed lines represent the fitting curves using $B (T) = A (1-T/T_c)^n$, with $n$ a fitting parameter. All the curves were well fitted using the expression with $n$
= 1.9 for $B_{\text{onset}}$ and $n = 1.4$ for $B_{\text{peak}}$ and $B_{irr}$. These values are similar to the values obtained for optimally Co doped BaFe$_2$As$_2$ single crystal [193].

In order to assess the nature of pinning mechanisms in more detail, it is useful to look at the variation of vortex pinning force, $F_p = B \times J_c$, with the magnetic field. In Figure 4-10, we plot the normalized pinning force, $F_{p}^\text{norm} = F_p / F_{p}^\text{max}$, as a function of the reduced field, $h = B/B_{irr}$, where $B_{irr}$ is the irreversibility field. The $B_{irr}$ is taken as value of the applied magnetic field at the point where $J_c$ is too small to be useful for any practical application. Here, we have used the criterion of $J_c < 100$ A/cm$^2$ for determining $B_{irr}$. It should be noted that the scaling of normalized pinning force has been done based on the field normalized by the irreversibility field, with $h = B/B_{irr}$, instead of the upper critical field, with $h = B/B_{c2}$, due to the fact that the difference
between $B_{c2}$ and $B_{irr}$ is sizable, and $B_{irr}$ is more significant in the low temperature regime in case of iron-based superconductors, MgB$_2$, and cuprates\textsuperscript{37, 33, 29, 42, 57, 58, [207]}. The temperature dependence of $B_{c2}$ and $B_{irr}$ obtained from the $\rho$ - $T$ curves (see the inset in Figure 4-9) clearly reveals that $B_{irr}$ is far below $B_{c2}$. Note that all the $F_p^{norm}$ curves for $9 \text{ K} \leq T \leq 14 \text{ K}$ collapse into one unified curve. We fit these data using the Dew-Hughes formula, $F_p = A h^p (1-h)^q$ \textsuperscript{[166]}, where $A$ is a constant, and $p$ and $q$ are two parameters whose values depend on the origin of pinning mechanism. The Dew-Hughes fit is shown by the black dashed-dotted line in Figure 4-10 with $p = 2.01$ and $q = 2.96$. The value $p/(p+q) \approx 0.4$ matches well with the peak positions in $F_p^{norm}$ versus $h$ plots. According to the Dew-Hughes model, in the case of $\delta l$ pinning for a system dominated just by point pinning, $p = 1$ and $q = 2$, with $F_p^{norm}$ occurring at $h_{max} = 0.33$. Pinning due to grain boundaries leads to $h_{max} \approx 0.2$, while in a system in which variation in the superconducting order parameter controls the pinning mechanism, $h_{max} \approx 0.7$ \textsuperscript{[166, 208]}. There is no maximum in the case of $\delta l$ volume pinning with $p = 0$ and $q = 2$. In the case of $\delta T_c$ pinning, the maximum of $F_p$ is expected to be located at higher $h$ values. For example, for point pinning, the maximum is expected at $h = 0.67$, with $p = 2$ and $q = 1$. For surface pins, the maximum exists at $h = 0.6$, $p = 1.5$, and $q = 1$, and for volume pins, $h = 0.5$ with $p = 1$ and $q = 1$. In our case, $h_{max} = 0.4$, indicating that point pinning alone cannot explain the pinning mechanism in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal. Similar analysis has been done for Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ ($h_{max} = 0.43$), BaFe$_{1.85}$Co$_{0.15}$As$_2$ ($h_{max} = 0.37$), and BaFe$_{1.91}$Ni$_{0.09}$As$_2$ ($h_{max} = 0.33$) by Sun et al. \textsuperscript{[154]}. They noticed that $B_{c2}$ and $B_{SMC}^{peak}$ decrease faster with decreasing temperature for BaFe$_{1.91}$Ni$_{0.09}$As$_2$ compared to Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ and BaFe$_{1.85}$Co$_{0.15}$As$_2$ single crystals, which is related to the inhomogeneity in BaFe$_{1.91}$Ni$_{0.09}$As$_2$. The fact that strongest pinning is in Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ among these three systems indicates that...
inhomogeneous distribution of dopants or As deficiency cannot play a crucial role in determining strong pinning in iron pnictide. Yang et al. proposed that, the obtained value of $h_{\text{max}} = 0.33$ should be attributed to small-size normal cores, as in the case of arsenic deficiency in (Ba,K)Fe$_2$As$_2$ single crystal [198]. In the case of BaFe$_{1.8}$Co$_{0.2}$As$_2$, a peak at $h_{\text{max}} \approx 0.45$ was suggested to be related to the inhomogeneous distribution of Co ions [132]. In particular, the fact that, in our case, the maximum in $F_p$ occurs at $h < 0.5$ indicates that the pinning centres in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ are of the $\delta l$ type, while for $\delta T_c$ pinning, it is expected that the maximum would occur at $h > 0.5$.

Figure 4-11 Temperature dependence of the normalized measured critical current density at 0.05 T (open circles) and 0 T (solid squares). Inset: Field dependence of $J_c$ at $T = 3$ K.

With the aim of understanding more about the origins of pinning in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal, the experimental results were analysed using collective pinning theory. According to the theoretical approach proposed by Griessen et al. in YBCO thin film [209], in the case of $\delta l$ pinning, the normalized critical current
density, $J_c(t)/J_c(0) \propto (1-t^2)^{5/2}(1+t^2)^{1/2}$, while for $\delta T_c$ pinning, $J_c(t)/J_c(0) \propto (1-t^2)^{7/6}(1+t^2)^{5/6}$, where $t = T/T_c$. It should be noted that the flux pinning is two-dimensional in such thin films, as the coherence length along the flux lines exceeds the thickness of film $^{56}$. Figure 4-11 shows a comparison between the experimental $J_c$ values and theoretically expected variation within $\delta l$ and $\delta T_c$ pinning mechanisms at 0.05 T (open circles) and 0 T (the so-called remanent state shown by solid squares). The $J_c(t)$ values have been obtained from $J_c(B)$ curves at several temperatures. A remarkably good agreement between the experimental results and theoretical $\delta l$ pinning curve is obtained. It is likely that pinning in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal originates from spatial variation of the mean free path. Our observation of the dominant $\delta l$ pinning is in good agreement with the reported $\delta l$ pinning mechanism for K$_x$Fe$_{2-y}$Se$_2$, FeSe$_{0.5}$Te$_{0.5}$, and FeTe$_{0.7}$Se$_{0.3}$ single crystals at low magnetic field.

In summary, BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal exhibits high pinning potential, although it has very small coherence length. It is possible that, its nearly isotropic properties are responsible for the high pinning potential value of this compound. Also we have observed second magnetization peak effect in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal. The onset and peak positions of the SMP move to lower magnetic field as the temperature is raised from 3 to 15 K. Analysis using the Dew-Hughes model suggests that point pins alone cannot explain the observed field variation of pinning force density. The maximum in $F_p$ at $h < 0.5$ indicates that the pinning centres in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ are of the $\delta l$ type, while for $\delta T_c$ pinning, it is expected that maximum would occur at $h > 0.5$. In addition, a good agreement between experimental and theoretical fitting using $\delta l$ pinning is obtained based on the collective flux pinning model.
CHAPTER 5

VORTEX-GLASS PHASE TRANSITION AND ENHANCED FLUX PINNING IN C⁴⁺ IRRADIATED BAFE_{1.9}NI_{0.1}AS₂ SUPERCONDUCTING SINGLE CRYSTALS

5.1 Introduction

The commercial applicability of Fe-based superconductors [36] relies on their ability to carry high current, which is determined by the effectiveness of pinning sites in these materials. Heavy ion irradiation and neutron irradiation are the most effective approaches to introduce effective artificial pinning centres for supercurrent enhancement in both conventional and high temperature superconductors [36, 149]. The vortex pinning in high temperature superconductors is controlled by dynamic and static disorder [210]. Dynamic disorder is caused by large thermal fluctuations, while static disorder is caused by structural imperfections such as twin boundaries, columnar defects, etc. The latter can be effectively engineered with heavy ion or neutron irradiation of the material.

Electron scattering on static disorder (or structural defects) is particularly interesting for the family of Fe-based materials exhibiting multiband superconductivity. In fact, the structural disorder is always present in the Fe-based superconductors, where chemical doping is needed to induce superconductivity. Another way to introduce defects into superconductors is by ion irradiation, which results in the formation of additional pinning centres and significantly increases the electron scattering. As a result, defects induced by irradiation are effective for enhancing the critical current density, \( J_c \), in superconductors. For instance, columnar defects created by heavy ion irradiation were found to be the most effective pinning...
sites for two-dimensional (2D) pancake vortices and are thus responsible for a significant $J_c$ enhancement in (highly anisotropic) high temperature cuprate superconductors [211, 212] (HTSs). Fe-based superconductors have revealed much smaller anisotropy ($\gamma = 1-8$ at $T \approx T_c$) [213, 214, 215, 216, 217], especially doped BaFe$_2$As$_2$ (Ba-122) superconductors with $\gamma \approx 1-3$. Very strong intrinsic pinning strength has been observed in K doped 122 single crystals with rigid vortices, mainly due to the small anisotropy [131]. As a result, the point defects induced by neutron irradiation are effective for pinning vortices and enhancing the critical current density, $J_c$, by a factor of 1.5-3 [144]. In the case of Fe-based superconductors, it was demonstrated that thermal neutron irradiation leads to $J_c$ improvement by a factor of 1.5-3 [144], while heavy ion irradiation using Au [145], Pb [148, 149], or Ta [143] ions increases $J_c$ by a factor of 3-10 as a result of columnar defects [143, 145]. Both thermal neutron and heavy ion irradiation, however, are high cost procedures for large-scale applications compared to light ion C$^{4+}$irradiation, which was successfully employed in this work for the first time.

In this Chapter, the influence of defects induced by light-ion (C$^{4+}$) irradiation on $T_c$, the irreversibility field, $B_{irr}$, the upper critical field, $B_{c2}$, and the pinning potential, $U_o$, in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ superconducting single crystal is discussed, and the vortex phase diagram for the sample before and after irradiation will be resolved.

### 5.2 Experimental details

Single crystals with the nominal composition BaFe$_{1.9}$Ni$_{0.1}$As$_2$ were prepared by a self-flux method [189]. The as-grown single crystals were cleaved and shaped into thin plates for measurements. Irradiation with 35.59 MeV C$^{4+}$ was carried out in a
direction perpendicular to the broad surface of the sample, using a square shaped beam 7 × 7 mm² in cross-section. The sample was irradiated for a total time of 3 min with ion dose of 3 × 10¹² ions·cm⁻². For the sake of consistency, irradiation and all the measurements were carried out on the same piece of single crystal sample. The sample was placed on a conductive sample holder with conductive C-tape in order to prevent charging and excessive heating during irradiation. The beam current was measured before and after irradiation with a Faraday cup, and the average beam current was approximately 10 nA. Magnetization of the sample was measured using magnetic and physical properties measurement systems (MPMS and PPMS, Quantum Design). The critical current density, \( J_c \), was calculated from the magnetic hysteresis data using an extended Bean model [136, 218]: \( 20\Delta m/ (a (1 - a/3b) (a < b)) \), where \( \Delta m \) is the height of the magnetization loop, and \( a \) and \( b \) are the length and width of the sample perpendicular to the applied magnetic field, respectively. The transport properties were measured over a wide range of temperatures and magnetic fields up to 13 T using the Quantum Design PPMS.

The magnetic flux distribution inside the studied samples was visualized with the help of the magneto-optical imaging (MOI) technique. MOI measurements were conducted at temperatures varying from 6.5 K to 15 K. Images were acquired by a computer-controlled charge-coupled display (CCD) camera. An external magnetic field (\( B_{\text{ext}} = 57 \) mT) was applied perpendicular to a FeGdY garnet film and the sample surface after zero-field-cooling of samples to the measurement temperature. The local critical current density was determined using numerical inversion of Biot-Savart’s law, where the measured \( z \)-component of the magnetic flux (\( B_z \)) is related to the in-plane critical current in the sample [219].
The GEANT 4 package was used for the Monte Carlo calculations to estimate the distribution of carbon ions and the redistribution of other ions caused by carbon ion collisions.

5.3 Results and discussion

Figure 5-1 (a) shows the magnetization loops at 2 K for the sample before and after irradiation. The magnetic moment of the sample is obviously enhanced after C\(^{4+}\)-irradiation over the entire range of magnetic fields investigated.

![Figure 5-1](image)

Figure 5-1 (a) Magnetization loops at 2 K before and after irradiation of the sample; (b) magnetic field dependence of critical current density before (dashed-dotted line) and after (solid line) C\(^{4+}\)-irradiation; (c) \(J_{c\text{-irr}}/J_{c\text{-un-irr}}\) ratio as a function of temperature at different applied fields.

Figure 5-1(b) shows the calculated \(J_c\) for the pristine and the C\(^{4+}\)-irradiated single crystal as a function of magnetic field with \(B//c\). The irradiated sample exhibits enhanced \(J_c\) performance, which is both field and temperature dependent. For instance, at low temperatures (2 K and 5 K), \(J_c\) of the C\(^{4+}\)-irradiated sample is enhanced at all
fields that were studied, while at higher temperature ($T = 10$ K), $J_c$ improvement is observed at $B < 4$ T. In general, the enhancement of current carrying ability for the sample before and after irradiation ($J_{c, \text{ irr}}/J_{c, \text{ un-irr}}$) is between 1.5 and 1 at magnetic fields smaller than 4 T, as shown in Figure 5-1(c).

As can be seen from Figure 5-1(b), $J_c$ is as high as $1.6 \times 10^9$ A/m$^2$ at 5 K and $B = 0.5$ T before irradiation, increasing to $2.3 \times 10^9$ A/m$^2$ after C$^{4+}$-irradiation. It has been reported that for BaFe$_{1.8}$Co$_{0.2}$As$_2$ crystals irradiated by neutrons with a dose of $4 \times 10^{17}$ cm$^{-2}$ (Ref. [144]), the $J_c$ increased from $3 \times 10^5$ to $7 \times 10^5$ A/cm$^2$ at $B = 0.5$ T ($J_{c, \text{ irr}}/J_{c, \text{ un-irr}} = 2.3$). These results are scalable with ours, taking into account the much lower ion doses of C$^{4+}$ ($10^{12}$ ions/cm$^2$) during the irradiation process. Therefore, we demonstrate that light C$^{4+}$ ion irradiation is also an effective approach towards enhancing the $J_c(B)$ performance in Fe-based superconductors. The peak effect, which is commonly observed in Fe-based superconductors, can be seen for both studied samples at $T = 10$ K (Figure 5-1 (b)). Note that peak position shifts to lower magnetic field after C$^{4+}$-ion irradiation, as indicated by the arrows in Figure 5-1(b).

Another feature of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal is that pristine and carbon irradiated single crystals show flux jump effect, which is more pronounced in the irradiated sample (Figure 5-1 (a)) due to the higher $J_c$. The size of flux jumps is smaller than that observed in Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$ single crystal [131, 220], with the flux lines fully penetrating into the whole sample.

Magneto-optical images of the irradiated and the reference samples measured at $T = 6.5$ K and $B_{\text{ext}} = 57$ mT are shown in Figure 5-2(a, b). Both samples have defects (bright areas marked by arrows), along which magnetic field penetrates inside the sample more rapidly. These defects are due to uneven cutting of the single crystals (at
the edges) and alteration of the layered structure of iron-pnictide superconductors [as
seen at the bottom right corner of the sample in Figure 5-2(a)]. The magnetic flux
penetration into the samples has been measured along the lines drawn in Figure 5-2(a,
b) (avoiding defect-occupied areas of the samples) at different temperatures. The
corresponding $B_z$ profiles are plotted in Figure 5-2(c). As can be seen, at constant
temperature, the magnetic flux penetrates deeper inside the reference sample
compared to the irradiated sample.

Figure 5-2 MOI images of (a) irradiated and (b) reference sample measured
at $T = 6.5$ K and applied magnetic field $B_{\text{ext}} = 57$ mT. (c) $B_z$ profiles taken
along the red lines in (a) and (b) at $T = 6.5$ K, 8 K, 12 K, and 15 K. Solid
(dotted) lines correspond to the irradiated (reference) sample. The inset in (c)
represents the slope of the flux penetration ($|dB/dx|$) inside the samples (at $B_z = 20$ mT) as a function of temperature. (Lines are only a guide for the eye.)
The slope of flux penetration $|dB/dx|$, inset in Figure 5-2(c), is steeper (higher) for the irradiated sample at all temperatures studied. This suggests that defects introduced by irradiation enhance flux pinning in this sample. The effect of these defects is reduced with increasing temperature, however, as can be seen from the shrinking difference between the $|dB/dx|$ values for reference and irradiated samples as $T \rightarrow 15$ K.

![Graph showing $J_c$ values](image)

Figure 5-3 $J_c(T)$ values for samples studied at $B_{ext} = 57$ mT, calculated from MOI images using the Biot-Savart law [219]

The $J_c$ values were calculated from MOI images using Biot-Savart law [219], and the corresponding values at each temperature are presented in Figure 5-3. Note that there is a good correlation between the results observed by quantitative MOI and magnetometry techniques (Figure 5-1). In accordance with the magnetometry technique, $J_c$ values are enhanced for the irradiated sample compared to the reference sample at all temperatures studied. $J_c$ enhancement is more significant, however, at low temperatures, and it becomes smaller with increasing temperature. This behaviour
is consistent with reduced pinning strength on defects introduced by irradiation as $T \rightarrow T_c$ (due to the increase in the coherence length, $\xi(T)$, and thermal fluctuations.

![Arrhenius plots of the resistivity for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal for $B = 0.1, 0.5, 1, 3, 5, 7, 9, 11, \text{ and } 13$ T with $B\parallel ab$. Inset: Temperature dependence of resistivity for zero magnetic field before and after irradiation.]

Figure 5-4 Arrhenius plots of the resistivity for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal for $B = 0.1, 0.5, 1, 3, 5, 7, 9, 11, \text{ and } 13$ T with $B\parallel ab$. Inset: Temperature dependence of resistivity for zero magnetic field before and after irradiation.

The temperature dependence of resistivity, $\rho(T)$, at zero magnetic field for the sample before and after C$^{4+}$-irradiation is shown in the inset of Figure 5-4. The resistivity decreases with decreasing temperature from 200 to 20 K for both samples, supporting their metallic behaviour above $T_c$. At 200 K, resistivity of the sample increases from $14.3 \times 10^{-5}$ $\Omega\cdot\text{cm}$ (before irradiation) to $31 \times 10^{-5}$ $\Omega\cdot\text{cm}$ (after irradiation), which is related to increased electron scattering on defects induced by C$^{4+}$-irradiation.

The transition temperature, $T_c$, was determined from the $\rho(T)$ curves measured at zero magnetic field. $T_c$ was 18.3 K, with a transition width ($\Delta T_c$) of 0.9 K for the sample without irradiation. Remarkably, the $T_c$ value decreased to 17.8 K (by 0.5 K
only) with almost the same $\Delta T_c$ (0.8 K) after C$^{4+}$-irradiation. The reduction of $T_c$ after ion irradiation is a common feature observed in many cuprate and pnictide superconductors [144, 221] and is related to such effects as interband scattering [222], a reduction in anisotropy [141], etc. The C$^{4+}$ irradiation, however, only caused small changes in $T_c$ and transition width in our sample.

Arrhenius plots of resistivity for the irradiated BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal in different magnetic fields, with $B//ab$, is shown in the main panel of Figure 5-4. It was reported that the broadening of resistivity transition in magnetic field is a direct consequence of thermal fluctuation in the vortex system [200]. Therefore, resistive transport measurements are commonly used to study vortices and vortex phase transitions [223, 224, 225, 226, 227]. Note that the calculations described below correspond to the irradiated sample only for simplicity. The results leading to vortex phase diagram for the reference sample, however, will be summarised at the end of this section.

According to vortex-glass theory [223], in vortex glass state and close to the glass transition temperature, $T_g$, the resistivity disappears as a power law

$$\rho = \rho_0 \left| \frac{T}{T_g} - 1 \right|^s$$  \hspace{1cm} 5-1

where $s$ is a constant, which depends on the type of disorder, and $\rho_0$ is a characteristic resistivity related to the normal state. According to Ref. [227], the temperature difference $T-T_g$ can be replaced by the energy difference $k_B T-U_0$, where $U_0$ is an effective pinning energy. This modifies equation 5-1 to

$$\rho = \rho_n \left| \frac{k_B T}{U_0(B,T)} - 1 \right|^s$$  \hspace{1cm} 5-2
where $\rho_n$ is the normal state resistivity. In this model, transition from vortex solid to vortex liquid state occurs when the two energy scales are equal, i.e. $U_0 (B, T_g) = k_B T_g$.

[226] The effective pinning energy was found empirically as

$$U_0(B, T) = U_B \left(1 - \frac{T}{T_c}\right) \text{ with } U_B = k_B T_c / (B/B_0)^\beta$$

where both $B_0$ and $\beta$ are temperature and field independent constants. By considering the pinning energy at glass temperature, i.e. $U_0(B, T_g) = k_B T_g$, the temperature dependence of vortex glass line is obtained as follows: [225, 226]

$$B_g(T) = B_0 \left(\frac{1-T/T_c}{T/T_c}\right)^{1/\beta}$$

It was shown that equation 5-4 describes the vortex glass transition for Y-123 [225, 226, 227, 228]. A useful scaling form for the resistivity was obtained by combination of eq. 5-2 and eq. 5-3

$$\rho = \rho_n \left| \frac{T_c-T_g}{T_g(T_c-T)} - 1 \right|^s$$

According to the vortex glass model 5-1, resistance goes to zero at glass temperature, $T_g$, as $\rho \propto (T-T_g)^s$. Consequently, $T_g(B)$ can be extracted by applying the Vogel-Fulcher relation: $(d \ln \rho /dT)^{-1} \propto (T-T_g)$ to resistive tails [227]. We can rewrite eq. 5-5 in the form:

$$\left(\frac{\partial \ln \rho}{\partial T}\right)^{-1} = \frac{T-T_g}{s} \left(\frac{T_c-T}{T_c-T_g}\right).$$

This expression differs from Vogel-Fulcher relation by a correction factor $A(T) = (T_c - T) / (T_c - T_g)$, which is close to 1 at temperatures sufficiently close to $T_g$. Therefore, the
usual Vogel-Fulcher relation can be applied to the resistivity tail for estimating $T_g$ directly.

Inset in Figure 5-5 shows the experimental data fitted by a straight line, which corresponds to equation 5-6. As can be seen, there is a good agreement between the data and Vogel-Fulcher relation at low temperature (below $T^*$, the crossover temperature from vortex glass state to high temperature regime). The intercepts of fitting lines with x-axis give $T_g$ values of 14.7 K and 15.5 K for the irradiated and pristine sample, respectively. It is likely that enhancement of point defects due to C$^{4+}$ irradiation results in reduction of $T_g$ for the irradiated sample. The effect of point disorder can be understood as promotion of increased vortex bending and meandering, and therefore, the critical current density was increased by C$^{4+}$ irradiation. Similar effects of point disorder on the location of solid-to-liquid transition have been reported for Re-123 superconductors, with Re a rare earth element [223, 229].

As shown in the inset of Figure 5-5, at $T > T^*$ there is a crossover from the low temperature (linear) regime, which corresponds to the vortex glass state, to high temperature regime, where resistivity corresponds to the Arrhenius plot [230]. This crossover occurs at temperature $T^*$, where the three-dimensional (3D) vortex glass correlation length becomes significant. In layered superconductors, above this temperature, ($T > T^*$), the vortex flux lines act as 2D pancake vortices [230]. The parameter $B_0$ in $B_g(T) = B_0 \left(\frac{1-T/T_c}{T/T_c}\right)^{1/\beta}$ equation 5-4 controls this (field-induced) change from 3D to 2D regime of vortex fluctuations. We have estimated values of $B_0 = 16.4$ and $\beta = 0.91$ by fitting the $B_g(T/T_c)$ data using equation 5-4 [the main panel of Figure 5-5].
In vortex–glass and modified vortex–glass models, \([141, 223, 225, 226]\) the 2D regime is achieved for fields larger than \(B_0 \approx \Phi_0 / (\gamma d)^2\), where \(\Phi_0\) is the flux quantum, \(d\) is the interplane spacing, and \(\gamma\) is the effective mass anisotropy. Using the above value of 16.4 for \(B_0\) and assuming \(\gamma = (\rho_c/\rho_{ab})^{1/2} = 12.3\) for un-doped BaFe\(_2\)As\(_2\) single crystal, as reported in Ref. \([231]\), we have estimated the interplane spacing \(d = 9.3\) Å. There is a qualitative agreement between our analysis of \(d\) (from fitted \(B_0\)) and the value found in Ref. \([39]\) for Ba-122 superconductor. This gives evidence that vortex lattice becomes 2D at temperatures higher than the crossover temperature \(T^*\), while it will become 3D at sufficiently low temperatures due to the finite interplane coupling. The exact explanation of nature of dimensional crossover in Ba-122 superconductor will require further study, however equation 5-5 suggests that there is scaling behaviour between the normalized resistivity \(\rho/\rho_n\) and scaled...
temperature $T_{sc} = [T (T_c-T_g)/T_g(T_c-T)]^{-1}$, which is plotted in Figure 5-6. The resistivity transitions at fields between 0 and 13 T scale into one curve at critical exponent $s = 2.6 \pm 0.4$, which is in agreement with the value of 2.8 observed from vortex-glass model. This resistivity scaling behaviour of the vortex liquid has been observed in several high-$T_c$ superconductors such as oxygen-deficient Y-123 single crystal [226, 227] and Tl-2212 thin film.

By solving eq. 5-2 for $U_0$, one obtains

$$U_0(B, T) = k_B T \left[ 1 + \left( \frac{\rho}{\rho_n} \right)^{1/s} \right]^{-1} \quad 5-7$$

Figure 5-6 Resistivity scaling according to Eq. (5.5) for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal for $0 \leq B \leq 13$ T.

According to eq. 5-7 the pinning potential can be calculated directly from experimental data, provided that one knows $\rho_n$ and the exponent $s$. The $s$ parameter is estimated from the inverse slope of resistivity in the vortex glass state, as mentioned
above, and $\rho_n$ is the normal state resistivity of the samples at $T = 20$ K. The calculated $U_0(B,T)$ for C$^{4+}$ irradiated BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal for fields between 0 and 13 T is shown in Figure 5-7.

In Figure 5-7, dashed-dotted lines fitting the low temperature part of the resistivity curves are well described by equation 5-3 with a field dependent $U_B$, where $U_B$ is the pinning energy at 0 K.

The extrapolation of the linear behaviour of $U_0(T)$ at temperatures close to $T_g$ and different fields merges in the point $T = T_c$ and $U_0 = 0$ [in agreement with Eq. (5.3)]. Thus, the field dependence of pinning energy $U_B$ can be estimated directly from the slope of these lines.

![Figure 5-7 Pinning potential $U_0(B, T)$ for irradiated BaFe$_{1.9}$Ni$_{0.1}$As$_2$ calculated according to Eq. (5.7) in fields in the range of $0 \leq B \leq 13$ T.](image)

The field dependence of the estimated $U_B$ values is shown in Figure 5-8. Note that different power law dependences are found at low and high magnetic fields. $U_B/k_B$
decreases slowly with increasing applied magnetic field for $B < 1 \, T$ and scales as $B^{-0.14}$, but it scales as $B^{-1.17}$ for $B > 1 \, T$ for irradiated sample. This result suggests that at low fields, the single vortex pinning may co-exist with collective creep, while at high magnetic fields, the collective creep dominates.

Another important point is that one can directly obtain the $T_g$ values by taking crossing points of the (solid) $U_0(B, T)$ lines and $U_0 = k_B T$ lines, as indicated by arrow in Figure 5-7. This criterion was used for the estimation of $T_g$ in modified vortex-glass model [226, 227]. According to eq. 5-2, $U_0 (B, T)$ is the average pinning energy in the system and gives vortex solid to liquid transition when $U_0 (B, T_g) = k_B T_g$.

Figure 5-8 Magnetic field dependence of $U_B/k_B$ as estimated from Eq. (5.3) and the slopes in Figure 5.7 for the irradiated and un-irradiated BaFe$_{1.9}$Ni$_{0.1}$As$_2$ sample. The solid lines are the fits to the data, giving the relation $U_B/k_B = B^n$ with $n = 0.14$ at $B < 1 \, T$ and $n = 1.17$ at $B > 1 \, T$. Inset: Field dependence of the estimated activation energy based on the thermally activated flux flow (TAFF) model for the irradiated sample.

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In many studies, the vortex liquid resistivity is described by a thermally activated flux flow (TAFF) model [200], \( \rho(T, H) = \rho_n \exp \left( \frac{U^*}{k_B T} \right) \) [232], where \( \rho_n \) is the normal state resistivity and \( U^*/k_B \) is the activation energy, where \( k_B \) is the Boltzmann constant. By using the TAFF model, we obtained the activation energy, \( U^*/k_B = \frac{\partial \ln \rho}{\partial (1/T)} \), which is shown in the inset of Figure 5-8. At \( B = 0.5 \) T, \( U^*/k_B \) was 2052 K and \( U_B/k_B \) was 203 K for the irradiated sample. Although \( U^* \) is larger than \( U_B \) by an about one order of magnitude, they have a similar magnetic field dependence.

In order to compare the \( U^* \) and \( U_B \) values, we took the derivative \([\partial \ln \rho/\partial (1/T)]\) of eq. 5-2 and used eq. 5-3. As a result, one can obtain the relation

\[
U^* = \frac{s[1+(\rho/\rho_n)^{1/s}]}{(\rho/\rho_n)^{1/s}} U_B = AU_B
\]

From using \( s = 2.6 \) and experimental data for \( \rho/\rho_n \approx 10^{-2} \), the coefficient \( A \) is on the order of 1. Therefore, our data analysis yields good agreement between both TAFF and vortex-glass models in the vortex liquid region.

The upper critical fields, \( B_{c2} \), for the samples were obtained from 90% of corresponding resistivity values near the transition to normal state. Using the estimated vortex glass line, \( B_g \), the reconstructed \( B-T \) phase diagram is shown in Figure 5-9. The slopes of \( B_{c2} \) and \( B_g \) are 3.5 and 1.9 T/K for \( B//c \) before irradiation, and they change slightly to 3.1 and 1.8 T/K after \( C^{4+} \)-irradiation, respectively. According to the collective pinning model [141], the disorder-induced spatial fluctuations in the solid-vortex lattice can be clearly divided into markedly different regimes according to the strength of applied field. Two different regimes are distinguishable: (1) vortex glass, which governs the region below \( B_g \); and (2) vortex liquid, which holds between \( B_g \) and \( B_{c2} \), where thermal fluctuations are important.
The vortex solid state is characterised by non-zero $J_c$, while the vortex liquid is dissipative at all currents. In a very clean system, the solid to liquid phase transition is most likely a first order melting transition. It turns into a second order vortex glass transition, however, for highly disordered systems including point defects or twin boundaries, or artificial defects resulting from ion irradiation [233]. Figure 5-9 indicates that $C^{4+}$ irradiated BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal exhibits very wide regions of vortex-glass phase, which originates from vastly enhanced vortex pinning. Especially at low temperature, the vortex glass line $B_g$ exhibits an upward curvature, which indicates even stronger pinning behaviour at low temperature. It should be noted that $B_{c2}$ and $B_g$ are slightly decreased after $C^{4+}$ irradiation. This is related to the reduction in electron mean free path due to the increase in scattering after $C^{4+}$ irradiation.
Figure 5-10 shows the distribution of carbon ions in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal from a Monte Carlo calculation. The results show that almost all C ions end up in a well-defined layer, at a depth of around 24 $\mu$m. This layer looks quite homogeneous for 500 carbon ions fired along the red arrow. As the beam of carbon ions is uniformly distributed across the sample surface, we expect a fairly homogeneous distribution of carbon in this layer.

The binding energy of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ is about 3 eV/atom, so most of the damage is done by primary carbon ions through primary knock-on collisions and none by the Ba, Fe, Ni, and As recoils, because their energy is below 3 eV, as shown in Figure 5-11(a). The energy carried by C$^{4+}$ ions into the irradiated layer is distributed to the BaFe$_{1.9}$Ni$_{0.1}$As$_2$ crystal lattice, and as a result, the atoms in that layer will recoil or be moved out of their lattice sites. Some of these atoms will fall back into a thermodynamic equilibrium position (self-annealing), but a number of them will remain in interstitial positions, locally destroying the BaFe$_{1.9}$Ni$_{0.1}$As$_2$.
lattice. To see which of the lattice atoms are more disrupted by carbon ions, the calculated distributions of individual atoms (Ba, Fe, Ni, As) which are knocked out of their lattice sites are shown in Figure 5-11(b).

![Figure 5-11](image)

Figure 5-11 (a) Distribution of the energy of carbon ions to the atoms/ions in their paths through collisions. (b) Calculated distribution of the individual Ba, Fe, Ni, and As atoms which are knocked out of their lattice sites.

Figure 5-11 (b) shows that most of the BaFe$_{1.9}$Ni$_{0.1}$As$_2$ lattice disruption is contained in and around the C$^{4+}$ irradiated layer, at a depth of around 24 μm, with little disruption between the entry surface and damaged layer. Also, the most disrupted (recoiled) atoms are Fe and As, as they have the highest concentrations and lower masses. The total number of vacancies produced by C-ions and Ba, Fe, Ni, and As recoils is around 2,300 vacancies/ion in the damaged layer. According to this calculation, the C-irradiation and the resulting C-irradiated layer constitute a 3D defect layer with a thickness of 1.5 μm at a depth of about 24 μm under the irradiated surface. The distribution of damage in the cross-section of this 3D layer has a Gaussian profile. This damage matrix is likely to form a network (connected regions) in the damaged layer. Therefore, the defect/vacancy region coexists with the superconducting region that was not destroyed during C-irradiation. This type of
defect distribution, which is very similar to those of extended defects, is distinct from the columnar defects caused by heavy ion irradiation [145].

In conclusion, we investigated the effects of $C^{4+}$ irradiation in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal. It was found that $C^{4+}$ irradiation causes little change in $T_c$, but it can greatly enhance in-field critical current density by a factor of up to 1.5, with enhanced flux jumping at 2 K. Also, the MOI results confirm the enhancement of $J_c$ for irradiated sample. Our results suggest that light $C^{4+}$ ion irradiation is an effective method for the enhancement of $J_c$ in Fe-superconductors compared to heavy ion irradiation and neutron irradiation. In addition, the glass transition that is introduced based on a modified model for vortex-glass transition can be applied to both pristine and the $C^{4+}$-irradiated BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal. For temperatures below the superconducting transition temperature, a scaling of all measured resistivity $\rho(B, T)$ and of the pinning potential $U_0(B, T)$ in magnetic fields up to 13 T with critical exponent $s = 2.6 \pm 0.41$ is obtained. The vortex phase diagram has been determined, based on the evolution of vortex-glass transition temperature $T_g$ with magnetic field and the upper critical field. Furthermore, a comparison has been made with the thermally activated flux flow behaviour which is usually employed to account for the resistivity in vortex liquid region. Monte Carlo calculations show that $C^{4+}$ ions end up in a well-defined layer at a certain depth, causing extended defects and vacancies within the layer, but few defects elsewhere on their paths.
CHAPTER 6

ANGULAR DEPENDENCE OF PINNING POTENTIAL AND UPPER CRITICAL FIELD IN UNDER-DOPED BAFE$_{1.9}$CO$_{0.1}$AS$_2$ SINGLE CRYSTAL

6.1 Introduction

The thermal activation behaviour of vortices in superconductors determines their magneto-transport properties, which are critical for practical applications. In highly anisotropic cuprates, a magnetic field perpendicular to the superconducting layers penetrates in the form of pancake vortices, while a parallel field creates Josephson vortices [234]. The interaction between pancake vortices and Josephson vortices creates vortex chains [235] or Josephson vortices decorated by pancake vortices [236] when the magnetic field is tilted. The pinning potential in cuprates is highly anisotropic and strongly field dependent due to the strong thermally activated behaviour of two-dimensional (2D) pancake vortices.

Iron based superconductors [36] exhibit relatively high transition temperature, $T_c$, very high upper critical field [114], and relatively low anisotropy [89, 213, 237, 238]. Among pnictide superconductors, Ba-122 compounds have typical 2D layered crystal structure. However, they show nearly isotropic superconductivity [213] and very high intrinsic pinning potential, which is weakly field dependent [131]. These unique futures make 122 superconductors more favourable for practical application than other pnictide superconductors.

Thermally activated flux flow has been studied in NdFeAsO$_{0.7}$F$_{0.3}$ [200], Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$ [131], BaFe$_{1.9}$Ni$_{0.1}$As$_2$ [239], and Tl$_{0.58}$Rb$_{0.42}$Fe$_{1.72}$Se$_2$ [240] single
crystals for $B//ab$ and $B//c$. Very recently, studies of the angular dependence of transport critical current density, $J_c$, have indicated that $J_c$ decreases monotonically with angle, $\theta$, for $\theta < 90^\circ$, where $\theta$ is defined as the angle between magnetic field and $c$-axis. The ratio of $J_c(B//ab)/J_c(B//c) = 7.5$ or $1.8$ at $B = 1$ T and $T = 4.2$ K for La-1111 [241] and Co-122 [242] thin film, respectively. It should be noted that the determination of angular dependence of pinning potential and upper critical field is important for understanding how $J_c$ changes with both angle and field. So far, there has been no report on the angular dependence of these parameters in any pnictide superconductors. In this Chapter, the angular dependence of the upper critical field and the pinning potential of under-doped BaFe$_{1.9}$Co$_{0.1}$As$_2$ single crystal are investigated by measuring the magneto-transport at different magnetic fields and angles. Furthermore, by scaling the angular dependence of the resistance, based on the anisotropic Ginzburg-Landau (GL) theory, anisotropy value ($\Gamma'$) has been determined for different temperatures.

6.2 Experiments

Single crystal BaFe$_{1.9}$Co$_{0.1}$As$_2$ was grown using the high temperature self-flux method. FeAs and CoAs were prepared by placing a mixture of As powder and Fe/Co powder in a quartz tube and reacting it at 600$^\circ$C for 10 hours after it had been heated to 600$^\circ$C for 17 hours. A mixture of FeAs/CoAs and Ba pieces was then placed in an alumina crucible. The whole assembly was sealed in a large quartz tube and heated to 1180$^\circ$C for 15 hours, which was followed by a reaction at 1180$^\circ$C for 10 hours [189]. The as-grown single crystal was cleaved and cut into a rectangular shape for measurements. The transport properties were measured over wide ranges of temperature and magnetic field up to 13 T with applied current of 5 mA, using a
physical properties measurement system (PPMS, Quantum Design). The current was applied in the \textit{ab}-plane. The angular dependence of resistivity was measured using 13 T PPMS, with the angle, $\theta$, varied from $0^\circ$ to $180^\circ$, where $\theta = 0^\circ$ corresponded to the configuration of $B//c$ and $\theta = 90^\circ$ to $B//ab$, respectively.

6.3 Results and Discussion

The temperature dependence of in-plane resistivity of under-doped BaFe$_{1.9}$Co$_{0.1}$As$_2$ single crystal is shown in Figure 6-1. The resistivity decreases with decreasing temperature from 300 K to 50 K, supporting metallic behaviour of this compound. The resistivity increases with further decreasing temperature, however, and shows an anomaly at 49 K due to a magnetic/structural phase transition [71]. The $T^\text{onset}_c$ and $T_c(0)$ were determined to be 17.4 and 15.2 K, respectively.

![Figure 6-1](image)

Figure 6-1 Temperature dependence of the in-plane resistivity of BaFe$_{1.9}$Co$_{0.1}$As$_2$. The inset shows an enlargement of the region with $T \leq 35$ K and a schematic diagram of the sample.
The angular dependence of resistance at $T = 17$ K is shown in Figure 6-2(a). All the curves show a symmetric dip-like structure, with a minimum at 90° and maximum resistance at 0° and 180°. The normal state resistance decreases with decreasing magnetic field and temperature, due to the enhancement of superconducting state. The angular dependence of resistance is not very sharp with varying field and temperature, possibly due to moderate anisotropy of BaFe$_{1.9}$Co$_{0.1}$As$_2$ sample, which has also been reported in Nd-1111 single crystals [243]. Similar behaviour was observed at $T = 15$ and 12.5 K.

![Figure 6-2 (a) Angular dependence of the magnetoresistance at T = 17 K. (b) Resistance as a function of B at T = 17 K.](image)

In layered superconductors, variation in the superconducting order parameter, $\psi$, can be described by $\delta \psi/\delta z$ when the order parameter is quasi-continuous across the neighbouring layers. Therefore, the anisotropic Ginzburg-Landau approximation, $B_{c2}^{\alpha\beta}/\sqrt{\sin^2 \theta + \Gamma^2 \cos^2 \theta}$, can be used to estimate anisotropy values for our sample. The angular dependence of resistance can be scaled as $R = R(0) \cdot f(B/B_{c2}^{\alpha\beta})$ [244]. Then, the resistance measured under different magnetic fields should collapse into one curve at a certain temperature if the $\Gamma$ parameter is properly scaled. The results of this scaling at $T=17$ K are shown in Figure 6.2(b). The estimated anisotropy value
is 1.8 for $T = 12.5$ K, 1.9 for $T = 15$ K, and 2.1 for $T = 17$ K. The obtained anisotropy values are very close to the reported value of 2 at $T = T_c$ for Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals [132]. The anisotropy values which are obtained from this rescaling analysis are more reliable compared to estimating the anisotropy from the ratio of $B_{c2}$ in the $ab$-plane to that along the $c$-axis.

The results of magnetotransport measurements at $\theta = 45^\circ$ are shown in Figure 6-3(a), where $\theta$ is the angle between applied magnetic field and $c$-axis. According to the thermally activated flux flow model, the resistance can be described as:

$$R = (2v_0LB/J) \sinh \left( \frac{J_{c0}BV}{T} \right) e^{J_{c0}BV/LT} \tag{6-1}$$

where $v_0$ is the attempt frequency for flux bundle hopping, $L$ is the hopping distance, $J$ is the applied current density, $J_{c0}$ is the critical current density at 0 T, and $V$ is the bundle volume [141]. If $J_{c0}BV/L << 1$, Equation 6-1 can be written as:

$$R = (2R_cU/T) e^{-U/T} \tag{6-2}$$

where $U = J_{c0}BV$ is the thermally activated energy and the critical resistance, $R_c = v_0LB/J_{c0}$. Assuming that $2R_cU/T$ is a temperature independent constant, defined as $R_0$, and $U = U_0(1-T/T_c)$ then Equation 6-2 can be simplified as the Arrhenius equation:

$$\ln R(T, B) = \ln R_0 - U_0/T \tag{6-3}$$

Therefore, the activation energy, $U_0(B)$, is the slope of lower part of the curve in Arrhenius plot.
Figure 6-3 (a) Temperature dependence of resistance at different magnetic fields for $\theta = 45^\circ$, where $\theta$ is the angles between applied magnetic field and $c$-axis. (b) Arrhenius plots of resistance at same angle in under-doped BaFe$_{1.9}$Co$_{0.1}$As$_2$ single crystal.

In order to study the flux motion in BaFe$_{1.9}$Co$_{0.1}$As$_2$ single crystal, electrical resistance is plotted as a function of $1/T$ at different magnetic fields up to 7 T and at different angles between the applied magnetic field and $c$-axis. Figure 6-3(b) shows Arrhenius plots of resistance for $\theta = 45^\circ$. The linear dependence of $\ln R$ on $1/T$ in lower part of the curves indicates that this part can be described by thermally activated flux flow model [200], Similar measurements were performed for several other angles, from $0^\circ$ up to $90^\circ$, and $U_0(B)$ was calculated from the corresponding Arrhenius plots.

Figure 6-4 (b) shows the angular dependence of $U_0$ at different magnetic fields up to 7 T. The best fit of the experimental data yields pinning potential values ranging from $U_0/k_B = 2900$ K to $1900$ K, where $k_B$ is the Boltzmann constant, for $B//c$ and $B//ab$, respectively, at the low magnetic field of 0.1 T. These values are comparable to the reported values of $U_0 = 3000$-4000 K for NdFeAsO$_{0.82}$F$_{0.18}$ single crystals [105]. For comparison, we also included the $U_0$ for Bi-2212 single crystal in
Figure 6-4(a). The $U_o$ value for under-doped BaFe$_{1.9}$Co$_{0.1}$As$_2$ is three times higher than for Bi-2212 single crystal [127]. This value is lower, however, than the reported value of 9100 K for Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$ single crystal [128] for $B//ab$. The $U_o$ value for our under-doped BaFe$_{1.9}$Co$_{0.1}$As$_2$ crystal with $B//c$ exhibits a weak power-law dependence ($U_o \propto B^{-0.15}$) for $B < 3$ T, where single vortex pinning dominates, followed by a power-law decrease with $U_o \propto B^{-0.81}$, indicating collective pinning at higher fields, $B > 3$T. The pinning potential values decrease when the sample is rotated from $B//c$ to $B//ab$. It should be noted that the $U_o$ decreases slightly for $0 < \theta \leq 45^\circ$, and then remains constant for $\theta \geq 45^\circ$. Twin boundaries, which can form below the temperature of structural transition from tetragonal to orthorhombic, could possibly act as barriers to vortex motion, resulting in the higher $U_o$ value for $B//c$ [245, 246] compared to that for $B//ab$.

![Figure 6-4(a) Field dependence of $U_o$ for BaFe$_{1.9}$Co$_{0.1}$As$_2$ single crystal compared with other values from the literature [105, 127, 128]. (b) Angular dependence of pinning potential for different magnetic fields.](image)

The critical current density, $J_c$, vs. magnetic field is shown in Figure 6-5(a). $J_c$ was calculated from the magnetic hysteresis data at $T = 2$ and 4.2 K by using the extended Bean model:
\[ J_c = 20 \times \Delta M/V/(a(1-a)/3b)) \] with \( a<b \)

Where \( a \) and \( b \) are the sample dimensions, \( V \) is the sample volume, and \( \Delta M \) is the height of the magnetization loop. The \( J_c \) is as high as \( 1.7 \times 10^5 \) and \( 1.5 \times 10^5 \) A/cm\(^2\) at \( T = 2 \) and 4.2 K in zero magnetic field, respectively. The \( J_c \) decreases with increasing magnetic field up to 1 T, and after that, it become nearly field independent, which is related to the relatively high pinning potential and weakly anisotropic properties in BaFe\(_{1.9}\)Co\(_{0.1}\)As\(_2\) single crystal.

![Figure 6-5](image.png)

Figure 6-5. (a) Field dependence of \( J_c \) at \( T = 2 \) and 4.2 K. (b) Angular dependence of \( B_{c2} \) as functions of temperature. (c) Angular dependence of \( dB_{c2}/dT \). (d) Angular dependence of \( B_{c2} \) (at \( T = 15 \) K).

The upper critical field, \( B_{c2} \), is characterized as the field at which resistivity becomes 90% of the normal state resistivity. Figure 6.5(b) shows \( B_{c2} \) as functions of temperature for different angles between field and \( c \)-axis. From Figure 6-5(c), \( B_{c2} \)
follow the same trend with angle for $\theta > 45^\circ$. The estimated slopes $d{B_c^2}_{ab}/dT$ is 4.8 T/K for $B//ab$. The $d{B_c^2}_{ab}/dT$ is in good agreement with the reported value of $d{B_c^2}_{ab}/dT = 4.9$ T/K [132]. The slopes decrease as the sample is rotated from 0° to 90° [Figure 6-5 (d)], which is similar to the trend of $U_o$ with increasing angle.

In summary, the angular dependence of $U_o$ and $B_c^2$ were investigated for under-doped BaFe$_{1.9}$Co$_{0.1}$As$_2$ single crystal. $U_o$ decreases while $B_c^2$ increase with increasing angle from $B//c$ to $B//ab$. The anisotropy parameter decreased from 2.1 to 1.8 as $T$ decreased from 17 to 12.5 K, using the anisotropic GL theory.
CHAPTER 7
THE FLUX PINNING MECHANISM, AND ELECTRICAL AND MAGNETIC ANISOTROPY IN Fe_{1.04}Te_{0.6}Se_{0.4} SUPERCONDUCTING SINGLE CRYSTAL

7.1 Introduction

Following the discovery of superconductivity in REFePnO (RE = rare earth, Pn = P or As) [8, 36, 247, 248, 249], doped AFe₂As₂ (A = alkaline or alkaline earth metal) [13, 39], LiFeAs [40], and (Sr₄M₂O₆)Fe₂Pn₂ (M = Sc, Ti, or V) [58, 250] families of iron pnictide superconductors, the observation of superconductivity in tetragonal FeSe [53] has opened up a new window of opportunity to further understand the mechanism of superconductivity in iron pnictides. The iron chalcogenide FeSe compounds are of great interest from the viewpoints of both vortex properties in the mixed state and practical application. This is largely due to the relatively simple structure and similarity in the Fermi surface (Eₕ) of these arsenic-free compounds to the other pnictide superconductors. The Eₕ surfaces of FeSe and FeTe contain cylindrical hole and electron sections at the centre and the corner of the Brillouin zone, respectively [251]. It has proven difficult, however, to grow homogeneous superconductive single crystals. Therefore, attention has been paid to some extent to the Te doped systems [103]. FeSe₁₋ₓTeₓ compounds have the tetragonal structure, where Fe (Se/Te) layers stack along the c-axis, and have critical temperature, T_c, as high as 15 K [103, 252, 253]. The antiferromagnetic order of FeTe is gradually suppressed by increasing x in FeTe₁₋ₓSeₓ, and the maximum T_c is achieved for x = 0.5 [254].
It is crucial to understand the pinning mechanism from both practical and fundamental points of view. There are two main interactions between vortices and pinning centres in type II superconductors: the magnetic interaction and the core interaction [141]. The magnetic interaction is due to the interaction at interfaces between superconducting and non-superconducting materials parallel to the applied magnetic field. The core interaction covers pinning due to the variation in transition temperature ($\delta T_c$) and pinning because of the variation in charge carrier mean free path near lattice defects ($\delta l$) [141]. For FeTe$_{0.5}$Se$_{0.5}$ single crystal, it has been found that the dominant pinning mechanism is $\delta l$ pinning, which is related to small bundle vortex pinning due to randomly distributed weak pinning centres [165]. Yadav et al. have studied the flux pinning force, $F_p$, in FeTe$_{0.6}$Se$_{0.4}$ and found that the obtained $h_{\text{max}} = 0.28$ (where $h_{\text{max}}$ is the field corresponding to maximum pinning force density, normalized with respect to the irreversibility field), which can be understood in terms of $\delta l$ pinning with a mixture of surface pinning and the point core pinning due to normal pinning centres, with different ranges of pinning interactions [168].

In this Chapter, the vortex pinning mechanisms, pinning potential, and anisotropy of Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ single crystal have been studied systematically by magnetic and transport measurements at different temperatures. The anisotropy value was determined by scaling the angular dependence of resistivity, based on the anisotropic Ginzburg-Landau (GL) theory.

### 7.2 Experimental

Single crystals of Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ were prepared by a self-flux method. Details of the single crystal growth are reported elsewhere [255]. The as-grown crystals were cleaved and cut into a rectangular shape for magnetic and transport measurements.
that were performed over wide ranges of temperature and magnetic field up to 13 T, using a physical properties measurement system (PPMS, Quantum Design). The angular dependence of the resistivity was also measured, with the angle, $\theta$, ranging from 0° to 180°, where $\theta = 0°$ corresponds to $B//c$ and $\theta = 90°$ to $B//ab$, respectively. The critical current density, $J_c$, was calculated using the Bean model.

### 7.3 Results and discussions

Figure 7.1(a) shows typical hysteresis $MH$ loops collected at several temperatures below $T_c$. The minimum magnetic moment located at nearly zero field represents the first magnetization peak. The field completely penetrates into bulk of the sample after zero field cooling [165]. The second magnetization peak (SMP) or fishtail effect can be seen at 4 K, 7 K, and 8 K. The onset ($B_{SMP}^{onset}$) and peak ($B_{SMP}^{peak}$) positions of the second magnetization peak are indicated by arrows in Fig. 7.1(a) for $T = 4$ K. Both the $B_{SMP}^{onset}$ and the $B_{SMP}^{peak}$ positions move to lower magnetic fields as the temperature increases from 4 to 10 K and completely disappear at $T = 11$ K. $J_c$ was calculated at various temperatures from these $MH$ loops by using the Bean model [136]. For a rectangular shaped crystal with dimensions $c < a < b$, when $B//c$, the in-field critical current density, $J_c(B)$, is given by

$$J_c(B) = \frac{20* \triangle m (B)}{a (1-a/3b)},$$

where $\triangle m$ is the difference between magnetizations measured during the return and forward legs of the $M$–$H$ loop at a particular applied magnetic field, and $a$ and $b$ are the length and width of the sample perpendicular to the applied field.
Figure 7-1 (a) M-H loops of BaFe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ single crystal, and (b) $J_c$ vs. field at several temperatures.

$J_c(B)$ (Figure 7-1 (b)) shows a fast decrease at low fields ($B < 0.5$ T), followed by weak field dependence at high fields at $T = 4$ K. The in-field $J_c$ is as high as $1.2 \times 10^9$ A/m$^2$ at 4 K and zero magnetic field. This value is slightly higher than the reported value ($1 \times 10^9$ A/m$^2$ at $T = 1.8$ K and low field) in a FeTe$_{0.6}$Se$_{0.4}$ single crystal [168, 256]. It is likely that this is due to the enhanced iron concentration in Fe$_{1.04}$Te$_{0.6}$Te$_{0.4}$ single crystal, which introduces more defects into the crystal structure and consequently, higher pinning potential into the system.

In order to understand the flux pinning mechanism which controls the vortex pinning force, it is useful to look at the variation of pinning force density with magnetic field. In the mixed state of type II superconductors, the pinning force should follow the same general relationship, if the flux pinning is dominated by a single mechanism [166]. The pinning force, defined as $F_p = \mu_0 B \times J_c$, can be calculated from the $J_c$ values shown in Fig. 7.1(b). In Figure 7-2, we plot the reduced
pinning force $f$ versus the reduced magnetic field $h$ ($f = F_p/F_{p,\text{max}}, \ h = B/B_{\text{irr}}$) at the same temperature as in Figure 7-1(b). There are various methods to determine the irreversibility field, $B_{\text{irr}}$, from the magnetization and resistivity measurements [257]. Here, we use $B_{\text{irr}}$ as the field at which $J_c(B)$ is extrapolated to $10^6$ A/m$^2$. The curves show a scaling behaviour at $T < 10$ K, indicating that a single pinning mechanism dominates at this temperature range. We fit these data within the Dew-Hughes scenario:

$$F_p \approx h^p (1-h)^q$$

(7.2)

The best fit of the curves (dashed-dotted line in Fig. 7.2) is obtained with an $f(h)$ dependence given by $h^{1.35}(1-h)^{3.1}$. The obtained $p$ and $q$ values are slightly lower than the reported values of $p = 1.54$ and $q = 3.8$ in FeTe$_{0.6}$Se$_{0.4}$ single crystal [168], possibly due to excess iron concentration in this compound. The value of $p/(p+q) \approx 0.3$ agrees well with the peak positions of these in $f$ vs. $h$ plot, and it is in good agreement with the reported value of $h_{\text{max}} \approx 0.28$ for FeTe$_{0.6}$Se$_{0.4}$ single crystal [168]. It should be noted that according to the Dew-Hughes theory [166], point pinning is expected to lead to $p = 1$ and $q = 2$, with $F_{p,\text{max}}$ occurring at $h_{\text{max}} \approx 0.33$, whereas, in a system dominated by grain boundary pinning, $h_{\text{max}} \approx 0.2$. In the case of pinning due to variation in the superconducting order parameter, however, $h_{\text{max}} \approx 0.7$. In this case, $h_{\text{max}} \approx 0.3$, implying $\delta l$ pinning with a mixture of point pinning and grain boundary pinning. This value is slightly higher than the reported value of $h_{\text{max}} = 2.8$ for FeTe$_{0.6}$Se$_{0.4}$ single crystal [168], likely because of enhanced point pinning centres due to the increased iron concentration in Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ single crystal.
Figure 7-2 Field dependence of the reduced pinning force, with the fitting results obtained from $h^P(1-h)^q$. Inset shows $F_p/F_{p,max}$ as a function of field.

The temperature dependence of resistivity of Fe$_{1.04}$Te$_{0.6}$Te$_{0.4}$ for $B$/$c$ is shown in Fig. 7.3(a), where the onset of $T_c$ gradually shifts to lower temperatures with increasing magnetic field. It is worth noting that the shape of $\rho_{ab}(T)$ with $B$/$c$ is comparable to those for (Ba, K) Fe$_2$As$_2$ and (Ba, Rb) Fe$_2$As$_2$ single crystals [258, 259], and it is different from the shapes for SmFeAsO$_{0.8}$F$_{0.2}$ and NdFeAsF$_{0.82}$F$_{0.18}$ single crystals [107, 260], where resistive tails were clearly observed for $B$/$c$. According to the thermally activated flux flow model, the resistivity can be described by the following Arrhenius equation:

$$\ln R(T,B) = \ln R_0 - U_o/T, \quad (7.3)$$

Therefore, the activation energy, $U_o(B)$, is slope of the lower part of curve in the Arrhenius plot. In order to study the flux motion in Fe$_{1.06}$Te$_{0.6}$Te$_{0.4}$ single crystal, the resistivity is plotted as a function of $1/T$ at different magnetic fields up to 13 T.
Figure 7-3 (a) Temperature dependence of resistivity under different magnetic fields for \(B//c\). (b) Arrhenius plots of resistivity for the same magnetic fields.

Figure 7-3(b) shows Arrhenius plots of resistivity for \(B//c\). The linear dependence of \(\ln \rho\) vs. \(1/T\) in the lower part of curves indicates that this part can be described by the thermally activated flux flow model [200]. Similar measurements were performed for \(B//ab\) and also for several angles, from 0\(^\circ\) up to 90\(^\circ\) for \(B = 6\) T, and \(U_o(B)\) was calculated from the corresponding data.

Figure 7-4 (a) Magnetic field dependence of \(U_o\) for \(B//ab\) and \(B//c\). (b) Angular dependence of \(U_o\) for \(B = 6\) T.
The magnetic field dependence of pinning potential for $B//c$ and $B//ab$ is shown in Figure 7-4(a). The best fit to the experimental data yields a value of the pinning potential of 404 K for $B//ab$ at $B = 0.1$ T. The activation energy drops very slowly with increasing applied magnetic field for $B < 5$ T, scaled as $B^{-0.1}$, and then decreases slowly as $B^{-0.61}$ for $B > 5$ T. This indicates that the pinning potential is almost field independent for $B < 5$ T. The angular dependence of the pinning potential for $B = 6$ T is shown in Figure 7-4(b). The pinning potential values increase when the sample is rotated from $B//c$ to $B//ab$.

Figure 7-5 Scaling of the resistance as a function of $B/(\sin^2 \theta + \Gamma^2 \cos^2 \theta)^{1/2}$, based on GL theory at $T = 14$ K. Inset shows the angular dependence of the resistivity at 14 K under different magnetic fields.

The angular dependence of resistivity for Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ at 14 K is shown in the inset of Figure 7-5. All the curves have a symmetric cup-like shape, and the minimum value is at $\theta = 90^\circ$, where $\theta$ is the angle between applied magnetic field.
and the $c$-axis. According to the anisotropic Ginzburg-Landau model, the effective upper critical field, $B_{c2}^{\text{GL}}(\theta)$, can be characterized as [141]

$$B_{c2}^{\text{GL}}(\theta) = B_{c2,ab} / (\sin^2 \theta + \Gamma^2 \cos^2 \theta)^{1/2}$$  \hspace{1cm} (7.4)$$

where $\Gamma$ is the anisotropy of the sample. As resistivity in the mixed state depends on the effective field [244], the angular dependence of resistivity can be scaled as $\rho = \rho_o \cdot f(H) / B_{c2}^{\text{GL}}$, where $\rho_o$ is the temperature independent part of resistivity. Then, the resistivity measured under different magnetic fields should collapse into one curve at a certain temperature if the anisotropy parameter is properly scaled. Good scaling behaviour can be obtained for Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ with $\Gamma = 2.5$ at $T = 14$ K, as shown in the main panel of Figure 7-5. This value is higher than the obtained value of $\Gamma = 2$ for Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ single crystal [125]. Variation in the value of anisotropy could be a result of different Fe concentration. It should also be mentioned that anisotropy values were obtained using different methods, GL theory in our work, and the ratio of upper critical field along the $ab$-plane and to that along the $c$-axis in Ref. [125].

In summary, we have studied the pinning mechanism, pinning potential, and anisotropy of Fe$_{1.04}$Te$_{0.6}$Se$_{0.4}$ single crystal. The obtained values of $p$ and $q$ based on the Dew-Hughes model indicate the presence of $\delta l$ pinning, with a mixture of surface and point core pinning in this compound. In addition, the pinning potential value was obtained using the thermally activated flux flow model. The anisotropy value was obtained using GL theory.
CHAPTER 8
ANISOTROPIC THERMALLY ACTIVATED FLUX FLOW AND THE VORTEX GLASS TRANSITION IN Fe1.06Te0.6Se0.4 SUPERCONDUCTING SINGLE CRYSTALS

8.1 Introduction

The capability to carry high transport current in magnetic field is one of the most significant aspects of superconductors. The limiting value of critical current is given by the balance between pinning force due to the spatial variation of condensation energy and, on the other hand, Lorentz force applied by the transport current. Flux creep and flux flow are the two distinguishable regimes of dissipation. Flux creep occurs when pinning force dominates and flux flow when the Lorentz force dominates. The activation energy for flux motion can be estimated from dc resistivity measurements. It is crucial to understand the thermally activated energy and the depinning critical current from both practical and fundamental points of view. Thermally activated energy (TAE) has been well studied in high temperature cuprates [127, 261, 262, 263, 264]. It is well known that strong thermal fluctuation of high temperature superconductors is due to very high transition temperature, short coherence length, and high anisotropy of these compounds, which result in broadening of superconducting transition with applied magnetic field. Iron-based superconductors show a relatively high transition temperature, $T_c$, and short coherence length. They reveal nearly isotropic superconductivity, however, which makes them distinct from cuprates. For iron-based superconductors, the thermal fluctuations of vortices can lead to thermally activated flux flow (TAFF), causing the resistance transition from the $R(T,B)$ curve to shift to lower temperatures and also broaden as the field increases. For
example, $\textit{REFeAsO}_{1-x}\textit{F}_x$ [105, 265, 266, 267, 268], where $\textit{RE}$ is a rare earth element, shows similar transition broadening to $\textit{YBa}_2\textit{Cu}_3\textit{O}_{7-x}$ (YBCO) with increasing field. On the other hand, thermal fluctuations are negligible in Ba-122 compounds, as resistive transition curves $R(T,H)$ shift to lower temperature [128], but do not broaden as the field is increased. The broadening is intermediate for iron chalcogenides. $\text{FeSe}_{1-x}\text{Te}_x$ compounds have tetragonal structure, where Fe (Se/Te) layers are stacked along $c$-axis, and have $T_c$ as high as 15 K [103, 252, 253]. The antiferromagnetic order of FeTe is gradually suppressed by increasing $x$ in $\text{FeTe}_{1-x}\text{Se}_x$, and the maximum $T_c$ is achieved for $x = 0.5^{13}$. It was reported, however, that $T_c$ can reach 37 K under pressure for FeSe compounds [269]. Possible superconductivity above 77 K in single unit cell FeSe films on SrTiO$_3$ (STO) substrate [270] has been reported very recently. The arsenic-free $\text{Fe}_{1+y}\text{Se}_{1-x}\text{Te}_x$ compounds are of great interest from the viewpoints of both vortex state and practical applications. This is due to their simple structure and nearly isotropic upper critical field. Also, the high critical current density of $J_c > 10^6 \text{ A} \cdot \text{cm}^{-2}$ under very high field of 30 T that has been recently achieved in FeSe$_{0.5}$Te$_{0.5}$ coated conductors [271] is another significant aspect of these compounds. In addition, as iron is the only magnetic element in such compounds, it provides a unique opportunity to study the effects of excess iron in Fe position on the vortex motion and thermally activated energy. Therefore, high quality single crystals of these compounds are perfect candidates to study vortex properties and thermally activated energy of iron chalcogenides.

In this Chapter, the thermally activated flux flow (TAFF) behaviour of $\text{Fe}_{1.06}\text{Te}_{0.6}\text{Se}_{0.4}$ single crystals is investigated in magnetic field up to 13 T, using the conventional Arrhenius relation and modified TAFF model. It will be shown that Arrhenius curve slopes are directly related to, but not equal to, the activation energies
of Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystals. Therefore, the use of a modified TAFF model, $\rho(T, B) = \rho_0 \exp(-U/T)$, is suggested, where the temperature dependence of prefactor $\rho_0 = 2 \rho_c U/T$ and nonlinear relation of thermal activation energy, $U(T, B)$, are considered. The modified TAFF method results are in good agreement with very high $J_c$ values from experimental data. It was found that Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ superconductor can be regarded as both a 3D- and 2D-like system, which is dependent on magnetic field direction in the TAFF region. The vortex phase diagram has been determined based on the evolution of vortex-glass transition temperature, $T_g$, with magnetic field and upper critical field.

8.2 Experiments

Single crystals of Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ were prepared by a self-flux method. Details of the single crystal growth are reported elsewhere [255]. The as-grown single crystal was cleaved and cut into a rectangular shape for transport and magnetic measurements. The transport properties were measured over a wide range of temperatures and magnetic fields up to 13 T, with applied current of 5 mA, using a physical properties measurement system (PPMS, Quantum Design).

8.3 Results and discussion

Figure 8-1 shows the resistivity $\rho(T, B)$ of Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystal near the $T_c$ for $B//ab$ and $B//c$. The onset of $T_c$ moves to lower temperature with increasing magnetic field for both $B//ab$ and $B//c$. The trend is more noticeable, however, for $B//c$ than for $B//ab$. 

Figure 8-1. Resistivity curves for Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystals under various applied magnetic fields for (a) $B//c$ and (b) $B//ab$.

Figure 8-2. Determination of the vortex glass transition temperature from Eq. (8.1) for Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ at different magnetic fields.

According to vortex phase transition theory [223], in vortex glass state and close to glass transition temperature, $T_g$, resistivity disappears as a power law

$$\rho = \rho_0 |T/T_g - 1|^\xi$$  \hspace{1cm} (8.1)
where $s$ is a constant and depends on the kind of disorder, and $\rho_0$ is a characteristic resistivity that is related to the normal state. Therefore, resistivity goes to zero at $T_g$. Consequently $T_g(B)$ can be extracted by applying the relation, $(\text{dln}\rho/\text{dT})^{-1} \alpha (T-T_g)/s$, to resistive tails. Figure 8.2 presents the resistivity of Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ based on vortex-glass model, Eq. (8.1), in the temperature range $T_g < T < T^*$, with intercept $T_g$ and $s = 2.3 \pm 0.1$. It is clear that resistivity can be well described by the vortex glass model. The estimated vortex glass transition line is shown in Figure 8.3.

The upper critical field, $B_{c2}$, is characterized as the field at which resistivity becomes 90% of normal state resistivity. Figure 8.3 shows $B_{c2}$ as functions of temperature for $B//ab$ and $B//c$. $B_{c2}$ exhibits a linear temperature dependence for both $B//ab$ and $B//c$. The estimated slopes for $B_{c2}$ are -9.4 and -6.5 T/K for $B//ab$ and $B//c$, respectively. $B_{c2}$ was estimated by using the conventional one-band Werthamer–Helfand–Hohenberg (WHH) theory: $B_{c2}(0) = -0.69 T_c (dB_{c2}/dT)$, assuming the upper critical field is limited by orbital pair breaking effect. The estimated values of $B_{c2}$ close to the zero temperature limit for $B//ab$ and $B//c$ are $\mu_0 B_{c2}^{ab} = 99.9$ T and $\mu_0 B_{c2}^{c} = 65$ T, respectively. The estimated $B_{c2}$ calculated from WHH theory is higher than Bardeen Cooper Schrieffer (BCS) paramagnetic limit, $B_{p}^{BCS}$, in the weak coupling regime. By using the weak coupling BCS formula, $B_{p}^{BCS} = 1.84 T_c$, we obtain $B_{p}^{BCS} = 28.3$ T and 26.7 T for $B//ab$ and $B//c$, respectively. The estimated $B_{c2}^{ab}$ and $B_{c2}^{c}$ from the WHH formula are 3.5 and 2.4 times these limits for $B//ab$ and $B//c$, respectively, indicating that Zeeman paramagnetic pair breaking possibly is essential for both directions. Also, it reveals the unconventional superconducting mechanism in this family. The anisotropy value, $\Gamma$, obtained using $\Gamma = B_{c2}^{ab}/B_{c2}^{c}$, is equal to 1.5. According to the collective pinning model [126], disorder-induced spatial fluctuations in solid-vortex lattice can be clearly divided into markedly different regimes according
to the strength of applied field. Two different regimes are distinguishable: (1) the vortex glass, which governs the region below transition field, $B_g$; and (2) the vortex liquid, which holds between $B_g$ and $B_{c2}$, where thermal fluctuations are important. As can be seen from Figure 8.3, the vortex-glass phase indicates that Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystal features only a narrow region of the vortex-liquid phase, which is denoted by $\Delta T$, with a $\Delta T^{\text{in}}$ and $\Delta T^{\text{ab}}$ of 3.1 ± 0.5 K and 2.6 ± 0.2 K, respectively, at magnetic field of 0 up to 13 T. This result suggests that the vortex-glass region depends weakly on magnetic field, which originates from vastly enhanced vortex pinning in the studied magnetic field levels.

Based on the TAFF theory [126, 261], resistivity in the TAFF regime can be written as:

$$
\rho = \left( \frac{2\nu_0 LB}{J} \right) \exp \left( -\frac{J_{c0} BV L}{T} \right) \sinh \left( \frac{JB VL}{T} \right)
$$

(8.3)

Where $\nu_0$ is the attempt frequency for a flux bundle of volume $V$, $L$ is the hopping distance, $B$ is the magnetic induction, $J$ is the applied current density, $J_{c0}$ is the critical current density in the absence of flux creep, and $T$ is the temperature. If $J BV L \ll 1$ and $J$ is small enough, then Equation (8.3) can be rewritten as:

$$
\rho (T, B) = \left( 2\rho_c U/T \right) \exp \left( -U/T \right) = \rho_0 f \exp \left( -U/T \right)
$$

(8.4)

Here $U = J_{c0} BLV$ is the thermal activation energy (TAE), $\rho_c = \nu LB/J_{c0}$, and $\rho_c U/T$ is considered as the prefactor $\rho_0 f$. Mostly, the TAE of cuprates and iron-based superconductors (FBSs) is analysed using Equation (8.4), assuming that $2\rho_c U/T$ is temperature independent. Then $U(T, B) = U_0(B) \left( 1 - t \right)$, where $t = T/T_c$, and $\ln \rho$ vs. $1/T$ becomes the Arrhenius relation, $\ln \rho(T, B) = \ln \rho_0(B) - U_0(B)/T$. Here, $B$ is the magnetic field.
field strength, and \( \ln \rho_0(B) = \ln \rho_{0f} + U_0(B)/T_c \). Moreover, it can be resolved that 
\[ \partial \ln \rho / \partial (1/T) = U_0(B) \]. Therefore, \( \ln \rho \) vs. \( 1/T \) should be linear in the TAFF regime where the slope is \( U_0(B) \), and its \( y \) intercept is represented by \( \ln \rho_{0f}(B) \). It is likely that \( U \neq U_0 \) and \( \rho_{0f} \neq \) constant, however, as the lowest temperature part of \( \rho(T) \) curve is used for determination of \( U_0 \) in Arrhenius model. It is suggested \([262]\) that temperature dependence of \( \rho_{0f} \) in Equation 8.4 should be taken into account for analysis. According to the condensation model, \( U_0 \propto B_c^2(t) \xi^n(t) \), where \( H_c \) is the thermodynamic critical field, \( \xi \) is the coherence length, \( t = T/T_c \), and \( 0 < n < 3 \) \([127]\), depending on the dimensionality of the vortex system. Since \( B_c(t) \propto 1-t \) and \( \xi(t) \propto (1-t)^{-1/2} \) near \( T_c \) then

\[ U(T, B) = U_0(B) (1-t)^q \]  
(8.5)

where \( q = 2-n/2 \), which shows dimensionality dependence. Generally, \( q = 1.5 \) is observed in high temperature superconductors showing 3D behaviour, whereas \( q = 2 \) represents 2D behaviour \([272, 273, 274]\). Combining Equations (8.4) and (8.5), it can be derived that

\[ \ln \rho = \ln(2 \rho_c U_0) + q \ln(1-t) - \ln T - U_0(1-t)^q/T \]  
(8.6)

where \( \rho_c \) and \( U_0 \) are temperature independent and \( T_c \) is obtained from Arrhenius fitting. Therefore, slope of the Arrhenius plot near \( T_c \) is given by:

\[ -\frac{\partial \ln \rho}{\partial (1/T)} = [U_0(1-t)^q - T][1 + \frac{qt}{1-t}] \]  
(8.7)

Equation (8.7) is known as the modified TAFF model, and the fit using this model is in better agreement with experimental results than Arrhenius model for cuprates and some iron-based superconductors \([261, 262, 267, 275, 276]\). According to Equation (8.7), the activation energy obtained from the slope of Arrhenius plot near \( T_c \) is
increased with respect to the actual value by $[1 + q t/(1-t)]$. As the $U_0$ is strongly temperature dependent near $T_c$, the enhancement is large.

Figure 8-3. Temperature dependence of $B_{c2}$ for $B//ab$ and $B//c$. $\Delta T$ indicates the vortex-liquid region.

Figure 8.4 presents the Arrhenius plots of $\rho (1/T)$ at different magnetic fields for $B//ab$ and $B//c$. The red solid lines show the results of linear fitting in the low-resistivity range. All the linear fittings cross at approximately $T_c$, which is about 14.6 and 15.1 K for $B//c$ and $B//ab$, respectively. The slope of these Arrhenius plots for low resistivity can be related to the activation energy. The insets show $\ln \rho_c$ vs. $U_0$, which are obtained from the linear fits of Arrhenius results. Based on $\ln \rho_0(B) = \ln \rho_{0f} + U_0 (B)/T_c$, $\ln \rho_{0f}$ and $T_c$ can be obtained by linear fitting.
Figure 8-4. Longitudinal resistivity in different magnetic fields for (a) $B_{//c}$ and (b) $B_{//ab}$. The corresponding solid red lines and blue dashed lines are fitting results from the Arrhenius relation and Equation (8.6), respectively. The insets show $\ln \rho_0$ vs. $U_0$ data, which were obtained by using Arrhenius plots. The green dashed line is the linear fit to inferred data.

The values of $T_c = 14.9$ and $15.8$ K for $B_{//c}$ and $B_{//ab}$, respectively, are in good agreement with the obtained values of $T_{cross}$, the points where the linear fits cross, within the range of error. It is likely that the obtained values of $U_0$ are not accurate enough, as they are only estimated based on the lowest temperature part of the $\rho(T)$ curve in a very narrow area, i.e., Arrhenius relation in the TAFF region. Then, the effects of nonlinear relationship of $U(T, B)$ against $T$ and temperature dependence of $\rho_c$ should be considered. Therefore, Equation (8.6) was fitted to the experimental data. The blue dashed curves in Figure 8.4 represent the results from Equation (8.6). All fits are in good agreement with the experimental data, and the results are more accurate than the Arrhenius model, which just covers a very narrow resistivity area at low temperature.

It should be emphasized that the actual value of $U_0$ is two times higher for $B_{//ab}$ than in the $B_{//c}$ direction. It is likely that coupling strength between FeAs planes,
which determines the pinning behaviour, is more effective than the actual defect structure [127].

Figure 8-5. $q$ as a function of magnetic field. $q$ is obtained from fitting resistivity in TAFF regime using Equation (8.6) for both $B//ab$ and $B//c$.

Figure 8-5 presents the magnetic field dependence of $q$, which is obtained from the best fits of experimental data to Equation (8.6) for $B//ab$ and $B//c$. The value of $q$ is $2.1 \pm 0.1$ for $B//ab$. For $B//c$, the value of $q$ in Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystals is about $1.5 \pm 0.1$, which is similar to estimated $q = 1.5$ in LiFeAs crystal$^{20}$, but is different from the value of $q = 2$ in Fe$_{1+y}$(Te$_{1+z}$S$_z$)$_2$$^{19}$, SmFeAs$_{0.9}$F$_{0.1}$ [267], and many cuprates [262, 264], which generally show 2-dimensional behavior with a similar scaling. Therefore, Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ superconductor has a small anisotropy like that of LiFeAs superconductor and can be regarded as a 3D-like system in the TAFF region for $B//c$. The different values of $q$ for $B//c$ and $B//ab$ indicate that the dimensionality for $B//ab$ is very much closer to two-dimensional behaviour than that for $B//c$. These results suggest that temperature dependence of pinning potential is correlated with
dimensionality behaviour in Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ single crystal. Therefore, one can tune the effective pinning potential with the direction of applied magnetic field with respect to $c$-axis or $ab$-plane directions, and the crossover from 2D to 3D can be found.

![Figure 8.6](image)

**Figure 8.6.** Magnetic field dependence of $U_0$ obtained from (a) modified TAFF model (Equation (8.3)) and (b) Arrhenius relationship for $B//ab$ and $B//c$. The dashed lines are power law fittings using $U_0(B) \propto B^{-n}$

Figure 8.6 shows the magnetic field dependence of $U_0$ obtained from (a) modified TAFF model using Equation (8.5) and (b) Arrhenius relationship for $B//ab$ and $B//c$. In both field directions, $U_0(B)$ indicates power law field dependence for both the modified TAFF model and Arrhenius relation. Using the modified TAFF model, for $B//ab$, $n = 0.51$ for $B > 5$ T and $n = 0.08$ for $B < 5$ T, while for $B//c$, $n = 0.98$ for $B > 5$ T and $n = 0.1$ for $B < 5$ T. It is likely that single vortex pinning is dominant at low magnetic field, as $U_0$ decreases very slowly with increasing magnetic field$^{21}$. On the other hand, $U_0$ becomes strongly field dependent for $B > 5$ T, indicating the crossover from single vortex pinning to a collective pinning regime, as vortex spacing becomes significantly smaller than the penetration depth in higher fields. The obtained values of $U_0$ using the Arrhenius relation are nearly five times smaller than the obtained values using modified TAFF model at low magnetic field. The higher value of $U_0$ obtained from the modified TAFF model is in good agreement with the high value of the
critical current density due to high pinning potential in this compound\textsuperscript{22,[271]. The trend in $U_0(B)$ is similar in both models, however. In other words, $U_0(B)$ is revealed to be weakly field dependent for $B < 5$ T, but it becomes strongly field dependent for $B > 5$ T. It is likely that the slopes of Arrhenius plots are directly related to, but not equal to, the real value of activation energy. Similar behaviour has been reported for YBa$_2$Cu$_3$O$_7$ single crystals\textsuperscript{1}.

Figure 8-7. $-d(ln\rho)/d(1/T)$ as a function of temperature in different magnetic fields for $B//c$ and $B//ab$. The solid red lines represents the $U_0$ obtained from linear fitting of Arrhenius relation, and the green dashed curves are plotted using Equation (8.6), with fitting parameters $U_0(B)$ and $q$ obtained from the modified TAFF model (Equation 8.5 in Figure 8.5). The insets show the inverse temperature dependence of $ln\rho$.

The values of $U_0$ are estimated from limited temperature interval where the data reveal linear behaviour on the Arrhenius plot. Even if the slope does not change significantly in this temperature interval, it doesn’t demonstrate that $U_0$ is temperature independent [127]. To investigate whether $U_0$ is temperature independent or not, $-d(ln\rho)/d(1/T)$ was plotted as a function of temperature in different magnetic fields for $B//ab$ and $B//c$ in Figure 8.7. In the normal state for $T > T_c$, $-d(ln\rho)/d(1/T)$ is almost temperature and magnetic field independent, but then for $T < T_c$, it gradually increases with the onset of superconductivity. Then, it is enhanced sharply in the
superconducting regime with increasing temperature, which is related to the TAFF regime. If \( U(T, B) = U_0(B) \) (1-\( t \)) and \( \rho_{0f} = \text{const} \), then \(-d(\ln \rho)/d(1/T) = U_0(B)\), and therefore, \( U_0(B) \) should be a set of horizontal lines. The horizontal red lines in Figure 8.7 represent the \( U_0(B) \) values, with each of them having a limited length. Each length covers the temperature interval that relates to the interval of 1/\( T \) for estimating \( U_0(B) \) in the Arrhenius plot. It should be noted that the slopes change with temperature without reaching a constant value. The \(-d(\ln \rho)/d(1/T)\) curve increases with decreasing temperature and almost crosses the centre of horizontal \( U_0(B) \) lines. This means that \( U(T, B) \neq U_0(B) \) (1-\( t \)) and \( \rho_{0f} \) is temperature dependent, while each \( U_0(B) \) value is only the average value of \(-d(\ln \rho)/d(1/T)\) in the temperature area of fitting, and the values of \( U_0 \) obtained from Arrhenius relation are estimated in a very narrow temperature interval. Therefore, it is likely that the \( U_0(B) \) values estimated from conventional Arrhenius model are not accurate enough. Then, temperature dependence of \( \rho_{0f} \) and the nonlinear relation of \( U(T, B) \) should be considered [262]. The dashed green lines in Figure 8.7 are plotted using Equation (8.7). The \( U_0 \) and \( q \) parameters were determined by fitting Equation (8.5) to the corresponding resistivity data in Figure 8.4. It is obvious that the modified TAFF model can effectively fit the upturn trend of \(-d(\ln \rho)/d(1/T)\) with decreasing temperature and can give a more accurate value of \( U_0(B) \) compared to the Arrhenius relation.

In summary, it is shown that Arrhenius curve slopes are directly related to, but not equal to, the activation energies in Fe_{1.06}Te_{0.6}Se_{0.4} single crystals. Therefore, use of the modified TAFF model is suggested, where the temperature dependence of \( \rho_{0f} \) and the nonlinear relation of \( U(T, B) \) should be considered. The modified TAFF method results are closer to the experimental data. It was found that there is a correlation
between the effective pinning potential, temperature, and magnetic field, which is governed by the dimensionality of Fe$_{1.06}$Te$_{0.6}$Se$_{0.4}$ crystal.
CHAPTER 9

STRUCTURAL, TRANSPORT, AND MAGNETIC, MAGNETORESISTANCE, AND ANOMALOUS ANGULAR DEPENDENCE OF MAGNETORESISTANCE IN BA (Fe1-xCrx)2As2 SINGLE CRYSTALS

9.1 Introduction

The structural and magnetic properties of pnictide superconductors have been subjects of interest since their discovery. As has been discussed in previous chapters, the $AFe_2As_2$ compounds show a structural phase transition from tetragonal to orthorhombic with nematic symmetry [277] (in which the nematic state refers to a unidirectional self-organized state which breaks the $C_4$ rotational symmetry of the underlying lattice), together with a magnetic transition from paramagnetic to a commensurate antiferromagnetic spin density wave (SDW) with an ordered moment of less than $1 \mu_B$ [70, 278, 279]. Superconductivity occurs when these transitions are suppressed through chemical doping or pressure [39, 280]. Replacement of Fe by Cr [281] or Mn [93], however, does not result in superconductivity, even though antiferromagnetic spin density wave and structural transition are suppressed. The absence of superconductivity in Cr doped Ba-122 single crystal has been reported by Sefat et al. [281]. Neutron diffraction results reveal that SDW and structural transition temperatures are the same for $0 \leq X \leq 0.305$, while the ordered moment of SDW state remains constant for these concentrations and is reduced for $x \geq 0.3$ [75]. This is in contrast to observations of a higher structural transition than the magnetic transition in the case of Co [71], Ni [282], Cu [282], Pd [48], and Rh [48] doping. The magnetic moment remains constant with increasing Cr concentration.
for \( x \leq 0.2 \), although the magnetic transition temperature decreases from 140 K for \( x = 0 \) to 56 K for \( x = 0.2 \). The magnetic moment is reduced with increasing Cr concentration for \( x = 0.335 \) and 0.305 [75]. As the moment decreases, a G-type antiferromagnetic (AF) order with strong magnetism appears, which prevents the occurrence of superconductivity in these compounds.

\[
\text{Ba(Fe}_{1-x}\text{Cr}_x\text{)}_2\text{As}_2 \quad (x = 0.303) \quad \text{single crystal is an unique compound among all the Cr doped BaFe}_2\text{As}_2 \quad \text{compounds, as it reveal a competition between the spin density wave and G-type antiferromagnetic behaviour, as shown in the phase diagram in Fig. 8.1. Transport and magnetic measurements show an interesting twofold symmetry for Ba(Fe}_{2-x}\text{Cr}_x\text{)As}_2 \quad (x = 0.303) \quad \text{compound which depends on the temperature and magnetic field. Neutron diffraction is a powerful tool to study the nature of magnetic structure and SDW in Ba(Fe}_{2-x}\text{Cr}_x\text{)As}_2 \quad (x = 0.303) \quad \text{single crystal. It allows us to determine the magnetic structure of Ba(Fe}_{2-x}\text{Cr}_x\text{)As}_2 \quad (x = 0.303) \quad \text{compound in undiscovered part of the phase diagram [75] (Figure 9-1).}
\]

![Phase diagram of \( \text{Ba(Fe}_{1-x}\text{Cr}_x\text{)}_2\text{As}_2 \) single crystal indicating SDW magnetic and structural phase transitions](image-url)
In the last six chapters, superconductivity and vortex pinning have been the main focus of thesis. It should be noted that understanding the antiferromagnetic order of iron ions itself is also crucial for both fundamental study and practical applications. It is very interesting to design new magnetic devices based on the spin-dependent transport properties of pnictide materials. The possibility of pnictide superconductors, especially BaFeCrAs$_2$, as half-metallic and antiferromagnetic, based on first principles calculations and the tight binding model has been reported by Hu et al. Ba$(\text{Fe}_1-x\text{Cr}_x)_2\text{As}_2$ is a good candidate to study the magnetic properties of this family, as it is the first doped compound with no superconducting phase, and magnetic phases are the only competitors as Cr concentration increases.

In this chapter, the structural, transport, and magnetic properties of the parent compound BaFe$_2$As$_2$ and non-superconducting Ba$(\text{Fe}_1-x\text{Cr}_x)_2\text{As}_2$ compounds are presented and discussed.

9.2 Experiments

High quality Ba$(\text{Fe}_1-x\text{Cr}_x)_2\text{As}_2$ single crystals were grown out from a mixed flux of FeAs and CrAs, as described in reference [281] by Oak Ridge National Laboratory group. Single crystals were mechanically cleaved and shaped into rectangular bars for magnetic and transport measurements. The transport and magnetic properties were measured over a wide range of temperature and magnetic fields up to 13 T, with applied current of 5 mA using a physical properties measurement system (PPMS, Quantum Design). The angular dependence of resistivity were investigated up to 13 T, where $\theta$, angle between the $ab$-plane and applied field, was varied from 0° to 360° and back to zero. The magnetic and crystal
structures transitions as a function of temperature were determined using neutron diffraction. Laue data was obtained on the KOALA neutron Laue diffraction instrument located on a thermal neutron guide at OPAL reactor source of the Australian Nuclear Science and Technology Organization.

9.3 Results and discussion

Room temperature single crystal X-ray diffraction data were collected for $x = 0$ compound. The crystal and structural data parameters are summarized in Table 9-1. The refined lattice parameters are $a = b = 3.9444(6)$ Å, $c = 12.907$ Å for the parent compound BaFe$_2$As$_2$.

Table 9-1 Crystal and structural data parameters for BaFe$_2$As$_2$.

<table>
<thead>
<tr>
<th>Formula</th>
<th>BaFe$_2$As$_2$</th>
<th>BaFe$_2$As$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature</td>
<td>100(2) K</td>
<td>273(2) K</td>
</tr>
<tr>
<td>Wavelength</td>
<td>0.71073 Å</td>
<td>0.71073 Å</td>
</tr>
<tr>
<td>Crystal system</td>
<td>Orthorhombic</td>
<td>Tetragonal</td>
</tr>
<tr>
<td>Space group</td>
<td>$Fm m m$</td>
<td>$I4/mmm$</td>
</tr>
<tr>
<td>Unit cell dimensions</td>
<td>$a = 5.5880(10)$ Å, $\alpha = 90^\circ$.</td>
<td>$a = 3.9444(6)$ Å, $\alpha = 90^\circ$.</td>
</tr>
<tr>
<td></td>
<td>$b = 5.5910(10)$ Å, $\alpha = 90^\circ$.</td>
<td>$b = 3.9444(6)$ Å, $\alpha = 90^\circ$.</td>
</tr>
<tr>
<td></td>
<td>$c = 12.925(3)$ Å, $\alpha = 90^\circ$.</td>
<td>$c = 12.907(3)$ Å, $\alpha = 90^\circ$.</td>
</tr>
<tr>
<td>Z</td>
<td>4</td>
<td>2</td>
</tr>
<tr>
<td>Density (calculated)</td>
<td>6.561 mg/m$^3$</td>
<td>6.597 mg/m$^3$</td>
</tr>
<tr>
<td>Theta range for data collection</td>
<td>3.15 to 28.59°.</td>
<td>3.16 to 35.81°.</td>
</tr>
</tbody>
</table>
Single crystal X-ray diffraction was also conducted at low temperature of 100 K. According to the refinement from the reflections of BaFe$_2$As$_2$, crystal structure changes from tetragonal with $I4/mmm$ space group at $T = 273$ K to orthorhombic crystal structure with $Fmmm$ space group at $T = 100$ K. The obtained lattice parameters are $a = 5.5880(10)$ Å, $b = 5.5910(10)$ Å, and $c = 12.925(3)$ Å at $T = 100$ K. Images from Ba (Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x = 0.303$) tetragonal/orthorhombic crystal were collected at temperatures from 5 K – 100 K. The intensities for each set were indexed and processed using the program LAUE123. Structural refinement was performed using CRYSTALS program suite. Since Laue is only sensitive to ratios between unit cell dimensions, the absolute lattice constants were adopted from the existing high resolution X-ray data, which was also found to be consistent with the tetragonal values measured using the monochromatic neutron diffractometer Wombat instrument at 100 K ($c = 12.98$ Å, $a = 3.96$ Å). Previous work suggested that a subtle orthorhombic transition occurred for this composition below 50 K, where both orthorhombic domains share almost identical lattice constants ($3.96 \pm 0.005$ Å). Clear experimental information regarding the 3-dimensional crystalline effect of this transition is lacking. Figure 8.2 shows the Laue data at 100 K and 4 K.

Most importantly, the systematic broadening of members of (hh0)\textsuperscript{T} family below 50 K is consistent with a subtle orthorhombic phase transition, however, it is clear that other peaks, including those in the perpendicular (h -h k) lines, are affected, suggesting that crystal strain in the perpendicular direction is an important consideration. The moderate increase in intensity at the index nominally assigned to (-101) is consistent with G-type AF transition reported for this material. Although
there is also a structural component amplified by lower-order wavelengths in unpolarised Laue experiment. Taken together, these results imply that transition is consistent with the proposed phase diagram, although 3D strain prohibits a straightforward refinement of the low-temperature structure. Nevertheless, existence of a symmetry-lowering phase transition is a crucial precondition for anisotropic in-plane resistivity. This focused attention on the in-plane resistivity below 50 K.

Figure 9-2 Neutron Laue pattern at a) 100 K and b) 4 K for Ba\((Fe_{1-x}Cr_x)_{2}As_2\) (\(x = 0.303\)). c) Full-width-at-half-maximum of (hh0) peak as a function of temperature, suggesting an orthorhombic transition/increase of strain below 50 K.

Figure 9-3 (a) shows in-plane magnetoresistance for Ba\Fe_{2}As_2 single crystal at several temperatures under a fixed magnetic field of 13 T upon rotating \(H\) within the \(ab\)-plane. A twofold symmetry is observed below the SDW transition temperature. The magnetic field dependence of twofold symmetry is presented in Figure 9-3(b).
The twofold symmetry is clearer for higher magnetic field and lower temperature. No sign of hysteresis has been observed for BaFe$_2$As$_2$ compound.

Figure 9-3 (a) Resistance measured in $ab$-plane as a function of angle $\theta$ under magnetic field of $H = 13$ T at different temperatures. (b) $ab$-plane angular dependence of resistance under different magnetic fields at $T = 2$ K. The angle was measured from $0^\circ$–$360^\circ$ and $360^\circ$–$0^\circ$.

Figure 9-4(a) shows the temperature dependence of resistivity for Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x = 0.073$). The resistivity is roughly temperature independent for $T < 125$ K and then increases gradually with temperature and shows a sharp upturn at $T = 93$ K, which is possibly related to changes in the scattering and a decrease in the number of carriers. Figure 9-4(b) presents angular dependence of resistivity for the same composition at $T = 2$ K and $15$ K under applied magnetic field of $13$ T. A twofold symmetry is observed at all temperatures below $93$ K. Very narrow hysteresis is detected, although just at $T = 2$ K and $H = 13$T, as indicated by the arrows in Figure 9-4(b).
Figure 9-4. (a) Temperature dependence of resistivity for $\text{Ba(Fe}_{1-x}\text{Cr}_x\text{)}_2\text{As}_2$ ($x = 0.073$). (b) Angular dependence of resistivity for the same compound at $T = 2$ and $50$ K under magnetic field of $13$ T.

Figure 9-5(a) shows the temperature dependence of resistivity at zero magnetic field for $\text{Ba(Fe}_{1-x}\text{Cr}_x\text{)}_2\text{As}_2$ ($x = 0.303$). The temperature dependence of resistivity shows metallic behaviour, and resistivity decreases with decreasing temperature between $300$ and $100$ K. The resistivity increases, however, with further decreasing temperature down to $2$ K, manifesting semiconducting behaviour. No sign of superconductivity was observed down to $2$ K for this compound, probably due to the enhanced impurity scattering associated with Cr doping in $\text{Ba(Fe}_{1-x}\text{Cr}_x\text{)}_2\text{As}_2$ single crystal, which result in commensurate magnetic structure in this compound. A similar case was reported for Cu doped BaFe$_2$As$_2$ single crystal, where absence of superconductivity is proposed to be related to the increased impurity scattering and commensurate magnetic structure associated with Cu substitution [283]. It is possible that strong impurity potential results from Cr doping, inducing localization in the system. For example, Berlijin et al. report that weak impurity potential elements such as Co substitution in 122-FeAs can lead to emergence of incoherent carriers and reduce nesting condition on the Fermi surface, which can affect the competition
between magnetism and superconductivity in under-doped compound. Therefore, the long-range antiferromagnetic order is suppressed and superconductivity appears [284]. The opposite effect can occur for the elements with strong impurity potential such as Zn and Cr.

Figure 9-5(b) shows in-plane resistivity at 2 K under different magnetic fields upon rotating the sample within $ab$-plane, as shown in the inset. For all cases, current is aligned in $ab$-plane, and field is applied parallel to $ab$-plane. Angle sweeps were carried out in a field of 13 T following an initial zero-field cooling (ZFC). Data were collected as the angle was increased from $0^\circ$ to $360^\circ$ and then rotated back to zero. The angular dependence of resistivity for $H > 6$ T shows two remarkable features, a sharp twofold symmetry and a field dependent hysteresis with different coercive angles. The maximum coercive angle is almost $90^\circ$. These results clearly demonstrate a resistive hysteresis for field $< 5$ T. A complete resistive hysteresis loop, which was measured from $45^\circ \rightarrow 135^\circ \rightarrow 45^\circ$ is shown in the inset of Figure 9-5(b).

![Figure 9-5](image)

Figure 9-5 Results of transport measurements for Ba (Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x = 0.303$): (a) Temperature dependence of resistivity at zero magnetic field; inset shows the geometry of measurement. In the case of applying magnetic field, both current and magnetic field are in $ab$-plane, and resistivity is measured by rotating the sample in $ab$-plane. (b) Angular dependence of in plane resistivity at $B = 2$ T, 6 T, 10 T, and $T = 2$ K; inset shows a complete resistivity hysteresis loop, which was measured from $45^\circ \rightarrow 135^\circ \rightarrow 45^\circ$. 
We now look at how the resistive hysteresis loop varies with temperature. Figure 9-6 shows the angular dependence of in-plane magnetoresistivity under a fixed magnetic field of 13 T. The sharp twofold symmetry curves are present for $T < 40$ K, however, they are absent for $T > 40$ K. This result indicates that the resistive hysteresis loop only appears below the temperature where a structural transition occurs. The width of the hysteresis loop becomes narrower with increasing temperature to 10 K, and it disappears at $T = 20$ K.

![Figure 9-6 Angular dependence of in-plane magnetoresistivity at different temperatures under a fixed magnetic field of 13 T for Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x = 0.303$).](image)

With the aim of understanding the metamagnetism behaviour of this compound, we have studied its effect on the magnetoresistance. Figure 9-7 shows the magnetoresistance of Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$, $x = 0.303$ single crystal at different temperatures between 2 K and 100 K for two standard configurations of $B//ab//I$ and $B//c//I$. The resistance decreases with increasing magnetic field for both configurations at $T= 2$ K. For $B//ab//I$ at $T=10$ K and 20 K, MR shows a wide peak around $B = 7$ T and 6 T, respectively. There is no evidence of a peak in the MR measurement for the $B//c//I$ configuration. The MR becomes field independent for $T \geq 50$ K for both configurations.
Figure 9-7 Magnetoresistance of Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$, $x = 0.303$ single crystal at different temperatures for $B//ab\parallel I$ (a) and $B//c\parallel I$ (b).

In order to discover if the resistive hysteresis loop is related to any magnetic transitions, the temperature and field dependence of magnetization was investigated at different temperatures and fields. Figure 9-8(a) shows the temperature dependence of magnetization under an applied magnetic field of 0.1 T.

The magnetization shows a sudden increase in magnetic moment below 100 K and a kink at 45 K. Figure 9-8 (b) shows the ferromagnetic state is present in the sample with a negative Weiss temperature, $\Theta$, of -310 K. Figure 9-8(c) illustrates the field dependence of magnetization at different temperatures. There are nonlinearities in magnetization for $T < 100$ K; however, magnetization shows linear behaviour for $T \geq 100$ K. It was suggested that nonlinearities in magnetization are related to metamagnetic transitions for $x \geq 0.18$ in Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ single crystals [281]. Metamagnetism is broadened as the temperature is increased. The inset of Figure 9-8(c) illustrates a small hysteresis loop at $T = 2$ K. The magnetization show a paramagnetic trend combined with small ferromagnetic behaviour.
Figure 9-8 Magnetization results of Ba (Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x=0.303$): (a) Temperature dependence of the zero field cooling (ZFC) and field-cooled (FC) magnetic moment under applied magnetic field of 0.1 T. (b) Inverse magnetic susceptibility as a function of temperature. The ferromagnetic state is present in the sample with a negative Weiss temperature, $\Theta$, of -310 K. (c) Magnetization hysteresis loop results at different temperatures. The inset shows hysteresis ferromagnetic loop at $T=2$ K.

In conclusion, the structural, magnetic, and transport properties of Ba Fe$_{1-x}$Cr$_x$)$_2$As$_2$ single crystal have been investigated. The angular dependence of resistivity manifests a twofold symmetry for the parent compound BaFe$_2$As$_2$ and all studied concentrations of Cr; however, hysteresis in the twofold symmetry is observed with increasing Cr concentration. A very sharp and wide hysteresis twofold symmetry is just observed for Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ ($x=0.303$) compound, which can be interesting for practical applications. The Laue data at 100 K and 4 K reveals the systematic broadening of members in (hh0) family below 50 K is consistent with a
subtle orthorhombic phase transition, however, it is clear that other peaks, including those in the perpendicular (h -h k) lines, are affected, suggesting that crystal strain in the perpendicular direction is an important consideration. Moderate increase in intensity at the index nominally assigned to (-101) is consistent with the G-type AF transition reported for this material.
CONCLUSIONS

This thesis provides a detailed study on the physical properties, pinning mechanism, and thermally activated flux flow of iron based superconductors. In particular, the properties of electron doped BaFe$_{2-x}$Ni$_x$As$_2$, BaFe$_{2-x}$Co$_x$As$_2$, and Fe$_{1+y}$Se$_{1-x}$Te$_x$ single crystals were intensively investigated. In addition, structural, transport and magnetic properties of parent compound BaFe$_2$As$_2$ and non superconducting Ba (Fe$_{1-x}$Cr$_x$)$_2$As$_2$ compounds are investigated. A brief summary of the key results presented in this work is given below.

1- Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ single crystals:

Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ single crystals were characterized by magnetotransport and magnetic measurements up to 13 T over a wide range of temperatures below and above the superconducting critical temperature, $T_c$. High pinning potential and nearly isotropic superconducting properties were observed for this compound. The second magnetization peak and flux jumping was detected in magnetic hysteresis loops. Analysis using Dew-Hughes model has suggested that point pins alone cannot explain the observed field variation of pinning force density. Also, based on the collective flux pinning model, field dependence of the magnetization shows that flux pinning in this compound is dominated by spatial variation in the charge carrier mean free path.

In order to enhance the critical current density, C$^{4+}$ ion irradiation was employed. It is found that C$^{4+}$ ion irradiation can enhance the critical current density by a factor of 1.5, while the transition temperature remains unchanged.
2- **Under-doped Ba(Fe_{1-x}Co_x)_{2}As_{2}** single crystals:

The angular dependence of pinning potential and upper critical field was investigated using magnetotransport measurements. Pinning potential values decreased, while the upper critical field increased by rotating the sample from $B//c$ to $B//ab$. The anisotropy parameter was determined using the anisotropic Ginsburg Landau theory. The anisotropy value decreased from 2.1 to 1.8 as the temperature decreased from 17 to 12.5 K.

3- **Fe_{1+y}Se_{1-x}Te_{x}** single crystals

The vortex pinning mechanism, pinning potential, and anisotropic properties were studied systematically by magnetotransport measurements at different temperatures on Fe$_{1+y}$Se$_{1-x}$Te$_{x}$ single crystals. The anisotropy was determined by scaling the angular dependence of resistivity, based on the anisotropic Ginsburg Landau theory. The anisotropy value of 2.5 at $T = 14$ K was obtained. According to Dew-Hughes model, the spatial variation in charge carrier mean free path is responsible for the vortex pinning in these compounds. Also, the thermally activated flux flow behaviour of this compound was investigated using conventional Arrhenius relation and modified thermally activated flux flow model. The results show that Arrhenius curve slopes are directly related to, but not equal to, the activation energy of Fe$_{1+y}$Se$_{1-x}$Te$_{x}$ single crystals. Therefore, use of a modified thermally activated flux flow is suggested where the temperature dependence of prefactor $\rho_{0f} = 2\rho_{c}U/T$ and the nonlinear relation of thermally activated energy are considered. The high value of pinning potential using the modified thermally activated energy method is in good
agreement with the high values of critical current density of these compounds obtained from experimental data.

4- BaFe$_2$As$_2$ and Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ single crystals

In order to understand the magnetic and magnetoresistance behaviour of non-superconducting Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ a detailed investigation was carried out using magnetic, magnetoresistance and neutron diffraction measurement under different magnetic field and temperatures. Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ is the first doped compound with no superconductivity phase and magnetic phases is the only competitor as Cr concentration increases. Transport and magnetic measurements show an interesting twofold symmetry for the parent compound BaFe$_2$As$_2$ and all the studied concentration of Cr; however, hysteresis in the twofold symmetry was observed with increasing Cr concentration. Especially, a very sharp and wide hysteresis twofold symmetry observed for Ba(Fe$_{1-x}$Cr$_x$)$_2$As$_2$ (x=0.303) compound which can be interesting for practical application. The Laue data at 100 K and 4 K reveals the systematic broadening of families in (hh0)$^T$ family beneath 50 K is consistent with a subtle orthorhombic phase transition, however, it is clear that other peaks including those in the perpendicular (h -h k) lines are affected, suggesting that crystal strain in the perpendicular direction is an important consideration. The moderate increase in intensity at the index nominally assigned (-101) is consistent with G-type AF transition reported for this material.
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