

On the Magnetic and Superconducting properties of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ Single crystals

BY

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This thesis, written by **Rami Mohammad Dakhalalah Hamad** under the direction his thesis advisor and approved by his thesis committee, has been presented and accepted by the Dean of Graduate Studies, in partial fulfillment of the requirements for the degree of **DOCTOR OF PHILOSOPHY IN PHYSICS.**



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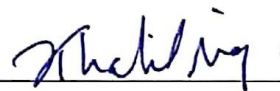


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2018

To

My Parents

My Wife & Children

My Brothers & Sisters

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All praise and thanks to Allah Almighty, most gracious, most merciful. Grace and peace on the master of prophets, Muhammad.

First and foremost, I must express my sincere gratitude to Professor Khalil A. Ziq, my advisor and Dr. Shankar Kunwar, who supervised part of this work. Their supervision, continuous support, and dedication made the graduate years a little more bearable. I will always remember their extended helping hand, encouragement and friendship.

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LIST OF ABBREVIATIONS

BSC	:	Bardeen, Cooper, and Schrieffer theory
TAFF	:	Thermal Activated Flux Flow regime
FC	:	Flux Creep regime
FF	:	Flux Flow regime
HTS	:	High Temperature Superconductor
STM/STP	:	Scanning Tunneling Microscopy/ Spectroscopy
XRD	:	X-Ray Diffraction
VSM	:	Vibrating Sample Magnetometer
ER	:	Empirical Relation
GL	:	Ginzburg-Landau model
WHH	:	Werthamer-Helfand-Hohenberg model
SPM	:	Second Peak Magnetization
ICDD	:	International Centre for Diffraction Data
FWHM	:	Full Width at Half Maximum
TAE	:	Thermal Activation Energy

ABSTRACT

Full Name : Rami Mohammad Dakhalalah Hamad
Thesis Title : On the Magnetic and Superconducting properties of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$
Single crystals
Major Field : Physics
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This thesis investigates factors affecting growth and physical properties of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ high quality single crystals. In particular; we investigated the effects of Fe-contents on some of their fundamentals properties in order to better understand the microscopic nature of superconductivity in these Fe-based superconductors. Understanding some basic properties of the vortex state, pairing symmetry and gap structure facilitates more understanding of microscopic nature of superconductivity.

We found that the activation energy decreases with increasing iron content reaching maximum value for the optimum iron content when ($X=1$). Increasing iron content cause disorder in the conducting layers and this may cause the decrease in activation energy. The low values of activation energy (U_o) reflects the low vortex-pinning in the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductors.

In particular; we investigated the activation energy in the various flux flow regimes, thermodynamical critical field, pinning forces critical currents, energy gaps and scaling behaviors.

Magnetic measurements revealed that higher iron content lead to a gradual increasing in critical current density (J_c) and pinning forces (P_f). The $P_f - H$ curves revealed two different trends. For temperatures, ($T < 10$ K), the pinning forces increased with increasing iron contents, whereas for temperatures ($T > 10$ K), the pinning forces decreased with increasing iron contents. We used different scaling models to investigate different pinning mechanism on $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ system.

We used the thermodynamic critical field to evaluate the energy gap and the coupling strength and compared the STM results we obtained on the optimal sample. The BS deviation function of the reduced thermodynamic critical magnetic field indicates weak coupling mechanism. Increasing iron content further weakens the coupling, and slightly reduces the energy gap.

ملخص الرسالة

الاسم الكامل: رامي محمد دخل الله حمد

عنوان الرسالة: دراسة الخصائص المغناطيسية وفائقة التوصيل في بلورات ($\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$) أحادية التركيب

التخصص: الفيزياء

تاريخ الدرجة العلمية: نوفمبر 2018

تعنى هذه الدراسة بدراسة تأثير محتوى الحديد في عينات ($\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$) فائقة التوصيل الكهربائي بنسب مختلفة من الحديد ($0.9 < X < 1.1$). لقد تم تمييز العينات باستخدام حيود الأشعة السينية (XRD). بالنظر إلى نتائج تحليل بيانات حيود الأشعة السينية، وجدنا أن التركيب لها رباعي الأوجه وأن المحور (c) له قيمة قصوى عند محتوى حديد ($X=1.02$)، بينما المحور (a) له قيمة قصوى عند محتوى حديد ($X=1.00$). تم إجراء القياسات الكهربائية على جميع العينات باستخدام جهاز قياس المقاومة الكهربائية عند درجات حرارة من (10 – 20) كلفن ومجالات مغناطيسية تصل إلى 1.5 تسلا). أمكن تحليل منحنيات التيار مع فرق الجهد، واستنتبنا منها قيم طاقة التنشيط الحراري الفعال التشخيصية في حالة المادة المختلطة. كما قمنا بحسابها بواسطة منحنيات ارهينيوس ووجدنا انها متوافقة مع بعضها البعض وتساوي تقريبا (100K). وان قيمها أكبر ما يمكن عند ($X=1.00$) وأنها تقل بازدياد محتوى الحديد. تم إجراء قياسات المغنطة على جميع العينات باستخدام جهاز قياس التمغنط للعينات المهتزة (VSM) عند درجات حرارة من (4 – 13) كلفن ومجالات مغناطيسية تصل إلى 9 تسلا) لبعض العينات وفي البعض الآخر تصل إلى 14 تسلا). أمكن تحليل منحنيات التمغنط، واستنتبنا منها الخواص غير المنعكسة مثل المجال المغناطيسي غير المنعكس، وشدة التيار الحرج، والقوى المثبتة عند درجات حرارة مختلفة. ومن الخواص المنعكسة تم حساب المجالات المغناطيسية اللاعكوسة والمعدلة والحرارة وهذه الأخيرة والتي لها علاقة بالخواص الحرارية للمادة. لقد وجدنا

أن سلوك التيارات الحرجة والقوى المثبتة، قد تأثرت بشكل كبير عند تغيير قيم محتوى الحديد بنسب مختلفة. ثم قمنا بمعايرتها باستخدام قيم المجالات المغناطيسية الالاعكوسة والمعدلة. يجدر بالذكر أن التيارات الحرجة المعاييرة، والقوى المثبتة المعاييرة وقد أظهرت سلوكاً موحداً مع المجالات المغناطيسية المعاييرة. لقد قمنا بدراسة العديد من المتغيرات بناءً على نتائج الخواص المنعكسة وغير المنعكسة، مثل: القفزات في الحرارة النوعية الإلكترونية وفجوة الطاقة ، ومدى تأثر كل ذلك عند تغيير قيم محتوى الحديد. حيث وجدنا أن فجوة الطاقة تقل بازدياد محتوى الحديد وقد فسرنا ذلك أنه قد يسبب ازدياد محتوى الحديد تشوها في التركيب البلوري للعينات والذي من الممكن أنه قد يسبب بدوره أجهاد داخلي على العينات والذي بتغيره تتغير الخصائص فائقة التوصيل للمادة بينما تزداد القفزات في الحرارة النوعية عند ازدياد محتوى الحديد والذي يشير لمدى حساسيتها لتغير محتوى الحديد حيث تتأثر بتغير التوزيع الالكتروني للمادة.

CHAPTER 1

INTRODUCTION AND LITERATURE REVIEW

1.1 Historical background of superconductivity

In 1911, Kamerlingh Omens and his group were investigating the variations of residual resistance of metals with impurity concentration at low temperature. When they cooled high purity mercury to (4K), they noticed a sudden drop in its electrical resistance to near zero Ohm [1]. Mercury, being a metal, should have some residual resistance at ($T=0K$) that increase with the increasing impurity concentration. The Sudden dropped of resistance to near zero indicated a transition beyond normal conducting process. This marked the discovery of new phenomenon called superconductivity, that opened up a new research area in condensed matter physics.

It took Walter Meissner and Robert Oschsenfeld another 22 years to discover a second characterizing magnetic property of superconducting materials. In 1933, Meissner and Oschsenfeld were trying to distinguish between super and perfect conductor. They cooled a superconducting material in the presence of a magnetic field blow its superconducting transition temperature (T_c). Meissner and Oschsenfeld found that below (T_c), the field is expelled out from inside of superconductor ($B_{ins}= 0$) unlike what is expected in perfect conductor. The phenomenon is later known as "Meissner effect" [2].

After discovering Meissner effect, in 1935 London brothers put two equations to explain the electromagnetic properties of superconductors and to the expulsion of the magnetic

field from the interior of superconductor. The London equations are based on a modification of the Maxwell equations. The equations predict how far the magnetic field can penetrate the superconductor [9]. The London brothers proposed a penetration depth (λ), that predicts the distance that the magnetic field penetrates the superconductor. Fifteen years later; in 1950 Pippard proposed another characteristic length that measures the spatial variation of the density of superconducting electrons around the penetrating magnetic field. This is known as the coherence length (ξ). Ginzburg and Landau introduced their theory in 1950 in which they used the ratio of both lengths, (ξ) and (λ) to distinguish between different types of superconductors. The ratio of penetration depth to coherence length gives new parameter (κ) known as Ginzburg-Landau parameter [10], as:

$$\kappa = \frac{\lambda}{\xi} \quad (1.1)$$

Abrikosov [11] showed that Ginzburg-Landau equation has solution in two different regions: Type I superconductors with (κ) value in range ($0 < \kappa < 1/\sqrt{2}$), and with positive surface energy in the interface between normal and superconducting states. Type II superconductors, with ($\kappa > 1/\sqrt{2}$) and negative surface energy in the interface between normal and superconducting states. Abrikosov also predicted the presence of a mixed state in type II superconductor, where the magnetic field penetrates in certain regions known as vortices.

John Bardeen, Leon Cooper, and John Schrieffer proposed the theoretical explanation of superconductivity in 1957 [3]. The main feature of the (BCS) theory is that two electrons in the superconducting state can form a bounded pair called a Cooper pair [49]. The two electrons, usually repel each other because of their alike charges, happens to attract each other due to the mediation of lattice vibration. The lattice structure is momentarily

deformed by a passing electron and the deformed region attract another electron of opposite momentum. This is commonly known as phonon mediated exchange interaction between the two electrons. The (BCS) theory explained superconductivity in elements and simple alloys at temperatures near to absolute zero. Moreover; the (BCS) theory predicts a maximum superconducting transition temperature close to (23K), while, at higher transition temperatures the theory fails to explain superconductivity.

Progress in enhancing the superconducting critical temperatures was greatly motivated by the idea of producing room temperature superconductors. In 1986 a new ceramic has been discovered with transition temperature in the mid (30's K). The ceramic is made from barium, lanthanum, copper and oxygen was discovered by Georg Bednorz and Alex Muller with its critical temperature about (35K) [4]. Soon after that, Paul Chu prepared a new ceramic material by replacing lanthanum by yttrium, which became superconducting at (93K) [5]. These discoveries marked a new era of high temperature superconductors.

1.2 Properties of Superconductors

There are two main properties of superconductors. The first one is the vanishing of the electrical resistance of a superconductor at and below the critical transition temperature and the second one is the Meissner effect in which the magnetic field (lower than a critical field H_{c1}) is expelled out of the superconductor. The Meissner effect can be explained by a super current circulating the surface of the superconductor that creates a magnetic field which completely cancels the applied magnetic field inside the superconductor [49]. The Meissner effect differentiates a superconductor from a perfect conductor.

1.2.1 Critical Parameters

Superconductors are characterized by several critical parameters. The critical temperature (T_c), the critical fields (H_c , H_{c1} and H_{c2}) and the critical current density (J_c). These parameters define the boundaries of the phase diagram within which a superconductor can exist. Figure (1.1) illustrates the three critical parameters.

The transition (critical) temperature is the maximum temperature below which the electrical resistance of material disappears, and the material transforms to the superconducting state [6, 7]. The upper critical magnetic field is defined as the minimum applied magnetic field at which the superconductivity is vanished. Normal conducting state of the material is restored when magnetic field is higher than the critical magnetic field [49]. At low temperature, the critical magnetic fields closely follow a parabolic temperature dependence known as Tuyn's law [8]:

$$H_c(T) = H_c(0) \left[1 - \left(\frac{T}{T_c} \right)^2 \right] \quad (1.2)$$

Where ($H_c(0)$) is the critical field at absolute zero and (T_c) is the critical temperature. Finally, the critical current is the maximum current that superconducting materials can carry, above which the superconductivity of the materials is destroyed even though it may be below its (T_c).

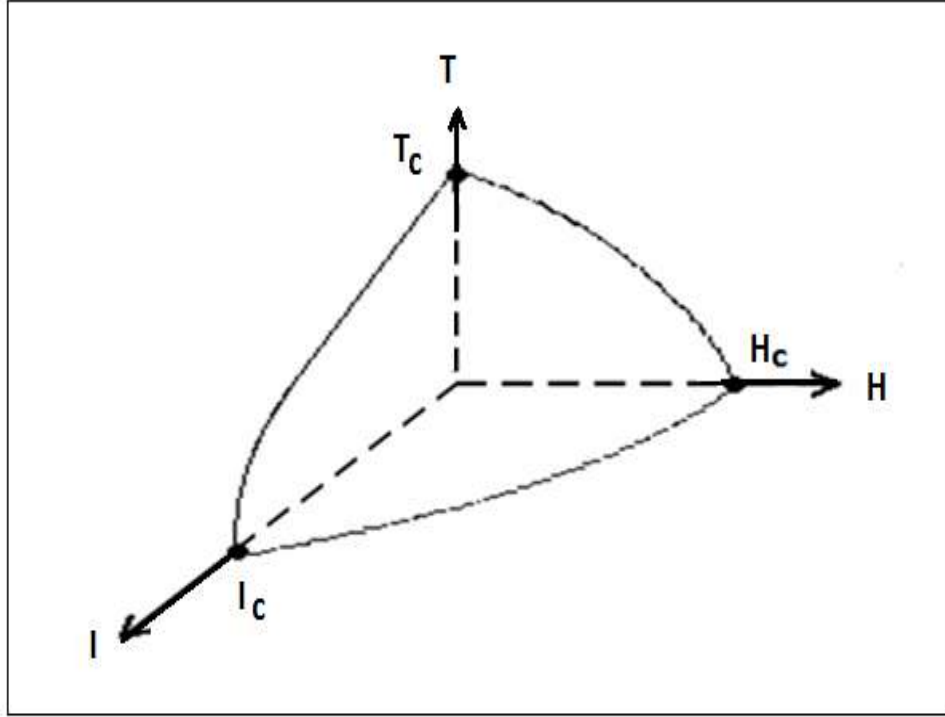


Figure (1.1): Critical Surface Phase Diagram with critical parameters [50].

For practical applications, it is desired to have materials with high critical parameters without destroying the superconducting state [12]. Experimentally, critical current density, (J_c) can be measured using transport measurements, a one microvolt ($1\mu\text{V}$) is often used to define the corresponding current as a critical current. Critical current density can be measured magnetic measurements, the hysteresis loops and Bean's model. In this thesis, we will be measuring the hysteresis loops for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples and using Bean's model to evaluate critical current density upon varying temperature, field and iron content.

1.2.2 Magnetic Properties

Superconducting materials are categorized into two main types according to their response to the applied magnetic field. Type I (Soft): are superconducting materials in which the superconductivity state is destroyed when the applied magnetic field exceeds the critical

value (H_c). While, the second is called type II (Hard) superconductors characterized by two critical fields (H_{c1}) and (H_{c2}) [12]. When the applied magnetic field is lower than the (H_{c1}); the material is in pure superconducting (Meissner) state. At higher fields, larger than (H_{c1}) but lower than the (H_{c2}) the material is partially penetrated by the magnetic field forming vortices distributed inside within the superconducting state. The state is known as a mixed or “vortex” state [6, 7]. When the applied magnetic field is higher than (H_{c2}), the superconducting state is destroyed, and material becomes normal conductor. The magnetization behavior of type I superconductor is presented in figure (1.2a) and for type II in figure (1.2b). The magnetic phase diagram of type I and type II superconductors is presented in figure (1.3) a and b respectively.

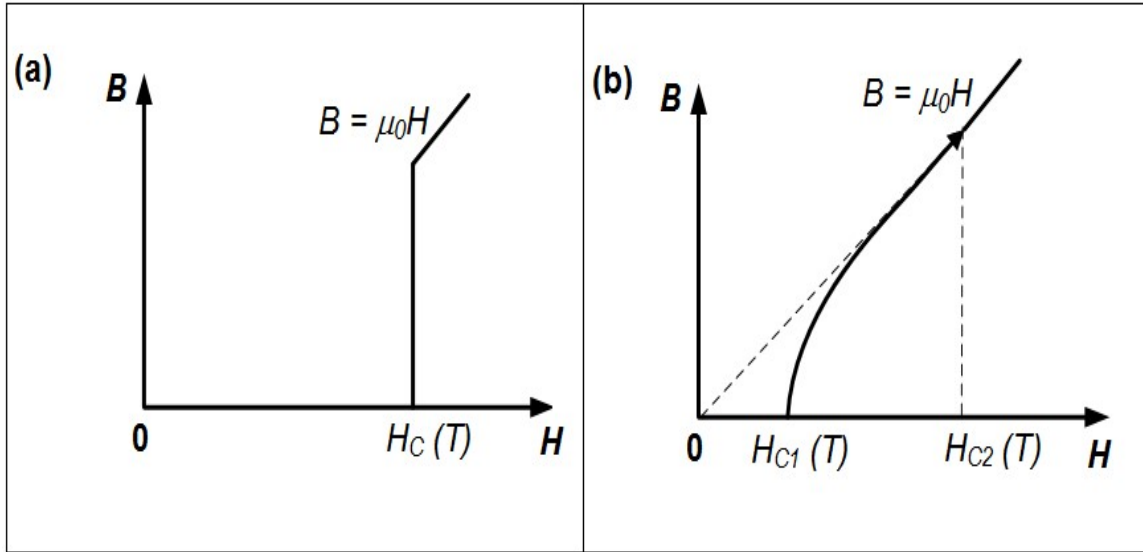


Figure (1.2): B versus H for types of superconductors in (a) type-1, below H_c , there is no magnetic field penetration while in (b) type-2, below H_{c1} , no magnetic field penetrates between H_{c1} and H_{c2} , magnetic field rises smoothly to $B = \mu_0 H$ [8].

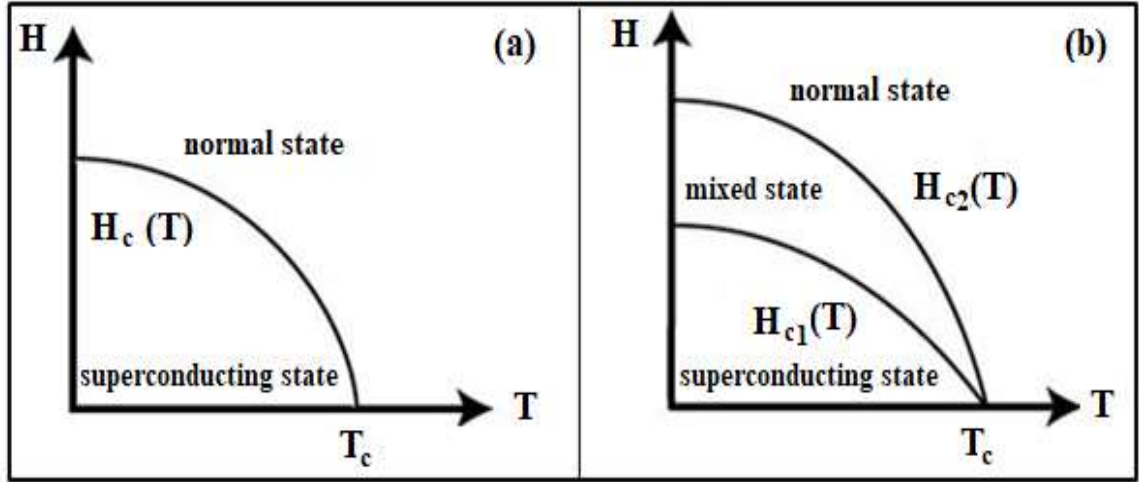


Figure (1.3): (a) Type I (Soft) superconductor, (b) Type II (Hard) superconductor [21].

1.3 The Discovery of Fe-based Superconductors

In this thesis, we investigate several superconducting properties of one member of the newly discovered Fe-based superconductors, namely $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$. The newly discovered materials combine two “often conflicting” important material properties; superconductivity and magnetism. In conventional superconducting materials, the magnetic nature of iron would prevent the electrons from forming Cooper pairs that constitute the building block of the superconducting state [13].

Hosono and his group has discovered the first superconducting material in this group in 2006 [14]. The group discovered superconductivity in $\text{LaFeAsO}_{1-x}\text{F}_x$ with critical temperature about (26K) [14, 15]. This new type of superconductors consists of conducting layers of iron and pnictides - group fifteen in the periodic table, like arsenic (As) and phosphorus (P). One of the drawbacks in these materials is the use of poisonous arsenic. Soon after that, superconductivity was observed in Arsenic free FeSe compound at about (8K) [16]. Despite the low transition temperature in this compound, its structure is simple and provides a base for understanding many features of high temperature

superconductivity. Its transition temperature can be improved by chemical substitution at the Se-site by Te [19]. Applying pressure has also been found to enhance the superconducting transition temperature [18]. Reducing the thickness of FeSe (few layers) also enhances (T_c) to near (100K) [17].

A general feature in all groups of Fe-based superconductors is the Fe–As layers which are the basic building unit of structure. A single layer consists of a square lattice of iron atoms connected with the As-atoms that are alternately displaced above and below the Fe-layer as shown in figure (1.4) [16]. A tetrahedral structure is formed from interpenetrating lattice of As and Fe atoms. Fe-based superconductors have been extended to variety of materials substitutions. Four main groups of iron-based superconductors, 1111, 122, 111, and 11 types were discovered. These groups may or may not contain oxygen. These groups are classified according to the existence or absence of a sandwiched layer in between the neighboring Fe–As layers, and the orientation of the neighboring Fe–As layers with respect to each other, these families are:

1- The 11-family compounds (FeSe) have no sandwiched layer and all layers are oriented in the same direction.

2- The 111-family compounds (LiFeAs) have a monatomic sandwiched layer and all layers are oriented in the same direction.

3- The 122-family compounds (BaFe_2As_2) have a monatomic sandwiched layer with alternating orientation layers.

4- The 1111-family compounds ($\text{RFeAsO}_{1-x}\text{F}_x$) have diatomic sandwiched layer and all layers are oriented in the same direction and R represents rare earth element.

One of the main issues in these new materials is the hope that understanding various properties of iron-based superconductors may provide a base for generalized theory of superconductivity, beyond the BCS theory. A new theory is essential to understand the mechanism of superconductivity in high temperature superconductors. Moreover, they may offer a new ground for practical applications of superconductivity [20].

In the present work, we will study the effect of excess of iron composition on the lattice structure and superconducting properties, such as upper critical field (H_{c2}), irreversibility field (H_{irr}), critical current density (J_c), pinning force (P_f), thermodynamic critical field (H_c), thermal activation energy (U) and energy gap (E_g) of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ compounds. Which can create an opportunity to open the door for using this simple structure type of compounds in wide applications.

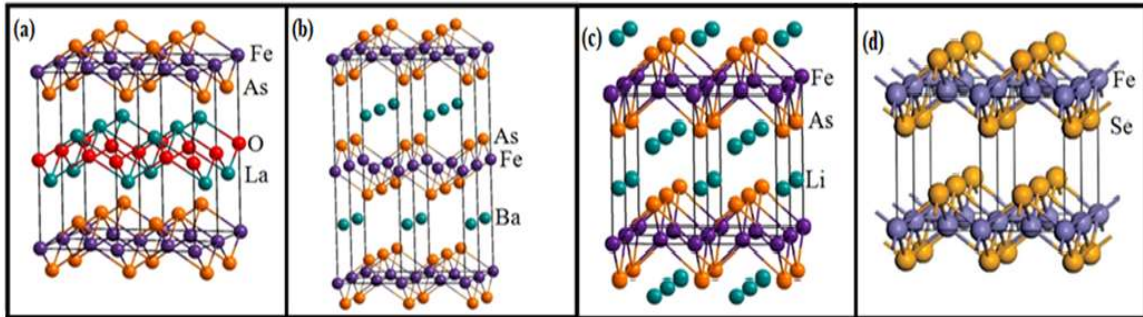


Figure (1.4): Groups of Fe-based superconductors, (a) 1111, (b) 122, (c) 111, (d) 11 type [16].

1.4 The Vortex State in HTS

The vortex state occurs in type II superconductor for applied fields between (H_{c1} and H_{c2}). The regions where the magnetic field penetrates the superconducting material are known as vortices. These vortices are approximately cylindrical shape with their axes parallel to the applied magnetic field. The radius of each vortex is equal to coherence length (ξ) its magnetic flux equals to the quantum flux (ϕ_0), where:

$$\phi_o = \frac{hc}{2e} = 2.07 \times 10^{-11} \text{G. m}^2 \quad (1.3)$$

Where (h) is the Planck's constant, (c) is the speed of light and (e) is the charge of the electron. Currents swirling around the vortices and generate magnetic fields parallel to the applied field. These small magnetic moments repel each other and move to arrange themselves in an ordered array known as a fluxon "hexagonal" lattice. The penetrated flux density has a maximum at the center of vortex and falls to a small value at a distance larger than the penetration depth (λ) away from the center. While the density of the superconducting electrons n_s falls to zero at the center of each vortex line as shown in figure (1.5) [21].

Mixed state bounded by both (H_{c1} and H_{c2}), Abrikosov presented formulas to calculate (H_{c1} and H_{c2}) by depending on values of (ϕ_o , ξ and λ) as:

$$H_{c1} \approx \frac{\phi_o}{\pi\lambda^2} \quad (1.4)$$

$$H_{c2} \approx \frac{\phi_o}{\pi\lambda\xi^2} \quad (1.5)$$

For practical applications, it is highly important to keep these vortices pinned in their sites in response to applied magnetic fields. Vortex motion can be effectively prevented by defects, impurities and grain boundaries [6, 7]. When the applied electrical current or temperature are increased, the vortices starts moving causing flux flow resistance that dissipate heat inside the superconductor. The presence of pinning centers prevents the motions of the vortices if the pinning forces exceeds the applied Lorentz force. The phase diagram reveals a rich region in the mixed vortex state in type II superconductors, as shown in figure (1.6). Vortex motion at temperatures close to critical transition temperature can

be explained using the Kim-Anderson model [53] when the pinning potential was taken into account. In addition, the specific features of vortex dynamics, such as the irreversibility line and a vortex glass transition, can be explained [54-57]. In single crystals of high (T_c) superconductors; the voltage generated due to motion of intragrain vortices [58]. Fisher *et al.* described theoretically the scaling behavior of the current–voltage relation [60, 62 and 53]. Koch *et al.* [61] showed the first experimental evidence for the continuous vortex glass transition at temperatures below (T_c) using the scaling argument of the nonlinear current–voltage curves in YBCO thin films. Gammel *et al.* [64] have found similar results in YBCO single crystal.

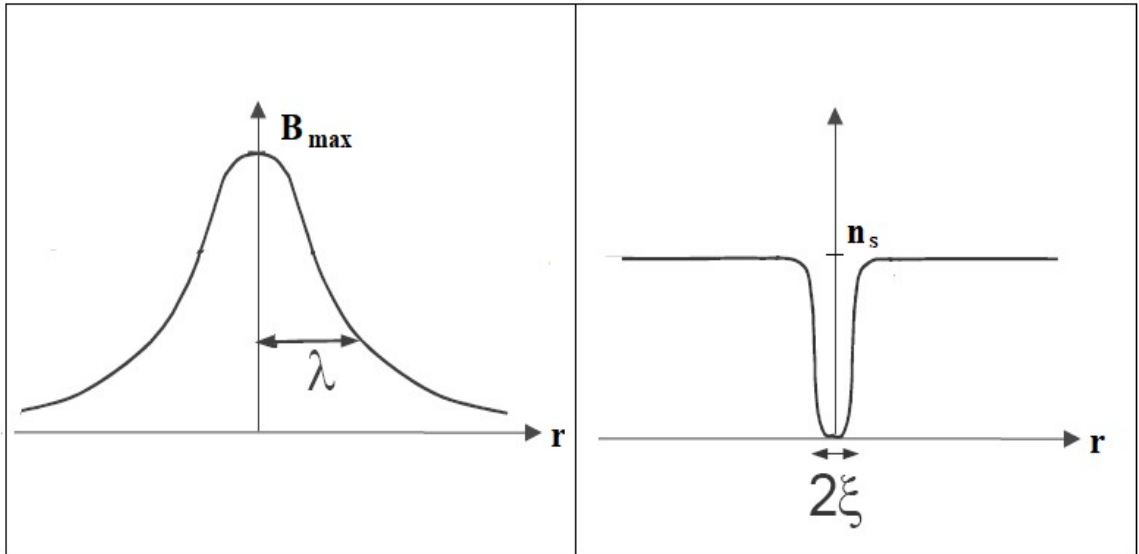


Figure (1.5): Variation of B and n_s with respect to distance from the center of vortex [21].

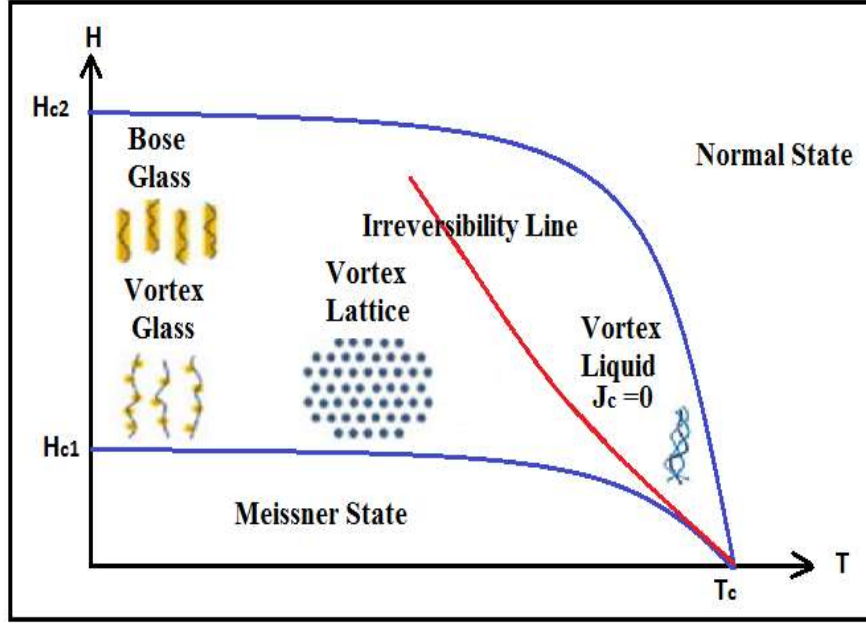


Figure (1.6): Representation of the magnetic phase diagram of type II superconductor [22].

There are different regimes according to status of vortices as thermal activated flux flow regime (TAFF), flux creep regime (FC) and flux flow regime (FF). The critical current density is used to distinguish various regimes. If the current density is lower than the critical current density, then this regime is called thermal activated flux flow regime, in which thermal excitations causes dissipation. If the current density is larger than the critical current density, then this regime is called flux flow regime (in such situation) the driving force on vortices is larger than the pinning force. Finally, if the current density approximately equals the critical current density, then this regime is called flux creep regime, which involves various vortices motion and several potential barriers [22].

1.5 The $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ Superconductor

FeSe is one of the simplest Fe-based superconductors. The parent compound superconducting FeSe has tetragonal structure consisting of two layers of FeSe and has no interlayer in between as shown in figure (1.7).

It revealed very interesting properties below and above its critical transition temperature (8K). Its critical transition temperature which is sensitive to the chemical doping [23, 24] and applied pressure [25, 26]. Ultrathin layers of FeSe revealed a noticeable rise in (T_c), about (100K) [17]. Partial substitution with Te atoms at Se sites enhances the critical transition temperature close to (15 K) in $\text{FeSe}_{1-x}\text{Te}_x$ when (X) value in range ($0.4 \leq X \leq 0.8$) [26]. Margadonna *et al.* [27] found that a pressure of only (2.6 GPa) increases the transition temperature to (27 K). The maximum improvement is reached when a pressure of (7 GPa) the critical transition temperature reached to (36.7K).

However, a further increase in pressure leads to a decrease in the critical transition temperature. At (14 GPa), the transition temperature is only dropping to (6K), the possible reason for the changes in its critical transition temperature may be related to changes in the band structure under pressure [28].

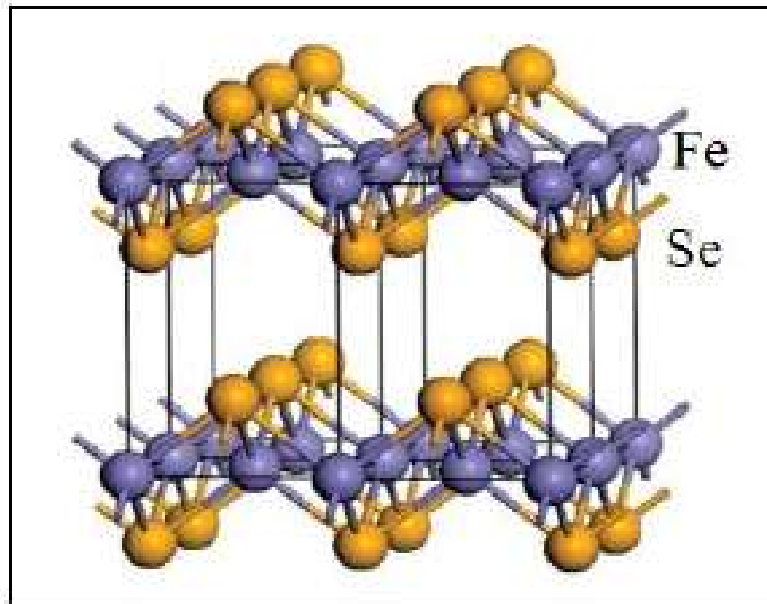


Figure (1.7): Tetragonal structure of pure FeSe superconductor [16].

1.6 Effect of Fe in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ Compounds

The structure of $\text{Fe}_y\text{Se}_{1-x}\text{Te}_x$ is relatively simple; it consists of iron-chalcogenide layers, which is like to Cu-layers found in cuprate high temperature superconductors. The optimum composition for highest T_c under normal condition is $\text{FeSe}_{0.5}\text{Te}_{0.5}$, with ($T_c \sim 15\text{K}$). Several groups have studied the effects of iron content in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ of single crystals on several transport and magnetic properties. Iron deficiency in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ thin film compounds led to formation of iron vacancies and causes variation in the charge carrier population. Zhuang *et al.* found that samples with iron deficiency leads to a higher (T_c), while excess of iron leads to deterioration of several superconducting properties [35]. The excess iron ($X > 1$) ions are occupying interstitial sites in the chalcogenide layer [36]. The interstitial sites of the (Se, Te) layers permit partial occupation of iron, which cause non-stoichiometric composition in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ [37]. Liu *et al.*, investigated the role of excess iron at the interstitial sites of (Se, Te) layers in the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ compounds and observed that the excess iron suppressed superconductivity and led to low charge carrier population [36]. Sudesh *et al.* studied the effect of variation of iron content on superconducting properties such as critical transition temperature (T_c), upper critical field at absolute zero ($H_{c2}(0)$) and irreversible magnetic field (H_{irr}) of polycrystalline $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples [29], and they found that the highest values of these properties were observed at ($X=1.00$).

Several reports showed that excess of iron (electron doping) cause internal stress, which may affect the critical transition temperature and other superconducting properties (Fig. 1.8). Furthermore, iron deficiency (hole doping) may cause internal strain, which has been found to improve the superconducting transition temperature in thin films. There are several contradicting reports on the effect of iron contents on the critical transition

temperature in both thin films and bulk samples [29-34]. Figure (1.8) shows the phase diagram (T_c , T_{SG} and T_{SDW}) for $\text{Fe}_y\text{Se}_{1-x}\text{Te}_x$ upon changing Fe, Se and Te concentrations.

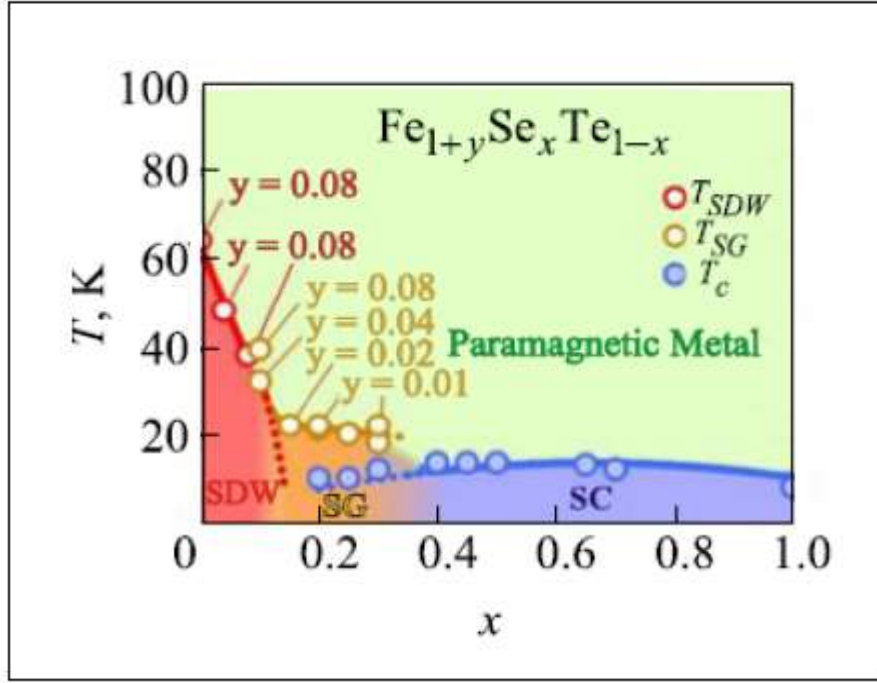


Figure (1.8): Phase diagram of $\text{Fe}_{1+y}\text{Se}_x\text{Te}_{1-x}$ [48].

1.7 Thermally Activated Flux Flow (TAFF)

Thermally activated flux flow behavior (TAFF) is the outcome of the thermal fluctuation in high temperature superconductor (HTS) when the vortices bundles jump the potential barrier between neighboring pinning centers. The resistivity in the (TAFF) can be expressed as [1]:

$$\rho_{TAFF}(T, H) = \rho_o e^{\left(\frac{-U}{k_B T}\right)} \quad (1.6)$$

$$\ln \rho_{TAFF}(T, H) = \ln \rho_o - \frac{U}{k_B T} \quad (1.7)$$

(U) can be obtained from the slope of linear part of Arrhenius plots.

Several models have been proposed to describe the behavior of the activation energy with temperature and field. One of the simple models is the condensation model, which expresses the activation energy as [38]:

$$U(T, H) = Af(H)^p \epsilon(t)^q \quad (1.8)$$

Where ($t = T/T_c$) is the reduced temperature, and A is constant. When the temperature is close to (T_c), the activation energy can be rewrite as:

$$U(T, H) = U_o H^p (1 - t)^q \quad (1.9)$$

In general (p and q) are field and temperature dependent [39-41]. There have been several reports that investigated the field and temperature dependent of the activation energy in high temperature superconductors. Plastrapal *et al.* proposed that power law dependence on field to explain characteristic thermal activation energy; ($U_o \sim H^\alpha$) [38] while Kucera *et al.* proposed that $U_o \sim H^{-1/2} (1-t)$. In general, the activation energy can be expressed as the following relation: $U(H, T) = U_o H^\alpha (1-t)^\beta$. For zero applied magnetic field $U(T) = U_o (1-t)^\beta$. Moreover, (α and β) are field and temperature dependent according to reference [39]. For a wide range of high temperature superconductors ($\alpha \sim 0.5$ and $\beta \sim 1$) [40 and 41].

1.8 Flux Pinning Mechanism in High Temperature Superconductor

Pinning force is a short-range force which created by defects, vacancies, boundaries and dislocations of materials. The imperfections of material in the mixed state of type II superconductors creates pinning centers [42] that in their role prevent the vortices from motion which mean they become trapped. This phenomenon can only occur in type II

superconductors. The applied magnetic field can penetrate the material, only when its value between (H_{c1} and H_{c2}) (Mixed state). As the field keeps increasing, the normal cores become closely packed and, in this case, the vortices have been affected by force due to the flow of current. The material loses its superconductivity when the force experienced by the vortices due to the flow of current which is larger than the critical current. Therefore, the material becomes non-superconducting [43]. So, the importance of the pinning force on the magnetic field as it reveals the field at which the flux leaves the pinning trap and the material becomes non-superconductor.

To investigate the flux pinning in high temperature superconductors (HTS), an improved method based on both the Dew-Hughes and Kramer models [44 and 45] has been developed. For (HTS), a pinning force density (P_f) is equal to the critical value of the Lorentz force:

$$P_f = J_c \times H \quad (1.10)$$

Where (P_f) is force per unit volume, (J_c) is the critical current density and (H) is an applied magnetic field. Knowing of the field and temperature dependence of pinning force is required to improve the critical current density. The superconductors are similar in their (P_f - H) dependence at different temperature. (P_f) equals zero at the irreversibility field because ($J_c \sim 0$) [46].

To understand the pinning mechanism in iron-based superconductors, it is essential to study the effect of temperature and applied field on the pinning force of vortices. According to Dew-Hughes model, the normalized pinning force at different temperatures can be written as a scaling law of:

$$f \approx h^p \times (1 - h)^q \quad (1.11)$$

Where, (p and q) are two parameters are describing the pinning mechanism which will explain by details in chapter six, and (h) is the reduced field, ($h = H/H_{irr}$), where (H_{irr}) is the magnetic field at which the pinning force vanishes, and ($f = P_f/P_{f\ max}$). Also, the (h_{max}) is the (h) value when pinning force maximum its value given by:

$$h_{max} = \frac{p}{p+q} \quad (1.12)$$

Many reports show values of (p , q and h_{max}) for several $\text{Fe}_y\text{Se}_x\text{Te}_{1-x}$ compounds. Such as the obtained values of (p , q and h_{max}) are 1.54, 3.8 and 0.28 respectively for $\text{FeSe}_{0.4}\text{Te}_{0.6}$ that indicate that vortex pinning is by a mixture of the surface and the point core pinning of the normal centers [65]. Das *et al.* reported values of (p , q and h_{max}) for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ are 1.65, 2.95 and 0.36 respectively, and they suggested that point pinning isn't enough to explain pinning mechanism in $\text{FeSe}_{0.5}\text{Te}_{0.5}$ compound [66]. The variations in the values of (h_{max}) for $\text{FeSe}_x\text{Te}_{1-x}$ compounds are result of change in the pinning force due to change in the Se and Te concentrations and sometimes these differences come from different methods that can be used in calculations, for example some people calculate (h) from (H/H_{c2}) and other from (H/H_{irr}).

1.9 Energy Gap in Superconductor

Energy gap in superconductor is defined as the binding energy of a cooper pair. Its typical value is several meV and is material dependent. According to (BCS) theory the upper limit of the energy gap at absolute zero is given by the following relationship. [47]:

$$E_g = 2 \Delta(0) \approx 3.528 k_B T_C \quad (1.13)$$

Where (k_B) is the Boltzmann's constant. (BCS) theory is only valid for the conventional superconductors and the value of the energy gap in such superconductors depends on their transition temperature [47]. The ratio, ($2\Delta/k_B T_c$) is commonly known as coupling strength. The maximum possible value of coupling strength for conventional superconductors is 3.528, which is called the weak coupling limit. Unconventional superconductors can have the coupling strength higher than the weak coupling limit and such coupling is termed as strong coupling. Several coupling strength related studies in iron-based superconductors provides contradictory results. Many reports found that some of the iron-based superconductors have weak coupling with coupling ratio while some of the iron-based superconductors have strong coupling ratio [67].

Energy gap in iron-based superconductor can be studied by Scanning tunneling microscopy/spectroscopy (STM/STS) and by using thermodynamic critical field (H_c), which can be found by [21]:

$$H_c = \frac{H_{c2}}{\sqrt{2}\kappa} \quad (1.14)$$

And energy gap value related to (H_c) as [71]:

$$E_g = 2 \Delta(0) = 2k_B T_c \sqrt{\left(\frac{\pi\gamma}{6}\right)\left(\frac{T_c}{H_c(0)}\right)} \quad (1.15)$$

Where (γ) is the electronic specific heat coefficient which varies from ~ 25 - 40 mJ/mol.K² [69, 70].

Recently, Kunwar *et al.* used scanning tunneling microscopy/spectroscopy (STM/STS) to measure the energy gap in FeSe_{0.5}Te_{0.5} superconductor. Their results revealed the presence

of two energy gaps, which are coexistence with the presence of weak and strong coupling mechanism in $\text{FeSe}_{0.5}\text{Te}_{0.5}$ superconductor. Escudero *et al.* found for same superconductor the ratio $(2\Delta/k_B T_c)$ has values between $(2.88 < 2\Delta/k_B T_c < 3.04)$, according to this $\text{FeSe}_{0.5}\text{Te}_{0.5}$ superconductor is in weak coupling limit [68].

1.10 Structure of the Thesis

The thesis is divided in seven chapters, summarized as follows: introduction and literature review are discussed in chapter one emphasizing on various superconducting properties of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ such as critical fields, critical current, pinning force and thermally activated flux flow. In chapter two, I present sample preparation, characterization and description of the $(I-V)$ isotherms and resistivity measuring system, vibrating sample magnetometer (VSM) and X-Ray Diffraction are also discussed. In chapter three, we start with initial characterization of the prepared single crystals. (XRD) pattern of different samples were presented and analyzed. The variations of the lattice parameters with iron content is also discussed. The $(I-V)$ isotherms and Arrhenius plots are discussed in chapter four. I also present variations of the thermal activation energies with temperature and applied magnetic field. Reversible and irreversible properties such as critical currents and pinning forces are discussed in chapter five. Also, different scaling procedures were discussed at the end of chapter five. Energy gap measurements are presented in chapter six. Summary of the main results of these studies and the possible extensions of this work is given in chapter seven.

1.11 Objective

In this thesis, we propose to investigate various electrical and magnetic properties of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single and poly crystals $(0.9 \leq X \leq 1.1)$. In particular; electrically resistivity (ρ),

upper magnetic field (H_{c2}), activation energies $U(H,T)$ and pinning force (P_f) will be investigated. Our main objectives are:

- a- To prepare high quality single crystals samples and polycrystalline samples of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$; near the optimal doping ($X \sim 1$).
- b- To investigate the effect of excess of iron content on the structure, flux flow and thermally activated flux flow of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ compounds.
- c- To investigate the effect of excess of iron content on several irreversible properties such as irreversibility field, critical current density and pinning forces.
- d- To study the behavior of scaling of the pinning force and critical current density and anisotropy changes with excess of iron contents.
- e- To obtain the evolution of the energy gap and the electronic density of states near the optimal doping.

CHAPTER 2

EXPERIMENTAL TECHNIQUES

In this thesis, we investigated the effects of iron contents on several electric and magnetic properties of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single and poly crystalline samples. The crystalline structure for all prepared sample were properly characterized using (XRD). Magnetic measurements were used to investigate critical current, pinning force and scaling behavior. Electric transport measurements have been used to study flux flow, thermal activated flux flow and activation energy near the superconducting transition under the influence of low applied magnetic field.

2.1 Samples Preparation

Solid state reaction has been used to prepare single and poly-crystalline samples of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ for $(0.9 \leq X \leq 1.1)$ using high purity Fe, Se and Te elements.

2.1.1 Polycrystalline Samples

High purity (99.99% or higher) Fe, Se and Te elements were used to prepare $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples. Stoichiometric ratios of the elements were used to prepare seven samples with $(0.9 \leq X \leq 1.1)$. All samples are given in the form $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ in table (2.1). The powdered elements are mixed and grinded using an agate mortar for about 10-20 minutes then pressed into pellets. The pressed pellets were encapsulated in a quartz tube and sealed at partial pressure of high purity Argon gas. All samples are initially annealed at 300 °C and 500 °C for 5 hours at each temperature, and then heated to 800 °C and held

for 10 hours. After that, the temperature was reduced to 400 °C at which the samples are annealed for 120 hours. After being cooled to room temperature; the samples were ground thoroughly then pressed in pellets, sealed in quartz tube under Argon and re-annealed at 400 °C for another 50 hours and cooled to room temperature. The annealing cycle is shown in figure (2.1).

Table (2.1): Samples used and the corresponding used masses to prepare one gram.

X	Fe (± 0.0001) g	Se (± 0.0001) g	Te (± 0.0001) g
0.90	0.3273	0.2571	0.4155
0.92	0.3322	0.2553	0.4125
0.95	0.3394	0.2525	0.4081
1.00	0.3488	0.2466	0.3989
1.02	0.3555	0.2464	0.3981
1.05	0.3621	0.2438	0.3940
1.10	0.3730	0.397	0.3873

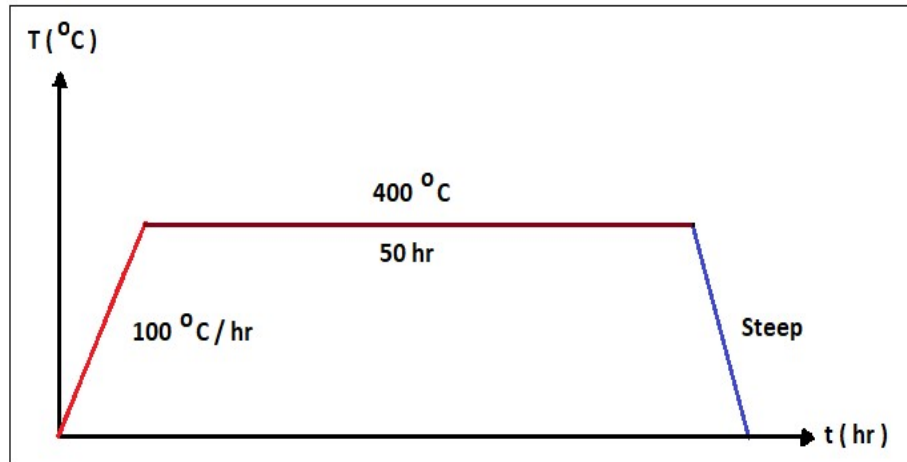
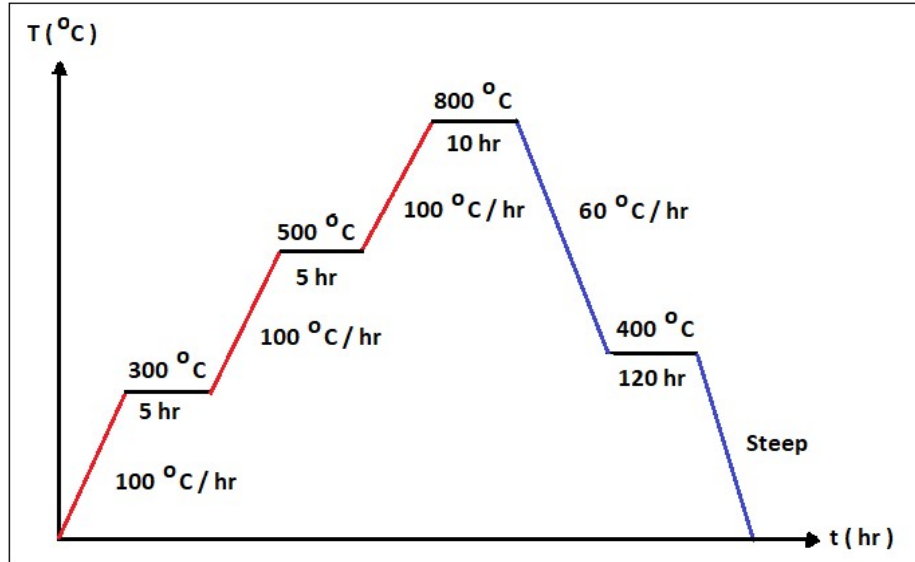


Figure (2.1): (a) First steps and (b) Second step annealing of polycrystalline samples.

2.1.2 Single Crystal growth

High purity ($\geq 99.99\%$) elements Fe, Se and Te have been used in different stoichiometric ratios to prepare $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$, stoichiometric ratios of the elements were used to prepare the samples with ($0.95 \leq X \leq 1.1$) (Table 2.1). The elements were grinded, pressed and sealed in a quartz tube partially filled with Argon gas. Single crystals were grown using Bridgman technique. The samples were annealed at 500 °C for 2 hours (T_{melting} for Se = 221 °C and T_{melting} for Te = 221 °C) and then at 1070 °C for 24 hours at this temperature the

whole material is homogenous and liquid. After that the samples were slowly cooled at a rate of 5 °C/hour from that temperature to 700 °C the reason of this slowly cooling to permit for single crystal growth. Then they were annealed at 700 °C for 5 hours to release internal stress and strains. This is followed by fast cooling to 400 °C and annealed for 2 hours to release more and more of internal stress and strains. Finally, the furnace was turned off and the samples are cooled to the room temperature as shown in the figure (2.2).

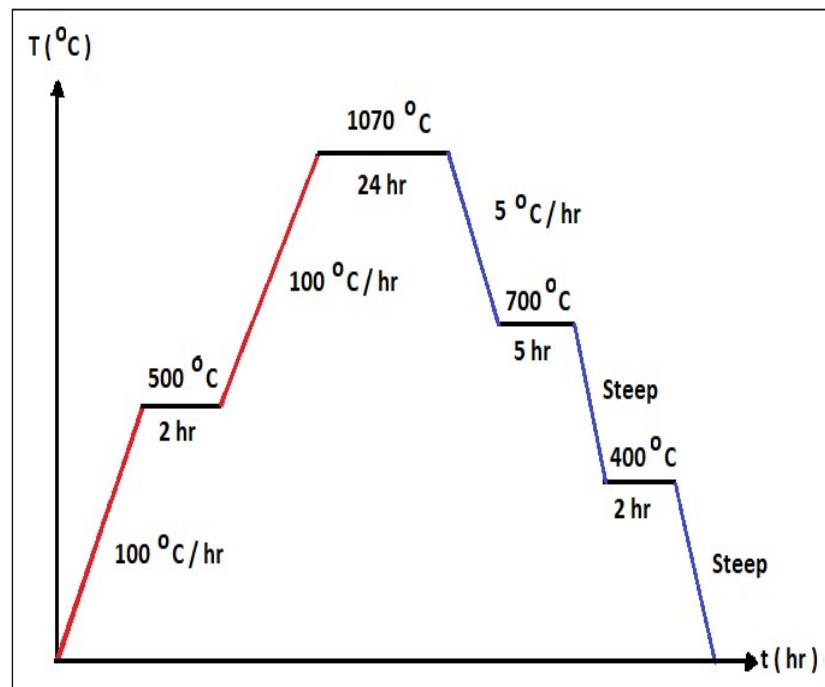


Figure (2.2): Various stages of annealing of single crystal samples.

2.2 Sample Characterization

The prepared samples were characterized by using different techniques. Four-point probe technique is used to measure the superconducting critical transition temperature (T_c) measurements and obtain the (I - V) characteristics of all samples. Powdered (XRD) technique was used to assist the crystalline structure.

2.2.1 T_c measurements by using the Four-Point Probe Technique

The four-point probe technique first used by Winner (1916) to measure the changes in resistivity with temperature [74, 75 and 76]. Resistances versus temperature were obtained by passing current ($I = 2$ mA) in order to determine (T_c) and transition width.

2.2.2 Characterization of the prepared samples by using X-Ray Diffraction (XRD) Technique

The prepared samples were characterized using the powdered X-ray diffraction (XRD). The (XRD) patterns are used along with Bragg's condition to calculate the lattice parameters [6,7]:

$$2d\sin\theta = n\lambda \quad (2.1)$$

The d -spacing for tetragonal structures d is given by [6, 7]:

$$\frac{1}{d^2} = \frac{(h^2+k^2)}{a^2} + \frac{l^2}{c^2} \quad (2.2)$$

A small part of sample was grind and loaded into the sample holder of Rigaku Ultima-IV X-ray diffractometer. $\text{CuK}\alpha$ radiation was used in the wavelength 1.54060 Å. The samples were step scanned between 0 and 80° at (2θ) scale while measuring the intensities of the diffraction peaks.

The (PDXL) software is used to analyze the (XRD) patterns, the powder diffraction database of the International Centre for Diffraction Data (ICDD) has been used to identify the impurity phases in all prepared samples.

2.3 Transport Properties

The electrical measurements were obtained using Lake Shore 7507 Hall-effect measurements system. The variations of resistance and the (I - V) characteristics with temperature are measured in the presence of magnetic field (0-2Tesla). The samples are mounted on the copper tail of a closed cycle cryostat (Advanced Research Systems). Lake Shore-340 temperature controller controlled the temperature. A constant current source (Keithley 220) provided constant currents up to 100 mA to the outer probes and nanovoltmeter (Keithley 2182) determined the voltage differences through the inner probes. The (I - V) characteristics were measured at different temperatures and different magnetic fields. All transport properties measurements were performed in the ab - plane.

A small drop of the silver paste was used to connect wires to the sample. The wires are firmly attached to the cold finger of the cryostat to reduce thermal heat flow to the sample. The pressure inside the cryostat was kept at $\sim 10^{-7}$ mbar to ensure good isolation and to avoid heat transfer from the surrounding to the sample. The resistivity was measured in the temperature range 10-300 K in various applied magnetic fields.

2.4 Magnetization Measurements

Magnetization measurements were performed using a computer-controlled PAR-Lakeshore (Model 4500/150A) variable temperature vibrating sample magnetometer (VSM). The maximum magnetic field is 9 Tesla, and a variable temperature range is 1.8 K to 300K. The VSM system consists of the following major parts:

- 1- Vibrating head and sample holder.
- 2- Variable temperature cryostat.

- 3- Temperature controller.
- 4- VSM controller.
- 5- Power supply and controller of the magnet.
- 6- PC with GPIB-PcII interface card which is connected to all controllers of system.

For detailed description of the system see reference [77].

CHAPTER 3

X-RAY DIFFRACTION ANALYSIS

In this chapter we investigate the structural properties for all samples by using X-ray diffraction.

3.1 Phase diagrams

Based on the three binary phase diagrams of (Fe-Se), (Fe-Te) and (Se-Te), that are shown in figures (3.1), (3.2) and (3.3) respectively, we can explain impurities appearance in XRD patterns for all the samples. Slowly cooling from 1075°C to 900 °C several phases and their mixtures such as FeSe, FeSe₂, Fe₃Se₄ and Fe₇Se₈ will form, which are as shown in the figure (3.1). Cooling FeSe_{0.5}Te_{0.5} to the temperature range of 810 °C to 650 °C is shown in the figure (3.2). In (Se-Te) binary phase diagram which is shown in figure (3.3), in the case of the formation of solid solution between the two elements over the entire range of concentration, no ordered state is found.

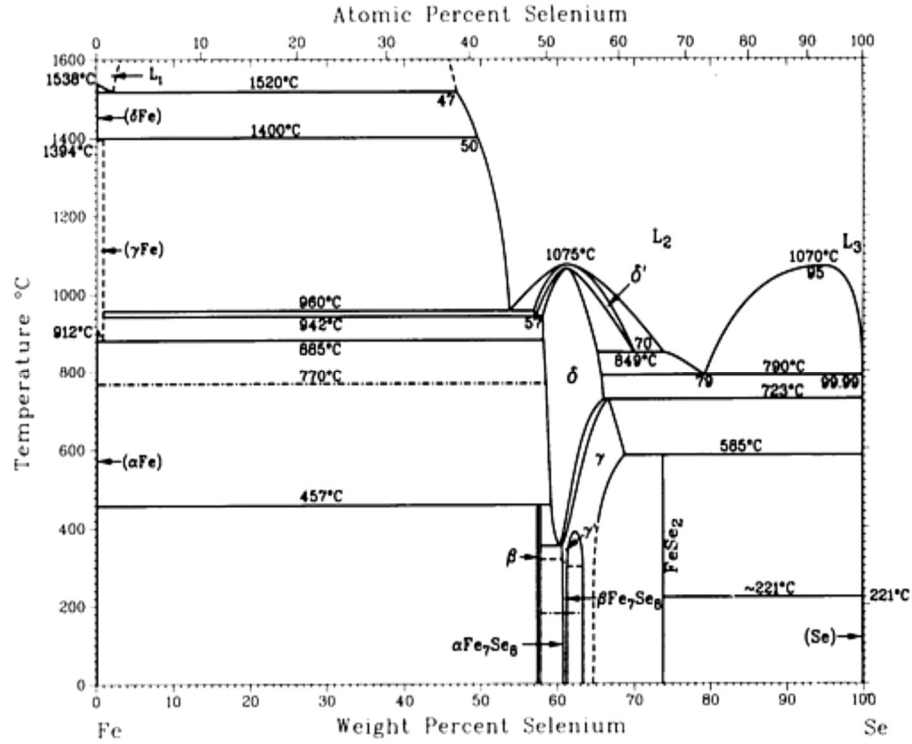


Figure (3.1): phase diagram of Fe-Se [80].

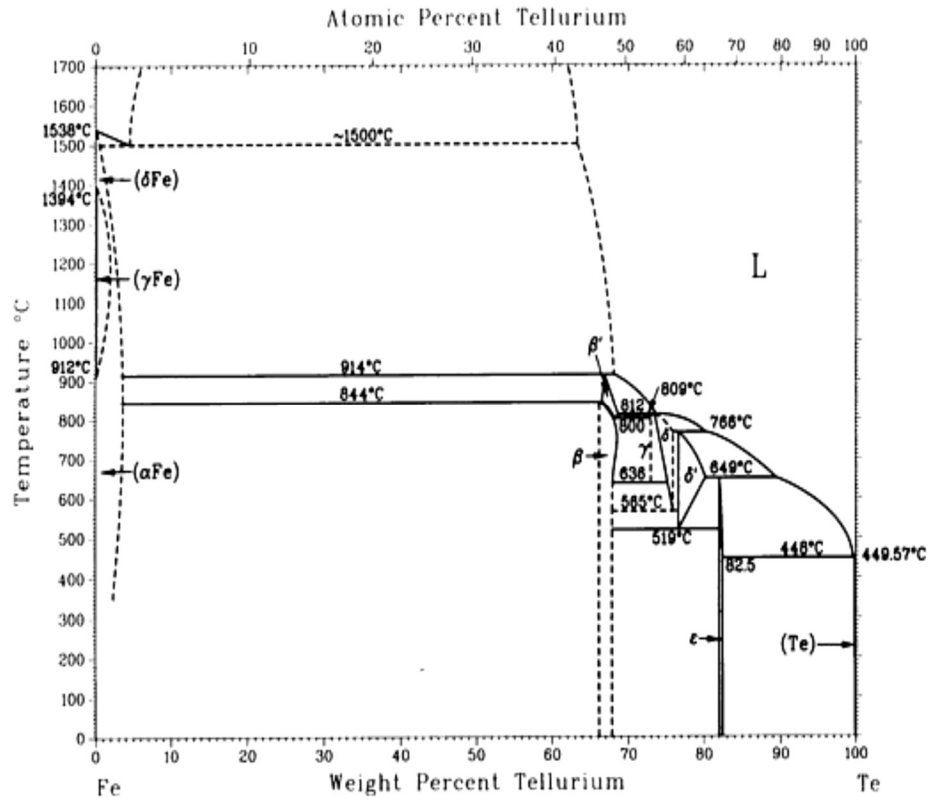


Figure (3.2): phase diagram of Fe-Te [81].

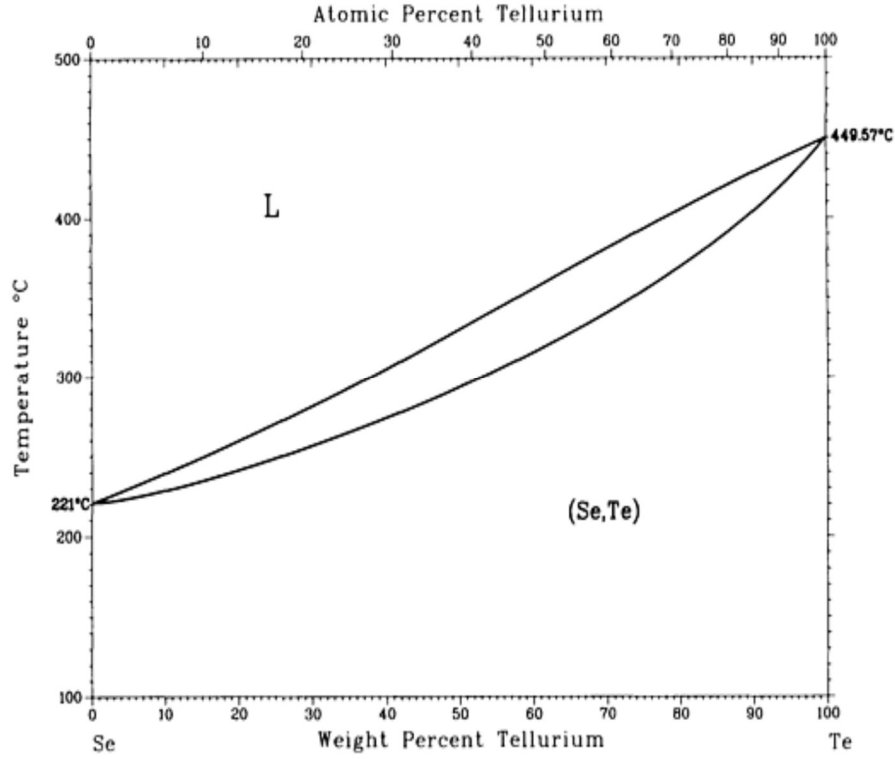


Figure (3.3): phase diagram of Se-Te [82].

3.2 Result and discussion of (XRD) for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples

The (XRD) measurements are performed at room temperature for two groups of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples. The first group is polycrystalline samples with different iron content ($0.9 < X < 1.1$) and the second group is single crystal samples with the iron content ($1 < X < 1.1$). Our aim is to study the effect of iron content in the samples.

3.2.1 (XRD) patterns for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples

X-ray diffraction patterns obtained for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples with ($X = 0.90, 0.92, 0.95, 1.00$) are shown in the figure (3.4) and for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples with ($X = 1.00, 1.02, 1.05$ and 1.10) are shown in the figure (3.5).

Figures (3.4) and (3.5) show the (XRD) patterns taken at room temperature for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples with ($0.9 \leq X \leq 1.1$). The majority of the diffraction peaks have been indexed with the tetragonal PbO structure with $P4/nmm$ space group. Samples with

large Fe-deficiency ($X \sim 0.9$) have several impurity phases such as Fe_7Se_8 , Fe_3Se_4 , FeO_2 , FeSe and Te . The presence of Fe_7Se_8 is in line with FeSe phase diagram shown in figure (3.1). However, we detected this impurity phase even in samples with excess of Fe [45]. The (XRD) figures revealed that the impurity phases decrease with increasing iron content in the samples reaching almost pure single phase for the optimal sample $\text{FeSe}_{0.5}\text{Te}_{0.5}$. And (XRD) analysis were presented in the tables (3.1).

Table (3.1): The lattice constants and the lattice volumes for the different $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples.

Sample	$c \pm \Delta c$ (Å)	$a \pm \Delta a$ (Å)	$V \pm \Delta V$ (Å ³)
0.90	5.931 ± 0.081	3.778 ± 0.003	84.655 ± 0.572
0.92	5.931 ± 0.058	3.788 ± 0.014	85.104 ± 0.731
0.95	5.956 ± 0.038	3.787 ± 0.008	85.400 ± 0.230
1.00	5.976 ± 0.061	3.779 ± 0.017	85.320 ± 0.829
1.02	5.974 ± 0.032	3.771 ± 0.014	84.944 ± 0.685
1.05	5.972 ± 0.024	3.767 ± 0.017	84.720 ± 0.930
1.10	5.966 ± 0.028	3.767 ± 0.023	84.686 ± 0.964

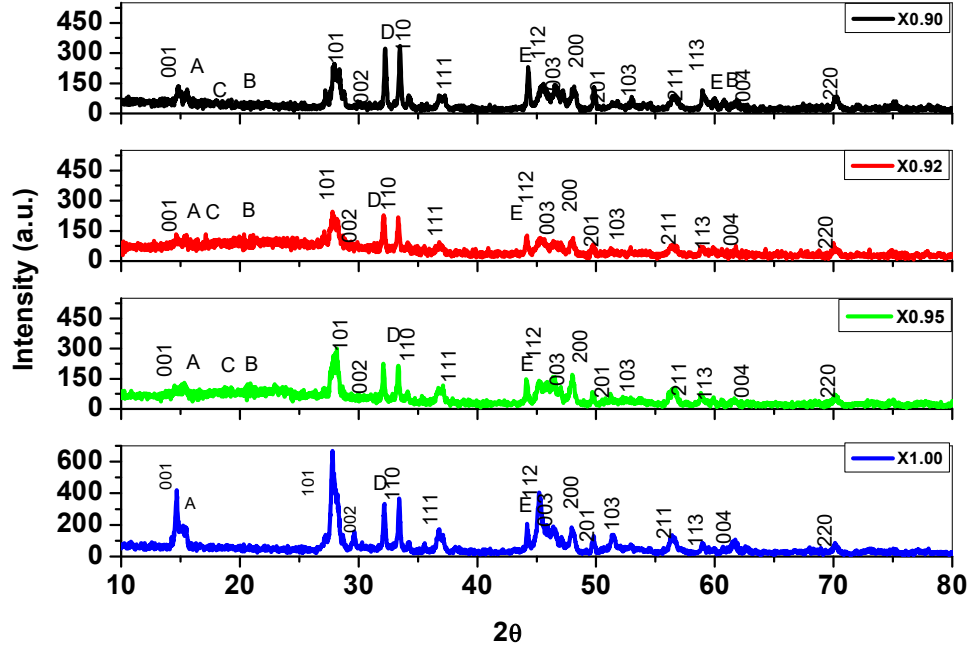


Figure (3.4): Room temperature (XRD) patterns for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples with x ($0.9 \leq x \leq 1.0$) and impurity phases A: FeSe, B: FeO₂, C: Te, D: Fe₇Se₈ and E: Fe₃Se₄.

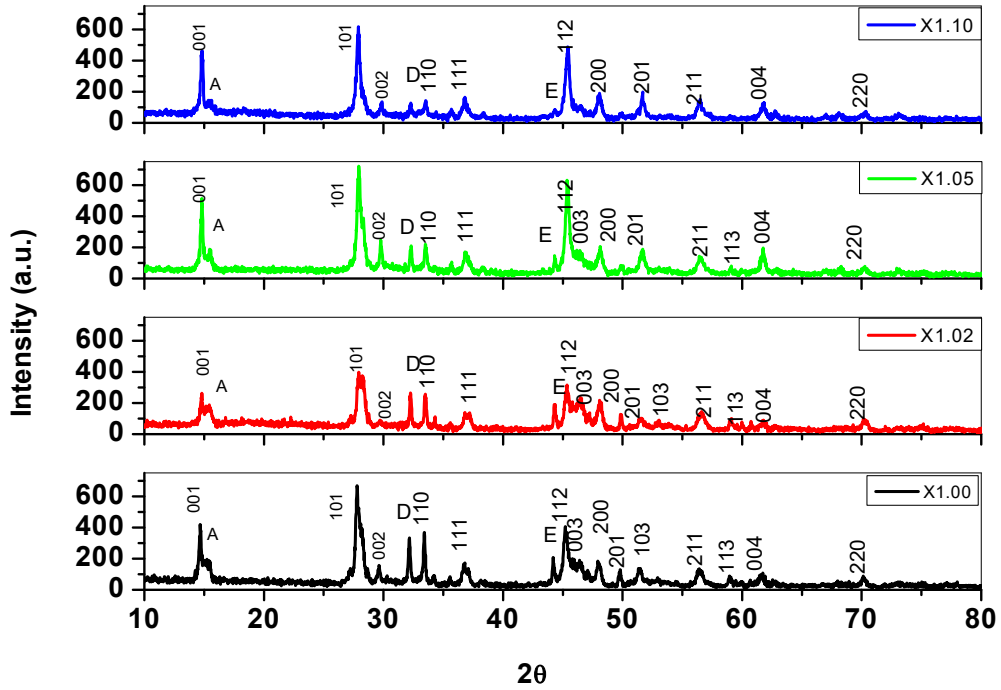


Figure (3.5): Room temperature (XRD) patterns for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples with x ($1.0 \leq x \leq 1.1$) and impurity phases A: FeSe, B: FeO₂, C: Te, D: Fe₇Se₈ and E: Fe₃Se₄.

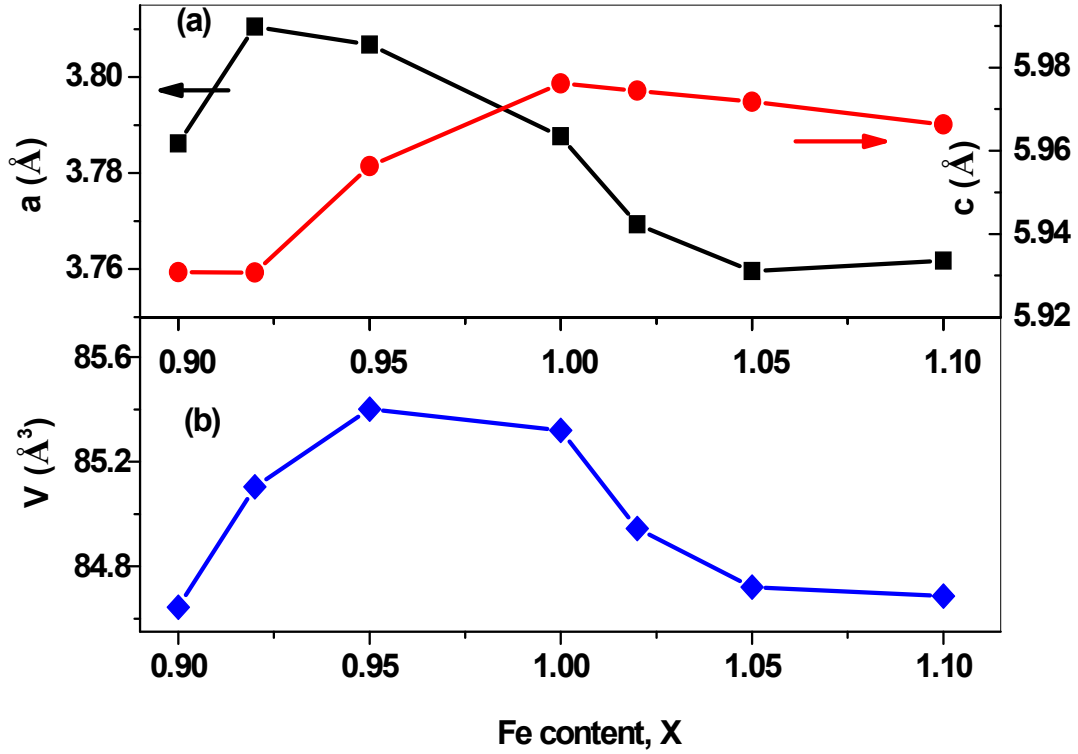


Figure (3.6): Variation of (a) lattice parameters with Fe content of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ and (b) the volume of the unit-cell with Fe content of the same sample.

Figure (3.6a) shows variation of lattice constants (a and c) with iron contents in samples of polycrystalline $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$, and figure (3.6b) shows the lattice volume variations with iron content in the same sample.

We found that the lattice parameters (c (Å)) reaches maximum near ($X=1.0$), while (a (Å)) is maximum near ($X=0.92$). The maxima volume of the unit cell occurs in between ($X=0.95$ and $X=1.0$), in the iron deficient region. Our study infers that Fe-deficiency may induce lattice strain while Fe-excess may induce internal stress. The competition effect of stress-strain may lead to the observed non-monotonic changes in the lattice parameters and in the unit cell volume [83].

3.2.2 Result and discussion for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystals samples

In an effort to reduce the impurity phases in polycrystalline $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples we prepared a set of single crystals, and we investigated their structure and several magnetic and transport properties. Single crystal before grinding indicating good quality of prepared of single crystals because diffraction peaks are coming from (00 l) namely (001), (002), (003), (004), with no impurities peaks. Figures (3.7) shows the (XRD) patterns taken at room temperature for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal sample before grinding.

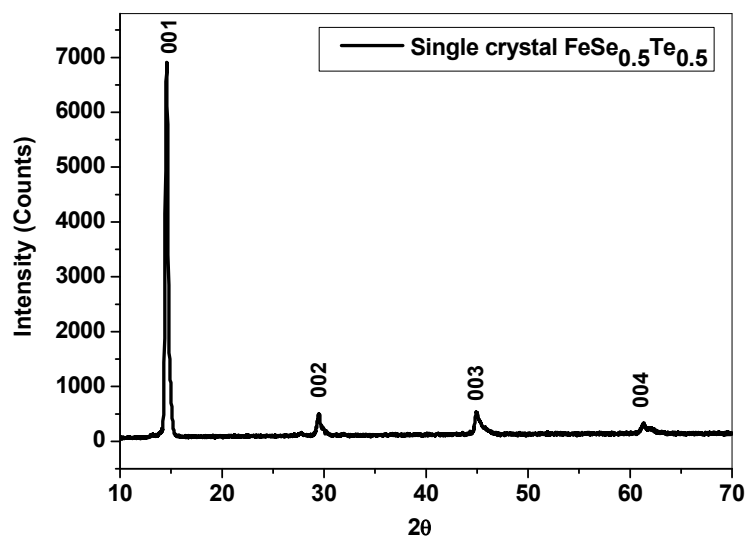


Figure (3.7): X-ray diffraction pattern of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal before grinding.

Prior to obtaining the (XRD)-patterns a small portion of the prepared single crystals were powdered and used for further (XRD) analysis. The X-ray diffraction patterns obtained for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal samples with ($x = 1.00, 1.02, 1.05$ and 1.10) are shown in the following figures (3.7– 3.10). Figures (3.8-3.11) show the (XRD) patterns taken at room temperature for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal samples with ($1 \leq x \leq 1.1$). The diffraction

peaks have been indexed with the tetragonal PbO structure with P4/nmm space group. Samples have several impurity phases such as Fe₃Se₄, FeSe and Te. However, comparing the powdered single crystals (XRD) patterns with the polycrystalline patterns, we found considerable reduction in the impurity phases in powdered single crystal patterns. XRD analysis are presented in the table (3.2).

Tab. (3.2): The lattice constants and the lattice volumes for the different Fe_xSe_{0.5}Te_{0.5} single crystal samples.

Sample	$c \pm \Delta c$ (Å)	$a \pm \Delta a$ (Å)	$V \pm \Delta V$ (Å ³)
1.00	6.022 ± 0.010	3.803 ± 0.017	87.10 ± 2.92
1.02	6.037 ± 0.018	3.783 ± 0.001	86.41 ± 2.55
1.05	5.981 ± 0.017	3.800 ± 0.023	86.36 ± 2.98
1.10	6.010 ± 0.010	3.790 ± 0.015	86.36 ± 2.64

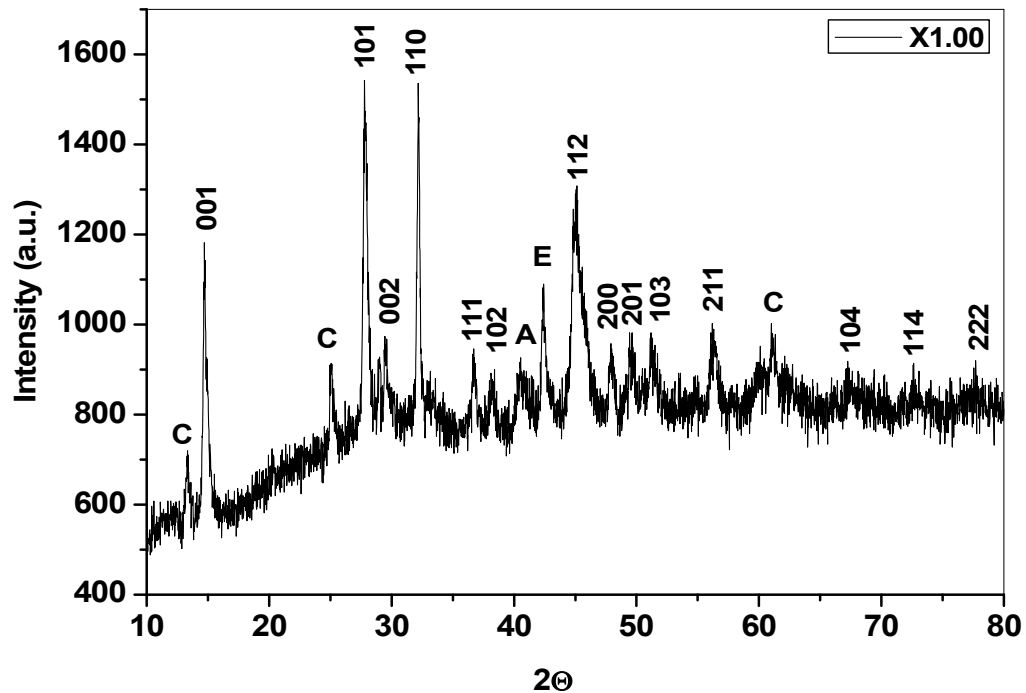


Figure (3.8): X-ray diffraction pattern of FeSe_{0.5}Te_{0.5} single crystal with the phases of the peaks and impurity phases A: FeSe, C: Te and E: Fe₃Se₄.

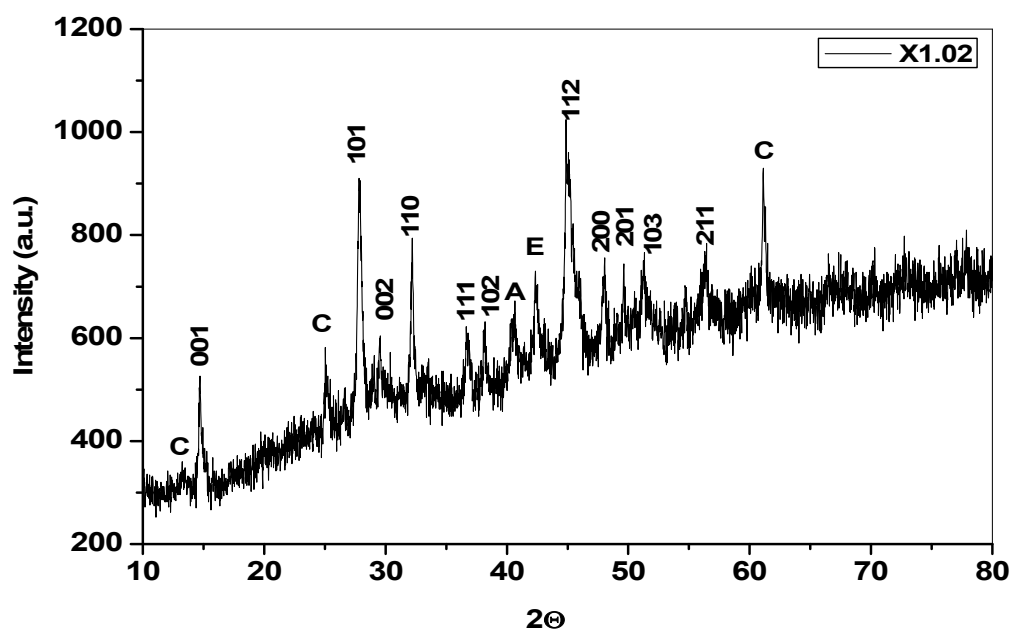


Figure (3.9): X-ray diffraction pattern of $\text{Fe}_{1.02}\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal with the phases of the peaks and impurity phases A: FeSe, C: Te and E: Fe_3Se_4 .

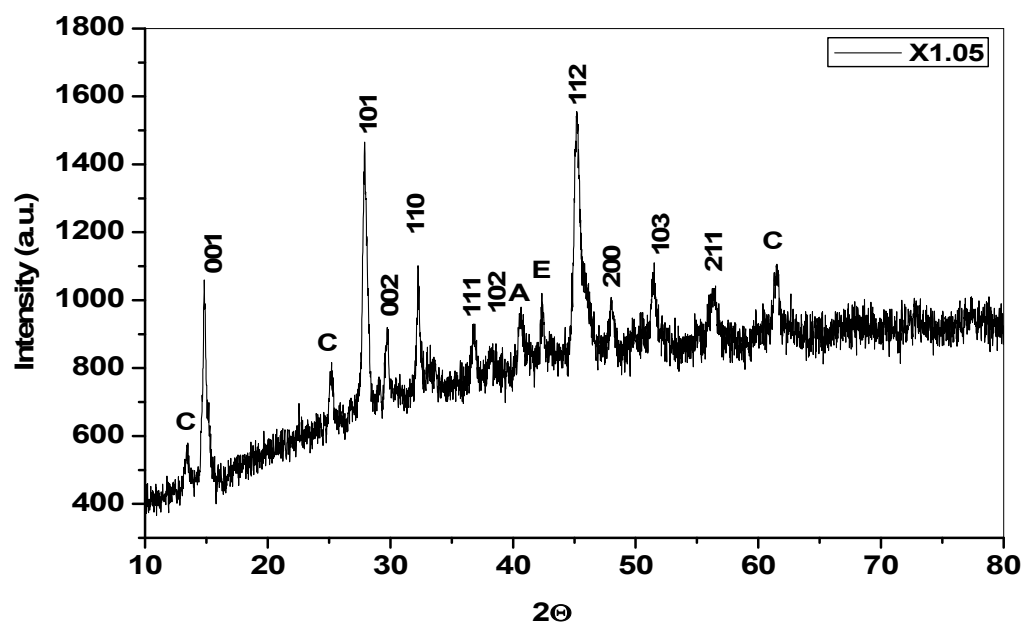


Figure (3.10): X-ray diffraction pattern of $\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal with the phases of the peaks and impurity phases A: FeSe, C: Te and E: Fe_3Se_4 .

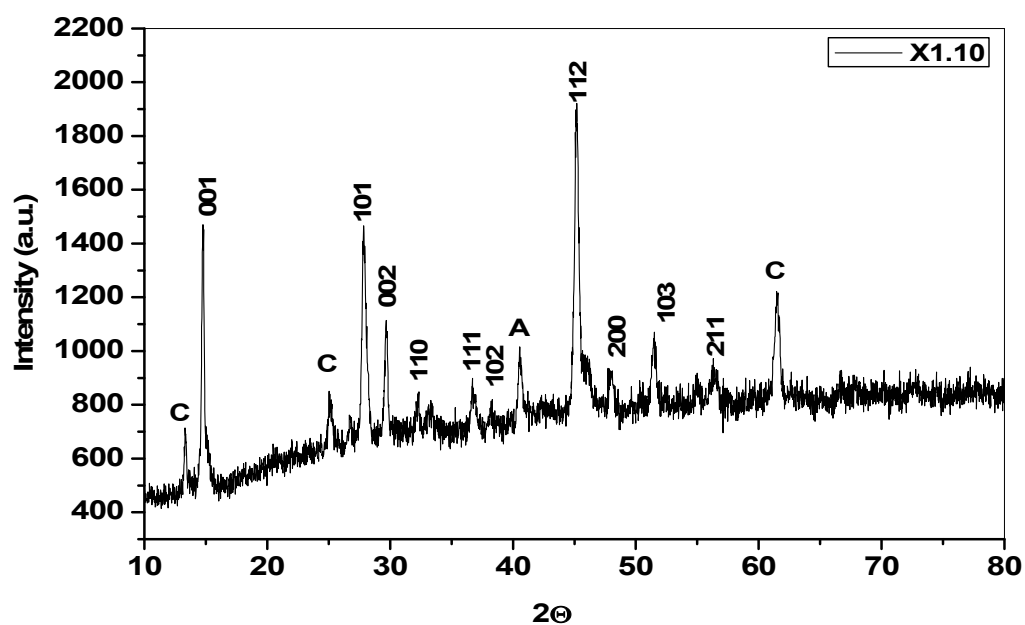


Figure (3.11): X-ray diffraction pattern of $\text{Fe}_{1.1}\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal with the phases of the peaks and impurity phases A: FeSe, C: Te and E: Fe_3Se_4 .

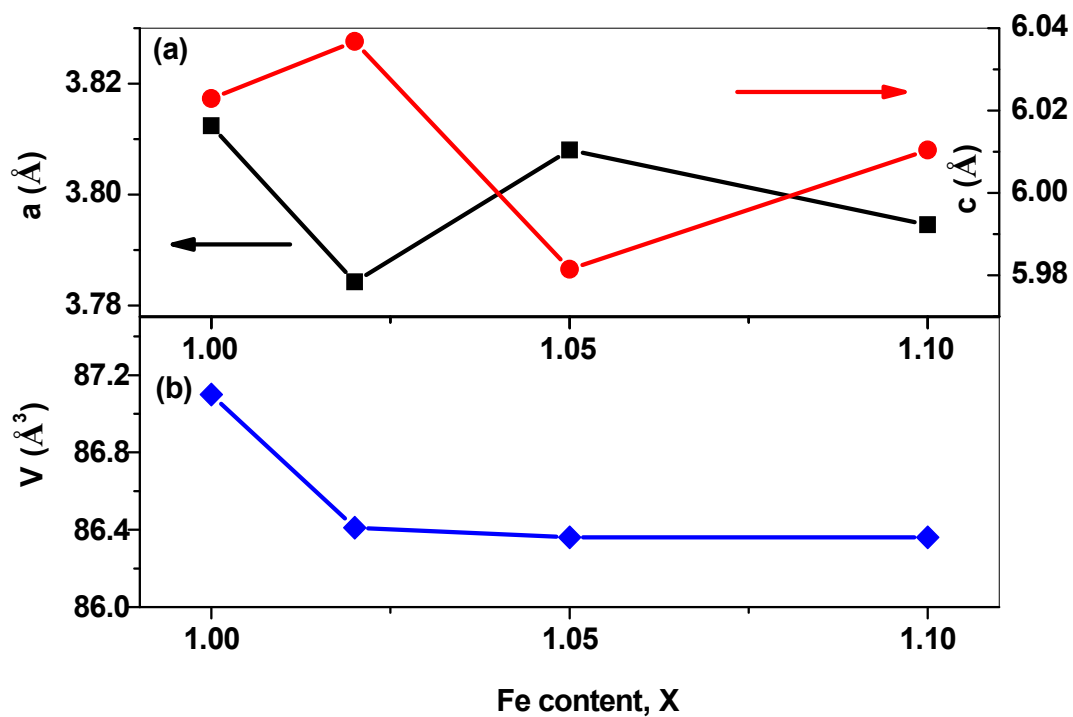


Figure (3.12): Variation of lattice parameters (a) and the volume of the unit-cell (b) with Fe content of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal samples.

The lattice constants and the unit cell volume are presented in table (3.2). The variations of the lattice constants (a and c) with Fe content in the single crystal samples of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$, and the variations of the lattice volume with Fe content in the samples of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ are shown in figure (3.12). We found that the lattice parameters (c (Å)) reaches maximum near ($X=1.02$), while (a (Å)) is maximum near ($X=1$). The maximum volume of the unite cell occurs at ($X=1$). The competition effect of stress-strain may lead to the observed non-monotonic changes in the lattice parameters and the volume of the unit cell [78].

Differences in internal strain and stress of single crystals and polycrystalline of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples will cause shift in lattice parameters [79]. The (FWHM) values are found different at same peak. For example, for the peak 110, at same Fe content ($X=1$), the (FWHM) values are 0.197 for single crystal and 0.242 for polycrystalline samples. This is clearly indicated by the observed sharp peaks in the (XRD)-patterns of the single crystals.

CHAPTER 4

THERMAL ACTIVATION ENERGY ANALYSIS

4.1 Introduction

The measurement/study of (I - V) isotherms of superconducting materials is common and popular technique to investigate several superconducting properties such as critical current density, pinning force, flux flow resistance and thermal activation energy.

Thermal Activation Energy (TAE) refers to the energy required for vortices to overcome the pinning potential barrier created by pinning centers in the superconducting state. In the presence of an applied magnetic field, or when a transport current passes through a superconductor, the Lorentz force causes variation in the effective potential barrier between the neighboring pinning potentials. The effective potential (U_{eff}) is calculated from the difference between the barrier height and the energy of the Lorentz force as shown in figure (4.1) [42]:

$$U_{eff} = U_o - U_L \quad (4.1)$$

Where (U_o) is potential barrier height, (U_L) is Lorentz potential, and (J_c) is commonly known as the de-pinning critical current density, which is the current requires to derive the vortex from its pinning center. Here we describe two cases in regard to the current density, (J). The first case is when there is no current has been applied ($J = 0$) to the system, which leads to application of no force on the system. In this case, due to thermal activated processes, the probability of vortex jumping over the potential barrier is the same in all

directions (isotropic), and the net vortex velocity is zero. For the second case the current density is non-zero. When a current has been applied ($J > 0$) to a system a driving force is being produced in the system. The action of the applied force it reduces the potential well on certain direction and increases it in the opposite direction. The benchmark of critical current density (J_c) can be used to distinguish three different regimes. When the current density is quite smaller than the critical current density ($J \ll J_c$), the dissipation is happened by thermal excitations and the regime is called the TAFF (Thermal Activation Flux Flow) regime. While at very high current densities ($J \gg J_c$), the pinning force is smaller than the driving force, as a result a diffusive vortex motion happens. Such regime of very high current density regime is called the (Flux Flow) regime. Finally, when current density is comparable to the critical current density ($J \sim J_c$), the regime is called flux creep regime, which includes different vortices motion and complicated potential barriers. Several experimental techniques can be used to evaluate the thermal activation energy. The (I - V) isotherms and the Arrhenius plot are commonly used techniques in this regard. In this chapter we used the (I - V) isotherms technique and Arrhenius plot to investigate the behavior of the activation energy near the superconducting transition temperature and in low magnetic field.

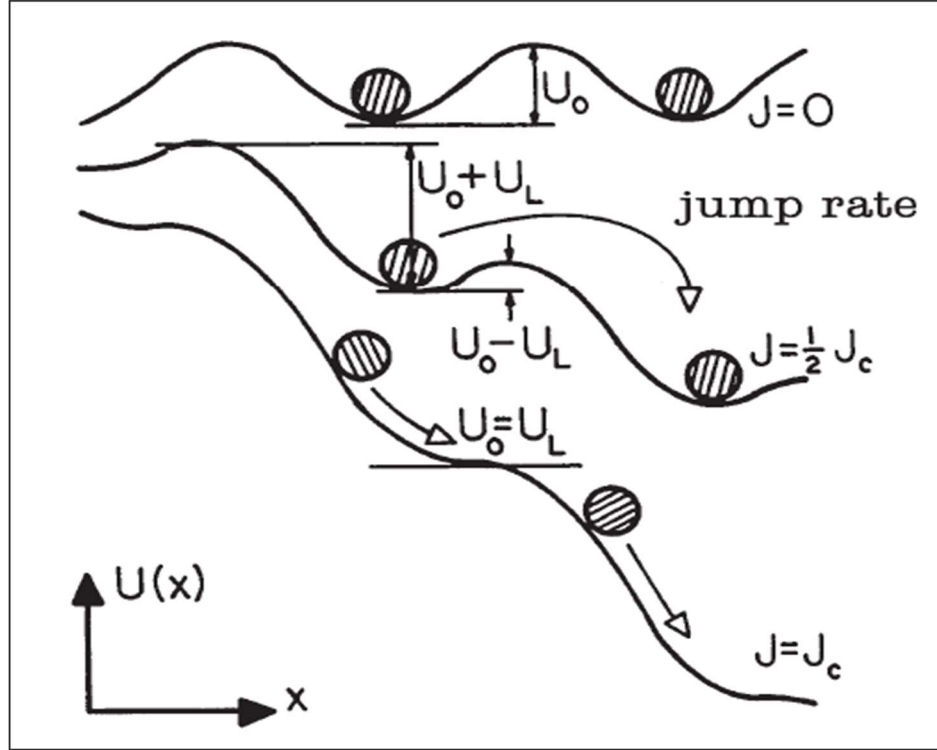


Figure (4.1): Pinning potential under the effect of different current densities [42].

4.2 The (I-V) Isotherms

The main characteristics of superconductor is the vanishing of the electrical resistance. An electric current can pass through the superconductor without energy losses, as long as it is below certain critical value known as the critical current (J_c). The value of the critical current for a superconductor depends on the temperature and the applied magnetic field.

But when a current passes through the mixed state of hard superconductors, this may cause vortex motion that give rise to flux flow resistance leading to energy losses [54]. Transport and dissipative properties in type II superconductors are dominant in the region close to critical temperature. Kim-Anderson proposed a model to explained flux creep and vortex motion close to critical temperature [53].

Vortex motion, the irreversibility line and a vortex glass transition have been extensively investigated in high temperature superconductors [55-59]. Fisher *et al.*, suggested the use of scaling behavior for the $(I-V)$ isotherms near critical transition temperature (high- T_c) to investigate the behavior of vortex state in high temperature superconductors [60 and 62]. The $(I-V)$ isotherms for thin films and single crystals of high- (T_c) superconductors, revealed a continuous vortex glasses transition at temperatures well below (T_c) .

The first experimental proof for the continuous vortex glass transition at temperatures below T_c , using scaling the $(I-V)$ curves, was performed by Koch *et al.* [61] on YBCO thin films, and on YBCO single crystals by Gammal *et al.* [64]. Sun *et al.* suggested a power-law ($V = \alpha I^\beta$) to describe the behavior of the $(I-V)$ isotherms. The exponent β is referred to the effective pinning barrier (U_{eff}).

In the mixed state, in type II superconductors, the vortex behavior can be classified into different regimes, namely, thermal activated flux flow regime (TAFF), flux creep regime (FC) and flux flow regime (FF). However, these regions are not sharply distinguished. For a given isotherm, the variations in the $(I-V)$ curves can be qualitatively used to distinguish between these regions as shown in figure (4.2) [65].

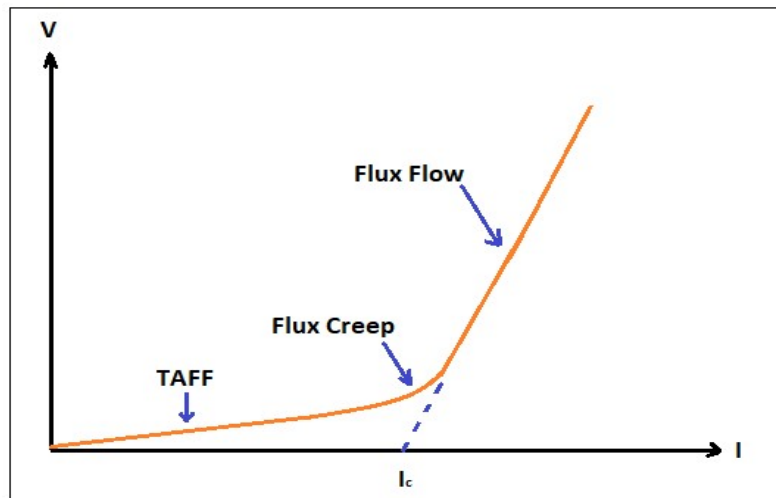


Figure (4.2): Schematic view of the $(I-V)$ curve with different vortices regimes.

The Fe-based superconductors have a moderate value of (T_c) with the added magnetic effects. Hence, they are in between conventional and high- (T_c) superconductors. Therefore, it will be interesting to study the effects of thermally activated flux flow on different transport properties. These properties are clearly affected by the dynamic behavior of vortices. The magnetic elements such as Fe may have additional effects on the current–voltage isotherms [64, 85].

In this thesis, we present the (I - V) isotherms of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystals near ($T_c \sim 14.5\text{K}$) under different values of magnetic field. We use the power-law relation to evaluate the variation of the activation energies (U) with temperature and field. Figures (4.3 a-d) show (I - V) curves of the sample $\text{FeSe}_{0.5}\text{Te}_{0.5}$ at magnetic field $H = 0, 0.25, 0.5$ and 1.4T respectively for temperatures range 13.4 - 15.4 K . The data are presented in a log-log scale which simplifies the evaluations of α and β in the power-law relation:

$$V = \alpha I^\beta \quad (4.2)$$

Where α is a constant, and β is given by:

$$\beta = U_{\text{eff}}/k_B T \quad (4.3)$$

And (U_{eff}) is the effective pinning potential [56, 57, 58 and 99]. The exponent β has been calculated from the slope of linear part of the (I - V) isotherms at ($H = 0, 0.25, 0.5, 1\text{T}$ and 1.4T). The activation energy (U_{eff}) was evaluated in both (TAFF) and (FF) regimes. The activation energy is essentially the pinning barrier introduced by defects and pinning centers in the sample. The results are plotted in figures (4.4) (a and b) as a variation of (T) for both (TAFF) and (FF) regimes at ($H = 0, 0.25, 0.5$ and 1.4T). The results are discussed in detail in the following section. Thermal activated flux flow (TAFF) occurs near $30\text{ }\mu\text{V}$, while flux flow (FF) regime is at voltage larger than $30\text{ }\mu\text{V}$.

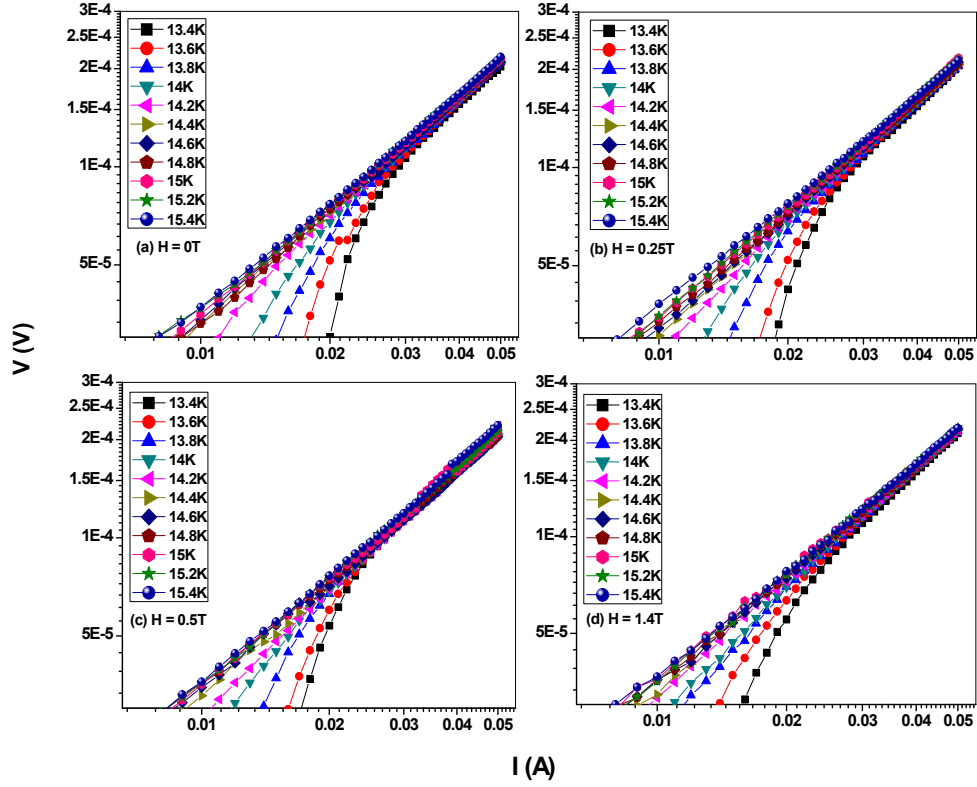


Figure (4.3): Selected (I - V) isotherms for single crystal $\text{FeSe}_{0.5}\text{Te}_{0.5}$ (a) at $H = 0\text{T}$, (b) at $H = 0.25\text{T}$, (c) at $H = 0.5\text{T}$ and (d) $H = 1.4\text{T}$, around its corresponding superconducting transition temperature.

4.2.1 Temperature Dependence

The figure (4.4) reveals that as the temperature reduces below the critical temperature, the potential pinning barrier (U_{eff}) continuously rises. The rise of (U_{eff}) in (TAFF) region is quicker than that in (FF) region. This is a direct reflection of the gradual rise in the exponent (β). Near (T_c), the value of the exponent (β) is close to one, indicating Ohmic (I - V) behavior. The deviation from Ohmic behavior occurs at (T_{nl}) a non-linear temperature about 14.2 K, yet it is about 0.3 K lower than the critical temperature ($\sim 14.5\text{K}$). The obtained pinning potential value at ($H = 1.4\text{T}$) for both regions descends to one tenths of its value at ($H = 0\text{T}$). Values of (β) were listed for optimum sample $\text{FeSe}_{0.5}\text{Te}_{0.5}$ in tables

(4.1) and (4.2) at ($H = 0T$) and ($H = 1.4T$) respectively. At temperature above (T_c), the exponent ($\beta \sim 1.1$), for both regions, indicating the non-linear characteristic of (I - V) curves which means the sample is no longer in the normal state. The fluctuations and thermal effects near (T_c) can cause deviations and limitations for Sun *et al.* model [56].

Table (4.1): Exponent values for different temperatures in both (TAFF) and (FF) regimes at H=0T.

T (K)	β in TAFF	β in flux flow
13.4	16.3798	2.7221
13.6	8.0485	2.4168
13.8	5.2259	2.0048
14.0	2.5881	1.5175
14.2	1.5729	1.3209
14.4	1.2196	1.1751
14.6	1.1683	1.1683
14.8	1.1431	1.1431
15.0	1.1173	1.1173
15.2	1.0702	1.0702
15.4	1.0671	1.0671

Table (4.2): Exponent values for different temperatures in both (TAFF) and (FF) regimes at $H=1.4T$.

T (K)	β in TAFF	β in flux flow
13.4	6.6037	2.9191
13.6	5.4789	2.6143
13.8	3.9308	2.0528
14.0	2.6469	1.5592
14.2	2.0215	1.3982
14.4	1.8983	1.3697
14.6	1.3562	1.2316
14.8	1.2006	1.2006
15.0	1.2087	1.2087
15.2	1.1953	1.1953
15.4	1.1191	1.1191

4.2.2 Magnetic Field Dependence

The variations of activation energies (TAFF) and (FF) regions with the magnetic field are presented in figures (4.5 a) and (4.5 b). At a given temperature, the obtained (U_{eff}) values in (TAFF) region are consistently higher than (U_{eff}) values in (FF) region. Also, values of (U_{eff}) are rapidly decreasing with increasing magnetic fields. This reflects the sensitivity of effective pinning potential to the applied magnetic field.

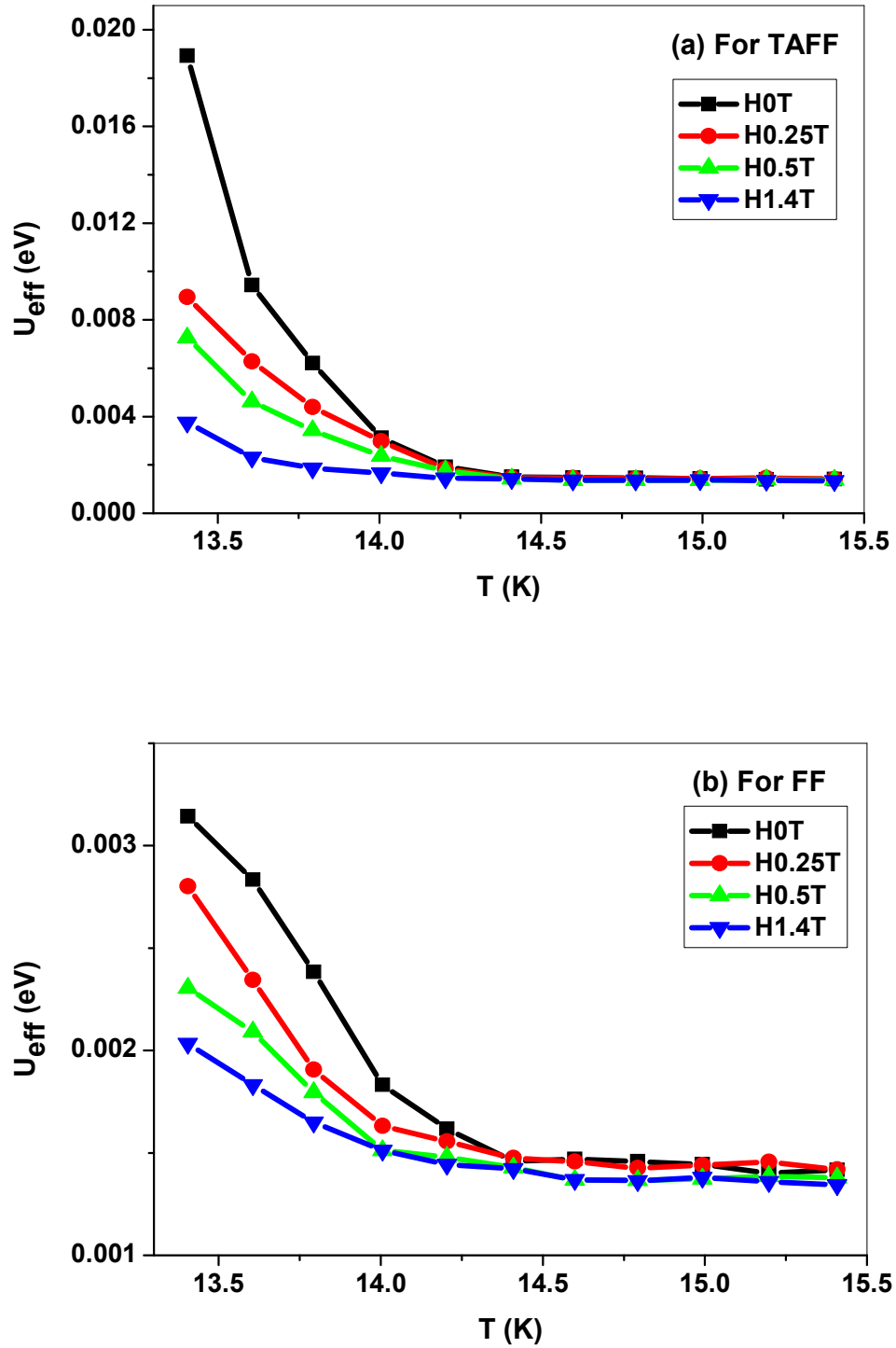


Figure (4.4): The variations of the effective pinning potentials (U_{eff}) versus temperature (a) in thermally activated flux flow region (TAFF) and (b) in flux flow region (FF) at different applied magnetic fields; $H=0, 0.25, 0.5$ and 1.4T for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal.

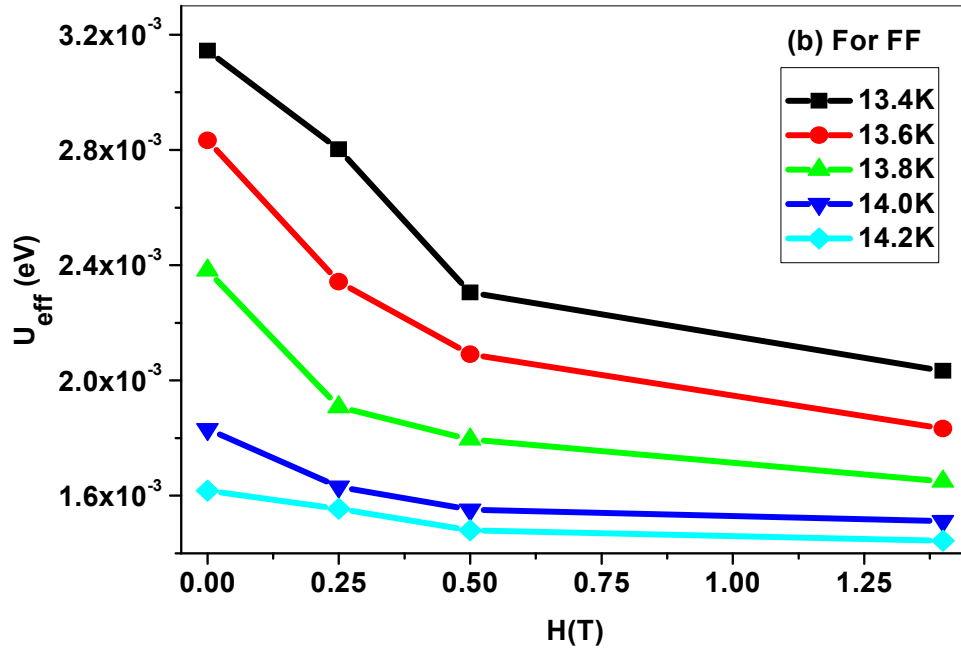
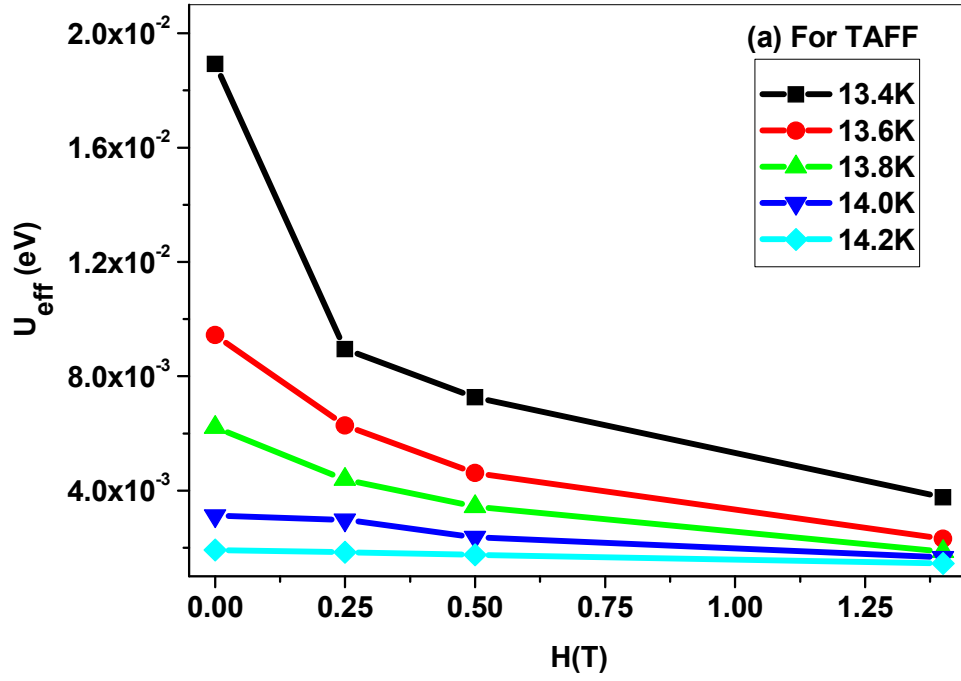


Figure (4.5): The variations of the effective pinning potential (U_{eff}) versus magnetic field (a) in thermally activated flux flow region (TAFF) and (b) in flux flow region (FF) at different temperatures for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal.

4.2.3 Characteristic Thermal Activation Energy (U_0)

The resistivity in (*TAFF*) regime is given by:

$$\rho = \rho_n \exp\left(-\frac{U}{k_B T}\right) \quad (4.4)$$

Where, (ρ_n) is the resistivity for normal state and (U) is the thermal activation energy, which is temperature and magnetic field dependent. Thermal activation energy shows parabolic behavior, it exhibits power law with magnetic field, which may depend on the type of flux pinning in the superconducting state [87].

Several researchers suggested that activation energy in high temperature superconductors is dependent on both temperature and magnetic field. Palstra *et al.* suggested that power law dependence on field to formulate characteristic thermal activation energy as ($U_0 \propto H^\alpha$) [51]. Yi Sun *et al.* found similar result for FeSe thin film [88]. Kucera *et al.* suggested that thermal activation energy for Bi-2212 thin films as ($U \propto H^{1/2} (1-t)$), where t is the reduced temperature ($t = T/T_c$) [40]. Wang reported that ($U \propto H^n (1-t)^{-\alpha}$) in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films with ($n = 1.5$) and ($\alpha = 0.57$) for ($H \parallel ab$) and ($n = 1.4$) and ($\alpha = 0.62$) for ($H \parallel c$) [89]. These reports, among many others, suggests that the thermal activation energy in presence of applied field can be expressed as:

$$U(H, T) = U_0 H^\alpha (1-t)^\beta \quad (4.5)$$

Where (α) and (β) are field and temperature dependent [70], values of ($\alpha \sim 0.5$) and ($\beta \sim 1$) for several superconductors [52, 41]. In figure (4.6), we presented the fitting of (U_{eff}) and T at constant (H), we calculated values of (U_0) for (*TAFF*) regime at ($H=0T$) and ($H=1.5T$)

and the results were listed in table (4.3). The figure (4.6) shows that all data converges to ($T_c=14.5\text{ K}$), in agreement with our earlier findings. And values of (α) and (β) equal 0.486 and 0.938 respectively for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal which agree with previous reported results.

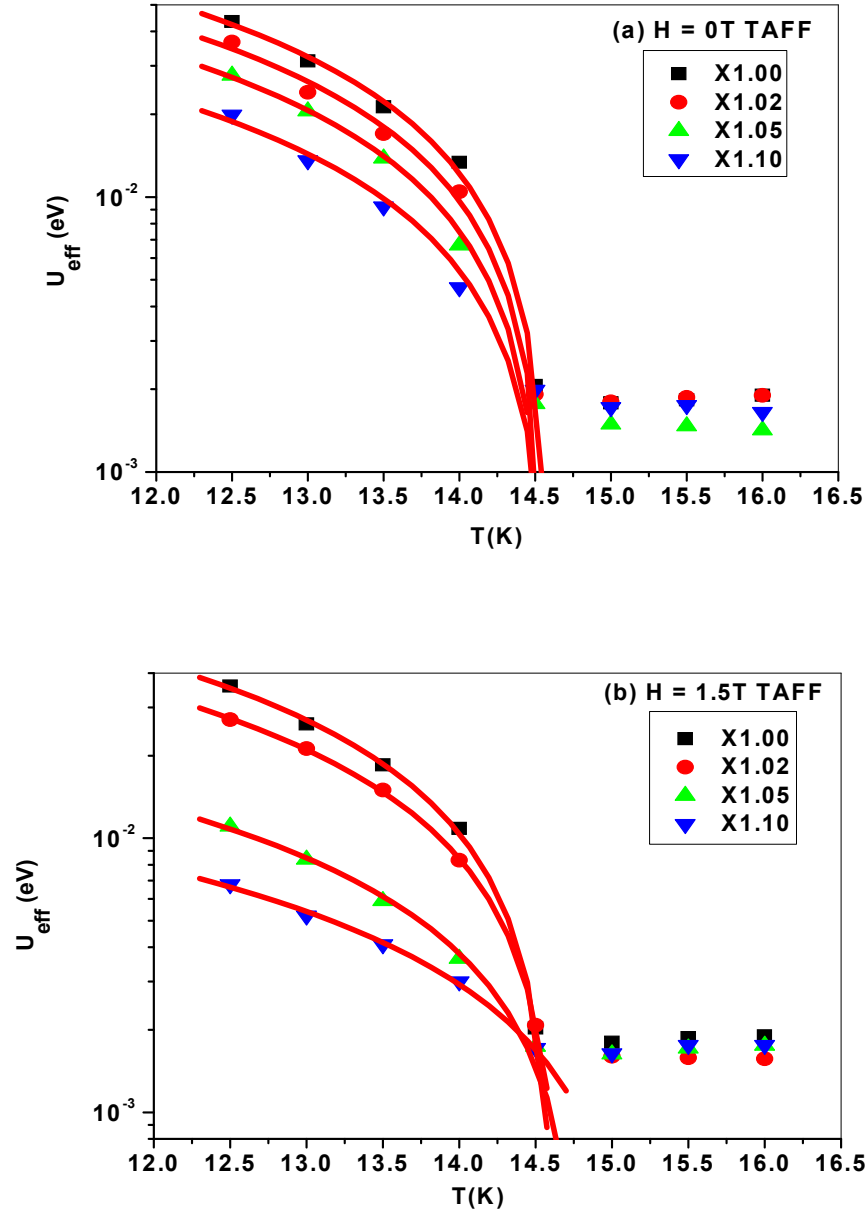


Figure (4.6): The log of effective pinning potential (U_{eff}) obtained are presented as variations with temperature for different iron content in (TAF) region in different applied magnetic field (a) at $H = 0\text{ T}$ and (b) at $H = 1.5\text{ T}$.

Table (4.3): The characteristic (TAFF) activation energy (U_o) for the different $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystal samples at $H = 0$ T and $H = 1.5$ T.

Sample	U_o (TAFF) (eV) at $H = 0$ T	U_o (TAFF) (eV) at $H = 1.5$ T
1.00	0.2943	0.2389
1.02	0.2420	0.1866
1.05	0.1915	0.0705
1.10	0.1307	0.0369

In figure (4.7) we plotted the activation energy values of table (4.3) with variation of iron content to study the relation between them. We found that the activation energy decreases with increasing iron content. Also, the (U_o) values are maximum for the optimum iron content when ($X=1.00$). The activation energy is found to be in the order of $100 k_B T$. Our result suggests more effective pinning centers in the samples with lower iron contents and the higher activation energy is desired for practical applications.

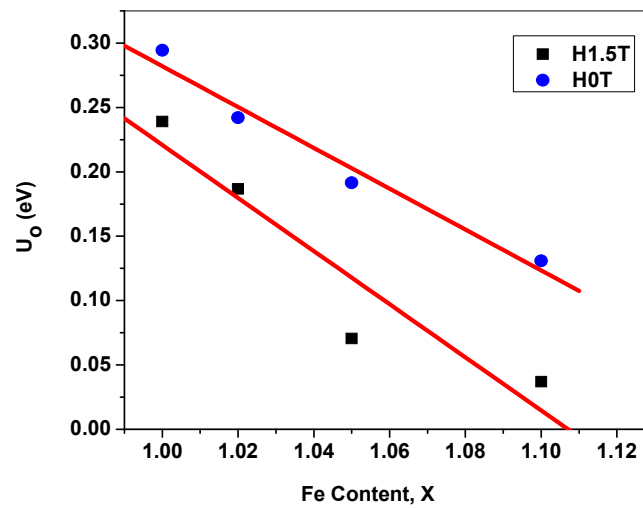


Figure (4.7) The characteristic pinning potential (U_o) obtained are presented as variations with different iron content in (TAFF) region in different applied magnetic field at $H = 0$ T and $H = 1.5$ T.

4.3 The Arrhenius Plots

To investigate the effect of variation of iron content on effective pinning potential U_{eff} of polycrystalline $Fe_xSe_{0.5}Te_{0.5}$ samples; we used Arrhenius plots of the resistivity in various applied magnetic fields. The resistivity values were measured for temperatures between 13.2 K and 15.4 K at different five applied magnetic fields in the range of (0 – 1) T for samples with different iron contents ($X = 0.95, 1.00, 1.05$ and 1.10). Figures (4.8) and (4.9) show the Arrhenius plot of $(\ln \rho)$ as a variation of $(1/T)$ for polycrystalline $FeSe_{0.5}Te_{0.5}$ and $Fe_{0.95}Se_{0.5}Te_{0.5}$ samples respectively.

Thermal activation energy values were calculated from slopes of Arrhenius plots and the results were plotted against applied magnetic field in figure (4.10). The figure (4.10) indicates higher effective pinning potential (U_{eff}) for optimum ($X=1.00$) and as iron content increases or decreases this will cause a reduction in effective pinning potential (U_{eff}). Furthermore, these values match with the activation energy at low field values for highly oriented $Fe_{1.03}Te_{0.55}Se_{0.45}$ superconductor with excess iron sample calculated by Junyie *et.al.* [71]. The smaller values of effective pinning potential at ($H = 1T$) compared to values at ($H = 0T$) which indicates the sensitivity of effective pinning to the applied magnetic field (as explained in figure 4.1), which make it easier for the vortices to move or jump over the potential barrier (42 - 110)

The calculated thermal activation energy values from Arrhenius plots are agreeing with the calculated values from ($I-V$) isotherms. Also, the (U_{eff}) values are sensitive to the criteria used for finding the critical temperature, such as, whether the critical temperature is measured when the resistivity drops to 0.1, 0.5 or 0.9 of its normal state resistivity (ρ_n) value. The activation energy values ($U(H, T)$) decrease quickly with the applied magnetic

field and they are showing the sensitivity of effective pinning to the applied magnetic field. We notice that (U) values are maximum for the optimum iron content at $(X=1.00)$ and reduce for the samples with higher/lower iron contents. The low values of $(U(T))$ about 10 meV indicate the low vortex-pinning behavior in the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductors.

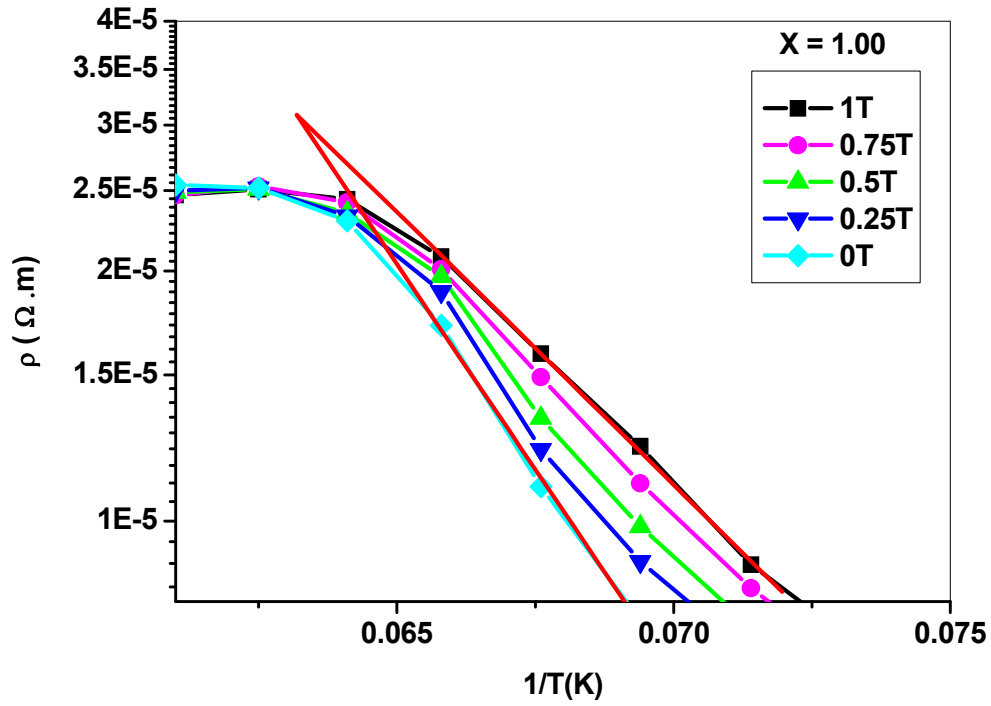


Figure (4.8): Arrhenius plot for the resistivity versus $1/T$ at different applied magnetic fields for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ polycrystalline sample. The two solid lines are a linear fit to the data.

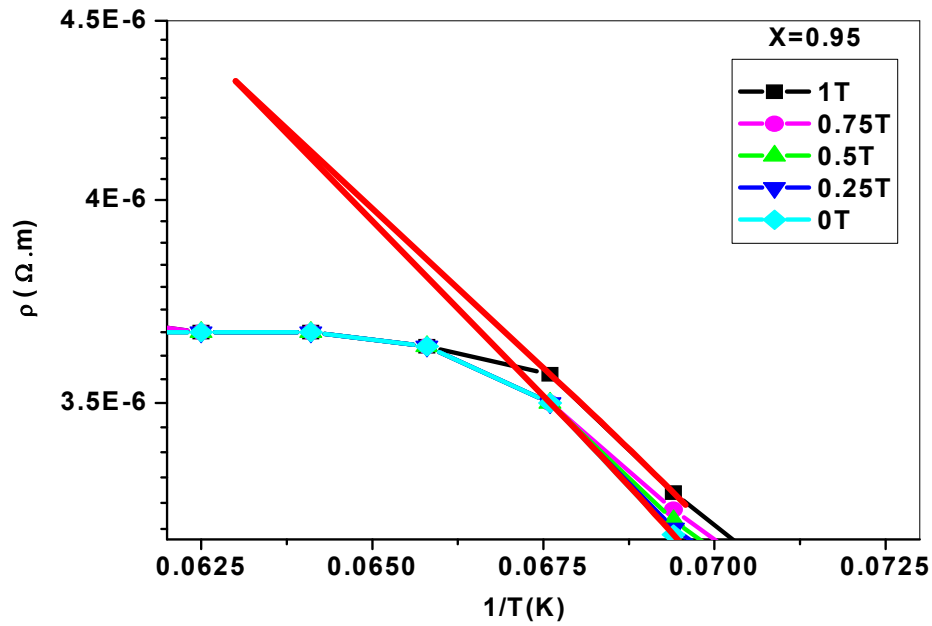


Figure (4.9): Arrhenius plot for the resistivity versus $1/T$ at different applied magnetic fields for $Fe_{0.95}Se_{0.5}Te_{0.5}$ polycrystalline sample. The two solid lines are a linear fit to the data.

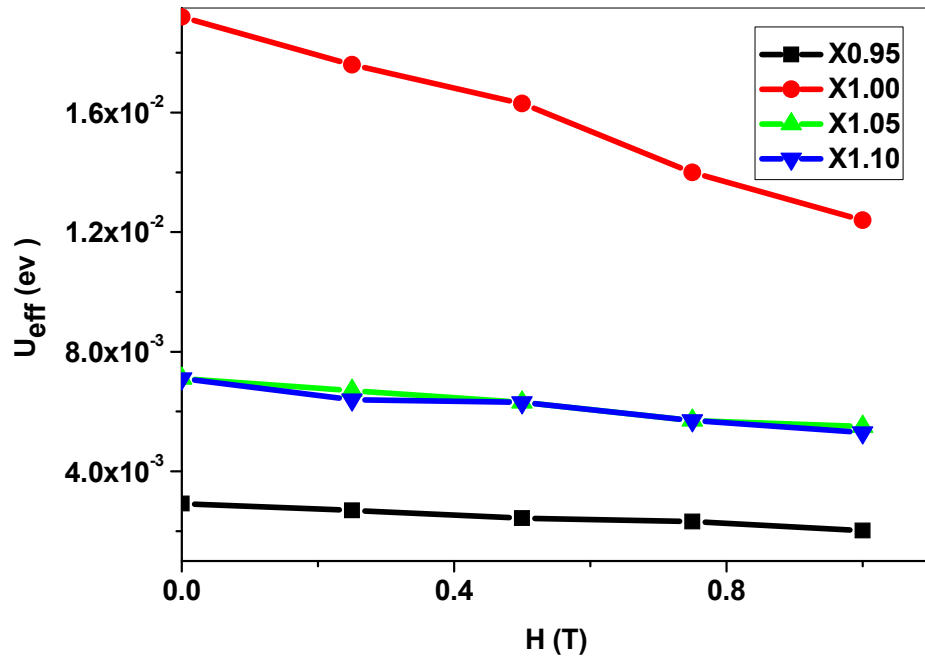


Figure (4.10): Thermal activation energy for the (TAFF) region (U_{eff}) as variation of applied magnetic field (H).

4.4 Upper Critical Magnetic Field

To study the effect of variation of Fe content on upper critical field, ($H_{c2}(T)$); we used Arrhenius plots as shown in figure (4.11) to estimate the phase transition at different magnetic fields of range of (0-1) T. We used three different criteria to identify the value of (T_c); namely, when the resistivity drops to 10%, 50% and 90% of its normal state resistivity (ρ_n) value. These criteria correspond to three different temperature in the flux flow region. The obtained ($H_{c2}(T)$) values reflect the effects of the flux flow on determining (H_{c2}). The (H_{c2}) values are presented in figure (4.12). The upper critical field at 0K, ($H_{c2}(0)$) has been estimated using three different models, namely, the empirical relation (ER), Ginzburg-Landau (GL) model and Werthamer-Helfand-Hohenberg (WHH) model, which are represented by the following equations respectively.

$$H_{c2}(T) = H_{c2}(0) [1-t^2] \quad (4.7)$$

$$H_{c2}(T) = H_{c2}(0) \left[\frac{1-t^2}{1+t^2} \right] \quad (4.8)$$

$$H_{c2}(0) = -0.693T_c \left(\frac{dH_{c2}}{dT} \right) \quad (4.9)$$

Where ($t = T/T_c$) is the reduced temperature [10, 11, 52 and 57]. The calculated values of ($H_{c2}(0)$) and the slope (dH_{c2}/dT) near (T_c) are listed in table (4.4).

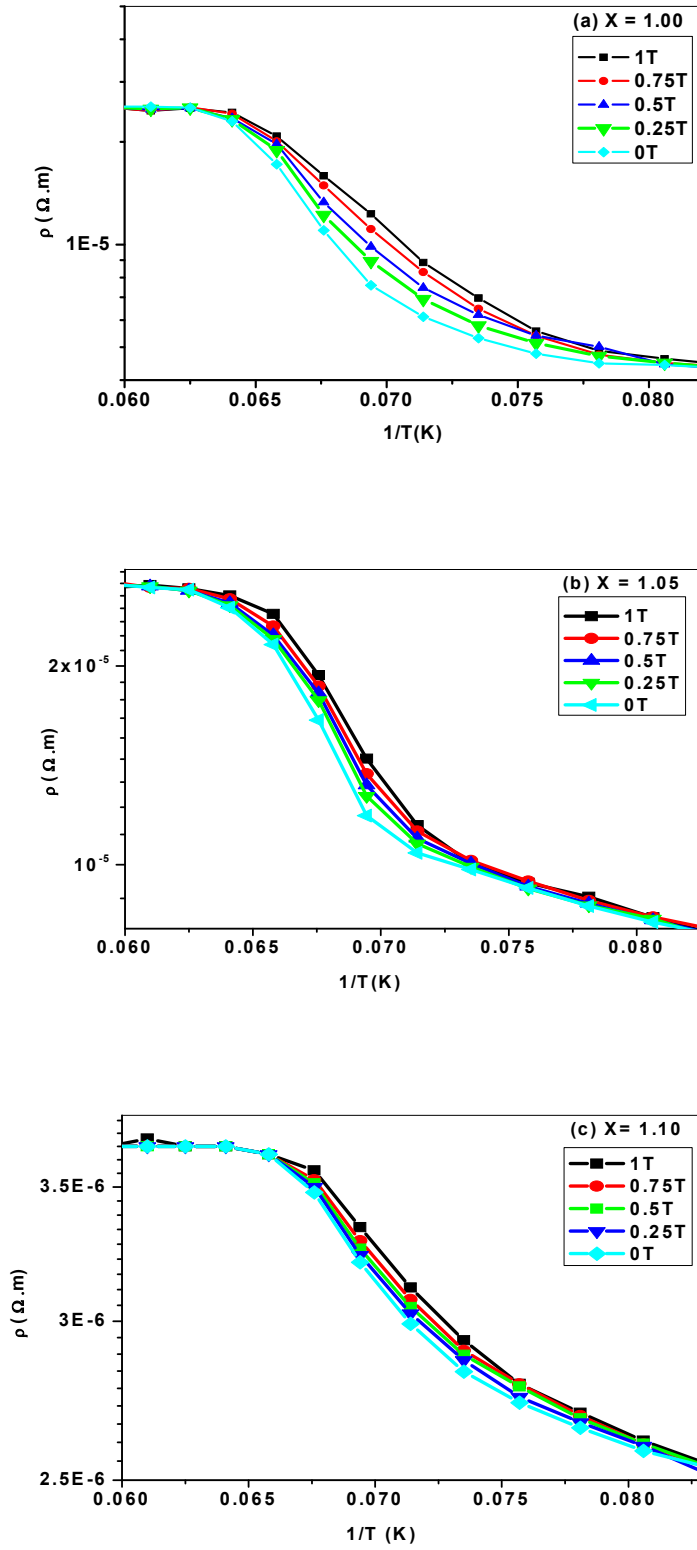


Figure (4.11): Arrhenius plot for the resistivity at different magnetic fields, (a) for $X=1.00$, (b) for $X= 1.05$ and (c) for $X= 1.10$.

Table (4.4): Calculated Upper critical field $H_{c2}(0)$ using the empirical relation and the slope near T_c for the different $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ polycrystalline samples.

Sample	10% ρ_n		50% ρ_n		90% ρ_n	
	$H_{c2}(0)$ (T)	$\frac{dH_{c2}}{dT}$ (T/K)	$H_{c2}(0)$ (T)	$\frac{dH_{c2}}{dT}$ (T/K)	$H_{c2}(0)$ (T)	$\frac{dH_{c2}}{dT}$ (T/K)
$\text{FeSe}_{0.5}\text{Te}_{0.5}$	13.4	-2.1	20.8	-2.8	43.7	-5.7
$\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$	27.8	-4.9	57.6	-8.0	63.7	-8.3
$\text{Fe}_{1.1}\text{Se}_{0.5}\text{Te}_{0.5}$	39.2	-8.3	29.5	-4.1	36.5	-14.4

Table (4.5): The calculated upper critical field $H_{c2}(0)$ for the different $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples by the GL and WHH models.

Sample	10% ρ_n (T)		50% ρ_n (T)		90% ρ_n (T)	
	GL	WHH	GL	WHH	GL	WHH
$\text{FeSe}_{0.5}\text{Te}_{0.5}$	25.8	19.8	40.5	28.4	86.3	60.1
$\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$	54.7	44.3	114.2	79.5	126.6	88.0
$\text{Fe}_{1.1}\text{Se}_{0.5}\text{Te}_{0.5}$	77.3	73.2	57.9	40.5	217.7	151.2

The data shown in Tables (4.4) and (4.5) reveals that the empirical relation gives the lowest ($H_{c2}(0)$) values followed by WHH then GL relations. The 10% drop criteria GL and WHH seems to yield ($H_{c2}(0)$) values in agreement with published data ~ 26 -77 T and 19-73 T respectively. Moreover; the 90% drop criteria in the normal resistivity gives the highest "unrealistic" estimates of the ($H_{c2}(0)$). The ($H_{c2}(0)$) values obtained from the empirical

relation are ~ 13.4 T for ($X = 1.0$) and showed a threefold (~ 39.2 T) increase for ($X = 1.1$) ($\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$) [78].

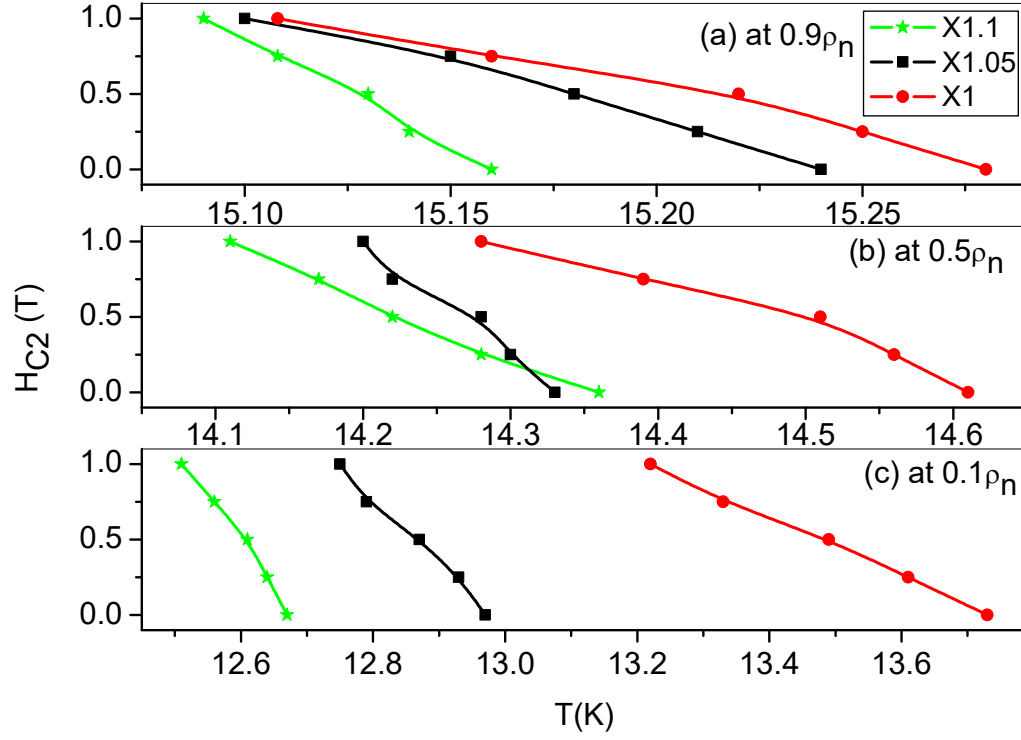


Figure (4.12): The variation of (H_{c2}) with temperature for $X=1.00$, 1.05 and 1.10 samples using (T_c) determined from different criterion of normal state resistivity (ρ_n): (a) $0.9 \rho_n$, (b) $0.5 \rho_n$ and (c) $0.1 \rho_n$ polycrystalline $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples.

CHAPTER 5

MAGNETIC PROPERTIES OF $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$

5.1 Hysteresis Loops

In this chapter we investigate the effects of iron contents on the magnetic properties of poly and single crystals $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$. In particular, we start by measuring the hysteresis loops and deduce several characterizing magnetic properties such as: the critical fields (H_{c1}) and (H_{c2}), full penetration magnetic field (H_p), irreversible magnetic field (H_{irr}) and the thermodynamic critical magnetic field (H_c). The remnant magnetization (M_r), which is related to the critical current at zero field and the critical current density (J_c), will also be determined from the width of the hysteresis loops at various temperatures. Finally, the pinning force (F_p) and its variations with the magnetic field and temperatures will be calculated based on Beans critical state model and the critical current density. Scaling behavior of the critical current density and pinning forces will also be discussed in this chapter [93].

The hysteresis loop at 4 K for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal is presented in figure (5.1). The initial negative magnetization drops linearly with applied field (in the Meissner state). Out of several methods that can be used to determine H_{c1} , we will use the initial value of the applied field at which the magnetization deviate from the linear slope ($-1/4\pi$). As the applied magnetic field increases above (H_{c1}), beyond the Meissner shielding, the applied magnetic field starts penetrating the superconducting sample, and consequently the magnetization gradually increase (i.e., $|M|$ decreases) reaching zero at the upper critical field (H_{c2}). The inflection point in the lower part of the magnetization curve is considered to be the full penetration field (H_p) (see the inset to

Fig. 5.1). The hysteresis completely disappears ($\Delta M = 0$) at the irreversibility field (H_{irr}), and magnetization becomes reversible, and gradually reaching zero at (H_{c2}). What we obtain from hysteresis loops are summarized as:

- 1- The area of hysteresis loop which represents the energy losses in the sample.
- 2- The remnant magnetization (M_r), which gives an idea about trapped flux, where (M_r) is the value of magnetization at a field equal to zero, ($H = 0$).
- 3- The critical current (J_c), which is the width of hysteresis loop, ($J_c \propto \Delta M$).
- 4- The irreversibility field (H_{irr}) at ($\Delta M = 0$).
- 5- The pinning forces (F_p) which are equal to ($J_c * H$).

The irreversible magnetization is commonly described by Bean's model [96]. Bean's model relates the width of the magnetization in the irreversible region of the hysteresis loop at a given field to the critical current [97].

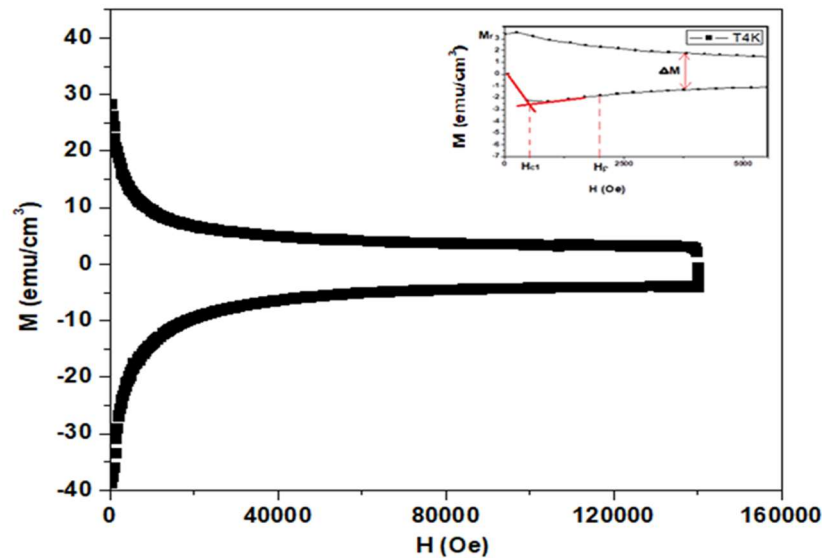


Figure (5.1): Hysteresis loop at 4K, inset: the determining of (M_r , H_{c1} , H_p and the width of the hysteresis loop).

5.2 Shape and Behavior of Hysteresis Loop

The shape of the hysteresis loop and the associated parameters can be affected by various factors as sample preparation and heat treatments, temperature, maximum applied magnetic field and iron content. In this section we want to investigate some of these factors and the corresponding effects on the hysteresis loops.

5.2.1 Effect of the Temperature on Hysteresis Loop

Figure (5.2) shows hysteresis loops for the single crystal samples of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ at different temperatures namely, 4K, 6K, 8K, 9K, 10K, 11K and 12K. It should be worth mentioning here that these all temperatures are below the critical temperature (T_c) and the loops are measured in (a maximum cycling magnetic field up to 14T when it parallel to (c) axis direction). From this figure, we notice that the hysteresis width gets narrower as the temperature increase, so by increasing temperature causes reduction in the effective activation energy which makes it easy for vortices to move around and this reduce the magnetization. Moreover, as the temperature increase the diameter of vortices (normal cores) increase on the expense of the superconducting volume, in other words the volume fraction of the superconducting material is reduced and hence reduces the magnetization.

The magnetization measurements were carried out for the samples with ($X = 1.00, 1.02$ and 1.05) at different temperatures below the (T_c), ranging from 4K to 12K. The typical hysteresis loops resulting from (M) versus (H) measurements are shown in figures (5.3a), (5.3b) and (5.3c) for the samples with different iron content; ($X=1.00, 1.02$ and 1.05) respectively.

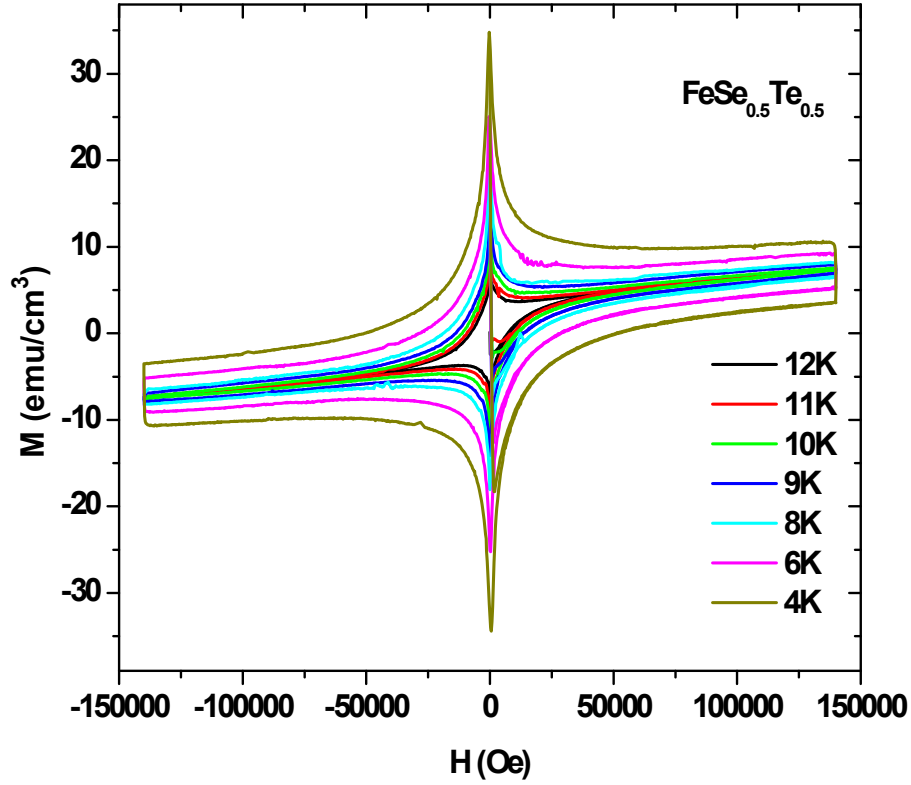


Figure (5.2): Hysteresis loops of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ at different temperatures.

5.2.2 Effect of the Iron Content on Hysteresis Loop

Figures (5.4a) and (5.4b) show hysteresis loops for the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples with iron content, ($X=1.00, 1.02$ and 1.05), at temperatures 4K and 6K , respectively. These figures show effect of iron content on hysteresis loops at constant temperature. From these figures, we notice that the hysteresis width at constant temperature gets narrower as the iron content increase in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples. This indicates reduction in superconductivity volume fraction of the material.

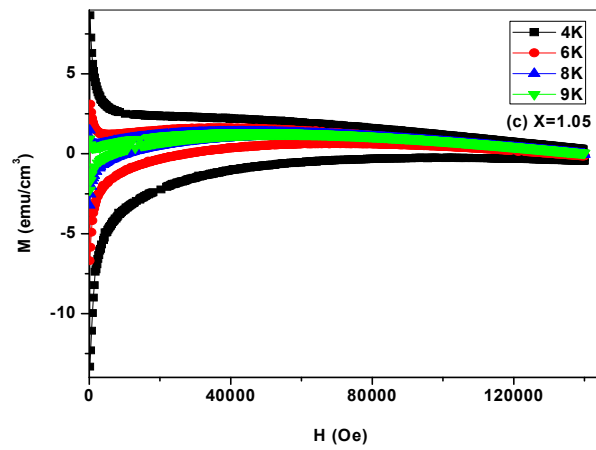
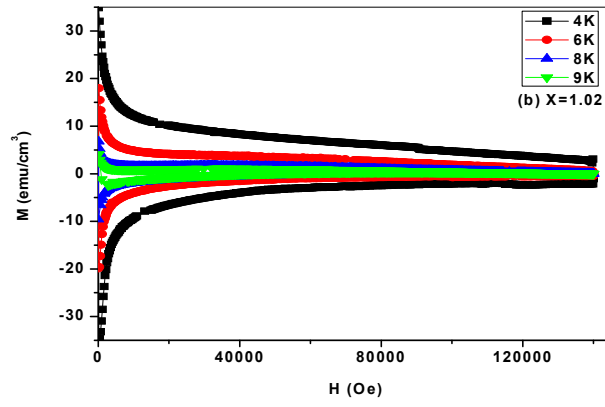
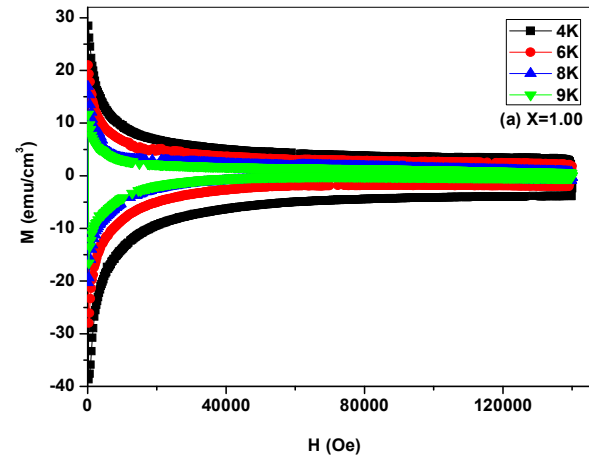


Figure (5.3): Hysteresis loops of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at different temperatures (4K, 6K, 8K and 9K) for (a) $X=1$, (b) $X=1.02$ and (c) $X=1.05$.

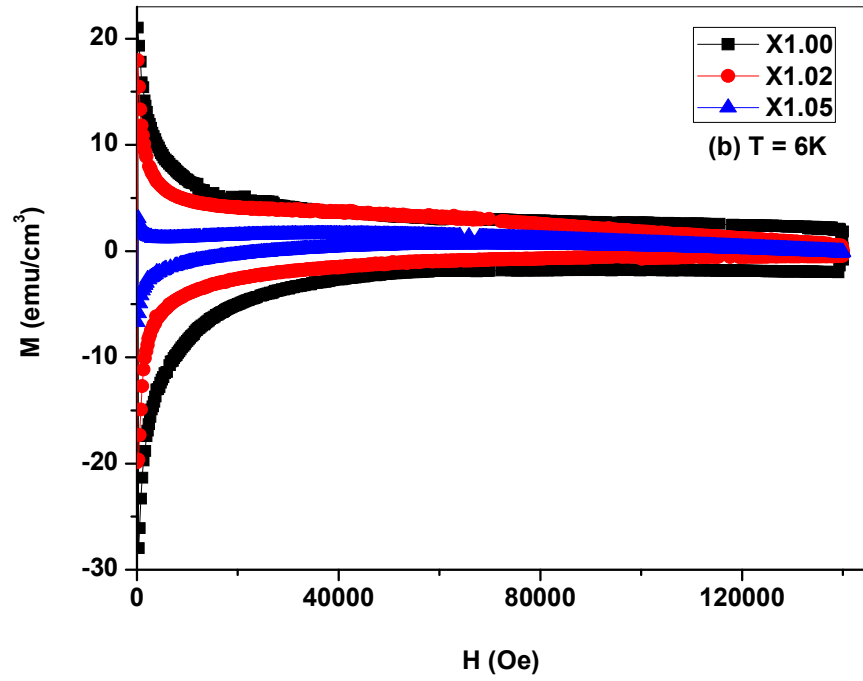
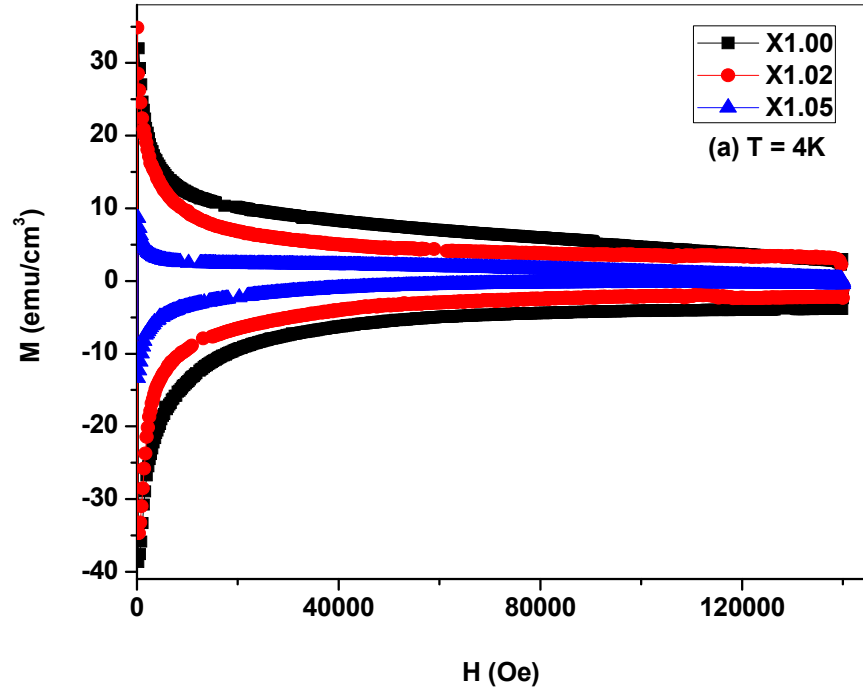


Figure (5.4): Hysteresis loops of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at (a) $T = 4\text{K}$ and (b) $T = 6\text{K}$.

5.3 Irreversible Properties

The irreversible superconducting properties are derived from the hysteresis loops of the magnetization [43]. In type II-superconductors, the flux lines do not move freely because they are pinned in their position by local pinning centers. The local pinning potential is non-uniform, and the vortices are taken away of their pinning centers in the irreversible part of hysteresis loops when applied magnetic field is less than irreversible applied magnetic field. Inside the vortices, the material is in normal state, while outside the vortices the material is in superconducting state with different free energy [95].

5.3.1 Bean's Model "The Critical State Model"

The measured width ΔM of the hysteresis loops is related to the critical current ($J_c(H, T)$) through Bean's "critical" state model. It can be used to explain many of the properties of type-II superconductors. The model was independently introduced by Bean (1962) and London (1963) and is usually referred to as Bean's Model. The model assumes that the critical current is flowing in the sample to prevent the penetration of flux into the sample when the magnetic field is increased above (H_{c1}) value. It describes the variations of magnetization within a superconducting sample. Bean based his model on some basic assumptions and it explains very well the mixed state of type II superconductors [97]. The assumptions incorporated in the model are as follows [96, 92].

- 1- There is no reversible magnetization; ($H_{c1} = 0T$). Only the critical state is considered.
- 2- There is a critical current density which can be carried by the superconductor.
- 3- Even very low electromagnetic forces induce the full critical current density.

4- The critical current density is independent of the external magnetic field.

5- In any region of a superconductor, where magnetic flux has penetrated, the critical current density is perpendicular to the magnetic induction (B), whereas everywhere else is valid ($J_c = 0$).

The sample is perfect diamagnetic if applied magnetic field is below (H_{c1}) because the slope of this section of hysteresis loop is negative. When the applied magnetic field is higher than the lower critical field value (H_{c1}), the magnetic flux start entering the sample uniformly, and as a result the magnetization start decreasing. The magnetization will continue decreasing as long as the field is increasing, until it reaches the upper critical field value (H_{c2}) of the magnetic field. At (H_{c2}), there is no magnetization ($M = 0$). The sample is completely in normal state. If the field is increased above (H_{c2}), ($\Delta M = 0$) the magnetization curve coincides with its earlier values in the reverse direction and there will be no hysteresis. For the samples with flat geometry and the field perpendicular to its plane, it can be shown that the critical current density at any field value is directly related to the width of the hysteresis loop by the following expression:

$$J_c = \frac{30\Delta M}{< d >} \quad (5.1)$$

Where ($\Delta M = M^+ - M^-$) (emu/cm³) is the height of hysteresis loop and ($< d >$) by (cm) is the average diameter of pellet which is used in magnetization measurement.

This model usually gives results that are comparable to the results that obtained by the direct transport method. Note that these measurements can also be affected by the flux creep since the magnetization can be time dependent.

In this section, we will present the critical current density estimated from the magnetization hysteresis loops using Bean's model by applying the equation (5.1). The results are shown in Figure (5.5) with its parts a, b, c and d for samples with different iron contents; ($X=1.00$, 1.02, 1.05 and 1.10), respectively at various temperatures. These figures reveal that the critical current density starts with large values at low magnetic field and decreases as the field increases until it reaches zero at (H_{irr}). Furthermore, as the temperature increases, (J_c) decreases until it becomes zero at (T_c). Figure (5.6), shows the relation of critical current density and magnetic field at 4 K for samples with different iron content.

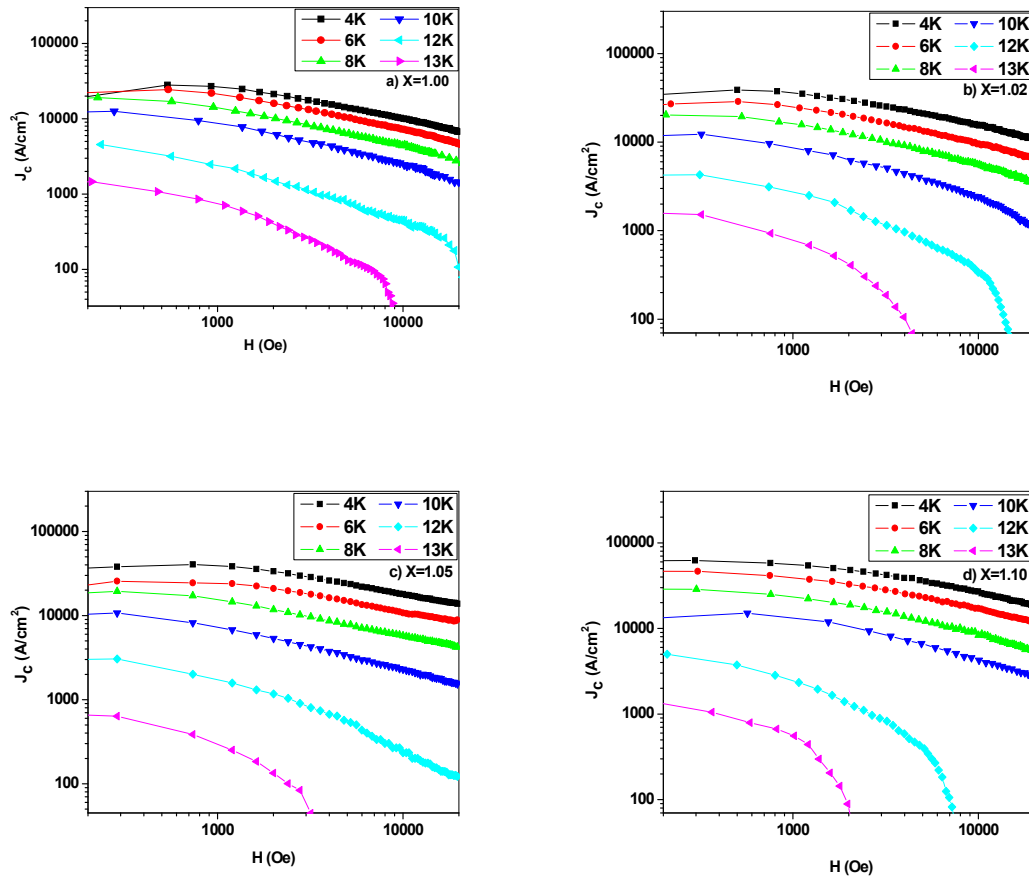


Figure (5.5): $J_c(H)$ curves at different temperatures for different iron content on log-log scale in (a) $X=1.00$, (b) $X=1.02$, (c) $X=1.05$ and (d) $X=1.10$.

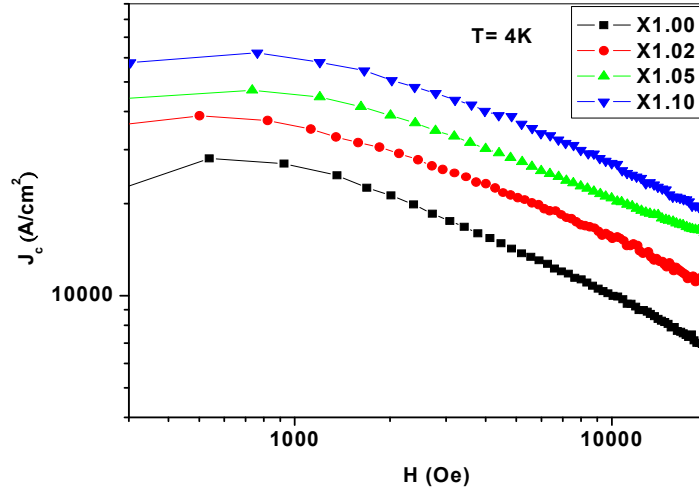


Figure (5.6): $J_c(H)$ curves for different iron content at $T = 4K$ on log-log scale.

The figure shows the sample with highest iron content ($X=1.10$) has the maximum critical current density. In another words increasing iron content ($1.00 \leq X \leq 1.10$) increases critical current density, which further indicates an increase in the pinning is associated with increasing iron content.

5.3.2 Pinning Force

Flux pinning is the phenomenon in which magnetic flux lines do not move (become trapped or pinned). Flux pinning is only possible when there are defects in the crystalline structure of the superconductor (usually resulting from grain boundaries or impurities) [21]. The pinning force (P_f) can be calculated when the applied field (H/c) directions using the data presented in figure (5.5) with the following relation.

$$P_f = J_c * H \quad (5.2)$$

Typical pinning force figures show that pinning force increases as the field increases until it reaches a maximum value (P_f^{max}). After that the force starts decreasing until it reaches

zero at (H_{irr}). Moreover, the pinning force decreases as the temperatures increases until it reaches zero at (T_c). The results for optimum single crystal with ($X=1.00$) are presented in figure (5.7), which shows the variation of pinning forces against the applied magnetic field up to 14T at different temperatures from 4K to 12K. These curves for $\text{FeTe}_{0.5}\text{Se}_{0.5}$ show second magnetization peak, which is to some extent overlapped with the first peak. Therefore, we cannot determine the first peak clearly to study the pinning mechanism at temperatures lower than 10K.

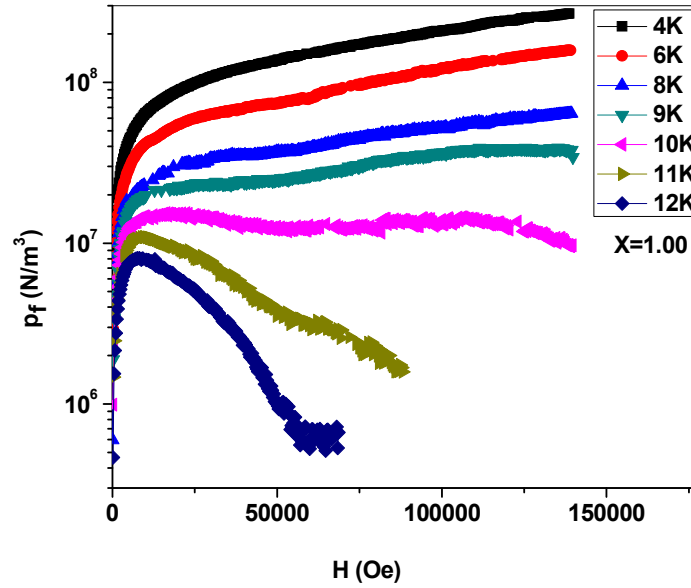


Figure (5.7): Pinning force of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ at different temperatures.

The pinning force plots for other samples with excess iron content ($X = 1.00, 1.02, 1.05$ and 1.10) are shown in figures from (5.8-a) to (5.8-d) respectively. At various temperatures, below critical transition temperature (T_c), pinning forces were plotted against the variation of applied magnetic fields up to 8.8 T. In all curves the pinning force shows a clear maximum at all temperatures ($10 < T < 13\text{K}$).

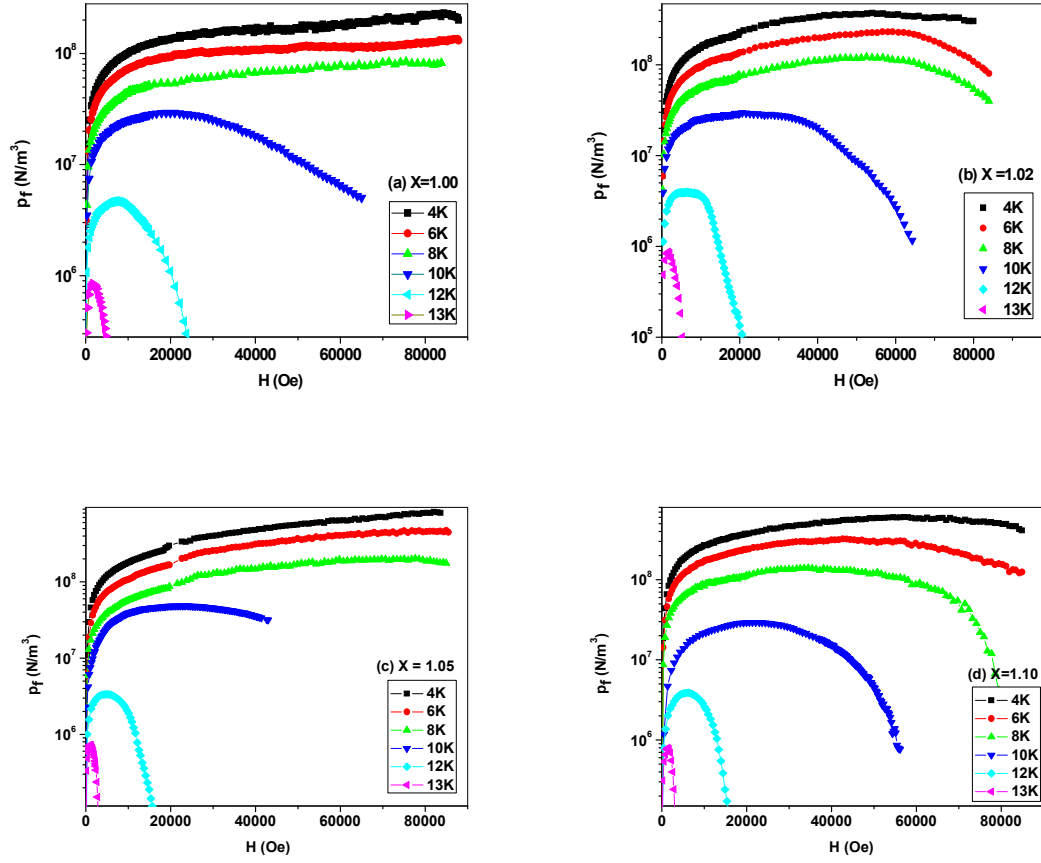


Figure (5.8): Log of pinning force of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at different temperatures for (a) $X=1.00$, (b) $X=1.02$, (c) $X=1.05$ and (d) $X=1.10$.

At a given temperature, the pinning force initially increases linearly, then gradually increases at high magnetic field with smaller (saturation) slope than the initial slope. Measurements were performed at different temperatures. The general behavior of the pinning force P_f is that; it decreases with increasing temperature. At temperatures close to ($T_c \sim 14.5\text{K}$) as shown in the figure 5.8, the width of the loop goes to zero at irreversible field, where the pinning force effectively goes to zero. Also, we can plot the variation of pinning force against the applied magnetic field for different iron contents. The plot at temperature 4K, 6K, 12K and 13K are shown in in figures (5.9), (5.10), (5.11) and (5.12) respectively. We found that the pinning force at low temperatures, namely 4K and 6K,

increases with increasing the iron content. Whereas at temperatures close to critical transition temperatures, namely 12K and 13K, the pinning force decreases by increasing the iron content.

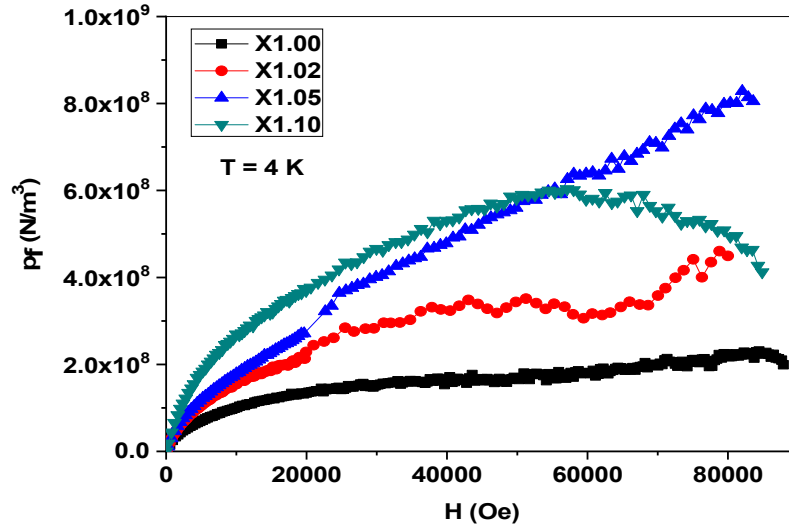


Figure (5.9): Pinning force of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at 4K temperature.

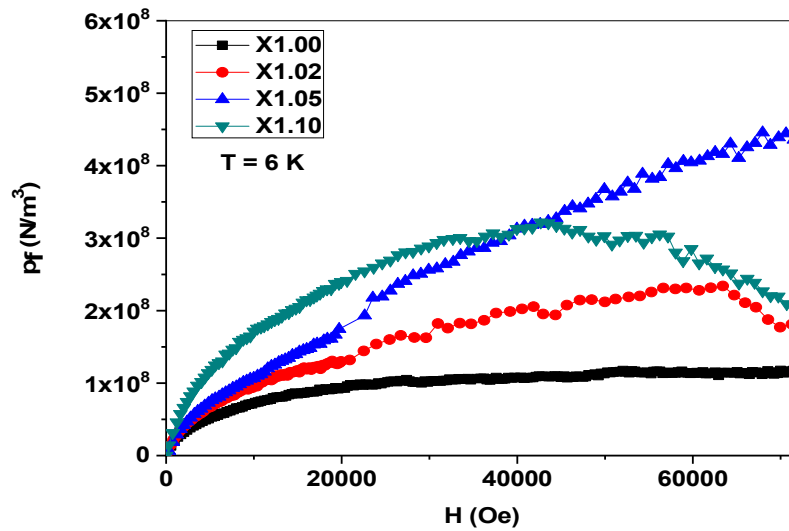


Figure (5.10): Pinning force of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at 6K temperature.

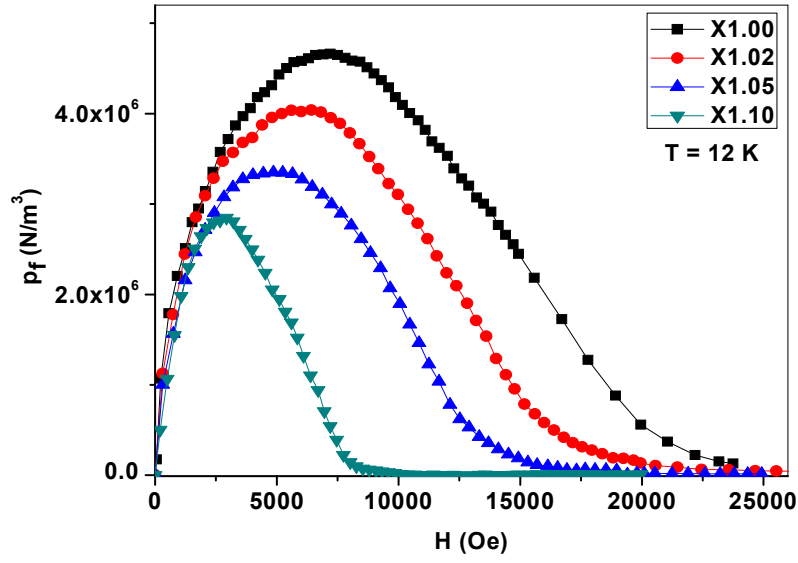


Figure (5.11): Pinning force of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at 12K temperature.

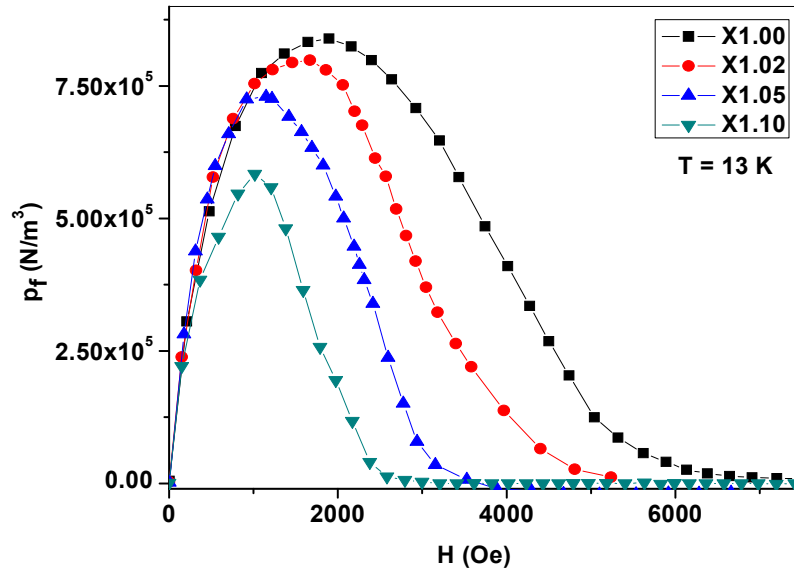


Figure (5.12): Pinning force of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at 13K temperature.

The maximum values of pinning force at different temperature for different iron contents are summarized in the table (5.1) and are plotted in figure (5.13). The (P_f) versus (T) curve infer to two distinct behaviors separated by the temperature of $\sim 10\text{K}$. This figure reveals

that the maximum pinning force increases as the iron content increases at same temperature. Moreover, the maximum pinning force decreases as the temperatures increases until it reaches zero at (T_c) at all iron contents. This indicates increasing iron content (X) in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ compounds cause more defects and new vacancies that probable can change the pinning mechanism.

Table (5.1): Maximum pinning force at different temperature for all samples.

T (K)	$P_f^{max}(X=1.00)$ (N/m ³)	$P_f^{max}(X=1.02)$ (N/m ³)	$P_f^{max}(X=1.05)$ (N/m ³)	$P_f^{max}(X=1.10)$ (N/m ³)
4	1.698×10^8	3.712×10^8	7.995×10^8	6.037×10^8
6	1.125×10^8	2.324×10^8	4.617×10^8	3.215×10^8
8	6.657×10^7	1.219×10^8	2.032×10^8	1.411×10^8
10	2.781×10^7	2.950×10^7	4.810×10^7	6.960×10^7
12	4.651×10^6	3.957×10^6	3.333×10^6	2.848×10^6
13	1.093×10^6	8.655×10^5	7.216×10^5	5.813×10^5

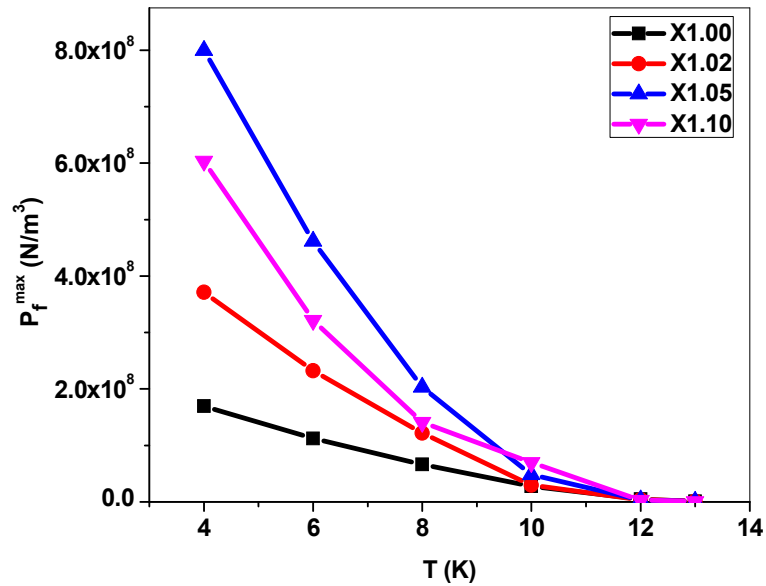


Figure (5.13): Variation of maximum pinning force of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at different temperatures.

5.3.3 Remnant Magnetization

Remnant magnetization is the residual magnetization in a sample after removing the applied external magnetic field. The remnant magnetization value can be determined from a hysteresis loop when ($H = 0$). Type II superconductors contains pinning centers due to defects and impurities in crystalline structure. These centers introduce a domestic medium at which flux lines are pinned in a relatively steady configuration. These trapped flux lines cause the remnant magnetization in zero applied magnetic field and hence, the remnant magnetization is a measure of the density of trapped flux inside the sample, which is proportional to the density of pinning centers [90].

In figure (5.14), we present the remnant magnetization for all samples at different temperatures. There is a little enhancement of remnant magnetization with increasing iron content at low temperatures, whereas the remnant magnetization decreases with increasing iron content at high temperatures.

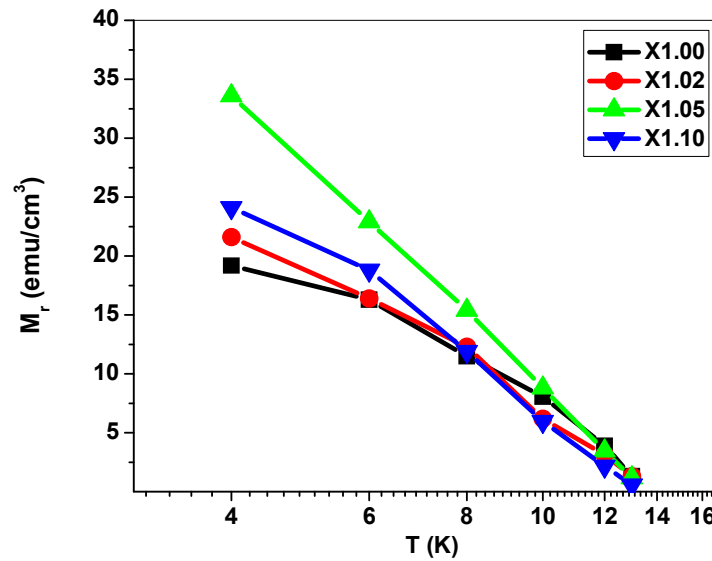


Figure (5.14): Remnant magnetization versus temperature for all samples.

5.4 Scaling of Pinning Force

5.4.1 Review of Scaling

Scaling pinning force is essential for two purposes. First, it can be used to understand behavior of the scaled pinning force to predict its behavior and the change in it. Second, it can be used to predict an isotropy of pinning. There are different models to scale the pinning force. Five of them uses two scaling parameters and the one of them uses one scaling parameter. The scaling of pinning force for different models are shown in table (5.2)

Table (5.2): Different pinning force scaling models.

No.	Scaling Pinning Force	Reduced magnetic Field
1	$f = P_f / P_f^{\max}$	$h = H / (H_{c2})^{0.25}$
2	$f = P_f / P_f^{\max}$	$h = H / H_{c2}$
3	$f = P_f / P_f^{\max}$	$h = H / H_{irr}$
4	$f = P_f / P_f^{\max}$	$h = H / H^*$
5	$f = P_f / P_f^{\max}$	$h = H / H_n$
6	$f = P_f / H_c^2$	$h = H / H_c$

Scaling of (P_f) versus H using $(P_f^{\max})$ and $((H_{c2})^{2.5})$ was first introduced by Fietz and Webb in 1967 for dilute Nb-based alloys [98]. Another scaling of (P_f) versus (H) using $(P_f^{\max})$ and (H_{c2}) was introduced by Kramer in 1972 [45]. The difficulty in these two-previous scaling is the magnitude of (H_{c2}) itself, which is very large for our optimum sample about 150T [29]. So other scaling models were appeared by scaling of (P_f) on $(P_f^{\max})$ and scaling of (H) by $(H_{irr}, H^*$ and $H_n)$. Where (H_{irr}) is the magnetic field at which the pinning force vanishes, (H^*) is the magnetic field at which the pinning force reaches its maximum value and (H_n) is the magnetic field at which the pinning force reaches its half maximum value.

Also, one of scaling model is the scaling of both (P_f) and (H) with respect to thermodynamic critical field (H_c). A scaling law of flux pinning of Fe-based superconductors can be introduced as a function of the irreversible reduce field ($h = H/H_{irr}$) and ($f = P_f/P_f^{max}$) [21] is given by,

$$f = m \times h^p \times (1 - h)^q.$$

Here, (m) is numerical parameters and exponents (p and q) are describing the pinning mechanism as shown in table (5.3).

Table (5.3): Different pinning mechanism.

No.	Function	Pinning Mechanism	h_{max}
1	$f_p = Ah^{0.5}(1-h)^1$	For magnetic interaction with normal volume pins.	0.333
2	$f_p = Ah^{0.5}(1-2h)$	For magnetic interaction leading to $\Delta\kappa$ pinning.	-----
3	$f_p = Ah^0(1-h)^1$	For core interaction with normal volume pins.	0
4	$f_p = Ah^1(1-h)^1$	For core interaction leading to $\Delta\kappa$ pinning by volume pins.	0.5
5	$f_p = Ah^{0.5}(1-h)^2$	For core interaction with normal surface pins.	0.2
6	$f_p = Ah^{1.5}(1-h)^1$	For core interaction leading to $\Delta\kappa$ pinning by surface pins.	0.6
7	$f_p = Ah^1(1-h)^2$	For core interaction with normal point pins.	0.333
8	$f_p = Ah^2(1-h)^1$	For core interaction leading to $\Delta\kappa$ pinning by point pins.	0.667
9	$f_p = Ah^2 \exp(-2h^{1.5})$	For collective pinning.	-----

The most famous pinning fitting models were introduced by Kramer and Dew-Hughes. Kramer model considers surface pinning with ($p \approx 0.5$, $q \approx 2$ and $h_{max} \approx 0.2$) [45]. While Dew-Hughes model considers surface pinning with ($p \approx 1$, $q \approx 2$ and $h_{max} \approx 0.333$) [44]. This discrepancy is one of the major problems for a consistent understanding of scaling laws for flux pinning.

5.4.2 Kramer Plots

Kramer curve is the name given to the plot of ($J_c^{0.5} H^{0.25}$) versus (H) which used to determine value of H_{irr} at given temperature [45]. The Kramer plots of all samples are shown in figures from figure (5.15) to figure (5.18).

The Kramer plots are approximately parallel to each other for temperatures less than 10K. This is an indication that they are having the same pinning mechanism at all temperatures. Also, we can see deviation of plots from linear behavior due to second peak magnetization (SPM) at higher fields. This effect diminishes at temperatures close to the transition critical temperature ($T_c \sim 14K$).

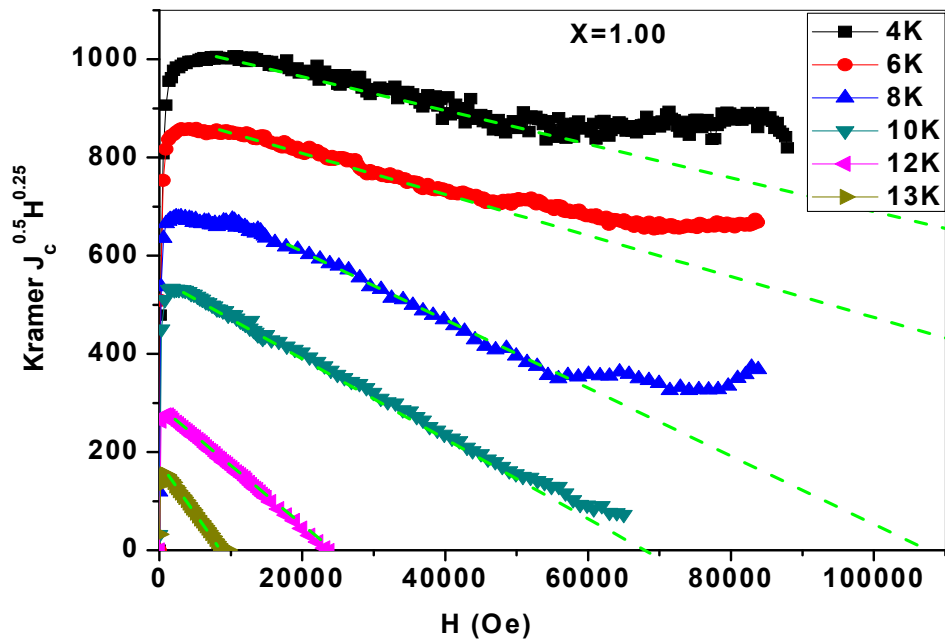


Figure (5.15): Kramer plots for the optimum $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample at various temperatures. The dashed lines indicate the Hirr values were determined certain temperatures.

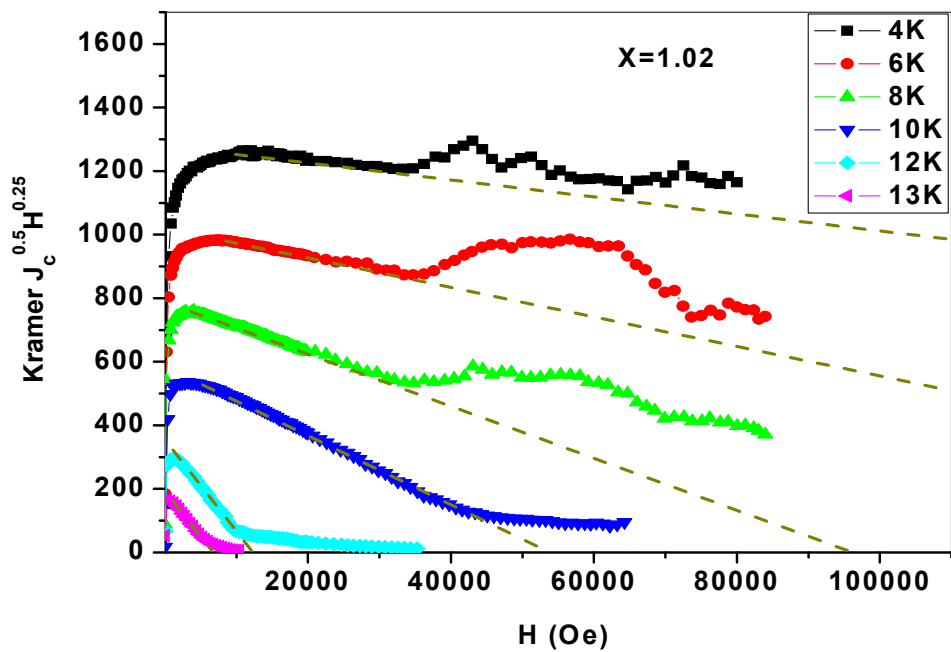


Figure (5.16): Kramer plots for the $\text{Fe}_{1.02}\text{Se}_{0.5}\text{Te}_{0.5}$ sample at various temperatures. The dashed lines indicate the Hirr values were determined certain temperatures.

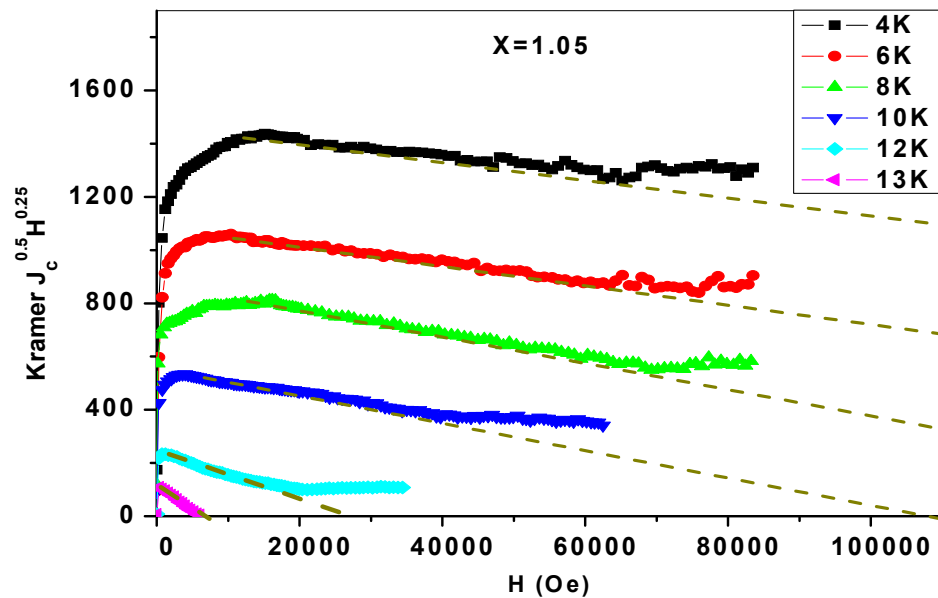


Figure (5.17): Kramer plots for the $\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$ sample at various temperatures. The dashed lines indicate the Hirr values were determined certain temperatures.

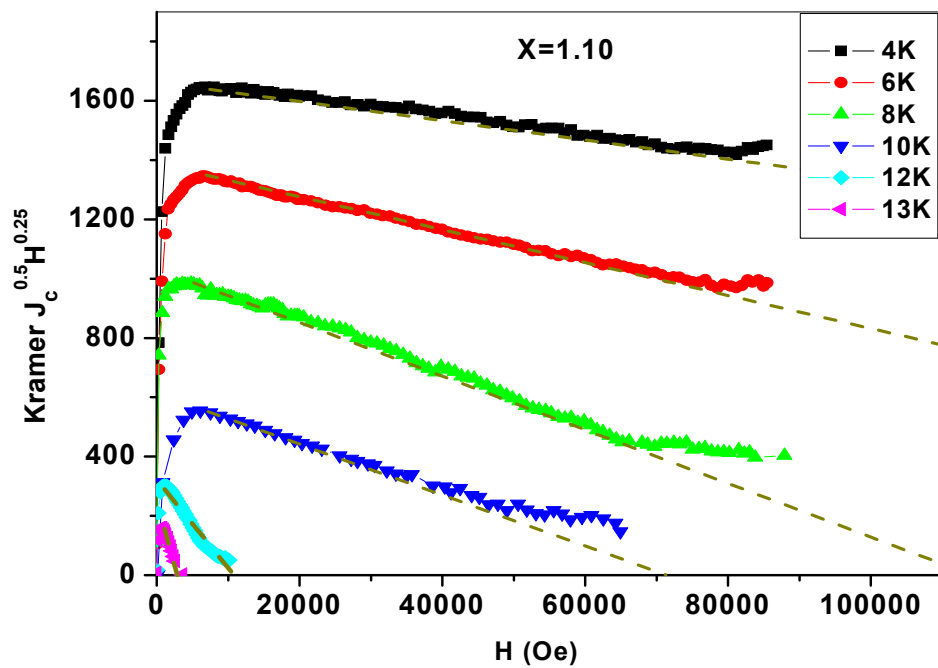


Figure (5.18): Kramer plots for the $\text{Fe}_{1.1}\text{Se}_{0.5}\text{Te}_{0.5}$ sample at various temperatures. The dashed lines indicate the Hirr values were determined certain temperatures.

5.4.3 Scaling Pinning Force Using H_{irr}

To understand the pinning mechanism in type II superconductors we need to understand the vortices and pinning centers. There are two main interactions between vortices and pinning centers, the magnetic interaction and the core interaction [46], which are summarized in table (5.3). The magnetic interaction is due to the interaction at neighboring interfaces between superconducting and non-superconducting materials that are parallel to the applied magnetic field. The core interaction is related to the pinning caused by the variations in transition temperature (δT_c) and the variation in charge carrier near lattice defects (δl) [46].

The reduced pinning force ($f_p = P_f / P_f^{max}$) versus the reduced field ($h_{irr} = H / H_{irr}$) [51] has been used to investigate the pinning mechanism in type II superconductors and its behavior at different temperature. The results are shown in figures (5.19) and (5.20) for three different temperatures 10K, 12K and 13K. We found that the curves overlap with each other. This indicates that over a wide range of temperatures the pinning force follows a scaling relation and this relation can be found by suitable fitting. If the temperature is changed, the maximum value of the pinning force takes place at the same value of the reduced field regardless of temperature. In table (5.4), we reported the values of the scaling components (p , q and h_{max}), which may explain the possible pinning mechanism.

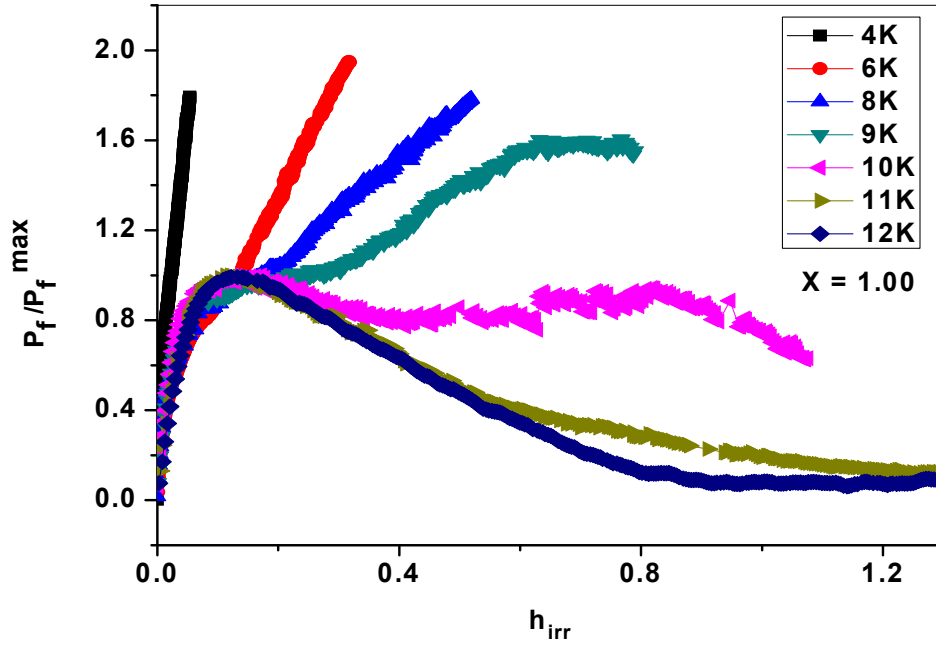


Figure (5.19): Reduced Pf curves as a function of reduced field for the $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample at various temperatures.

Table (5.4): Value of scaling components (p and q) and ($h_{\max}$).

Sample	p	q	$h_{\max}$	R^2
$\text{FeSe}_{0.5}\text{Te}_{0.5}$	0.641	1.502	0.299	0.991
$\text{Fe}_{1.02}\text{Se}_{0.5}\text{Te}_{0.5}$	0.545	1.194	0.313	0.986
$\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$	0.617	1.141	0.351	0.984
$\text{Fe}_{1.10}\text{Se}_{0.5}\text{Te}_{0.5}$	0.946	1.648	0.365	0.982

As we see in figure (5.19), for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample at low temperatures and high magnetic fields, fluctuations and second peak magnetization (SPM) cause some deviations from the universal behavior. This causes difficulties and makes complication in vortices and hence causes difficulties in pinning mechanism. Therefore, in figure (5.20) we will concentrate

in pinning mechanism at temperatures in the vicinity of the critical temperature (T_c). Excess of iron causes more defects and vacancies, which will affect the pinning mechanism.

According to Das *et al.*, the dominant pinning mechanism for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ samples is (δl) pinning, which is associated to small bundle of vortex pinning caused by randomly distributed weak pinning centers [66]. While pinning mechanism in $\text{FeSe}_{0.4}\text{Te}_{0.4}$ compound has been investigated by Yadav *et al.* and they found that the ($h_{max} = 0.28$) (where h_{max} is the reduced field at maximum pinning force, that normalized with respect to the H_{irr}). The value of (h_{max}) can be explained in terms of (δl) pinning with a combination of surface pinning ($h_{max} = 0.2$) and the point core pinning due to normal pinning centers ($h_{max} = 0.33$), with different ranges of pinning interactions [65].

Our results for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ and $\text{Fe}_{1.02}\text{Se}_{0.5}\text{Te}_{0.5}$ are agree with published results that pinning mechanism is (δl) pinning because the value of (h_{max}) lies in range ($0.2 < h_{max} < 0.33$). Whereas the pinning mechanism for $\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$ and $\text{Fe}_{1.1}\text{Se}_{0.5}\text{Te}_{0.5}$ is the mixture of point core pinning due to normal pinning centers ($h_{max} = 0.33$) and core interaction leading to $(\Delta\kappa)$ pinning by volume pins. Increasing iron content (X) in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ compounds adds more defects and new vacancies, that probably changes the pinning mechanism.

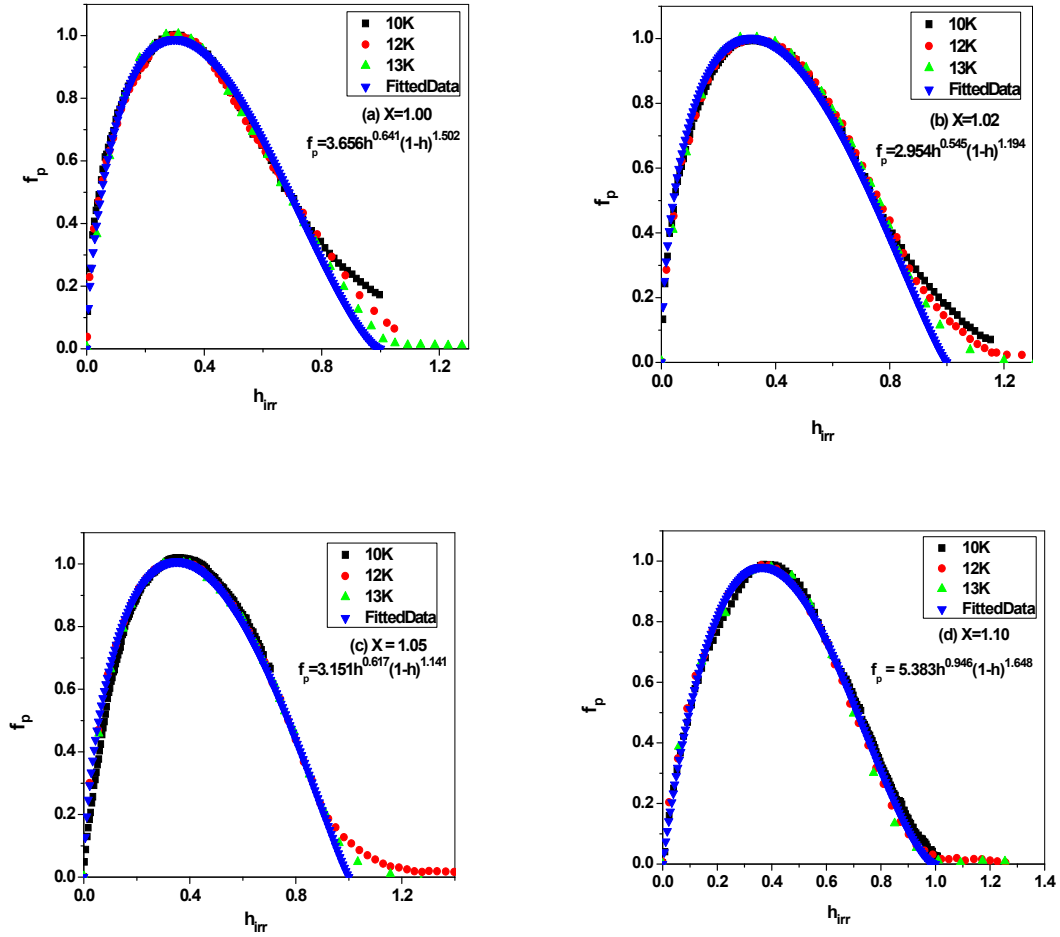


Figure (5.20): Reduced Pf curves as a function of reduced field for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at temperatures close to (T_c) for (a) $X=1.00$, (b) $X=1.02$, (c) $X=1.05$ and (d) $X=1.10$.

5.4.4 Modified Scaling Pinning Force Using H_n

Figure (5.21) presents the volume pinning force as a function of $(h_n=H/H_n)$, where (H_n) is defined as the field in which pinning force drops to half of its maximum value at fields above the peak field. This type of scaling used to investigate anisotropy and proposed firstly by Eisterer during his studying for the volume pinning force in MgB_2

superconductors [94]. Studying anisotropy by concentrating on other factors besides the pinning mechanism affect and make shift on (h_{max}) in normalized volume pinning force which includes anisotropy factor, (γ) that denotes the anisotropy of the upper critical field, (H_{c2}^{ab}/H_{c2}^c) and the percolation threshold, (p_c) which is defined as the minimum fraction of a superconducting material for a continuous superconducting current path. Due to limited effect of anisotropy factor and the percolation threshold (typical constant values $\gamma=4$ and $p_c=0.25$) on pinning behavior in the modified scaling law, Eisterer suggested that only $(h_{(n)max})$ has an important effect in pinning mechanism and he explained theoretically that their a shift in peak position to distinguish the pinning mechanism [94]. The shift in peak position is about 3% (from 0.341 for the clean limit to 0.352) for the dirty limit in grain boundary pinning type. But the shift in peak position is about 6% (from 0.465 for the clean material to 0.491) for the dirty material in point pinning type.

The values of (h_{max}) and $(h_{(n)max})$ refer to the peak position for $(h_{irr}=H/H_{irr})$ and $(h_n=H/H_n)$ respectively of reduced pinning force, which are reported in table (5.5) with different temperatures near critical transition temperature and different iron content.

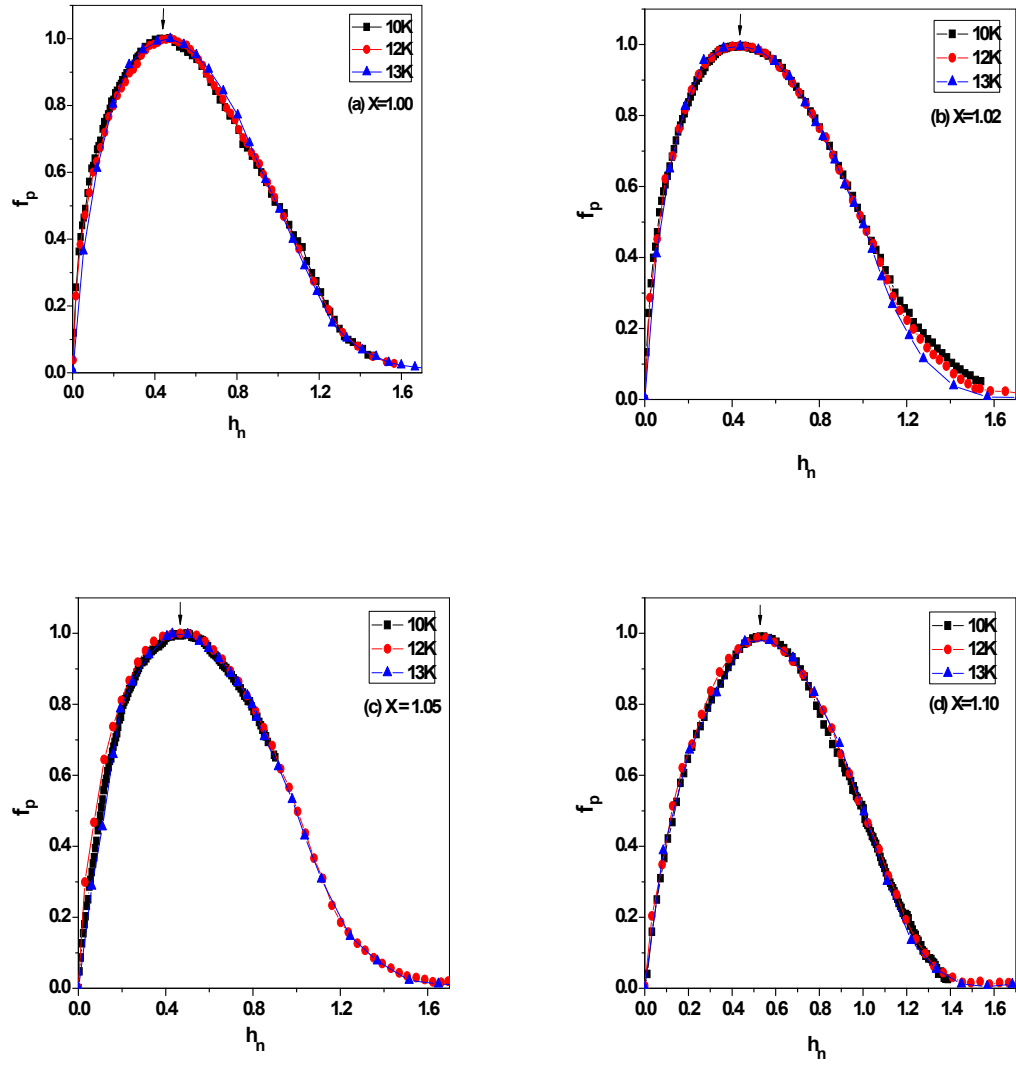


Figure (5.21): Normalized pinning force as a function of modified reduced field for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at temperatures close to (T_c) for (a) $X=1.00$, (b) $X=1.02$, (c) $X=1.05$ and (d) $X=1.10$.

Table (5.5): Value of $(h_{\max})$ and $(h_{(n)\max})$ for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples at $(T=T_c)$.

T(K)	X=1.00		X=1.02		X=1.05		X=1.10	
	$h_{\max}$	$h_{(n)\max}$	$h_{\max}$	$h_{(n)\max}$	$h_{\max}$	$h_{(n)\max}$	$h_{\max}$	$h_{(n)\max}$
10	0.308	0.447	0.322	0.462	0.359	0.495	0.371	0.548
12	0.305	0.434	0.318	0.451	0.353	0.487	0.368	0.537
13	0.301	0.428	0.312	0.438	0.345	0.466	0.363	0.522

Table (5.5) shows that the value of (h_{max}) and $(h_{(n)max})$ decreases as the temperature increases for the samples with optimum ($X=1.00$) and excess ($X>1.00$) iron contents. Also, the value of (h_{max}) and $(h_{(n)max})$ increases as the iron content (X) increases, indicating their reduction in anisotropy from clean limit to dirty limit. It is remarkable that as iron content increases, the sample becomes less anisotropic as compare to the samples with optimum iron content ($X=1.00$). The isotropic superconducting materials are useful for practical applications. Increasing iron content enhances the isotropic property of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductor with slightly compromising in its transition temperature.

CHAPTER 6

ENERGY GAP AND THE THERMODYNAMIC CRITICAL MAGNETIC FIELD

Pairing or coupling mechanism in superconductors can be understood from superconducting gap in superconductors, by finding the ratio, $(2\Delta(0)/k_B T_c)$, which is used to measure strength of coupling of cooper pairs. If the ratio, $(2\Delta(0)/k_B T_c)$ is lower than 3.53, this type of coupling is called the weak coupling or BCS type of coupling and if the value of the ratio is higher than the value 3.53, it is the region of strong coupling. Unclear results about the coupling strength for iron-based superconductors were showed by several studies. Some of these studies revealed that the iron-based superconductors have BCS type of weak coupling strength with the value of $(2\Delta(0)/k_B T_c)$ lower than 3.53 [105–107]. While other studies show that the value of $(2\Delta(0)/k_B T_c)$ is higher than 3.53 for some iron-based superconductors, which mean that they have a strong coupling strength [108].

6.1 Thermodynamic Critical Magnetic Field (H_c)

Thermodynamic critical magnetic field, (H_c), is a magnetic field with its value lying between upper and lower critical field values ($H_{c1} < H_c < H_{c2}$). These critical parameters related to (H_c , H_{c1} and H_{c2}) are shown in figure (6.1) with the following relations [63]:

$$H_{c2} = \sqrt{2} \kappa_2 H_c \quad (6.1)$$

$$H_{c1} = \frac{H_c \ln \kappa_1}{\sqrt{2}} \quad (6.2)$$

Where (κ_1 and κ_2) are the temperature dependence Ginzburg-Landau (GL) Parameters. At the critical temperature (T_c) they are identical ($\kappa_1 = \kappa_2 = \kappa$), deviations occur at lower temperatures. Values of (κ) for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample were measured at different pressures by Martinez *et al.* and they were listed in table (6.1) [109].

Table (6.1): Values of κ for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample at different pressures.

P (MPa)	0	158	158	264	580	633
κ	6.9(5)	9.2(0)	13.6(1)	10.2(9)	19.6(1)	9.2(5)

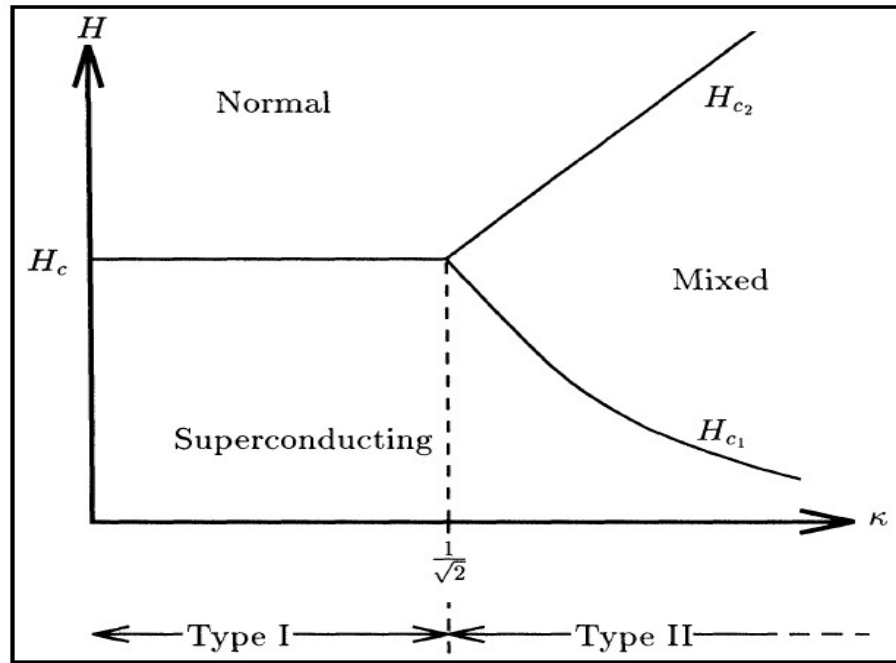


Figure (6.1): It illustrates the critical magnetic fields in both types of superconductor [63].

The importance of (H_c) is that it can be related to several thermodynamical properties of the superconducting material. For example; it is related to the amount of energy required to change the phase of the material from the superconducting to the normal. Such energy, which is required to change from superconducting to normal state, is commonly known as the free energy of superconducting state or the condensation energy of superconductor.

There are several procedures to evaluate the thermodynamic critical field for type II superconductors. The upper critical magnetic field method, specific heat analysis method, and the condensation energy method are the most established methods for the purpose. In this research, we used condensation energy method to evaluate (H_c) of $\text{Fe}_x\text{Fe}_{0.5}\text{Te}_{0.5}$.

According to thermodynamic principle, when a material is subjected to a magnetic field it will magnetized, and its free energy that is proportional to the area under the reversible magnetization curve in the Hysteresis curve. The difference between free energy of the normal state and the free energy of the superconducting state related to thermodynamic critical magnetic field as [8, 21]:

$$G_n - G_s = -\frac{H_c^2}{8\pi} \quad (6.3)$$

Where (G_n , G_s) represent the Gibbs free energy in the normal and superconducting states respectively. The thermodynamic critical magnetic field (H_c) can be calculated from area under the reversible magnetization curve as [21 and 100]:

$$\int_0^{H_{c2}} M dH = -\frac{H_c^2}{8\pi} \quad (6.4)$$

The reversible magnetization is approximated by the equilibrium magnetization, in a procedure suggested by Bean. The equilibrium magnetization (M_{eqm}) is calculated from average of increasing field magnetization (M_{inc}) and decreasing field magnetization (M_{dec}) as [21, 71, 101]:

$$M_{eqm} = \frac{M_{inc} + M_{dec}}{2} \quad (6.5)$$

We found hysteresis loop along with equilibrium magnetization area under curve, which is shown in figure (6.2). The area is used to find the thermodynamic critical magnetic field (H_c) using equation (6.4).

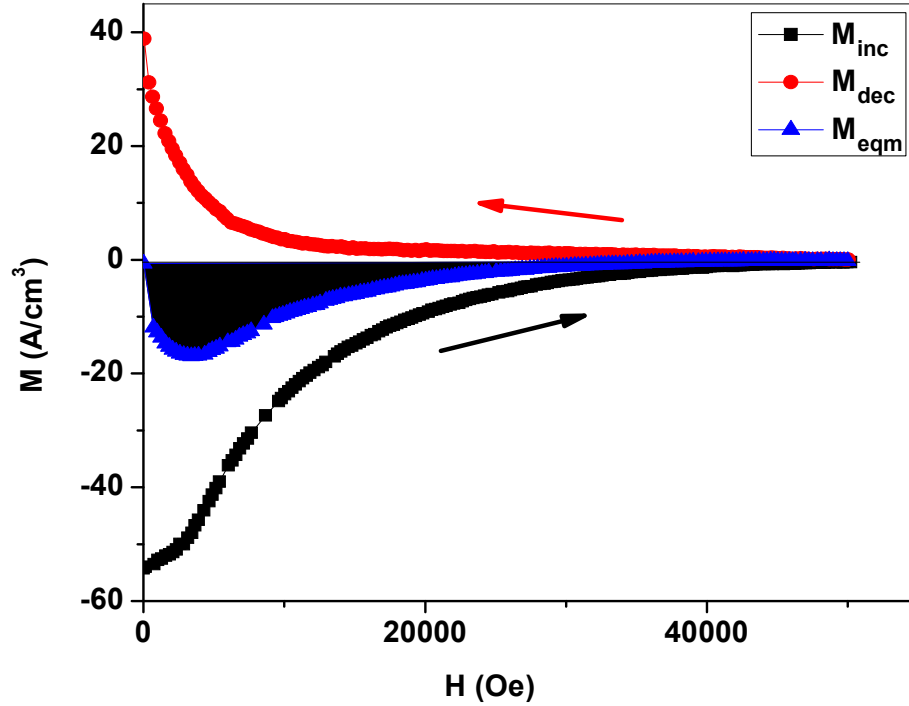


Figure (6.2): Hysteresis loop and the equilibrium magnetization (M_{eqm}) at $T = 12$ K, for optimum $FeSe_{0.5}Te_{0.5}$ sample.

6.2 Thermodynamic Critical Field of the Investigated Samples

Hysteresis loops were measured for all $Fe_xSe_{0.5}Te_{0.5}$ samples in the temperature range 4 - 13 K and in the magnetic fields up to 8.8 T. Some samples were measured in the fields up to 14 Tesla. The equilibrium magnetization values are obtained using eqn. (6.5). The obtained results for the optimal sample $FeSe_{0.5}Te_{0.5}$ are shown in figure (6.3). The area under curves were estimated numerically at different temperatures and the obtained (H_c) values for all $Fe_xSe_{0.5}Te_{0.5}$ samples are plotted versus (T) and ($1-t^2$), where (t) is the reduced temperature, presented in figures

(6.4 a) and (6.4 b) respectively. ($H_c(0)$) and slope are obtained from the linear fitting for plot of (H_c) versus (T) curve near (T_c). Same procedure is repeated for other samples and the result is shown in figures (6.4 a) and (6.4 b). The results of studied samples are summarized in table (6.2).

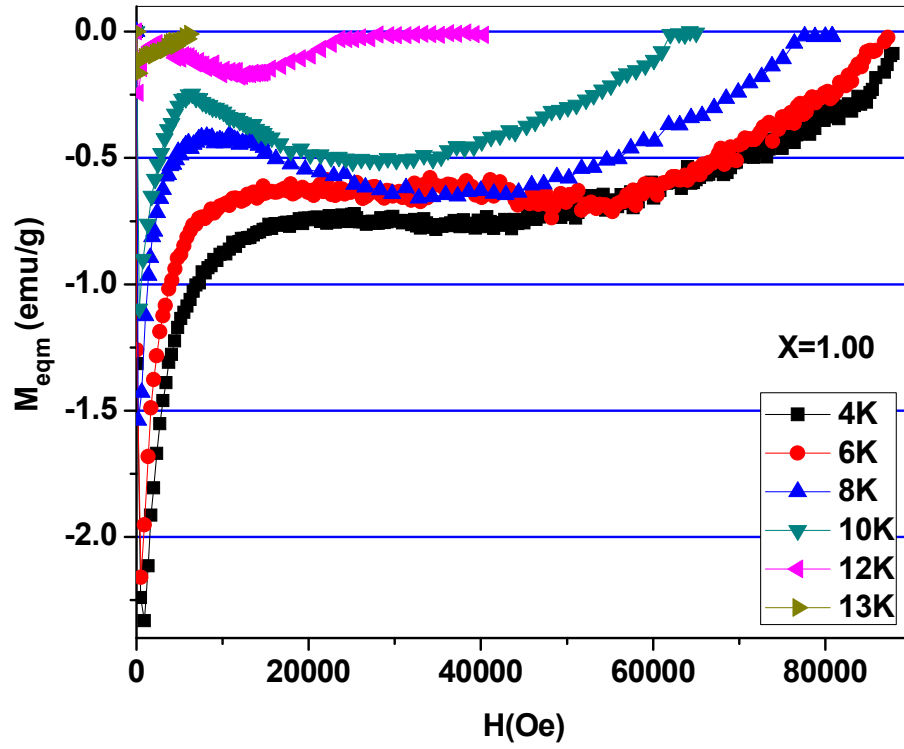


Figure (6.3): Equilibrium magnetization versus the applied field of the $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample at different temperatures.

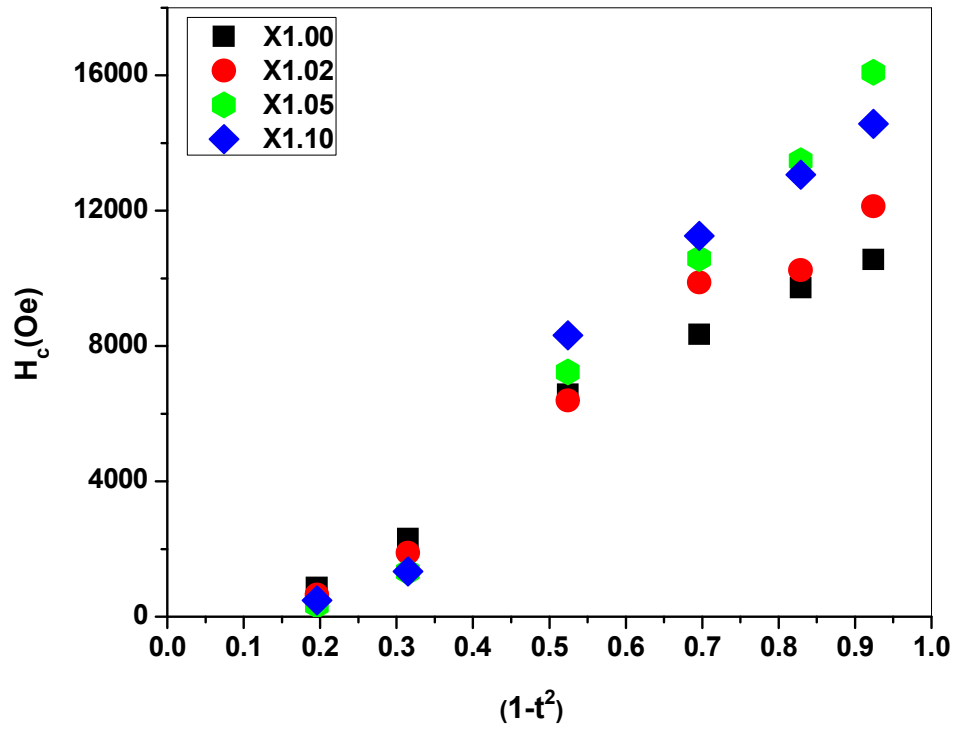
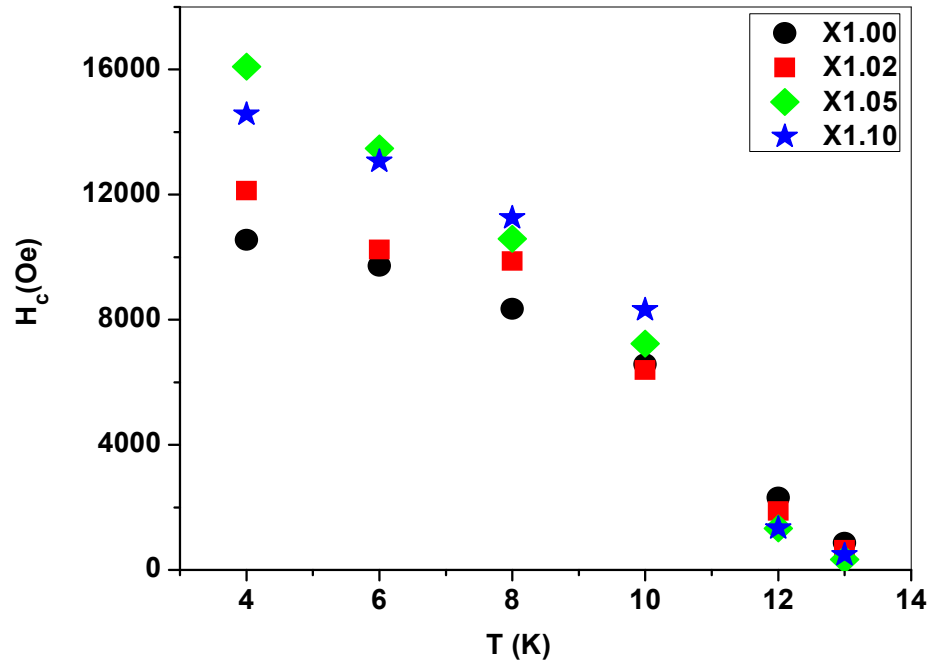


Figure (6.4): (a) H_c vs. T (a) and (b) H_c versus $(1-t^2)$ for all $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples.

Figures (6.4 a) and (6.4 b) used to evaluate the energy gap by first evaluating $(H_c(0))$ and the slope of (H_c) vs. (T) near (T_c) . The obtained values for $(H_c(0))$ and the slope $\left(\frac{dH_c}{dT}\right)_{T_c}$ for all samples are given in table (6.2). The results indicate that the slope and $(H_c(0))$ are increasing with increasing iron content reaching their maximum values at $(X=1.05)$.

Table (6.2): $H_c(0)$ and $\left(\frac{dH_c}{dT}\right)_{T_c}$ values for all samples.

Sample	X=1.00	X=1.02	X=1.05	X=1.10
$H_c(0)$ (Oe)	13704.8	15898.3	21304.5	17591.1
$\left(\frac{dH_c}{dT}\right)_{T_c}$ (Oe/K)	1935.2	1962.1	2395.1	2736.5

6.3 Energy Gap Calculations

The binding energy of cooper pairs is manifested in the form of the energy gap. The energy gap can be calculated from the thermodynamic critical field at $(T = 0)$, with the help of equation (6.6) [71]:

$$E_g = 2 \Delta(0) = 2k_B T_c \sqrt{\left(\frac{\pi\gamma}{6}\right)\left(\frac{T_c}{H_c(0)}\right)} \quad (6.6)$$

Where γ is the electronic specific heat coefficient, (γ) varies from 25 mJ/mol.K² [70] to 39 mJ/mol.K² [69]. The wide range in (γ) -values yield a range of the energy gap for different samples. Those values are listed in table (6.3). The coupling strength ratio $(2\Delta(0)/k_B T_c)$, is also given in the table. The coupling strength values we obtained for different samples are much less than the value predicted by the BCS theory ~ 3.5 and we found that the ratio decreases as the Fe-content increases.

Table (6.3): Energy gap values for all samples.

Sample	X=1.00	X=1.02	X=1.05	X=1.10
2Δ (0) (meV)	3.721	3.456	3.001	3.331
$\left(\frac{2\Delta(0)}{k_B T}\right)$	2.978	2.766	2.402	2.666

The optimum value of energy gap for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ is 3.4meV. This is consistent with the published data (3.8 meV) reported by Escudero *et al.* for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample [68] by using PCS junction method. Recently, we used (Kunwar *et al.*) used scanning tunneling microscopy/spectroscopy (STM/STS) to evaluate the energy gap for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ superconductor and obtained one of the values of ($\Delta \sim 3.1 \pm 0.8$ meV) [67]. Escudero reported 3.04 meV for the gap and the ratio ($2\Delta/k_B T_c$) is 2.746; which is comparable to the values that we obtained in this section. Sales *et al.* measured the energy gap value for optimum $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample by using specific heat measurements. They obtained ~ 3.25 meV and the ratio ($2\Delta/k_B T_c \sim 2.6$) that is close to our results [69].

Data from table (6.3) shows that the values of the energy gap generally decrease as the iron content (X) increases, which could infer to the excess of iron causes the lattice distortion. Several reports suggest that the excess of iron causes internal stress which eventually may cause the change in fundamental superconducting properties such as the critical transition temperature [48] and energy gap. It seems that excess of iron is affecting the cooper pair coupling.

6.4 BCS Coupling

According to BCS theory, the thermodynamic critical magnetic field (H_c) follows approximately parabolic temperature dependence [8]:

$$H_c(T) = H_c(0)[1 - t^2] \quad (6.7)$$

$$\left(\frac{H_c(T)}{H_c(0)}\right) = [1 - t^2] \quad (6.8)$$

Where ($H_c(0)$) is the value of (H_c) at absolute zero and T_c is the temperature at which the transition takes place in zero field and $t = \left(\frac{T}{T_c}\right)$ is the reduced temperature. The difference between the two sides of equation (6.8) separates BCS weak coupling (with negative D values) from strong coupling behavior (positive D values) for ($H_c(T)$) and it can be written as:

$$D = \left(\frac{H_c(T)}{H_c(0)}\right) - [1 - t^2] \quad (6.9)$$

The deviation is a function of the reduced thermodynamic critical magnetic field ($H_c(T)/H_c(0)$) and the reduced temperature (t). The calculated values of the deviation (D) for all samples is presented in figure (6.5) against the square of the reduced temperature.

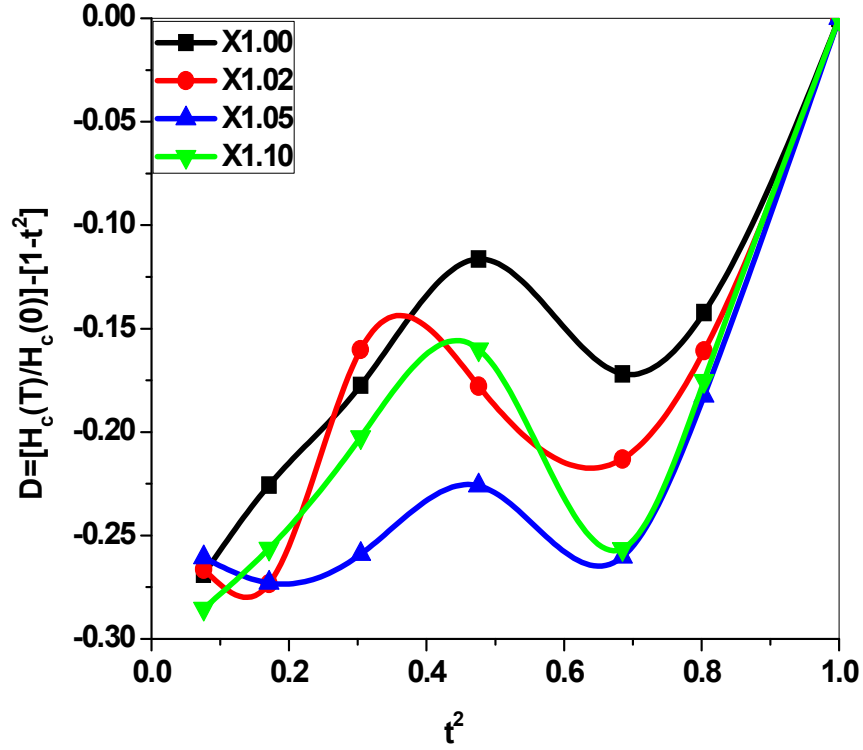


Figure (6.5): The deviation function of the thermodynamic critical field versus t^2 for all samples.

Figure (6.5) shows that for all samples, coupling lies overall under ($D = 0$) line which indicates weak coupling mechanism. Increasing iron content further weakens the coupling, and slightly reduces the energy gap.

The slope of the (H_c) near (T_c) can be used to evaluate the jump in the specific heat (ΔC).

The jump of the specific heat at (T_c) is given as [102]:

$$\Delta C = \left(\frac{VT_c}{4\pi}\right) \left(\frac{dH_c}{dT}\right)^2_{T_c} \quad (6.10)$$

where (V) is the molar volume and it equals molar mass/density. The approximated density of samples is 5.676 g/cm^3 .

The expected value of the jump in the specific heat according to BCS theory is given by [100]:

$$\Delta C_{BCS} = 1.43\gamma T_c \quad (6.11)$$

By using the values of slopes of (H_c) near (T_c) in equation (6.10) to calculate the jump in the specific (ΔC) , while equation (6.11) is used to calculate (ΔC_{BCS}) expected from BCS theory. Values for all samples were listed in table (6.4)

Table (6.4): The jump in the electronic specific heat values for all samples.

Sample	X=1.00	X=1.02	X=1.05	X=1.10
ΔC (mJ/mol K)	0.98	0.99	1.49	1.96
ΔC_{BCS} (J/mol K)	0.52	0.52	0.51	0.51
$\Delta C/\gamma_n T_c$	2.73	2.75	4.17	5.56

The BCS values of (ΔC) is close to 0.5 J/mol K, while the calculated values based on the thermodynamic critical field is significantly larger than the BCS values. Moreover, (ΔC) increases with increasing Fe-contents, almost doubled for (X=1.10).

The calculated (ΔC) value for optimum sample equals 0.976 mJ/mol K that agrees with reported value 0.9 mJ/mol K by Escudero [68]. Sales *et al.* obtained very small value of the jump for the optimum sample and; they found $(\Delta C \sim 0.26$ J/mol. K) [69]. The large discrepancy between the reported values of (ΔC) might reflect the sensitivity of the specific heat jump to defects and method of sample preparation. Defects may affect the coupling mechanism and the electronic contribution to the specific heat. According to $(\Delta C/\gamma_n T_c)$

values for all $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples, which are larger than 1.43, and these values increased by increasing iron content, this suggests an enhanced coupling strength.

CHAPTER 7

CONCLUSION AND RECOMMENDATION

7.1 Conclusion

In this work, we investigated the effects of Fe-contents in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single and polycrystalline samples. X-ray diffraction patterns have been used to analyze various structural phases and to calculate their lattice parameters. The diffraction peaks have been indexed with the tetragonal PbO structure with P4/nmm space group. The maximum lattice parameters (c (Å)) reaches maximum near ($X=1.02$), while (a (Å)) is maximum near ($X=1$), and the maximum volume of the unite cell occurs at ($X=1$). The electrical measurements were performed at temperatures lower than (T_c) and under the magnetic field up to 1.5 T. The (I - V) isotherms are related by the Sum Model [$V = \alpha I^\beta$] based power-law, which has been used to estimate the effective activation energies (U_{eff}) at various temperatures and applied magnetic fields. We conclude that activation energy values decreased by increasing iron content. Also, the activation energy (U_o) values are maximum for the optimum iron content when ($X=1$), the activation energy is found to be in the order of $100k_B T$. The variations of the resistivity with temperatures and fields have been used to calculate the activation energies using Arrhenius plots. The (U_o) values for the optimum iron content at 0 T and 1.5 T magnetic field equal 0.2943 eV and 0.2389 eV respectively and the values agrees with the one from (I - V) results. We noticed that activation energy (U_o) values are maximum for the optimum iron content when ($X=1$) and decrease for the samples when iron content increases. Increasing iron content cause disorder in the conducting layers and

this leads to decrease in activation energy. The low values of activation energy (U_o) reflects the low vortex-pinning in the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductors.

The magnetic measurements follow the same procedure as above to describe all magnetic results. We started our measurement with hysteresis loops and probable factors that can affect the loops. We calculated the irreversible properties, specially the critical current density (J_c) and the pinning force (P_f), by using Beans model. The ($J_c - H$) figures (5.5) revealed that the critical current density starts with large values at low fields and decreases as the field increases until it reaches zero at (H_{irr}). Furthermore, as the temperature increases, (J_c) decreases until it becomes zero at (T_c). Higher iron content lead to a gradual increasing in (J_c) values. The ($P_f - H$) figures (5.8) showed pinning force behavior with temperature variation for different iron contents. We distinguished two different trends around 10 K. For temperatures, ($T < 10$ K), pinning forces were increased by increasing iron contents, whereas for temperatures ($T > 10$ K), the pinning forces were decreased by increasing iron contents. We used different scaling models to investigate different pinning mechanism on $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ system. The reversible magnetization has been used to calculate the thermodynamic critical magnetic field (H_c). From the critical field (H_c) behavior for an iron content at different temperatures, we calculated the energy gap, the strength of cooper pair coupling, and their relationship with different values of iron contents.

7.2 Recommendations for Future Work

1. One may consider investigating various properties of single crystals on the lower concentrations of iron on $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ system. Anisotropic properties of the magnetization, critical current density and pinning forces may also be investigated.

2. One may think of examining several properties of the effects of elemental substitution of other elements (A) on the iron site of $\text{Fe}_x\text{A}_{1-x}\text{Se}_{0.5}\text{Te}_{0.5}$ system.
3. The effects of external pressure on the magnetic, transport and structural properties on $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystals system.

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THERMALLY ACTIVATED FLUX FLOW IN $\text{FeSe}_{0.5}\text{Te}_{0.5}$ SUPERCONDUCTING SINGLECRYSTAL

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Abstract. The current–voltage (J - E) isotherms of single crystal $\text{FeSe}_{0.5}\text{Te}_{0.5}$ sample have been measured at several temperatures near the transition temperature (T_c) and under applied magnetic fields (H). A power law ($E \sim J^\beta$) has been used to fit the data and evaluate the activation energy $U_0(T)$ using $\beta = U_0/k_B T$. At low current density ($J \ll J_c$), the initial behaviour is associated with thermally activated flux Flow ($TAFF$) while at $J > 1 \times 10^4$ V vortex flux flow (FF) behavior is expected. The effects of applied magnetic field on FF and $TAFF$ also been investigated. We found that $U_0(FF)$ was reduced with by about an order of magnitude in magnetic fields as low as ~ 1.5 Tesla -the reduction in $U_0(TAFF)$ is even faster than in $U_0(FF)$ -hence reflecting the low pinning nature (defects, vacancies etc.) of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ superconductor.

1. Introduction

The vanishing electrical resistance and diamagnetism below T_c is one of the main characteristics of superconductor. In superconductors, current transports occur with no energy loss for the currents lower than the critical current (J_c). However, it also depends on the temperature and the applied magnetic field. But in type-II superconductors, as the applied magnetic field increases above certain critical field H_{c1} , the vortex state complicates the picture. The vortex motion activated thermal energy may lead to “flux flow resistance” which could cause the energy dissipation.

Transport studies and dissipative properties in type II superconductors are usually limited to the region near T_c . Flux creep and vortex motion close to T_c can be explained using the Kim-Anderson model [9, 13], if the pinning potential was taken into account. Vortex dynamics, the irreversibility line and a vortex glass transition have been studied in high temperature superconductors [6-11]. From (I - V) measurements of thin films and single crystals high T_c superconductors several researchers suggested the continuous vortex glasses transition at temperatures below T_c . Fisher *et al.* suggested a scaling behaviour for the (I - V) curves obtained at various temperatures close to T_c [1, 2 and 3] and Koch *et al.* [2] showed experimentally the first evidence for the continuous vortex glass transition at temperatures below T_c by scaling the nonlinear (I - V) curves in YBCO thin films.

Similar results have been found in YBCO single crystal by Gammel *et al.* [5]. Sun *et al.* [7] have suggested a power-law relation as $V = \alpha I^\beta$ to describe the (I - V) curves. Where the exponent β is related to the pinning barrier U_0 . Based on the status of vortices the mixed state in type II superconductor contains different regimes; thermal activated flux flow regime (TAFF), flux creep regime (FC) and flux flow regime (FF) according to status of vortices. Although there is no clear-cut distinction between each regime, the variations of the current density with temperature is used as a guide in this respect [12]. The newly discovered Fe-based superconductors have an intermediate transition temperature where thermal fluctuations are in between conventional and high T_c superconductors. Hence, it will be interesting to investigate the effects of thermally activated flux flow on various transport properties of these new materials. These properties are strongly influenced by the dynamic behaviour of vortices in the superconductor and can be studied by measuring the current-voltage curves [4, 5].

In this paper, we report our (I - V) isotherms of single crystal $\text{FeSe}_{0.5}\text{Te}_{0.5}$ near T_c ($\sim 14.5\text{K}$) in the presence of applied magnetic field. We discuss our results in terms of the power law form, then present the variation of the activation energies U_0 (pinning potential barrier) with temperature.

2. Experimental Details

High purity ($\geq 99.99\%$) elements Fe, Se and Te have been used in 1:0.5:0.5 atomic ratios to prepare $\text{FeSe}_{0.5}\text{Te}_{0.5}$. The elements were grinded, pressed and sealed in a quartz tube partially filled with Argon gas. The tube was annealed at 500°C for 2 hours and then at 1070°C for 24 hours. The temperature was reduced at a rate of 4°C/h to 700°C and kept for 5 hours, then reduced to 400°C , at kept and annealed 2 hours, then furnace cooled to room temperature. Four-probe (dc) technique was used to measure the current-voltage (I - V) characteristics using Lake Shore 7507 measuring system. Keithley 220 constant current source has been used to supply the current, and the voltage was measured using Keithley 2182 nano-voltmeter. The sample was cooled in a closed cycle cryostat (Advanced Research Systems) and the temperature was controlled using Lake Shore 340 temperature controller.

3. Results and discussions

From x-ray diffraction pattern for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystal, the structure of the sample is a tetragonal structure with cell parameters $a = 3.797 \text{ \AA}$, $b = 3.797 \text{ \AA}$ and $c = 5.984 \text{ \AA}$. The lattice parameters are agreeing with the published data.

(I - V) isotherms of single crystal $\text{FeSe}_{0.5}\text{Te}_{0.5}$ have been measured at two different values of magnetic field $B=0\text{T}$ and $B=1.5\text{T}$. Figure 1 (a) and (b) shows a representative (I - V) isotherms of the sample at magnetic field $B=0\text{T}$ and $B=1.5\text{T}$ respectively. The data is presented on a log-log graph that simplify evaluating the α in the power-law behaviour $V = \alpha I^\beta$ where α is a constant, $\beta = U_0/k_B T$ and U_0 is the pinning barrier [7,8,10]. The exponent β is calculated from the slope of linear part of the (I - V) isotherms obtained at $H=0\text{T}$ and 1.5T and was used to evaluate the activation energies $U_0(\text{TAFF})$ and $U_0(\text{FF})$. The activation energy is essentially the pinning barrier provided by defects and pinning centres in the sample. The results are presented in Figure 2 (a) and (b) as a variation of $1/T$ for both TAFF and FF regimes at $H=0\text{T}$ and $H=1.5\text{T}$. Thermal activated flux flow (TAFF) occurs near $1\mu\text{V}$, while flux flow (FF) regime is at much higher voltage. The figures reveal that as the temperature decreases below T_c , U_0 continuously increases. The increase in TAFF regime is much faster than in FF regime. In other words, the gradual increase in exponent follow gradual increase in the pinning barrier with decreasing temperature. The exponent β approaches unity (Ohmic behaviour) near T_c . The deviation from Ohmic behaviour occurs at a non-linear

temperature $T_{nl} \sim 14.2\text{K}$, which is about 0.3K below the T_c ($\sim 14.5\text{K}$). The pinning potential at $H=1.5\text{T}$ for both cases drops to about one tenths of its initial value obtained at $H=0\text{T}$.

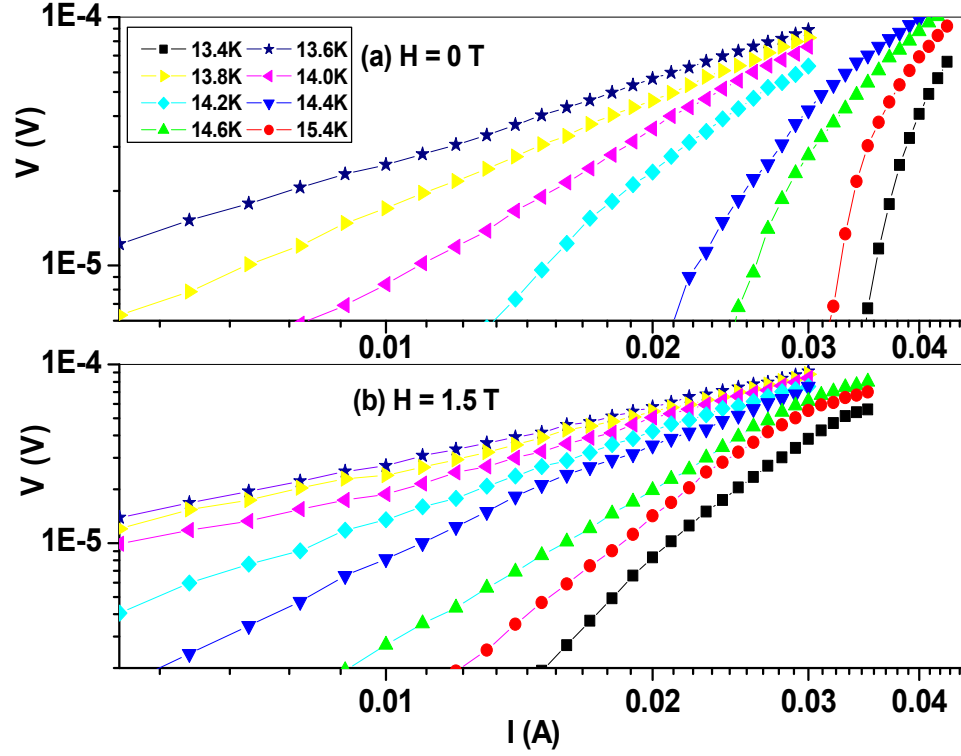


Figure 1. Selected (I-V) isotherms for single crystal $\text{FeSe}_{0.5}\text{Te}_{0.5}$ (a) at $B = 0\text{T}$ and (b) at $B = 1.5\text{T}$ around its corresponding superconducting transition temperature.

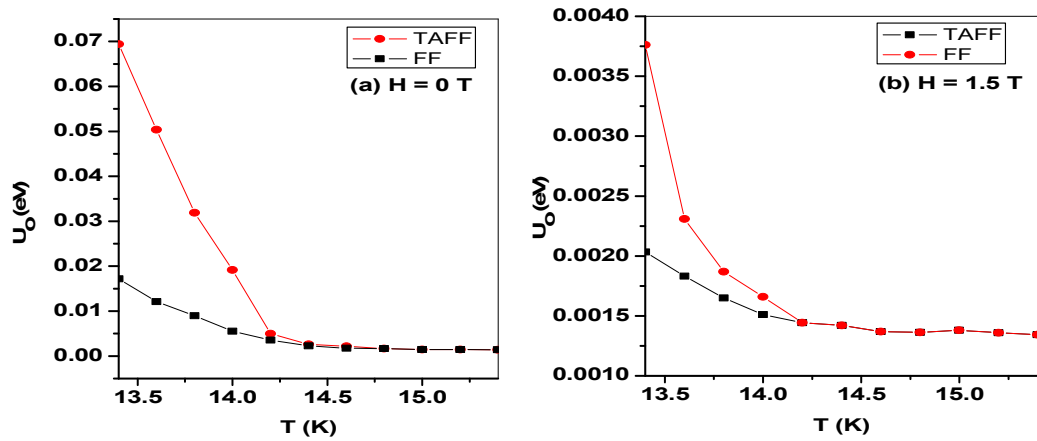


Figure 2. The pinning potential U_0 obtained for $H=0\text{T}$ (a) and for $H=1.5\text{T}$ (b) are presented as variations with temperature in both thermally activated flux flow (TAFF) and flux flow (FF) regions.

Above T_c the exponents $\beta \sim 1.4$, for both regimes; which illustrates that the I-V curves are nonlinear material is still not in the normal state. Fluctuations and thermal effects near critical temperature may cause such deviations and pose limitations for Sun *et al.* model.

4. Conclusion

Current-voltage isotherms of the single crystal $\text{FeSe}_{0.5}\text{Te}_{0.5}$ exhibit a power law behaviour in both regimes namely, TAFF and flux flow. The exponents β obtained of (from- TAFF and flux flow (FF) regime are found to decreases linearly as T increases. The pinning barrier U_0 for TAFF is about 0.85eV and for FF is about 0.11eV. Both energy barriers decrease by about an order of magnitude in 1.5T. These values indicate that the pinning forces are relatively week in $\text{FeSe}_{0.5}\text{Te}_{0.5}$ single crystals.

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Appendix – Paper (2)

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Effects of iron contents on the vortex state in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$

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Abstract.

We investigate the effects of iron content on the upper critical field (H_{c2}) and the activation energy $U(T)$ in thermally activated flux flow in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ near the superconducting transition temperature T_c . The variations in $H_{c2}(T)$ with temperature are analyzed using Ginzburg-Landau (GL), Werthamer-Helfand-Hohenberg (WHH) models along with the empirical relation (ER). The obtained values of $H_{c2}(0)$ depend strongly on the model and the criteria used to determine the transition temperature. However, the general trend is that that $H_{c2}(0)$ increases with increasing the Fe-content. The activation energy $U(T)$ is maximum for $x=1$ and rapidly suppressed by excess or deficiency of iron. The low values of $U(T)$ (~ 10 meV) reflect the low vortex-pinning nature (due to defects, vacancies etc.) in the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductor.

Introduction

The discovery of the iron-based superconductors sparked a new research interest in superconductivity and its relation with magnetism [1]. Among the wide range of Fe-superconducting, materials that have been discovered so far, the FeSe material received special attention since its initial discovery [2]. Despite the low T_c of FeSe (~ 8 K), the material uncovered a fertile ground with a variety of important phenomena such as nematic phase to name a few [2, 3]. The critical transition temperature is sensitive to the chemical doping and applied pressure [4, 5]. Ultrathin layers of FeSe showed a remarkable increase in T_c , approaching 100 K in single-layer FeSe films [6].

Partial substitution at Se sites with Te leads to an increase in T_c to ~ 15 K in $\text{FeSe}_{1-x}\text{Te}_x$ where $0.4 \leq x \leq 0.8$ [5]. The interstitial sites of the (Te, Se) layers in the tetragonal structure of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ allow for a partial occupation of an excess of Fe atoms, resulting in a non-stoichiometric compound. Excess of Fe (effectively electron doping) may induce internal stress and affect the transition temperature and other superconducting properties. Moreover; Fe deficiency (hole doping) induces internal strain, which has been found to enhance the superconducting transition temperature in thin films. There are several contradicting reports on the effect of Fe-contents on the transition temperature in thin films as well as in bulk samples [7-12].

Long annealing time at elevated temperatures helps in uniformity of the distribution of Te and Se atoms in the crystal matrix, reduces defects and induces superconductivity [13–15]. This is in line with the recent finding on the improved homogeneity in samples and the corresponding phase diagram for $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$ samples re-annealed at 700 °C for 2-days [16].

In this work, we found that near $x=1.0$, the onset of transition is not sensitive to iron contents after prolonged annealing at 400 °C. This motivated us to investigate the effects of Fe-contents on the flux flow behavior and the activation energy near T_c . We also investigated the Fe-effects on the upper critical field (H_{c2}) near T_c . The vortex state and the associated effects of Fe-contents on pinning forces may help in understanding superconducting properties in these new materials and their applications.

Experimental details

High purity (99.99%) Fe, Se and Te elements are mixed in the stoichiometric ratio to prepare $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples with $0.9 \leq x \leq 1.1$. Samples are mixed, grind then pressed into pellets then sealed in a quartz tube under a partial pressure of high purity Argon gas. All samples are annealed for 5 hours at 300 °C and 500 °C, then at 800 °C for 10 hours. The temperature is reduced to 400 °C and annealing is continued for 120 hours. The samples were ground thoroughly then pressed into pellets and received second annealing at 400 °C for 50 hours in a sealed quartz tube. The prolonged annealing at 400 °C helps in stabilizing the tetragonal structure [10].

For XRD analyses, we used RigakuMiniFlex-600 benchtop X-ray diffraction analyzers. The four-probe method was used to measure the resistivity using Lake Shore 7507 Hall-effect measurements system. The Keithley-220 constant current source is used along with Keithley-2182 nano-voltmeter. The temperature was controlled using Lake Shore-340 temperature controller.

Results and discussion

Figure 1 shows the XRD patterns taken at room temperature for selected $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples. The diffraction peaks were indexed using the PbO-tetragonal structure with P4/nmm space group. Fe-deficiency ($x < 1$) samples have several impurity phases such as Fe_7Se_8 , Fe_3Se_4 , FeO_2 , FeSe , and Te . The presence of Fe_7Se_8 is in line with the FeSe phase diagram. Traces of this impurity phase are also present in samples with an excess of Fe [10]. The impurity peaks decrease with increasing Fe content in the prepared samples. The lattice parameters and the unit-cell volume are presented in table 1. The variations of the lattice constants a and c with Fe content along with the variations of the unit-cell volume with Fe content are shown in figure 2.

Table 1. The lattice constants and the lattice volumes for the different $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples.

Sample	$c \pm \Delta c$ (Å)	$a \pm \Delta a$ (Å)	$V(\text{Å}^3)$
0.90	5.931 ± 0.081	3.778 ± 0.025	84.644
0.92	5.931 ± 0.058	3.788 ± 0.014	85.104
0.95	5.956 ± 0.038	3.787 ± 0.008	85.400
1.00	5.976 ± 0.061	3.779 ± 0.017	85.320
1.02	5.974 ± 0.032	3.771 ± 0.014	84.944
1.05	5.972 ± 0.024	3.767 ± 0.017	84.720
1.10	5.966 ± 0.028	3.767 ± 0.023	84.686

The lattice parameter (c) is maximum near $x=1.0$, while the lattice parameter (a) is maximum near $x=0.95$. The volume of the unit-cell reached a maximum value in the iron deficient region between $x=0.95$ and $x=1.0$. Fe-deficiency may induce lattice strain while Fe-excess may induce internal stress, the competition effect of stress-strain may have caused the observed non-monotonic changes in the lattice parameters and unit-cell volume [18].

The variations of the resistivity ρ with the temperature at different applied magnetic fields are presented as Arrhenius plots in figure 3. The figure reveals a small reduction in the onset of superconducting transition (T^{on}) with increasing Fe contents; namely; $T^{\text{on}} = 15.3\text{K}$, 15.2 and 15.1 for $x=1.0$, 1.05 and 1.1 respectively. The transition width, ΔT increases with increasing Fe-contents in agreement with results obtained in $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ single crystals.

To study the effect of variation of Fe content on upper critical field, $H_{c2}(T)$; we used Arrhenius plots (Fig. 3) to estimate the phase transition in different magnetic field (0-1.5) T. We used three different criteria; namely, when the resistivity drops to 10%, 50% and 90% of its normal state resistivity (ρ_n) value. These correspond to three different temperature in the flux flow region. The obtained $H_{c2}(T)$ values reflect the effects of the flux flow on determining H_{c2} . The H_{c2} values are presented in figure 4.

The upper critical field at 0 K, $H_{c2}(0)$ has been estimated using three different models: the empirical relation (ER): $H_{c2}(T) = H_{c2}(0) [1-t^2]$, Ginzburg-Landau (GL) model $H_{c2}(T) = H_{c2}(0) [(\frac{1-t^2}{1+t^2})]$ and Werthamer-Helfand-Hohenberg (WHH) model $H_{c2}(0) = -0.693T_c (\frac{dH_{c2}}{dT})$, where $t = T/T_c$ is the reduced temperature [17-20]. The calculated values of $H_{c2}(0)$ and the slope (dH_{c2}/dT) near T_c are listed in Table 2.

Table 2: Calculated Upper critical field $H_{c2}(0)$ using the empirical relation and the slope near T_c for the different $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples.

Sample	10% ρ_n		50% ρ_n		90% ρ_n	
	$H_{c2}(0)(\text{T})$	$\frac{dH_{c2}}{dT}(\text{T/K})$	$H_{c2}(0)(\text{T})$	$\frac{dH_{c2}}{dT}(\text{T/K})$	$H_{c2}(0)(\text{T})$	$\frac{dH_{c2}}{dT}(\text{T/K})$
$\text{FeSe}_{0.5}\text{Te}_{0.5}$	13.4	-2.1	20.8	-2.8	43.7	-5.7
$\text{Fe}_{1.05}\text{Se}_{0.5}\text{Te}_{0.5}$	27.8	-4.9	57.6	-8.0	63.7	-8.3
$\text{Fe}_{1.1}\text{Se}_{0.5}\text{Te}_{0.5}$	39.2	-8.3	29.5	-4.1	36.5	-14.4

Table 3: The calculated upper critical field $H_{c2}(0)$ for the different $Fe_xSe_{0.5}Te_{0.5}$ samples by the GL and WHH models.

Sample	$H_{c2}(0)$ at 10% $\rho_n(T)$		$H_{c2}(0)$ at 50% $\rho_n(T)$		$H_{c2}(0)$ at 90% $\rho_n(T)$	
	GL	WHH	GL	WHH	GL	WHH
$FeSe_{0.5}Te_{0.5}$	25.8	19.8	40.5	28.4	86.3	60.1
$Fe_{1.05}Se_{0.5}Te_{0.5}$	54.7	44.3	114.2	79.5	126.6	88.0
$Fe_{1.1}Se_{0.5}Te_{0.5}$	77.3	73.2	57.9	40.5	217.7	151.2

The data shown in Table 2 and 3 reveals that the empirical relation gives the lowest $H_{c2}(0)$ values followed by WHH then GL relations. The 10% drop criteria GL and WHH seems to yield $H_{c2}(0)$ values in agreement with published data $\sim 26-77T$ and $19-73 T$ respectively. Moreover; the 90% drop criteria in the normal resistivity gives the highest "unrealistic" estimates of the $H_{c2}(0)$. The $H_{c2}(0)$ values obtained from the empirical relation are $\sim 13.4T$ for $x=1.0$ and showed a threefold ($\sim 39.2T$) increase for $x=1.1$ ($Fe_xSe_{0.5}Te_{0.5}$) [21].

Thermally activated flux flow (TAFF) of vortices causes flux flow resistance below the superconducting transition temperature, hence causing broadening in the transition width. Applied magnetic field also broadens the transition width as shown in figure 3.

The activation energy for the TAFF can be evaluated from the Arrhenius plot of the resistance. Namely;

$$\rho = \rho_n \exp\left(-\frac{U}{k_B T}\right)$$

where ρ_n is the normal state resistivity and $U(H, T) = U_0 H^{-q}(1 - t)$ is the activation energy of the flux flow, U_0 and q are constants and sample dependent. Figure 4 shows the Arrhenius plot of $\ln \rho$ versus $1/T$, which is used to evaluate $U(H, T)$. Figure 5 shows the activation energy for the TAFF regime $U(H, T)$ versus magnetic field H . The figure indicates low values for $U(H, T)$ indicating low pinning in this Fe-based superconductor. Moreover, these values agree with low field values of the activation energy for highly oriented $Fe_{1.03}Te_{0.55}Se_{0.45}$ with excess Fe sample obtained by Junyie *et.al.* [22].

The small U value obtained at $H = 1\text{ T}$ compared to 0 T indicates the sensitivity of effective pinning to the applied magnetic field. We notice that U values are maximum for the Fe content $x=1.00$ and decrease for the samples with higher Fe content.

Conclusions

Various samples of $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductor have been prepared under prolonged annealing. The tetragonal structure is used to obtain the variations of lattice constants (a and c) with Fe content. The maximum lattice constant c reaches saturation near $x=1.0$, while the a -parameter and the volume of the unit-cell are maxima between $(0.95 < x < 1.0)$. The transition width obtained from resistivity measured in different magnetic fields revealed an increase in the transition width with Fe-contents. The $H_{c2}(0)$ values are evaluated from fitting the H - T phase diagram using two fluid model formula, GL formula, and WHH formula. The values obtained are model-dependent and sensitive to the criteria used in determining the superconducting transition temperature T_c . The activation energy $U(H, T)$ drop rapidly with the applied magnetic field indicating the sensitivity of effective pinning to the applied magnetic field. We notice that U values are maximum for the Fe content $x=1.00$ and decrease for the samples with higher Fe content. The low values of $U(T)$ ($\sim 10\text{ meV}$) reflect the low vortex-pinning nature in the $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ superconductor.

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Figure 1. Room temperature XRD patterns for $\text{Fe}_x\text{Se}_{0.5}\text{Te}_{0.5}$ samples with x ($0.95 \leq x \leq 1.05$) and impurity phases A: FeSe, B: FeO_2 , C: Te, D: Fe_7Se_8 and E: Fe_3Se_4 .

Figure 2. Variation of lattice parameters (a) and the volume of the unit-cell (b) with Fe content.

Figure 3. Arrhenius plot for the resistivity at different magnetic fields. The solid lines are a linear fit to the data.

Figure 4. The variation of (H_{c2}) with temperature for $x=1.00$, 1.05 and 1.10 samples using T_c determined from different criterion of normal state resistivity ρ_n : (a) $0.9 \rho_n$, (b) $0.5 \rho_n$, and (c) $0.1 \rho_n$.

Figure 5. The activation energy for the TAFF regime U versus magnetic field B .

Figure 1.

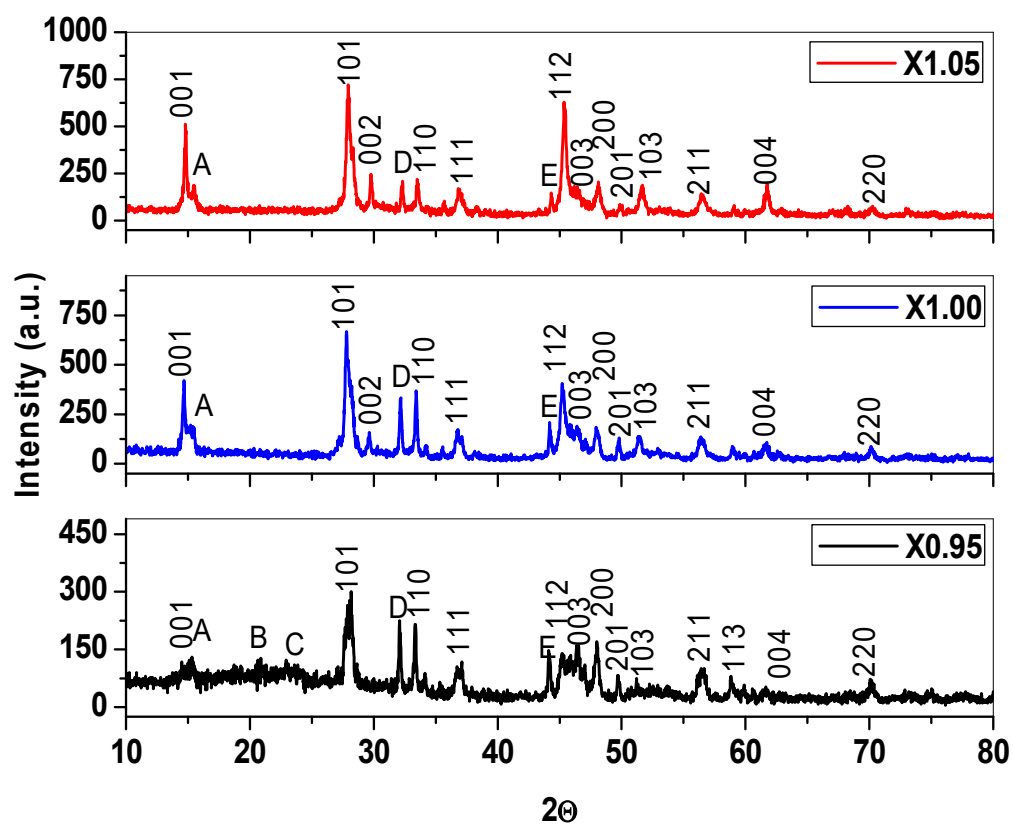


Figure 2.

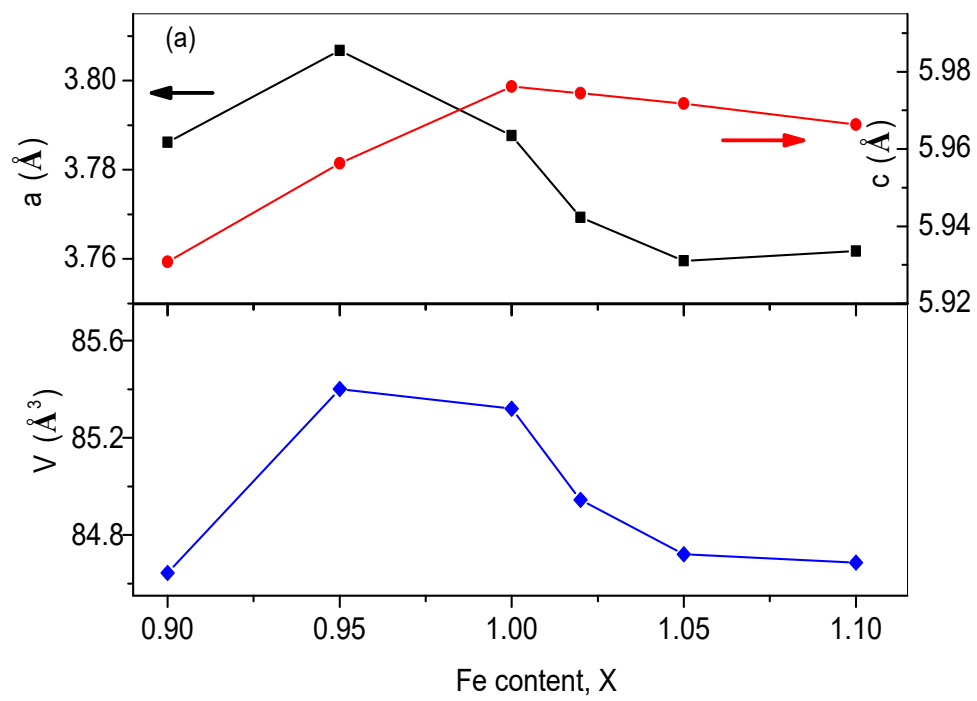


Figure 3.

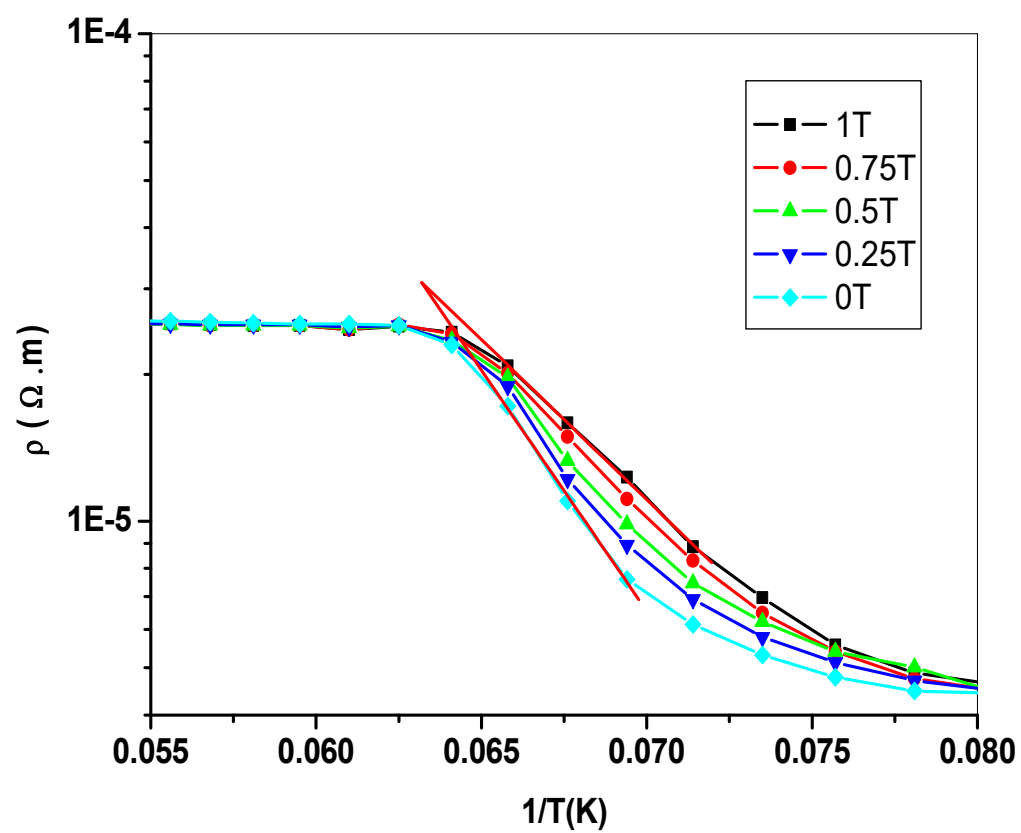


Figure 4.

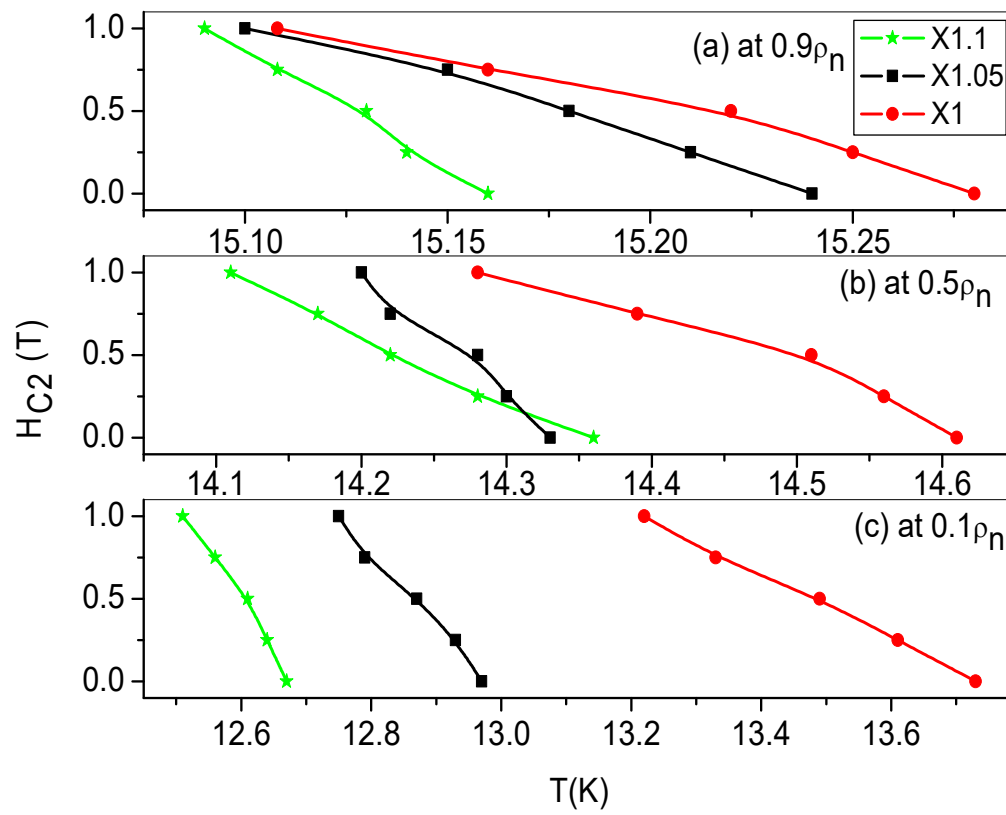
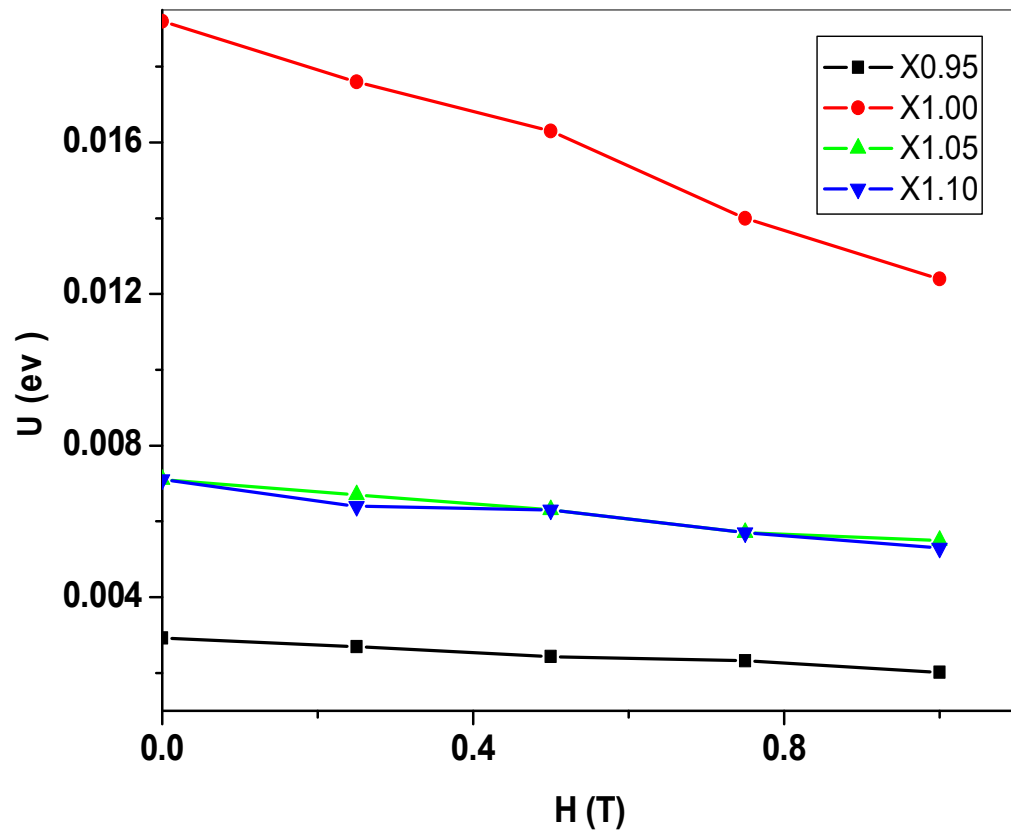


Figure 5.



Appendix – Paper (3)

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Coexistence of Weak and Strong Coupling Mechanism, in an Iron-Based Superconductor $FeSe_{0.5}Te_{0.5}$: Possible Signature of BCS-BEC Crossover

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Abstract

In this work, we present the scanning tunneling microscopy/spectroscopy (STM/STS) studies of an iron-based superconductor, $FeSe_{0.5}Te_{0.5}$. We obtained differential conductance (dI/dV) spectra and extracted the value of the superconducting order parameter (Δ) using extended Bardeen Cooper Schieffer (BCS) phenomenology for anisotropic s-wave pairing. We observed widespread values of Δ from 0.6 to 4.5 meV, with distinct two peaks in the histogram around 1 and 3 meV. The corresponding values of $2\Delta/k_B T_c$ for the peaks are 1.5 and 5.0, respectively, indicating coexistence of weak and strong coupling mechanism. We also found two different coupling mechanisms through Δ to Fermi energy ratio (Δ/E_F) of the material; which infers to the possible composite superconductivity in the realm of BCS-Bose Einstein Condensate (BEC) crossover.

1 Introduction

Iron-based superconductor was first discovered in 2008 with transition temperature (T_c) = 26 K in one of the iron pnictides, $La(O_{1-x}F_x)FeAs$ [1]. The T_c of these materials was immediately increased up to 56 K. The discovery sparked great interest in the field of condensed matter physics [2–6]. Today, the iron-based superconductors are one of the most studied unconventional high- T_c superconductors. Despite extensive studies on these materials, there still remain some unresolved issues, and one of such issues is the microscopic understanding of superconducting mechanism. More explicitly, the origin and type of the glue that couples two electrons together to form a Cooper pair is not clear yet. Pairing mechanism or coupling mechanism in superconductors is directly related to the superconducting gap, Δ . The Cooper pair coupling in conventional superconductors is caused by lattice excitations and it manifests itself in the form of the ratio, $2\Delta(0)/k_B T_c$ 3.53. Such a coupling is called the weak coupling or BCS type of coupling and the value 3.53 is considered to be the weak coupling limit. If the value of the ratio is higher than

the weak coupling limit, i.e., 3.53, it is considered to be the region of strong coupling. Therefore, the ratio $2\Delta/k_B T_c$ is taken as a measure of the coupling strength.

Not only for iron-based superconductors, the coupling mechanism is still under debate for the poster child of high- T_c superconductors, i.e., cuprates. In our earlier work, we studied the coupling mechanism of one of the electron doped high- T_c cuprates and observed the strong coupling of electronic origin [7, 8]. Iron-based superconductors have many similarities with cuprates, such as: both are layered materials with superconducting planes separated by charge tuning layers, both have high transition temperature, and both have the ground state of magnetic origin. Therefore, the understanding of coupling mechanism in iron-based superconductors may enhance our understanding of high- T_c superconductors as a whole.

In iron-based superconductors, different studies revealed quite ambiguous results as far as the coupling strength is concerned. For example: several studies found that some of the iron-based superconductors have BCS type of weak coupling mechanism with the value of $2\Delta/k_B T_c$ lying within the BCS limit [9–11]. Other studies observed that the value of $2\Delta/k_B T_c$ for some iron-based superconductors is much higher than 3.53, inferring to a strong coupling mechanism [12].

We report here the STM/STS measurements of one of the simplest iron-based superconductors, $FeSe_{0.5}Te_{0.5}$. We obtained tunneling conductance spectra and extracted superconducting gap Δ from those spectra to analyze the coupling strength.

2 Experimental Methods

The parent compound of our sample is iron selenide (FeSe) which has a tetragonal PbO structure with Fe-based planner sub-lattice. It has a layered crystal structure belonging to the $p4/nmm$ space group. The crystal of FeSe is composed of a stack of edge-sharing $FeSe_4$ -tetrahedra layer by layer. Substituting Se with Te enhances superconducting T_c and goes up to 15 K under normal conditions, without changing the crystal structure [13, 14]. Our samples were optimally doped $FeSe_{0.5}Te_{0.5}$ with $T_c \sim 14$ K. The crystals were cleaved in ultra-high vacuum for the clean surface and inserted into the STM head held at temperature 5 K.

3 Results and Discussion

Figure 1 shows an atomically resolved typical topographic 12×12.8 nm² image of the sample surface. The image was taken with a bias voltage of 11.7 mV and tunneling current of 0.96 nA. We found the cleaved surface as a square lattice with average values of lattice constants in the perpendicular directions as 3.8 and 3.9 Å, respectively. The values are close to the expected distance of Fe-Se bonds [15–17]. Measurement of $(dI/dV(r, eV))$, the differential conductance spectrum (commonly known as STM spectrum or tunneling spectrum) is one of the special features of STM and is called tunneling spectroscopy. We obtained varieties of spectra through tunneling spectroscopy: from spectra with well-

defined coherence peaks -to- spectra with ill-defined coherence peaks -to- the spectra with no coherence peaks. Figure 2a shows the typical tunneling spectra with well-defined coherence peaks whereas Fig. 2b shows the evolution of spectra from ill-defined coherence peaks to the spectra without any coherence peaks. All the spectra are obtained at different location within $12.8 \times 12.8 \text{ nm}^2$ area of the sample surface and each curve is the average of at least four neighboring tunneling spectra.

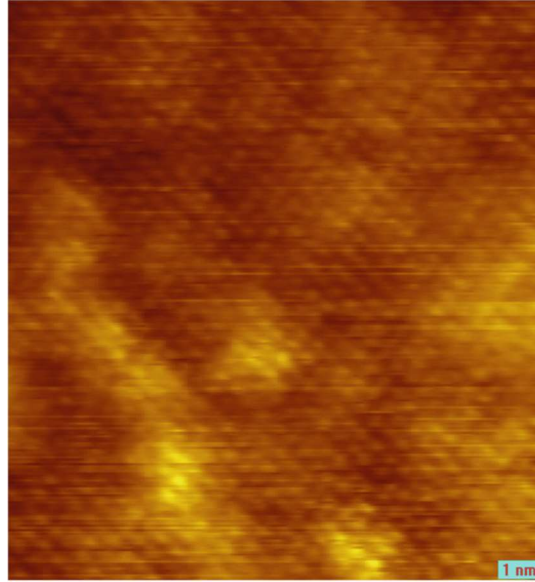


Fig. 1 Atomically resolved typical topographic $12.8 \times 12.8 \text{ nm}^2$ image of the sample surface

The tunneling spectra are quite inhomogeneous in space. Not only in the appearance of the spectra, the inhomogeneity extends to the size of the gap as well. The inhomogeneous nature of tunneling spectra is one of the signature properties of unconventional superconductors and is considered as a result of out-of-plane scattering [18, 19]. Inhomogeneity is clearly visible in Fig. 3 which shows the waterfall image of an arbitrary line cut through the sample. The spectra in the waterfall image was taken through $64 \text{ \AA}$ horizontal straight line, with one spectrum taken at each angstrom. The waterfall image shows the evolution of spectra from well-defined coherence peaks to spectra with no coherence peaks in a short distance. The spectra with ill-defined coherent peaks and the spectra with no coherent peaks are quite repeatable and are location specific. Such spectra were reported earlier [16, 17].

The important physical quantity in superconductivity is the order parameter Δ , whose structure and value are closely related to the pairing mechanism. Scanning tunneling microscopy as a local probe played a crucial role on the direct measurement of Δ . At low temperature ($\sim 5 \text{ K}$), the value of Δ can be directly measured as half of the energy difference between the two coherence peaks. But the accuracy of the measurement can be challenged if the coherence peaks are not sharp as in our case. Therefore, in order to extract the accurate value of Δ from the tunneling spectra, we used a simple phenomenological picture to capture the spectral line shape. This model captures the momentum space (k-space) variation

in the superconducting gap due to its anisotropic s-wave symmetry [20–22].

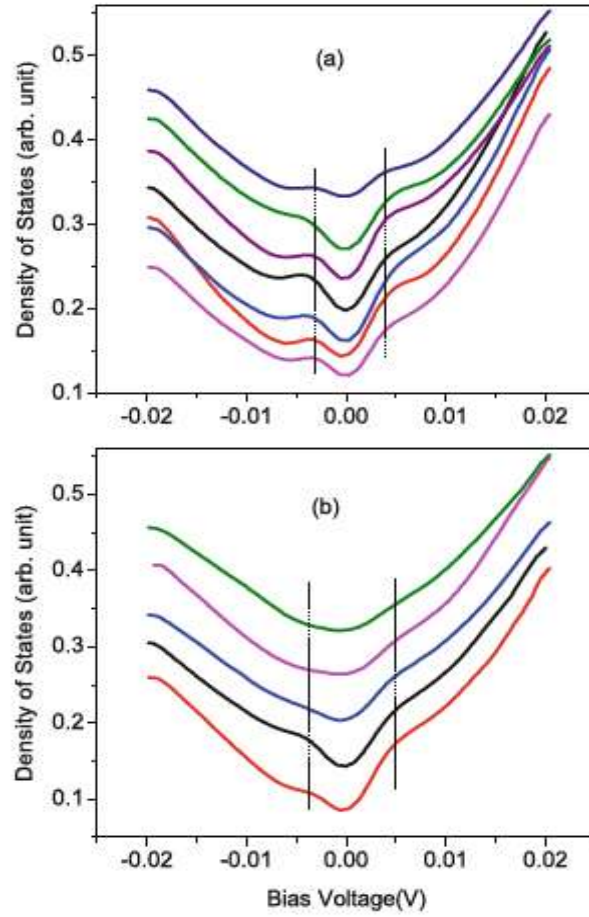


Fig. 2 (Colour online) Typical STM spectra obtained at 5 K at different locations on a sample (a) with distinct coherence peaks (b) spectra from ill-defined coherence peaks to spectra with no coherence peaks. Each curve is the average of at least four neighbouring spectra.

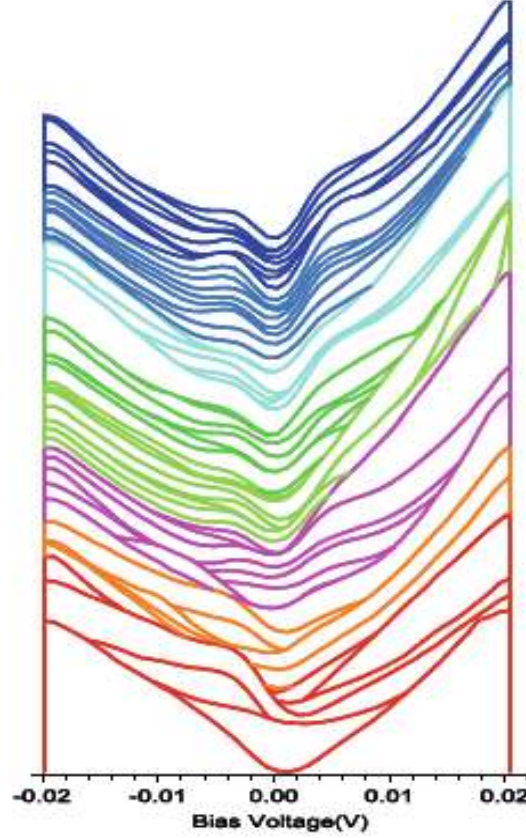


Fig. 3 (Colour online) Waterfall image of an arbitrary line cut through the sample. The spectra in the waterfall image was taken through 64 Å horizontal straight line, with one spectrum taken at each angstrom.

The tunneling conductance spectrum is equivalent to the integrated local density of quasiparticle electronic states. Using extended BCS theory for anisotropic s-wave pairing, the differential conductance, i.e., quasiparticle density of states, can be written as,

$$\frac{dI(V, T)}{dV} = \alpha \int_{-\infty}^{\infty} dE \frac{df(E + V, T)}{dV} \int_0^{2\pi} d\theta \operatorname{Re} \left(\frac{|E - i\Gamma|}{(E - i\Gamma)^2 - \Delta^*(T)^2} \right).$$

Where, $f(E, T)$ is Fermi-Dirac distribution function at temperature T , E is the quasiparticle energy, V is bias voltage, Γ is quasiparticle scattering rate, and α is proportionality constant.

We obtained the values of Δ and Γ by best fitting our experimental data with the theoretical quasiparticle density of states for the anisotropic s-wave pairing of gap function, $\Delta^*(\theta) = \Delta [0.7 + 0.3 \cos(4\theta)]$. The typical fits between theory and experimental tunneling spectra of well-defined coherence peaks are shown in Fig. 4a. We considered that the gap values from positive bias and negative bias portions of the spectra are identical if not the same. Therefore, we confined our analysis only to negative bias portion of each spectrum. The part of the spectrum, with energies higher than the coherent peak, is irrelevant for the measurement of Δ and has been ignored. Figure 4b shows the typical

fit between theory and tunneling spectra of ill-defined coherence peaks or the spectra without any coherence peaks. We found that the theoretical energy spectrum for quasiparticle density of states for asymmetric gap function fits extremely well with our tunneling spectrum. The value of best fitting parameter χ^2 is impressively low (for most of the spectra: from lower 10^{-7} to lower 10^{-5}) indicating the relevance of BCS phenomenology to explain these superconductors. Not only the pairing mechanism but also the pairing symmetry of the order parameter for iron-based superconductors is under discussion. First predicted by Mazin *et al.*, the unconventional s-wave symmetry is getting consensus from many different studies [23–28]. Impressive agreement of our data for anisotropic s-wave symmetry supports the node-less s-wave symmetry of order parameter for FeSe-based superconductors.

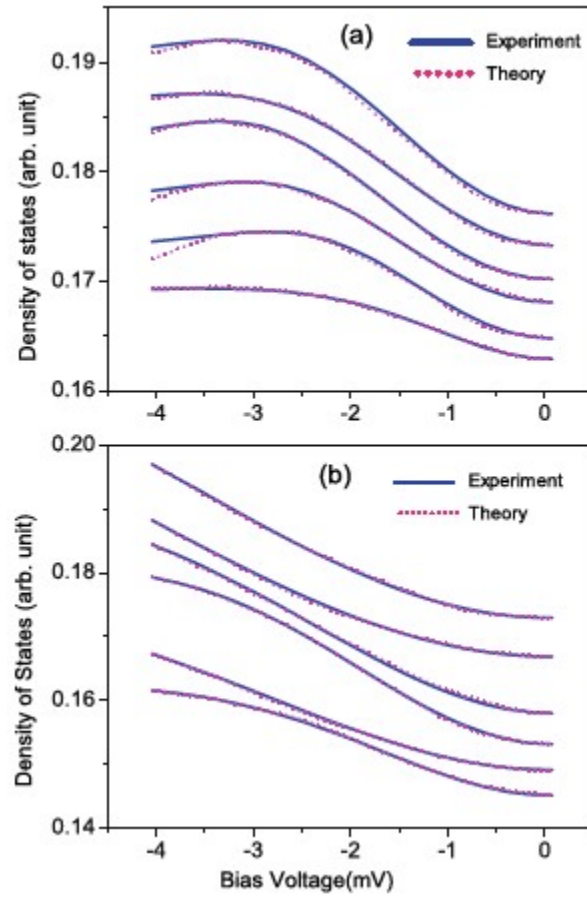


Fig. 4 (Colour online) Typical STM spectra (solid curve) in the negative bias region with their corresponding theoretical fits (dotted curve) (a) fits of spectra with coherence peaks (b) fits of spectra with ill-defined coherence peaks or no coherence peaks. The value of the spectra in the vertical axis (density of states) is being shifted from their original value to make them clearer to viewers.

One of the features of the tunneling spectra is the broadened coherence peaks in the quasiparticle density of states. The appearance of density of state is directly related to the Fermi surface topography. The Fermi surface topography of iron-based superconductors changes significantly in different systems depending on the atomic structure. It varies from hole pocket dominated to both hole and electron pockets, and to

only electron pockets in the FeSe-based systems [29, 30]. It is interesting that some of the tunneling spectroscopic studies (either on monolayer thin films or on single crystals) on FeSe-based superconductor observed double-gap features on a single tunneling spectrum where as other tunneling studies observed varieties of spectra from v-shaped one to single-gaped ones [31–33]. Angle-resolved photo-emission spectroscopy (ARPES) measurements on either single crystal or on thin films of the material reported single-gap feature [34–37]. Therefore, different shapes of quasiparticle density of states revealed by different studies infer to the complexity and exotic nature of the material.

Broadened coherence peaks in our tunneling spectra could be explained by impurity effect and thermal broadening process. Deficiencies at Se-sites are crucial to the superconducting phase in pure FeSe-based superconductors. These deficiencies act as scattering centers which cause the broadening in quasiparticle density of state. In our sample, Se or Te-site deficiencies cause the broadening of tunneling spectra and lattice mismatch between them, although very little, further enhances the broadening. The presence of high impurities manifest in the form of broadened coherence peaks and the broadening of tunneling spectra can be quantified through the quasiparticle scattering rate, T , which is found to be 2.7 ± 1.7 meV. Such a high value of gamma infers to higher number of scattering centers in the sample and could explain the broadened and ill-defined coherence peaks.

We found two values of superconducting order parameter Δ and they are: 1.0 ± 0.3 meV and 3.1 ± 0.8 meV, respectively. The histogram of the order parameter Δ with two distinct peaks are shown in Fig. 5a. As the coherence peaks are very sensitive to heat the thermal energy around 5 K is capable of some broadening in coherence peaks. Although the coherence peaks are widened or ill-defined due to quasiparticle scattering and thermal broadening, the difference in gap size at 5 K and lower temperatures is within the error limit [31, 38, 39]. Therefore, the value of Δ (5 K) should not be significantly different from Δ (0).

From the values of Δ , we calculated the values of the ratio $2\Delta/k_B T_c$ to measure the coupling strength. We found that the values of the ratio have two corresponding values: 1.6 ± 0.6 and 5.1 ± 1.3 , respectively. The histogram of $2\Delta/k_B T_c$ is shown in Fig. 5b which clearly shows the two distinct peaks separated by the BCS predicted values for weak coupling limit, i.e., 3.53. Considering $2\Delta/k_B T_c$ 3.53 as the weak coupling limit, our study infers that there is coexistence of weak and strong coupling in this superconductor. The material has very low Fermi energy (E_F) = 4 ± 2.5 meV around the gamma-point of the Fermi surface which is the major contributor of tunneling current [38, 40, 41].

The corresponding ratios of mean values of superconducting order parameter to the Fermi energy (Δ/E_F) are 0.2 and 0.8, respectively. The ratio Δ/E_F clearly separates the regions lower and higher than 0.5. The region with value higher 0.5 is considered to be the region of strong coupling, i.e., BEC type whereas the region with the value less than 0.5 is considered to be the region of weak coupling, i.e., BCS type; thus, inferring to the field of possible BCS-BEC crossover [42].

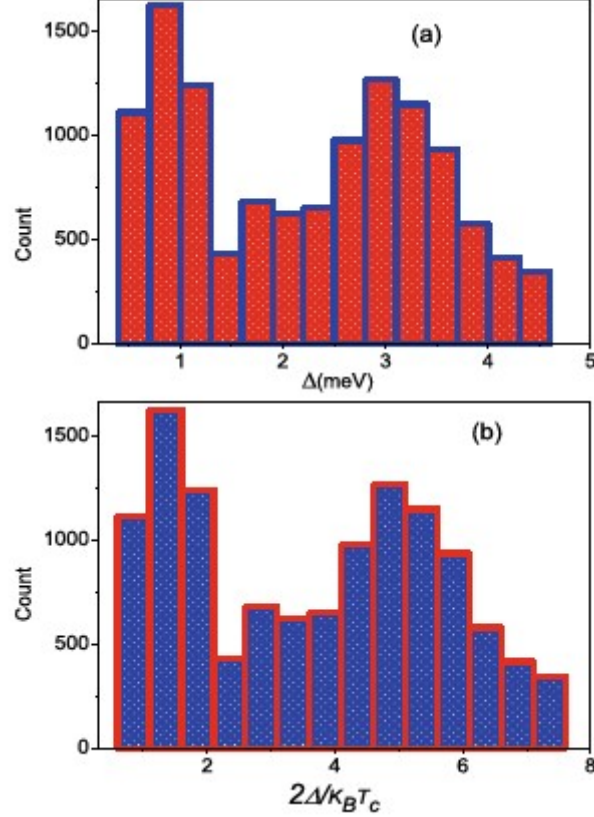


Fig. 5 (Colour online) Histograms: (a) the histogram of Δ with explicit two peaks. (b) the corresponding histogram $2\Delta/k_B T_c$

4 Conclusion

STM/STS studies on the single crystal $FeSe_{0.5}Te_{0.5}$ revealed a single-gap feature in the tunneling conductance spectra. Using BCS phenomenology for anisotropic s-wave pairing, we extracted the values of superconducting order parameter (Δ) and found two sets of values 1.0 ± 0.3 meV and 3.1 ± 0.8 meV, respectively, with the corresponding values of $2\Delta/k_B T_c$ clearly indicating the coexistence of strong and weak coupling mechanism in the superconductor. We found two mean values of the ratio Δ/E_F as 0.2 and 0.8 inferring to two different regions of coupling. With these findings, we can conclude that weak and strong coupling mechanism coexist in $FeSe_{0.5}Te_{0.5}$ superconductor, and it could be the possible composite superconductivity that lies in BCS-BEC crossover region.

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