The Effects of Radiation Damage on the Vortex Dynamics of High Temperature Superconductors

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THE EFFECTS OF RADIATION DAMAGE ON THE VORTEX DYNAMICS
OF HIGH TEMPERATURE SUPERCONDUCTORS

by
Andra Petrean

A Dissertation
Submitted to the
Faculty of The Graduate College
in partial fulfillment of the
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THE EFFECTS OF RADIATION DAMAGE ON THE VORTEX DYNAMICS OF HIGH TEMPERATURE SUPERCONDUCTORS

Andra Petrean, Ph.D.
Western Michigan University, 2000

This work focuses on the interaction between magnetic vortices in high temperature superconductors and defects introduced by irradiation with protons and heavy ions. The study concentrates on the high temperature superconductor YBa$_2$Cu$_3$O$_{7-x}$. This material is used to investigate the role of disorder in phase transitions, a topic of great interest in Solid State Physics. In addition, this study analyzes the interaction between vortices and columnar defects in various doses and defect morphologies, including the thermal stability of columnar defects.

We present experimental evidence of the vortex glass phase in proton irradiated YBa$_2$Cu$_3$O$_{7-x}$. For the first time, evidence for a vortex glass transition is detected in an untwinned single crystal of YBa$_2$Cu$_3$O$_{7-x}$ with induced point-like disorder. The first order melting transition, present before the irradiation, is suppressed by proton irradiation and a second order vortex glass transition appeared at lower temperatures. Our results suggest that sufficiently high pinning disorder is required in order for the vortex glass phase to be observed.

We then present a study of the effects of heavy ion irradiation on the phase diagram. The effects of the irradiation dose, irradiation ion type, and defect orientation on the pinning are investigated. For irradiation parallel to the c-axis of the crystal, we found that the columnar defects inhibit vortex motion when vortices are parallel to the defects, and they promote vortex motion for vortices perpendicular defects. Also for irradiation in this configuration, we learned that Uranium-induced
defects are more efficient pinning sites than Gold-induced defects. For irradiation parallel to the ab-plane, we found that the columnar defects inhibit vortex motion for vortices parallel to the defects.

Finally, the study discusses the thermal stability of columnar defects introduced by irradiation with Gold in YBCO single crystals. The dependence of the critical current density on the annealing temperatures is correlated with changes in the microstructure of the defects, as seen from transmission electron microscopy measurements. We found an increase in the critical currents at 77K with annealing, which emphasizes the high suitability of irradiated samples in applications that operate at liquid nitrogen temperatures.
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CHAPTER I

INTRODUCTION

The Phenomenon of Superconductivity

Superconductors are materials that have the extraordinary ability to carry electrical currents without any resistance when cooled below their transition temperature. Without resistance, electrical signals are not dissipated in the form of heat, and therefore all manners of devices and components become far more efficient. Superconductors also have remarkable magnetic properties, which create the possibility of entirely new applications.

After superconductivity was discovered in 1911 [1], practical applications were devised using this new phenomenon, such as superconducting magnets, superconducting generators, magnetic shielding devices, infrared sensors, and microwave devices. Magnetic resonance imaging (MRI) is playing an ever-increasing role in diagnostic medicine. The intense magnetic fields needed for these instruments are only feasible through the use of conventional superconducting magnets. Despite the fact that there are many technological applications of superconductors, their use is still limited because they need liquid helium (which is expensive and rare) as the cooling medium. The discovery of high temperature superconductors in 1986 [2] creates the possibility of even more technological applications. Due to the fact that the new materials can stay superconducting up to much higher temperatures, liquid nitrogen (which is cheaper and easier to use) can be used for cooling instead of liquid helium, increasing the applicability of these materials.
The technological applications of high temperature superconductors are hindered by their low critical current densities. The critical current is the maximum current the material can carry before losing its superconducting properties. Most technological applications require large current densities in high applied magnetic fields. The magnetic behavior of the high temperature superconductors has been the subject of intense study since their discovery. When a high temperature superconductor is placed in a magnetic field, the magnetic flux penetrates the sample in the form of flux bundles, called vortices. The vortices move through the superconductor if an electrical current larger than the critical current is applied. The motion of the vortices causes electrical resistance. However, the vortices can be pinned by defects in the sample. By increasing the number of defects in the sample, the critical currents can be increased. The interaction between the vortices and the defects is not yet completely understood. Radiation damage has been extensively used to investigate this interaction because it allows control over the number and type of defects introduced [3-10].

Historical Background

After superconductivity was discovered, continuous progress was made: new phenomena related to superconductivity were discovered, theory was developed, and materials having a higher transition temperature were created. The perfect diamagnetism of superconductors was discovered in 1933 [1], and the microscopic theory of superconductivity was completed by Bardeen, Cooper, and Schriefer (BCS) in 1957 [2, 3]. In 1985, the highest transition temperature was at 23 K for the intermetallic Nb₃Ge [4].
In 1986, after a systematic search, Bednorz and Muller discovered that the layered copper-oxide material La$_{2-x}$Ba$_x$CuO$_{4-y}$ becomes superconducting at temperatures above 30 K [5]. This discovery gave research in superconductivity enormous new momentum, and led to the conception of a whole new generation of cuprate superconductors, now commonly named the high $T_c$ superconductors. The YBa$_2$Cu$_3$O$_{6.6}$ compound, which went superconducting at ~ 93K, was discovered in 1987 [6]. Transition temperatures up to 133 K [7] and as high as 164 K [8] under hydrostatic pressure, have recently been reported. Even higher transition temperatures seem possible, which would make potential applications of superconductivity economical, due to the reduced costs for coolants. Moreover, the intensive research of the new superconductors has led to the discovery of many new phenomena of fundamental interest to solid state physics.

Much of the research on cuprate superconductors has focused on YBa$_2$Cu$_3$O$_{6.6}$ (YBCO), which is the first copper-oxide material discovered to be superconducting above the boiling point of nitrogen [6]. Figure 13 shows the crystal structure for YBa$_2$Cu$_3$O$_{6.6}$. Like most of the cuprate high-$T_c$ superconductors, YBa$_2$Cu$_3$O$_{6.6}$ has a unit cell containing planes of CuO$_2$, which are believed to be responsible for the superconductivity in this material. Layers of BaO and chains of CuO, which act as reservoirs of charge carriers for the CuO$_2$ planes, separate the CuO$_2$ planes. The layered structure of these materials leads to an anisotropy of the superconducting parameters. The anisotropy of the superconducting state can be expressed in terms of $\gamma=(m_c/m_{ab})^{1/2}$, where $m_{ab}$ and $m_c$ denote the effective masses of the quasi-particles for motion in the CuO$_2$ layers and perpendicular to these layers, respectively. The anisotropic character of these materials is apparent in transport measurements and will be discussed later in this chapter.
Fundamental Parameters

When a superconductor is cooled below its transition temperature $T_c$, in a magnetic field, the magnetic flux is expelled. Thus, in a weak magnetic field, a superconductor exhibits perfect diamagnetism, a phenomenon called the Meissner effect. The Meissner effect implies that in a magnetic field, the superconductors develop a surface current, which gives rise to magnetic fields that cancel the external field, leaving a field-free bulk. The surface currents that shield the superconductor of the outside magnetic field are present over a distance $\lambda$, called the penetration depth.

The phenomenology of superconductivity is described by the Ginzburg-Landau model [9]. This theory describes superconductivity in terms of a superconducting wave function $\psi$, and studies its variations in time and space. The wave function can be written as $\psi = |\psi|e^{i\phi}$, where $|\psi|$ is the density of the superconducting electrons, and $\phi$ is the phase. The density of the superelectrons cannot change arbitrarily with position, but decays over a distance called the coherence length $\xi$. The Ginzburg Landau parameter is the ratio of the penetration depth and the coherence length, $\kappa = \lambda/\xi$. The value of the Ginzburg Landau parameter dictates whether the superconductor is of type I or type II. For $\kappa < 1/\sqrt{2}$ the superconductor is of type I, while for $\kappa > 1/\sqrt{2}$ the superconductor is of type II.

Type I Superconductors.

The magnetic phase diagram of a typical type I superconductor is shown in Figure 1. Here $H_c$ represents the critical field, above which superconductivity is destroyed. The material is superconducting and in the Meissner state for fields and temperature values below and to the left of the $H_c$ vs. $T$ curve, while at higher fields
and temperatures the material is in its normal state. The experimental results for the 
$H_c$ vs. $T$ curve can be approximately described by the quadratic temperature 
dependence, $H_c(T) = H_{c0}[1-(T/T_c)^2]$, where $H_{c0}$ is the critical field at zero temperature.

![Magnetic Phase Diagram of Type I Superconductors.](image)

**Figure 1.** Magnetic Phase Diagram of Type I Superconductors.

**Type II Superconductors.**

The standard magnetic field temperature ($H$-$T$) phase diagram for a 
conventional type II superconductor is shown in Figure 2. In this phase diagram three 
phases may be distinguished: 1) In the normal phase above the upper critical field 
$H_{c1}(T)$, the superconductor behaves like a normal metal; 2) Below the lower critical 
field $H_{c2}(T)$, the superconductor is in the Meissner phase, i.e., the magnetic field is 
totally expelled from the interior of the superconductor; 3) Between $H_{c2}(T)$ and
$H_c(T)$, it is in the mixed state where the magnetic field penetrates the superconductor, in the form of flux bundles, called vortices. Each vortex carries exactly one magnetic flux quantum $\Phi_0=\hbar c/2e$. The magnetic field is screened from the rest of the superconductor by a local supercurrent, circling around the vortex.

![Magnetic Phase Diagram of Type II Superconductors.](image)

The vortices (see Figure 3) can be described as containing a core region of radius $\xi$, where the density of superconducting electrons is zero at the center and increases exponentially over the characteristic distance $\xi$. Within the core the magnetic field is at its maximum value and decays over a distance $\lambda$ from the core center. Both the penetration depth $\lambda$ and the coherence length $\xi$ depend on the temperature: $\lambda(T)=\lambda(0)/(1-T/T_c)^{1/2}$ and $\xi(T)=\xi(0)/(1-T/T_c)^{1/2}$, where $\lambda(0)$ and $\xi(0)$
represent the penetration depth and the coherence length, respectively, at zero temperature.

![Vortex Line With Nonsuperconducting Core](image)

Figure 3. Vortex Line With Nonsuperconducting Core \( \xi \).

The repulsive interaction between vortices makes the vortices arrange themselves in a triangular lattice (see Figure 4), called the Abrikosov lattice [10]. The intervortex spacing \( a_n \) is approximately given by \( a_n = (\Phi_0 / B)^{1/2} \), with \( B = H \). As the applied magnetic field is increased, the density of vortices increases, the distance between them decreases, until the vortices overlap and the superconductor is driven into the normal state. An applied current \( J \) exerts a Lorentz force on the vortex, given by:

\[
\vec{F}_L = J \times \frac{\Phi_0}{c}
\]  

The Lorentz force causes the vortices to move and if there is some viscous force opposing their motion through the material, work must be done in order to maintain this motion. This work can only be supplied by the transport current, and energy must be expended in driving the current through the material. This movement
will therefore create resistance and dissipation and an ideal type II superconductor does not have zero resistance in the vortex lattice mixed state, because an applied current will induce a flow of the whole vortex lattice.

![Figure 4. Abrikosov Lattice.](image)

Real superconductors, however, always contain imperfections in the crystal that tend to pin the vortices. If the Lorentz force is less than the pinning force, the vortices remain stationary. Because of this pinning, vortex motion is inhibited and superconductivity is restored.

**High $T_c$ Superconductors.**

All high $T_c$ superconductors are type II superconductors. However, their vortex phase diagram is much more complex than that of conventional type II superconductors. Figure 5 displays some of the pertinent features of the phase diagram of high $T_c$ superconductors.
Figure 5. Magnetic Phase Diagram of High Temperature Superconductors.

The vortex liquid, situated at high temperatures, in the mixed state, encompasses a large portion of the phase diagram. This liquid phase is characterized by the absence of a shear modulus, which prevents effective flux pinning. Thus the development of a critical current density is shifted to lower temperatures where the liquid undergoes a phase transformation into a vortex solid, analogous to the water to ice transition. This melting transition has been the focus of many studies, and it has been investigated with both dynamic [11-15] and thermodynamic measurements [16-18]. The presence of disorder drastically affects the phase diagram. In clean samples, the transition from the vortex lattice to the vortex liquid was shown to be first order.
The introduction of disorder into the system suppresses the first order phase transition [21-23] and suppresses the formation of a vortex lattice. Various glassy vortex states have been suggested in samples containing disorder, including the vortex glass [24], Bose glass [25, 26], splayed glass [27], and polymer glass [28] states, depending on the type of disorder. Phase transitions are an area of fundamental interest in solid-state physics. Due to the fact that the phase transitions in these materials are strongly affected by the introduction of defects, high T_c superconductors are ideal for studying the role of disorder in phase transitions (for a review, see [29]).

Characteristics of Clean Crystals of YBa\(_2\)Cu\(_{1-x}\)O\(_{y}\)

Temperature Dependence of the Resistivity

The enhanced mobility of the vortices at high temperatures where the vortices are in the liquid state produces dissipation that is easily seen in current-transport measurements. The resistive transition becomes broader in the presence of an applied magnetic field and the zero resistance point shifts to lower temperatures with increasing field. Figure 6 shows an example of the resistive broadening for magnetic fields H =0, 0.5, 1, 2, 3, 4, 5, 6, 7 and 8T applied parallel to the c-axis (top panel) and to the ab plane (bottom panel) of an untwinned single crystal. Note that only the 0T and the 8T curves are explicitly indicated on the figure, due to the lack of space. As H is varied from 0 to 8T, the curves shift to lower temperatures. (e.g., the 0.5T lies to the left of the 0T curve and the 1T curve lies to the left of the 0.5T curve). The figure shows some of the general features of the resistive transition. The transition becomes broader and "fan-shaped" with increasing magnetic field.
Figure 6. Resistivity in Applied Magnetic Fields for $H \parallel c$ (Top Panel) and for $H \parallel ab$ (bottom panel) for an Untwinned Single Crystal of YBa$_2$Cu$_3$O$_{7-x}$ for $H = 0, 0.5, 1, 2, 3, 4, 5, 6, 7$ and $8T$. 

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The broadening of the transition above $\rho > 0.5 \rho_n$, where $\rho_n$ is the normal state resistivity, is usually attributed to thermodynamic fluctuations in the order parameter. In high $T_c$ superconductors, the effects of fluctuations are much enhanced due to the high values of $T_c$, short coherence lengths, and the large anisotropies. The Ginzburg number $G_i$ is a measure of the importance of thermal fluctuations and it gives the temperature range over which thermal fluctuations are important by $\Delta T \approx G_i$. The Ginzburg number is given by:

$$G_i = \frac{1}{2} \left( \frac{\rho T_c}{H_i^2} \right)^2$$

(1.2)

where $H_i$ is the thermodynamic critical field, and $G_i(H) = G_i \left( \frac{H}{H_{ci}} \right)^{\frac{1}{2}}$, where $H_{ci}$ is the upper critical field. In conventional type II superconductors $G_i \approx 10^{-7}$, while in YBCO $G_i$ is much larger, $G_i \approx 10^{-3}$. In YBCO, $T_c \approx 90$ K, $\xi_{ab} \approx 16 \text{ Å}$, $H_{ci} \approx 120$ T.

The anisotropy parameter $\gamma$, defined earlier in this chapter, can also be written as:

$$\gamma = \frac{1}{2} \left( \frac{\lambda}{m_{ab}} \right)^{1/2} = \frac{\lambda_c}{\lambda_{ab}} = \frac{\xi_{ab}}{\xi_c}$$

(1.3)

where $\lambda_{ab}$, $\lambda_c$, $\xi_{ab}$, and $\xi_c$ are the penetration depth and the coherence length in the ab plane and along the c axis, respectively. For YBCO $\lambda_{ab}(0) \approx 1000 \text{ Å}$, $\lambda_c(0) \approx 7000 \text{ Å}$, $\xi_{ab}(0) \approx 16 \text{ Å}$, and $\xi_c(0) \approx 3 \text{ Å}$ and $\gamma \approx 7$. Due to the anisotropy of the material, the broadening of the transition is more pronounced when the magnetic field applied is parallel to the c axis than when it is parallel to the ab-plane, as seen in Figure 6.

For both $H || c$ and $H || ab$, the resistivity in applied magnetic field shows a smooth decrease at higher temperatures, followed by a sudden kink at lower temperatures where the resistivity drops very abruptly to zero. The kink in the
resistivity is associated with a first-order melting transition of the vortex lattice to a vortex liquid, usually observed only in clean single crystals. By determining the melting temperature at each magnetic field, the position of the melting line in the phase diagram can be found. As seen in Figure 7, the melting line is shifted to much higher temperatures when the orientation of the magnetic field is changed from $H \parallel c$ to $H \parallel ab$. For both orientations of the field, the melting line can be fitted to an equation of the form

$$H = H_n \left( 1 - \frac{T}{T_{c0}} \right)^\alpha$$

where $T_{c0}$ is the zero-field transition temperature, and $H_n$ and $\alpha$ are fitting parameters.

![Figure 7. Melting Line as Determined From the Vortex Melting Temperature for $H \parallel c$ and $H \parallel ab$.](image-url)
As discussed by Blatter et al. [28], the power-law fit given in equation (1.4) is a compact form for expressing the behavior of the melting line, where \( \alpha \) is an "effective" exponent. Near \( T_c \), at low fields, theory [30, 31] predicts an exponent \( \alpha \approx 2 \) for the melting transition. However, at high fields, this exponent is expected to become closer to unity [30]. Paulius et al. [32] found values of \( H_{hc} = 77 \pm 5 \text{T} \), \( \alpha_c = 1.23 \pm 0.03 \) for \( H \parallel c \) and \( H_{nab} = 611 \pm 22 \text{T} \), \( \alpha_{ab} = 1.24 \pm 0.04 \) for \( H \parallel ab \). The ratio \( H_{nab}/H_{hc} \) yields the anisotropy ratio \( \gamma \). An anisotropy of \( \gamma = 7.6 \pm 0.6 \) was found in the previous work [32], consistent with the range of values (5 \( \leq \gamma \leq 10 \)) obtained by other techniques [33-38].

Current-Voltage Characteristics

At high transport currents, where the pinning forces are negligible when compared to the Lorentz force, the forces on an isolated vortex are given by:

\[
\frac{Jc}{\Phi_0} f(t)v_n = 0 \tag{1.5}
\]

where \( f(t) \) is a coefficient that describes the frictional drag force exerted on the vortex and \( v_n \) is the vortex velocity. In this model, the vortex velocity is proportional to the applied current density and the dissipative flux flow resistivity is given by:

\[
\rho_{ff} = \frac{v_n B}{Jc} = \frac{B\Phi_0}{fc^2} \tag{1.6}
\]

If the drag force does not depend on the Lorentz force, the resistivity is then current independent, and therefore Ohmic. In this regime the flux flow resistivity \( \rho_{ff} \) is related to the normal state resistivity by \( \rho_{ff} = \rho_n B/H_{c2} \), by the Bardeen-Stephen model [39], which yields \( \rho_{ff} = \rho_n B/H_{c2} \), where \( B \) is the magnetic induction and \( H_{c2} \) is the upper critical field.
Vortices are affected by defects in the superconductor, which act as pinning centers. For low transport currents, the pinning force may approach the Lorentz force, and equation (1.5) is no longer true, because the pinning force must be included. The low current regime is described by the thermally activated flux flow (TAFF) model [40], which is represented in terms of a perturbation to the flow velocity of the vortices. According to this model, the retardation of vortex motion is described by an energy barrier to motion $U_p$, which is large compared to the thermal energy $k_B T$. The vortex is retarded by an amount $\delta v << v$, reducing the resistivity as

$$\frac{\rho}{\rho_n} \frac{\delta v}{v}$$

where the fraction $\delta v/v$ depends on the applied current. At low current densities, when the Lorentz force is less than the pinning force, the TAFF model assumes a thermally-activated vortex motion described by $\delta v/v = \Lambda \exp (U_p/k_B T)$, where $\Lambda \gg 1$. and equation (1.7) becomes

$$\rho = \frac{\rho_n}{A} e^{-\Lambda > 1}$$

At low current densities the TAFF model predicts ohmic behavior, with a small resistivity $\rho << \rho_{\text{eff}}$. For large current densities the Lorentz force overwhelms the pinning force such that $\delta v/v \to 0$ and flux flow resistivity is restored. Figure 8 describes this behavior.

At high temperatures, where the thermal energy is higher than the pinning energy, the vortex is in the flux flow state, and the current-voltage characteristics are Ohmic, as described by the Bardeen-Stephen model.

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Figure 8. Current-Voltage Characteristics in the Vicinity of the Melting Transition.

Figure 9. The Tail of the Resistivity for $H = 4T \parallel c$ in Applied Currents of $I = 0.1$, 1, and 5 mA. The Arrow Represents the Kink in the Resistivity at the Melting Transition.
When the temperature is lowered and the thermal energy is comparable with the pinning energy, the current-voltage characteristics develop an S-shape, as the flow crosses over from TAFF flow at low currents to flux flow at high currents, as shown in Figure 9.

Above the kink in the resistivity, indicative of the liquid-to-solid transition, the resistivity is Ohmic, while below the kink, the resistivity is highly non-Ohmic, and a higher resistivity is observed for higher currents.

Angular Dependence of the Resistivity

The resistivity of the material can be measured while fixing the temperature and changing the angle $\theta$ between the applied field and the c-axis of the crystal. The angular behavior of the resistivity $\rho(\theta)$ for an untwinned YBCO single crystal is shown for $H=0.5\,\text{T}$ in Figure 10.

![Angular Dependence of the Resistivity](image.png)

Figure 10. Angular Dependence of the Resistivity.
Also seen in the figure is the configuration of the applied magnetic field \( H \) and of the applied current \( J \). The magnetic field is rotated in a plane perpendicular to the current \( J \), such that the Lorentz force is always at its maximum value. The magnetic field was tilted from \( 90^\circ \) (\( H \parallel b \)-axis) to \( 0^\circ \) (\( H \parallel c \)-axis) at a temperature \( T=91.03 \text{K} \), in an applied field \( H=0.5 \text{T} \). The resistivity is a smooth function of the angle. Note that the resistivity is largest when the field is aligned parallel to the \( c \)-axis and is a minimum when the field is parallel to the \( b \)-axis. This angular dependence of the resistivity arises from the anisotropy of the material. In \( \text{YBa}_2\text{Cu}_3\text{O}_y \), the mass anisotropy ratio \( (m_c/m_{ab}) = \tau \) lies in the range 25-100 [33-38] and it depends on the oxygen concentration.

**Vortex-Defect Interaction**

The superconducting coherence length at zero temperature for \( \text{YBa}_2\text{Cu}_3\text{O}_y \) crystals for fields parallel to the \( c \)-axis is \( \xi_0=16 \text{Å} \) [35]. The coherence length varies with the temperature as \( \xi=\xi_0(1-T/T_c)^{1/2} \), and therefore the coherence length for \( \text{YBa}_2\text{Cu}_3\text{O}_y \) varies from \( \approx 16 \) to 100 Å throughout most of its superconducting temperature range, up to 0.98 \( T_c \). The normal vortex radius is equal to the coherence length. When the normal (non-superconducting) core is created, the free energy per unit length increases by the vortex condensation energy:

\[
\frac{1}{8\pi} \int \mathbf{h} \cdot d\mathbf{A} = \frac{H_c^2}{8\pi} \pi \xi^2
\]

(1.9)

where \( H_c \) is the thermodynamic critical field. Therefore the magnetic flux vortex can be pinned by a defect in the superconductor. This is because the vortex requires energy to penetrate the superconductor and drive the core normal. If the vortex
penetrates a region where a defect exists, it doesn't have to drive the region of the defect normal and thus saves energy. Figure 11 shows a schematic diagram of magnetic flux vortices in a superconductor with point and columnar defects. The most effective pinning occurs when the size and shape of the defect matches that of the vortex.

![Vortex Pinned by Point Defect](image)

![Magnetic Vortices](image)

![Current](image)

![Vortex Pinned by Columnar Defect](image)

Figure 11. Schematic Diagram of Magnetic Flux Vortices in a Type II Superconductor With Point and Columnar Defects.

How a vortex is pinned by a defect also depends on the volume of the intersection between the vortex and the defect, so the shape of the defect plays an important role. The shape of the defects can range from point-like to columnar. Point defects are introduced by irradiation with protons [21, 23, 41-43] or electrons [22, 44-46] and columnar defects are introduced by irradiation with heavy ions [32, 47-50].
As seen in Figure 12 the intersection volume between a point defect and a vortex has no dependence on the angular orientation of the vortex with respect to the point defect. In contrast, in the case of columnar defects, the intersection volume varies with the angular orientation of the vortex, going from a maximum when the defect and the vortex are aligned with each other, to a minimum when they are perpendicular to each other. From the angular dependence of this interaction important properties about the vortex and the defect can be extracted, such as the vortex stiffness and the pinning strength of the defect.

![Figure 12. Schematic Diagram of Magnetic Flux Vortices in a Type II Superconductor With Point and Columnar Defects.](image)

The pinning strength can also be affected by the size of the defect. Irradiation with different types of ions produces correlated defects of different sizes and morphologies. The density of defects also affects the pinning strength. By changing the total irradiation dose we can control the density.
Scope and Summary of the Thesis

The purpose of this work is to gain more insight into the dynamics of the vortices in YBCO crystals with defects. The experiments conducted reveal the great versatility and potential of these systems. Some of the experimental work is geared towards the study of phase transitions, which are of great interest in solid state physics. Other work deals with the interaction between vortices and columnar defects for various doses, defect morphologies, and orientations. Experimental work has also focused on a more applied aspect, the thermal stability of columnar defects.

Chapter II reviews the experimental details of our work and describes the technical details of how we obtained our data. We discuss the crystal growth, sample preparation, irradiation procedures, and measuring techniques.

In Chapter III we focus on the experimental evidence of the vortex glass phase in proton irradiated YBa$_2$Cu$_3$O$_{6.5}$. The chapter recapitulates the theory of the vortex glass and gives a description of the procedure that was used to perform the scaling analysis of the resistivity. The resulting scaling exponents and the phase diagram are presented. For the first time, evidence for a vortex glass transition is detected in an untwinned single crystal of YBa$_2$Cu$_3$O$_{6.5}$ with induced point-like disorder. Our results suggest that sufficiently high pinning disorder is required in order for the vortex glass phase to be observed. A summary of this work was published in Physical Review Letters [21].

Chapter IV studies the effects heavy ion irradiation on the phase diagram. We study the effects of the irradiation dose, irradiation ion type, and defect orientation on pinning. Columnar defects were introduced in YBa$_2$Cu$_3$O$_{6.5}$ in two different orientations: (1) parallel to the c-axis by irradiation with 1.4 GeV Uranium ions at three different doses and with 3.9 GeV Gold ions; (2) parallel to the ab plane by
irradiation with 1.4 GeV Lead ions. We present a comparison between the three different Uranium doses. We find a consistent change of slope at the matching field and a remarkable pinning anisotropy. We also compare the samples irradiated with Uranium and Gold at the same dose. We find that Uranium ions produce much stronger pinning centers than Gold ions. The differing pinning capabilities of the two types of ions are discussed in terms of the morphology of the defects they produce. We conclude with the results of the irradiation parallel to the ab plane. The irradiation parallel to the ab plane is the first experiment of this kind and we focus on the effects of the irradiation on the irreversibility line.

Chapter V studies the thermal stability of columnar defects introduced by irradiation with 3.9 GeV Au\(^{27+}\) in YBCO single crystals. The dependence of the critical current density on the annealing temperatures is correlated with changes in the microstructure of the defects, as seen from transmission electron microscopy (TEM) measurements. We find that the defects induced by irradiation are stable upon thermal annealing up to temperatures of 600 °C and that the critical current density at temperatures \(T \geq 45\) K can actually increase with thermal annealing. This work is currently in preparation to be submitted to Physical Review B.


CHAPTER II

EXPERIMENTAL METHODS

The YBa$_2$Cu$_{x}$O$_{y}$ Compound

Figure 13 shows the crystal structure of YBa$_2$Cu$_{x}$O$_{y}$, which was determined using X-ray diffraction [1] and neutron diffraction [2, 3].

![Figure 13. Structure of the YBa$_2$Cu$_{x}$O$_{y}$ Unit Cell.](image)

The material has an orthorhombic unit cell with axes $a=3.83\text{Å}$, $b=3.88\text{Å}$, $c=11.68\text{Å}$. It can be seen in Figure 13 that the structure has a planar nature. It
consists of a regular stacking of CuO, BaO, and Y layers. In the top and bottom planes, the oxygen is present only along the b direction, forming the so-called Cu-O chains. When the YBa$_2$Cu$_3$O$_{6.8}$ compound is grown, it starts out with a tetragonal unit cell, where there are no Cu-O chains, but only copper atoms in the top and bottom planes of the unit cell. As the samples are cooled, they absorb more oxygen, and at 680 K they undergo a transition from the tetragonal to the orthorhombic structure. The oxygen concentration plays an important role in the superconducting properties of the YBa$_2$Cu$_3$O$_{6.8}$ compound. This compound is superconducting for $0 \leq \delta \leq 0.6$. The transition temperature also depends on the oxygen concentration. The oxygenation state with the highest transition temperature is obtained for $\delta \approx 0.06$, and it represents the optimally doped state [4]. An important defect is related to the phase transition from the tetragonal (at high temperatures) to the orthorhombic structure at low temperatures. In order to reduce the stress associated with the CuO chains the a and b direction interchange at so-called twin boundaries (see Figure 14). The twin boundaries consist of strain fields of $\approx 30$ Å in size along the <110> direction.

Crystal Growth

Growth in Gold Crucibles

Single crystals of YBa$_2$Cu$_3$O$_{6.8}$ were grown using the self-flux method [5]. We begin with high purity compounds, BaCO$_3$ (99.999 % pure), CuO (99.999 % pure) and Y$_2$O$_3$ (99.999 % pure). A total of about 7.5 grams of these ingredients are weighed with an Y : Ba : Cu molar ratio of 5:27:68. The mixture is then ground with an agate mortar and pestle until a uniform powder is obtained. The powder is then pressed with a hydraulic press to a pressure of $\approx 3000$ psi into a 3/4" diameter, 1/4" tall pellet. The pellet is then placed in the center of a 3"x3"x1.5" gold crucible. The
The crucible is placed in a box furnace, centered at 10" from the furnace door, in order to avoid temperature gradients. The crucible is heated up from room temperature to a maximum of 983 °C and cooled slowly. Table 1 shows the temperature program used to grow the crystals in the gold crucibles.

Table 1
Temperature Program for Growth of Single Crystals of YBa$_2$Cu$_4$O$_{y-6}$ in Gold Crucibles

<table>
<thead>
<tr>
<th>Ramp (°C/hour)</th>
<th>Temperature Level (°C)</th>
<th>Dwell Time (hours)</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>861</td>
<td>0.0</td>
</tr>
<tr>
<td>50</td>
<td>889</td>
<td>7.0</td>
</tr>
<tr>
<td>50</td>
<td>917</td>
<td>9.0</td>
</tr>
<tr>
<td>110</td>
<td>983</td>
<td>1.1</td>
</tr>
<tr>
<td>1</td>
<td>866</td>
<td>0.0</td>
</tr>
<tr>
<td>200</td>
<td>24</td>
<td>END</td>
</tr>
</tbody>
</table>

The program takes about 6 days to complete. When the program ends, the gold crucible is pulled out of the furnace and the single crystals are visible in the crucible. The cylindrical pellet in the center of the crucible has partially melted and resolidified and it remains at the center of the crucible. The crystals grow everywhere on the bottom of the crucible, but the highest concentration is in the corners of the crucible. The crystals are extracted from the crucible by turning the crucible upside down and gently tapping from the back. Besides the YBa$_2$Cu$_4$O$_{y-6}$ single crystals, a lot of flux and CuO single crystals fall out of the crucible. However, the YBa$_2$Cu$_4$O$_{y-6}$
single crystals are easy to identify by their typical rectangular parallelepiped shape and black shiny surfaces.

**Growth in Yttrium-Stabilized Zirconia Crucibles**

We begin with the same high purity compounds used in growing single crystals in the gold crucible. A total of about 3.5 grams of these ingredients are weighed with a Y:Ba:Cu molar ratio of 1:4:10. The mixture is ground with mortar and pestle until a uniform powder is obtained. The Zirconia (ZrO$_2$) crucible is placed tilted, with one of the short edges lying on the edge of another larger Zirconia crucible and the powder is then loosely placed in the high end of the tray. The crucibles are then placed in the center of the box furnace (the back of the tray should be at about 12" from the furnace door), and heated up to a maximum of 980 °C. Table 2 shows the temperature program used to grow the crystals in Zirconia crucibles.

This program takes less than 2 days to complete, and it is very useful when new samples are needed fast. When the program ends, the Zirconia crucible is pulled out of the furnace and the single crystals are visible at the low end of the crucible. The crystals grow mostly in the corners of the crucible. The Zirconia crucible is not pliable, like the gold crucible, and tapping the crucible in order to extract the crystals may break the crucible. We use a scalpel in order to dislodge the samples from the crucible. In using this program, the YBa$_2$Cu$_3$O$_{6.5}$ single crystals are not as embedded in flux and they are therefore relatively easy to harvest. However, the samples grown in the Zirconia crucibles are thinner, and often curved, which makes it difficult to detwinn them. Therefore, the crystals grown in the Gold crucibles were better suited for our uses.
Table 2

Temperature Program for Growth of Single Crystals of
YBa$_2$Cu$_3$O$_{6.8}$ in Zirconia Crucibles

<table>
<thead>
<tr>
<th>Ramp (°C / hour)</th>
<th>Temperature Level (°C)</th>
<th>Dwell Time (hours)</th>
</tr>
</thead>
<tbody>
<tr>
<td>step (maximum power)</td>
<td>300</td>
<td>0.0</td>
</tr>
<tr>
<td>100</td>
<td>880</td>
<td>0.0</td>
</tr>
<tr>
<td>25</td>
<td>980</td>
<td>1.0</td>
</tr>
<tr>
<td>6.2</td>
<td>880</td>
<td>0.0</td>
</tr>
<tr>
<td>100</td>
<td>25</td>
<td>END</td>
</tr>
</tbody>
</table>

The single crystals grown through both the techniques described above are oxygen deficient, their superconducting transition is about 30K wide with an onset at ~80K, as measured by a low field SQUID (Superconducting Quantum Interference Device) magnetometer [6]. The samples require annealing in oxygen gas to optimize their superconducting properties. We clean the crystals of any flux from their surfaces with a razor and ethyl alcohol. The samples need to be clean, such that the oxygen can more readily enter the samples. The crystals are put in a quartz crucible, and placed in a quartz tube inside a tube furnace and oxygen gas flows through the tube at a rate of ~ 2 cm$^3$/min. The temperature is set to 420 °C and held there for approximately ten to fourteen days. The crystals are then slowly cooled to room temperature. This procedure yields optimally doped samples.
Crystal Detwinning

After the annealing the crystals typically display a $T_c \approx 93$ K, but contain planar defects in the form of twin boundaries. As explained earlier in this chapter, at the tetragonal to orthorhombic phase transition, in order to reduce the stress associated with the CuO chains, the a and b direction interchange and create the twin boundaries (see Figure 14).

Figure 14. Schematic of the Twin Boundary Plane in YBa$_2$Cu$_3$O$_{6.8}$.

The figure shows the Cu and O atoms and their orientation in the top and basal planes of the unit cell of YBa$_2$Cu$_3$O$_{6.8}$. The twin boundaries are planar structures that typically extend through the entire thickness of the crystal. The twin boundaries in the as-grown crystals can be viewed with a polarized light microscope, as seen in Figure 15. In the left part of the figure we show a twinned crystal, while on the right, an untwinned one. When viewed with a polarized light microscope, the twin...
boundaries separate two different colored regions where the basal plane is rotated by 90 degrees.

Twinned Untwinned

Figure 15. Polarized Light Microscope Views of a Twinned and Untwinned Crystal.

The twin boundaries act as highly anisotropic defect planes, and unless the twin boundaries are removed, the effects due solely to the defects introduced by irradiation are difficult to isolate. Therefore, it is necessary to remove the twin boundaries. The twin boundaries are removed by applying uniaxial pressure in the ab plane [7, 8]. The crystal is placed in a detwinning device where pressure is applied along one axis of the crystal. An adjustable micrometer (shown in Figure 16) controls the applied pressure. The device is placed in a furnace and heated to 420°C in flowing oxygen gas. The oxygen mobility varies with the temperature [4]. The samples have already been annealed for 10-14 days at 420 °C, and by performing the detwinning at the same temperature, we ensure that the oxygen concentration in our samples stays the same.
The applied uniaxial pressure is typically of the order of $\sim 10^7$ N/m$^2$. The pressure favors the domains that have their a-axis parallel to the direction of compression. By applying uniaxial pressure, the domains with the a-axis aligned with the uniaxial stress direction extend throughout the entire crystal, hence removing the twin boundaries.

For heavy ion irradiation experiments, these crystals must be thinned to less than $\approx 30$ µm along the c-axis (for the typical irradiation energies usually employed in our experiments) in order to ensure that the heavy ions produce continuous and straight defect tracks throughout the sample. The crystal is thinned by mounting it on a metal holder, and then grinding it, using polishing discs and diamond-in-oil suspensions. Using this method, flat millimeter-sized crystals less than 30 µm thick can be obtained.
Transport Measurements

Transport measurements are conducted using the standard four-probe technique. Low resistance contacts are necessary in order to conduct this type of measurements. Four gold pads, approximately 2000Å thick, are deposited on the crystal by gold evaporation in an evaporation chamber. When first deposited, the gold pads have a high contact resistance. Hence the gold pads are subsequently sintered at 420°C in flowing oxygen for approximately six hours to ensure good bonding of the gold to the ceramic crystal surface. Gold wires 25 μm in diameter are attached to the gold pads using silver epoxy, then cured for 5-10 minutes at relatively low temperatures, usually around 150°C, resulting in final contact resistances of about 1 Ω.

The cryostat used for characterizing the single crystals at low temperatures (described in more detail elsewhere [9]) is shown in Figure 17. The cryostat is a home built 4He system capable of achieving temperatures as low as 0.47 K, but adapted for use in our experiments with 4He. The main sections of the 4He cryostat are shown in the figure. It is a top loading system, with a sample chamber diameter of 0.75". The midsection of the sample chamber is surrounded by and in contact with a 1 K pot, which can hold 1 liter of liquid helium, used mainly for low temperature experiments below 1 Kelvin. The tail of the sample chamber is built of non-magnetic stainless steel and it is isolated from the liquid helium bath by an inner vacuum can.

Two NbTi superconducting magnets surround the tail of the cryostat: A 1.5 Tesla transverse split coil magnet resides in the 2.75" diameter bore of an 8.0 Tesla longitudinal magnet. The two superconducting magnets supply the orthogonal fields, which are controlled separately to give the desired magnitude and orientation of the applied field. The following steps define the cooling operation down to the 70 –
Figure 17. The Cryostat System.
100 K temperature regime typically employed in studying the high temperature superconductor YBa$_2$Cu$_3$O$_{6.8}$. The sample chamber, inner, and outer vacuum cans are evacuated with a rotary pump/diffusion pump system to about $10^{-6}$ torr. The bath of the dewar is then filled to capacity with liquid nitrogen to pre-cool the superconducting magnets. Subsequently, the liquid nitrogen is removed by supplying a back pressure of nitrogen gas. When all the liquid is displaced, the bath is flushed several times with $^4$He gas to ensure that no liquid nitrogen remains at the bottom of the dewar. Next, the bath is filled with liquid $^4$He to capacity. The sample chamber is filled with $^4$He gas to a pressure of $\sim 100$ mm Hg, and cooling occurs via conduction and radiation from the liquid $^4$He in the bath. Temperature control is maintained locally by a non-inductively wound phosphor bronze heater wire wrapped around a copper cap which surrounds the sample.

The tail of the resistivity probe consists of a Cernox thermometer calibrated from 300K to 0.33 K, an eight pin receptacle for the sample, and a cap heater consisting of a copper cap wrapped non-inductively with 36 gauge Phosphor Bronze heater wire. Some of the key features of the experimental setup are good thermal contact between the sample and the thermometer, and slow cooling rates, which ensure that the sample and the thermometer are at the same temperature.

The sample holder consists of a G-10 insulating body and Phosphor Bronze posts (see Figure 18). The YBCO single crystals for resistivity measurements are typically mounted on a sapphire or glass substrate with silver epoxy. The substrate with the sample is then attached to the G-10 disk with varnish. Contacts from the sample to the posts are made with 25 µm gold wire which are attached with silver epoxy to the sample, and attached to the posts with indium solder. The G-10 disk with the sample attaches to a commercially available 8-pin receptacle at the bottom of
the resistivity probe. Connection from the sample to the top of the probe is made with 24 twisted pairs of 38 gauge copper wire, terminating at a hermetically sealed 26 pin connector at the top. An O-ring slip connection, which connects the probe to the cryostat, enables the probe to be rotated 360°. This rotation allows an extra degree of freedom for the sample orientation with respect to the transverse magnetic field provided by the 1.5T superconducting split coil magnet.

![Crystal mounted on holder.](image)

Figure 18. View of the Sample and Sample Holder.

We use the standard four-probe geometry for measuring the AC resistivity. AC current is generated with a function generator. The superconducting sample is mounted in series with a 1 kΩ resistor. The resistance of the superconducting sample is typically 0.1 Ω at room temperature and it drops to zero below the transition temperature. Since the total resistance of the superconducting sample plus the 1 kΩ resistor varies by less than 0.1% due to the change in temperature, this series arrangements enables the function generator to provide a constant current source to the sample. An additional 0.1 Ω resistor in series with the sample is used to monitor the sample current independently. The sample voltage leads are each connected to a
lock-in amplifier. For DC resistivity measurements, the function generator and the associated resistors are replaced with a DC current source and the DC voltage is measured using a nanovoltmeter, making sure to reverse the current direction several times to avoid thermal drift voltages. The AC measurements are performed at low frequencies (23 Hz) in order to avoid the impedance effects present at higher frequencies. The advantage of the AC measurements consists in the fact that the lock-in amplifier can average out spurious noise signals from the measurement.

The data acquisition system consists of a desktop computer connected with the laboratory instruments via an IEEE 488.2 GPIB interface, running either C++ or LabView data acquisition programs.

Crystal Irradiation

In order to study the interaction between vortices and defects we introduce point defects by irradiation with protons and columnar defects by irradiation with heavy ions. The proton irradiations were performed using Western Michigan University's tandem accelerator. The heavy ion irradiations were performed at two facilities: The 36' diameter ATSCAT chamber at the Argonne Tandem Linear Accelerator System (ATLAS), located at Argonne National Laboratory, and the N3 chamber at the National Superconducting Cyclotron Laboratory (NSCL) at Michigan State University. The basic chamber configuration is shown in Figure 19. The samples are placed on a ladder, which is located at the center of the chamber. The ladder consists of several rectangular aluminum plates stacked vertically on top of one other, as seen in Figure 20. Samples are mounted onto the aluminum plates, except for the topmost plate, which contains a fixed diameter hole drilled into the plate.
A Faraday cup is placed at the back end of the chamber and connected to an electrometer to measure the beam current. The Faraday cup is used to record the beam current through the hole on the ladder mount. During irradiation, the beam current impinging on the samples on the ladder is also monitored with an electrometer. The ions striking the samples produce an ejection of surface electrons from the target. To counter this effect and to obtain an accurate record of the beam current, a suppressor plate with a collimator hole is placed in close proximity in front of the target samples. Monitoring the beam current on the conducting suppressor plate helps to steer the beam onto the target. The Faraday cup is geometrically...
suppressed; i.e., any ejected electrons will be collected by the long cylindrical wall of the cup, thereby yielding an accurate reading.

![Ladder, front view with hole, radius r, and crystals]

Figure 20. Sample Ladder Configuration.

It is very important that the beam is uniform over the diameter of the collimator hole, because we estimate the dose that hits our samples based on the average number of ions that go through the hole. We use a gold foil situated before the collimator in order to diffuse the beam and prevent "hot spots"- areas with higher ion concentration. We check the uniformity of the beam by exposing a piece of Gafchromic™ radiographic film, which is sensitive to radiation exposure. The film changes color in the area that was exposed to radiation. If the beam is not uniform, a reddish mark on the film will indicate the hot-spot area.

After sample alignment has been completed, the chamber is closed and pumped down to \(-10^{4}\) torr. The ions enter the chamber with a net positive charge. Given the ion charge \(Q\), the beam flux can be calculated in the following manner: The beam is adjusted such that the entire beam barely passes through the hole with radius \(r\) on the topmost plate of the ladder, and the current \(I_{fc}\) is measured by the
downstream Faraday Cup. The ion flux \( F \), defined as the numbers of ions per cm\(^2\) per second, is obtained from this current:

\[
F = \frac{I_\mu}{ZeA} = \frac{I_\mu}{Ze(\pi r^2)}
\]

where \( Z \) is the atomic number of the irradiation ion, \( e \) is the elementary charge, and \( A \) is the area through which the ions pass, i.e., the area of the hole in the topmost plate on the ladder. In order to obtain a particular irradiation dose, \( n \), where \( n \) is the columnar defect density expressed in particles per cm\(^2\), the irradiation has to take place over a time \( t \) such that \( n = Ft \). Therefore, the time \( t \) to obtain a particular irradiation dose is:

\[
t = \frac{n}{F} = \frac{neZA}{I_\mu}
\]

An accurate determination of the dose is very important, as the vortex-defect interaction is drastically affected by the ratio between the number of defects and the number of vortices. In chapter IV we will show the results of an experiment in which three irradiation doses were used, and their results compared.

Figure 21 shows a low magnification Transmission Electron Microscopy (TEM) image of an end-on view of the typical damage structure introduced by heavy ion irradiation in \( \text{YBa}_2\text{Cu}_3\text{O}_{6-\delta} \), as shown in [10].

Each black dot corresponds to a single damaged defect track created by a high-energy ion passing through the crystal. It can be seen that the distribution of ion tracks is random and fairly uniform in the region shown. Figure 22 shows TEM micrographs of a tilted (~15° away from the c-axis) view of the columnar defects formed by irradiation.
Defects were created by 1.3 GeV uranium ions.

Figure 21. TEM Image of Columnar Defects in YBCO. Looking Along the Defect Direction.

Defects were created by 1.3 GeV uranium ions.

Figure 22. TEM Image of Columnar Defects in YBCO. Tilted View.
The ion-induced defects are typically cylinders [11] of amorphized material with cylindrical strain fields around them [12]. The amorphous nature of the defects introduced in the material can be seen in the TEM image shown in Figure 23. It can be seen in the figure that the regular structure, indicative of the underlying crystalline order in the unirradiated region, is replaced by an amorphous structure in the ion-induced defect area.

Defects were created by 1.3 GeV uranium ions.

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Figure 23. TEM Image of the Amorphous Structure of the Columnar Defects in YBCO. Looking Along the Defect Direction.

One advantage of introducing defects through irradiation is that the defect concentration can be controlled by the irradiation dose. Moreover, the irradiation
allows us to control the type (point defects versus columnar defects) of disorder introduced by choosing the particles and energy used for the irradiation. In addition, we can study the effects of gradually increasing the disorder into the system. In the case of heavy ion irradiation, which introduce columnar defects, the irradiation dose can be related to a dose equivalent 'matching field' $B_\phi$, where the number of columnar defects equals the number of vortices. Thus the magnetic field range over which pinning is enhanced can be tailored. Most previous experiments on the effect of heavy ion irradiation in YBCO have been performed on twinned samples. Since twin boundaries can also act as correlated pinning sites [13], they can compete with the induced columnar defects, and complicate the separation of the two contributions to vortex pinning. Using untwinned crystals eliminates the interfering effects of twin boundaries.


CHAPTER III

EXPERIMENTAL EVIDENCE FOR THE VORTEX GLASS PHASE IN UNTWINNED, PROTON IRRADIATED YBa$_2$Cu$_4$O$_{6.5}$

Introduction

The vortex melting transition and the nature of the solid in high temperature superconductors are strongly affected by disorder. In clean samples, the transition from the vortex lattice to the vortex liquid [1-3] was shown to be first order [4-7]. In samples containing disorder, various glassy vortex states have been suggested, including the vortex glass [8], Bose glass [9, 10], splayed glass [11], and polymer glass [12] states, depending on the type of the disorder. In the presence of isotropic random point defects, a vortex glass phase is predicted [8].

Earlier experimental work [13-18], on thin films and twinned single crystals of YBa$_2$Cu$_4$O$_{6.5}$ reported evidence of the vortex glass phase. However, these types of samples contain correlated defects in the form of twin boundaries or screw dislocations [19]. Therefore, it is difficult to discern if a vortex glass state, due to point-like defects, or a Bose-glass state, due to correlated defects, is present in these earlier studies. Recent transport experiments on twinned crystals are consistent with a second order Bose-glass transition [20, 21]. Studies on untwinned single crystals with point-like disorder induced by proton [22] and electron [23] irradiation did not find evidence of a vortex glass phase, raising the question of whether the vortex glass phase exists in these systems [24, 25].

In this work we report evidence for a vortex glass in a proton irradiated YBa$_2$Cu$_4$O$_{6.5}$ untwinned crystal. Our work differs from earlier studies in three
essential ways. First, our crystal was untwinned and showed a strong first order melting transition before irradiation. This demonstrates the absence of any significant pinning centers, which might compete with the induced point-like defects in the irradiated crystal. Second, we used a higher proton dose, $3 \times 10^{16}$ p/cm$^2$, than was used in earlier works. The appearance of the vortex glass at these doses, but not at lower ones, suggests that there is a threshold of pinning disorder which must be exceeded in order for the glass to be observed. Finally, in our experiment only part of the crystal was exposed to proton irradiation while the remaining part was protected by a mask. This composite sample allowed simultaneous measurement of the irradiated and unirradiated portions of the same crystal.

Theory

In 1989, Fisher [8] argued that, although the translational order of the vortex lattice is destroyed in the presence of random disorder, a new phase would arise at low temperatures. In this phase, called the vortex glass, the vortices are fixed in a random pattern, determined by the vortex-vortex and vortex-defects interactions. Even though the pattern of the vortices has no long-range positional order, underlying this pattern is a long-order range in the phase of the superconducting wave function, which is associated with the fact that the vortices are immobile. Immobile vortices yield no dissipation, which means that the vortex glass phase is a true superconducting state.

An analogy [26] between the various ordered phases of type II superconductors and systems of magnetic spins can help in the understanding of the nature of the order parameter in the vortex glass phase (see Figure 24). In this analogy, the gradient of the phase $\phi$ of the superconducting wave function plays the role of the spin.
orientation. The analogue of the Meissner phase is the ferromagnet, as both phases are described by homogenous wave function. The magnetic equivalent of the vortex lattice is an antiferromagnet. In both cases, the wave functions possess long range translational order. Additionally, there is a more subtle long-range order in the phase $\phi$ of the superconducting wave function, which reflects the order of the lattice. The vortex glass phase is directly analogous to the order in the spin glass phase: both phases have no apparent long range order, but possess an underlying order in the phase of their wave functions, due to the fact that all spins or vortices are frozen into their positions. The disordered vortex liquid phase at high temperatures is analogous to the paramagnetic phase, because they both lack any long-range order.

As explained by [26], dissipation in the vortex glass phase is caused by loop-like excitations arising from thermal activation. In the presence of the point-like defect, part of the vortex will form a kink, because the vortex is trying to accommodate itself on the point-like defect. The vortex with a kink can be considered a straight vortex plus a loop-like vortex. When a current is applied, there are two forces that act on this loop: an elastic force directed inward, that tends to minimize the loop, and the Lorentz force directed outward, that tends to expand the loop. For loops larger than a critical size the Lorentz force is larger than the elastic force. In order to cause dissipation, the loops need to grow through thermal activation beyond this critical length, after which they can expand to infinity. The critical length corresponds to a barrier height over which the loops have to be excited to cause dissipation. The barrier height scales with the current density $J$ as $U \propto 1/J^\eta$, which leads to an activated behavior of the resistivity:

$$\rho \propto \exp\left(-\frac{U}{k_B T}\right) \propto \exp\left[-\left(\frac{J}{J_c}\right)^\eta\right]$$

(3.1)
Figure 24. Analogy Between Systems of Magnetic Spins (Left) and the Ordered Phases of Type II Superconductors (Right).

where $J_r$ is a temperature dependent proportionality constant and $\mu$ is a constant with a value between 0 and 1. In the limit of zero current, the resistivity is indeed zero and the vortex glass is truly superconducting.

As explained by [12], critical scaling behavior occurs near all continuous phase transitions and is associated with the existence of a characteristic length scale known as the correlation length, which diverges at the transition. Critical fluctuations set in upon approach of a continuous phase transition, i.e., as a function of time, some local order or disorder of the oncoming phase appears and disappears in domains with a typical size given by the correlation length.

As the transition from the vortex liquid phase to the vortex glass phase is a second-order (continuous) phase transition, critical scaling behavior is predicted in the vicinity of the transition. The relevant characteristic length scale, the vortex glass correlation length $\xi_{\text{Vg}}$, is expected to diverge critically in temperature at the glass transition temperature $T_\text{g}$, according to a power law:

$$\xi_{\text{Vg}} \propto \left| T - T_\text{g} \right|^{-\nu}$$  \hspace{1cm} (3.2)

where $\nu$ is the static critical exponent ($\nu > 0$). Further, a critical slowing down is expected for the dynamics near a continuous phase transition. This means that a fluctuation of size $\xi_{\text{Vg}}$ relaxes in a time $\tau_{\text{Vg}}$, which critically diverges according to:

$$\tau_{\text{Vg}} \propto \xi_{\text{Vg}}^{-\nu} \propto \left| T - T_\text{g} \right|^{-\nu}$$  \hspace{1cm} (3.3)

where $\nu$ is the dynamic critical exponent ($\nu > 0$). Note that the critical exponents $\nu$ and $\nu$ are predicted to have universal values for the vortex glass, regardless of the microscopic details of the disordered type II superconductor. The critical exponents
are predicted to be independent of the magnitude and orientation of the applied magnetic field.

Critical scaling means that near the transition, macroscopic quantities such as the resistivity or the current-voltage characteristics can be expressed in terms of a leading power of the vortex glass correlation length. Because of this, universal scaling laws exist, which relate these quantities at various temperatures. The physical picture behind the scaling law is that the applied current density \( J \) defines the characteristic length scale over which the vortices are probed. This length depends on \( J \) as:

\[
L = \left( \frac{ck_BT}{\Phi_0 J} \right)^{\frac{1}{\nu}}
\]  

(3.4)

where \( \Phi_0 = \hbar c/2e \) is the magnetic flux quantum, \( k_B \) is the Boltzmann constant, \( T \) is the temperature.

Above \( T_c \), the system is a liquid over large distances (\( L > \xi_{VG} \)), and is critical over small distances (\( L < \xi_{VG} \)). Below \( T_c \) the system shows a glassy response at large scales (\( L > \xi_{VG} \)), and is critical over small distances (\( L < \xi_{VG} \)). At the glass transition \( T_c \), the current voltage characteristic is algebraic. \( E \propto j^{s+1/2} \), where \( E \) represents the electrical field. Above \( T_c \) the system is a liquid over large distances and the resistivity scales as: \( \rho \propto (T-T_c)^s \), (where \( s \) is another critical exponent, related to \( \nu \) and \( z \) through \( s = \nu(z-1) \)), and critical at small distances. Below \( T_c \), there is a glassy response over large distances. \( E \propto \exp[-c(J/\mu)] \), and again a critical behavior at small scales.

The crossover densities \( j_c^L \) separating the critical behavior at small scales (probed by large current densities) from the liquid/glassy response at large scales (probed by small current densities) scale as \( j_c^L \propto (T-T_c)^\nu \) and \( j_c^L \propto (T_c-T)^\nu \). Figure 25...
summarizes the behavior of the current-voltage characteristics (or current density-electrical field characteristics) of a superconductor in the vicinity of the phase transition, as shown by Blatter [12].

Figure 25. Current-Voltage Characteristics of a Superconductor in the Vicinity of a Second Order Phase Transition.


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Experimental Details

The sample was grown using a self-flux method [27] and detwinned using a thermo-mechanical technique [28]. The sample dimensions were 1000 µm (l) x 235 µm (w) x 200 µm (t). A series of four voltage contacts between two current contacts was used to monitor the voltage across the unirradiated and irradiated regions of the crystal (see Figure 26). Current was applied to the ab-plane of the crystal. The resistivity was measured using a standard four-probe ac technique at a frequency of 23 Hz and a typical current density of 2 A/cm². Current-voltage characteristics were measured with a dc current supply and a nanovoltmeter. The crystal was placed in the bore of a 1.5 T split coil transverse magnet residing inside the bore of an 8 T solenoid magnet. By varying the magnitude of the two magnetic fields independently, various orientations of the magnetic field H with respect to the crystal can be achieved.

![Sample Configuration and Electrical Contact Geometry.](image)

The proton-irradiation was performed at room temperature using Western Michigan University’s tandem accelerator. One region of the crystal was covered with a 500 µm thick NdGaO₃ mask while the remaining region was irradiated parallel...
to the c-axis of the crystal with 9 MeV protons at a low beam flux ($\approx 9 \times 10^{11} \text{ p/cm}^2/\text{sec}$) to minimize heating. The total dose was $3 \times 10^{16} \text{ p/cm}^2$. TRIM [29, 30] calculations reveal that 9 MeV protons have a range of $\approx 280 \ \mu\text{m}$ in YBa$_2$Cu$_3$O$_{7-\delta}$, larger than the 200 $\mu\text{m}$ thickness of our crystal. The defects produced are point defects ($\approx 70\%$) and small clusters ($\approx 30\%$) of about 30 $\AA$ in size [31]. The dose we used produces a defect density of $\approx 5 \times 10^{17}$ defects/cm$^2$. Our TRIM calculations show that the defects are mainly displacements of O and Cu atoms (73%), but Y and Ba atoms are also displaced, accounting for the remaining 27% of the atomic displacements in the YBa$_2$Cu$_3$O$_{7-\delta}$ crystal. Previous studies have shown that roughly 30% of the total defects anneals out at room temperature [32]. TRIM calculations and TEM studies [33] reveal that the defect distribution is random, therefore uncorrelated.

Effects of the Irradiation

The unirradiated region of the crystal had a superconducting transition temperature of $T_{\text{c0}}=93.1\text{ K}$ with width $\Delta T_{\text{c0}}(10-90\%) \approx 400 \text{ mK}$. In the irradiated region, $T_{\text{c0}}$ dropped to 92.3 K and the width $\Delta T_{\text{c0}}$ increased slightly to $\approx 500 \text{ mK}$. Figure 27 shows the temperature dependence of the resistivity in the unirradiated (top panel) and irradiated (bottom panel) regions, for $H \parallel c$. measured with a current density of 2 $\text{ A/cm}^2$. In the unirradiated region, the resistivity in applied magnetic fields displays a "kink" at low temperatures where it drops sharply to zero. The kink is sharp over the entire range of magnetic fields investigated ($\Delta T_{\text{kink}}(H=4\text{T}) \approx 200 \text{ mK} < \Delta T_{\text{c0}}$), and is associated with a first order transition from the vortex liquid to the vortex solid [34], usually observed only in clean single crystals [1-3]. Similar resistivity curves were obtained for $H \parallel ab$. 

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Figure 27. Normalized Resistivity of the Unirradiated (Top Panel) and Irradiated (Bottom Panel) Regions of the Crystal as a Function of Temperature.
In the *irradiated* region (as seen in Figure 27) the kinks in the resistivity have completely disappeared, indicating that the first order phase transition has been suppressed. Similar behavior was observed for $H \parallel ab$.

The angular dependence of the resistivity, $\rho(\theta)$, where $\theta$ is the angle between $H$ and the $c$-axis, reveals the dimensionality of the defect structure. In untwinned samples $\rho(\theta)$ in the vortex state is a smooth function of $\theta$, with a maximum at $H \parallel c$ and a minimum at $H \parallel ab$, reflecting the intrinsic superconducting anisotropy (see Figure 28, top panel). In contrast, in twinned crystals, $\rho(\theta)$ shows a pronounced minimum at $H \parallel c$, as seen in Figure 28 bottom panel, when the vortices are aligned with the twin boundaries [35].

![Diagram of magnetic field and current](image)

The Magnetic Field $H$ is Rotated in a Plane Perpendicular to the Current $J$

![Graph of resistivity with and without correlated disorder](image)

**Figure 28.** The Angular Dependence of the Resistivity for an Untwinned (Top Panel) and a Twinned (Bottom Panel) Crystal.

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Figure 29 shows the normalized resistivity as a function of angle $\theta$ between $H$ and the c-axis of the crystal, for the unirradiated (open circles) and the irradiated (closed circles) portions at $H = 1 \text{T}$ and $T/T_c = 0.978$. It can be seen that the unirradiated region displays a $\rho(\theta)$ typical of a clean, untwinned crystal. In the irradiated region, the magnitude of $\rho(\theta)$ is decreased due to increased pinning, but the qualitative shape of the curve remains unchanged. Thus, unlike twin boundary pinning, we can infer that the defects induced by proton irradiation are isotropic.

Figure 29. The Normalized Resistivity as a Function of Angle $\theta$ Between $H$ and the c-axis of the Crystal.
Technical Details of the Scaling Procedure

In the presence of weak random point defects the vortices in YBa$_2$Cu$_3$O$_{7-\delta}$ are predicted to go into a vortex glass phase [8]. As the temperature decreases, and the electrical resistivity approaches zero, it has been predicted that the resistivity should scale as:

$$\rho = \rho_0(1 - l_g)$$  \hspace{1cm} (3.5)

where $s$ is a field-independent constant, $T_g$ is the glass temperature and $\rho_0$ is a factor.

We performed fits on our experimental data (resistivity versus temperature) to this power law behavior, where the values of $\rho_0$, $T_g$, and $s$ are unknown. One difficulty in fitting the data arises from the fact that the range of temperatures where the data actually fits a power law is hard to determine. Instead, it is visually much easier to determine the range where the data has a linear dependence.

By taking the derivative of equation (3.5) we get

$$\frac{dp}{dT} = \rho_0 s(T - T_g)^{-1}$$  \hspace{1cm} (3.6)

and then by dividing equation (3.5) by equation (3.6) we get

$$\frac{\rho}{(dp/dT)} = \frac{T - T_g}{s} = \frac{1}{s}T - \frac{T_g}{s}$$  \hspace{1cm} (3.7)

where the slope = 1 / s, and the intercept with the y-axis occurs at $T = T_g$. From the equations above it can be seen that the range over which $\rho / (dp / dT)$ vs. $T$ is a straight line will be the range where the resistivity displays the power law dependence. Moreover, from the slope of this line we can determine $s$ and from the intercept we can determine $T_g$ (see Figure 30).
Figure 30. Initial Estimate of the Linear Range and of Parameters $s$ and $T_g$.

However, in order to calculate the derivative of the resistivity with respect to the temperature, one has to perform some smoothing of the data to eliminate the experimental noise. The noise is not very significant in the resistivity data, but it can be very noticeable in the derivative. The smoothing performs a "moving average" of the curve. For each data point, a window of $N$ points surrounding this point is taken, a linear average on these points is performed, and one new data point is obtained. This data point reflects the behavior of the $N$ data points centered on the one data point we initially considered. The operation is done on all the data points in the curve and the resulting data is the "smoothed" data (see Figure 31).
First we smoothed the resistivity and the temperature data, then we calculated the derivative of the smoothed data with respect to the smoothed temperature, and then we smoothed the derivative data. The smoothing was performed on a window of N points, where N is an odd number, which we set to 5 in our analysis. With every smoothing, two data points from both ends of the smoothing range are averaged out (see Figure 31).

A few problems arise due to this smoothing. First, some of the data situated at both temperature ends of the range on which the smoothing is performed is averaged out. The power-law behavior is manifest over a fairly small temperature range, which means that usually we are dealing with a small number of data points and losing some of the data points can truly affect our analysis.

Also, smoothing the data can alter it. Even if the N points that are taken to be averaged have a power law dependence, by performing the smoothing, a linear average of them is calculated. The resulting data point will be distorted from the power-law dependence displayed by the N initial ones. So we conclude that by smoothing the data in order to eliminate some of the noise, the data can be altered and the values obtained for $T_g$, $s$, and $\rho_0$ are not very accurate. Nevertheless, this method is useful to give an indication of the range where the power law behavior is present and to give an approximate value for $T_g$.

The range in which one tries to do the fits is one of the most critical aspects of the glass phase analysis. If the range chosen is larger than the real one, our analysis shows that the value obtained for $s$ will be larger than the real value of $s$. If the range chosen is smaller than the real one, true values for the parameters can be obtained only if the range contains a large number of data points; otherwise the noise in the data is too large to obtain reliable values.
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**Figure 31. Example of Smoothing Procedure**

For our experimental data, the temperature range where we obtained a good fit to the straight line was fairly large. This fit was used to determine $T_c$ and then we performed fits to the power-law behavior on the original, raw data (no smoothing performed). The values obtained for $s$ and $\rho_0$ by performing the fits on the original, raw data are therefore very accurate, as no data altering or averaging is performed.

The curve-fitting program uses the least-squares method, which works by minimizing the square of the error between the original data and the values predicted.
by the equation. The power law fits can be conducted in a number of ways. We can let all three parameters $\rho_0$, $s$, and $T_e$ vary or we can keep one of these parameters fixed and let the other two vary. We found that if we let all three parameters vary, the results obtained can be misleading. We observed that in this case the fitting program has a tendency to minimize one of the three existing parameters. We learned that having one of the three parameters fixed and letting the other two vary is more efficient and gives more accurate and reproducible results. Identical fits to $\rho = \rho_0(T - T_e)^s$ were obtained by fixing either $T_e$ or $s$ while allowing the other two parameters to vary. Systematically incrementing $T_e$ caused $s$ to decrease monotonically and the fit to shift from predominantly above to predominantly below the data. Errors in $T_e$ and $s$ were determined by requiring the average deviation in the fit to $\rho$ in the low temperature range to be less than $\sim 10\%$. Figure 32 shows the effect of varying $s$.

![Figure 32](image)

Figure 32. The Effect of Varying the Parameter $s$ on the Resistivity Scaling.

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The solid line is a fit to equation (3.5) using the average value $s = 5.3$ and the dashed lines are for $s = 5.3 \pm 1$.

Results of the Scaling Analysis

Figure 33 shows $\rho(T)$ for $H \parallel c$ in the irradiated region, where the solid lines are fits using equation (3.5). Identical fits to $\rho = \rho_o (T - T_c)$ were obtained by fixing either $T_c$ or $s$ while allowing the other two parameters to vary.

Table 3 shows the values for $\rho_o$, $s$, and $T_c$ obtained for magnetic fields applied parallel to the c axis ($H \parallel c$) and parallel to the ab plane of the crystal ($H \parallel ab$).

Table 3

Values Obtained for $T_c$, $s$, and $\rho_o$ From the Power Law Fits.

<table>
<thead>
<tr>
<th>H (T)</th>
<th>$T_c$ (K)</th>
<th>$s$</th>
<th>$\rho_o$ ((\mu\Omega\text{-cm}))</th>
<th>$T_c$ (K)</th>
<th>$s$</th>
<th>$\rho_o$ ((\mu\Omega\text{-cm}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>88.40</td>
<td>5.34</td>
<td>2.24</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>2</td>
<td>85.90</td>
<td>5.00</td>
<td>0.249</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>3</td>
<td>83.60</td>
<td>5.07</td>
<td>0.0534</td>
<td>89.70</td>
<td>5.66</td>
<td>19.13</td>
</tr>
<tr>
<td>4</td>
<td>81.50</td>
<td>5.03</td>
<td>0.0206</td>
<td>89.10</td>
<td>6.11</td>
<td>4.83</td>
</tr>
<tr>
<td>5</td>
<td>79.60</td>
<td>4.84</td>
<td>0.0137</td>
<td>88.70</td>
<td>5.44</td>
<td>3.54</td>
</tr>
<tr>
<td>6</td>
<td>77.50</td>
<td>5.22</td>
<td>0.00334</td>
<td>88.30</td>
<td>5.06</td>
<td>2.70</td>
</tr>
<tr>
<td>7</td>
<td>75.70</td>
<td>5.23</td>
<td>0.00195</td>
<td>87.80</td>
<td>5.37</td>
<td>1.20</td>
</tr>
<tr>
<td>8</td>
<td>73.70</td>
<td>5.50</td>
<td>0.000611</td>
<td>87.50</td>
<td>4.96</td>
<td>1.47</td>
</tr>
</tbody>
</table>

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Figure 33. Scaling of the Resistivity Data.
Figure 34 shows the scaling parameter $s$ as a function of the applied magnetic field for $H \parallel c$ (closed circles) and $H \parallel ab$ (open circles). We obtained a field independent value of $s = 5.1 \pm 0.5$ for the field range from 1 T to 8 T for $H \parallel c$ and $s = 5.4 \pm 0.9$ for $3 \ T \leq H \leq 8 \ T$ for $H \parallel ab$. For $H \parallel ab$ the melting temperature is much closer to $T_{cl}$ than for $H \parallel c$. Because of the smaller fitting range, the uncertainties in $s$ are higher for $H \parallel ab$. Below 3 T, we are unable to obtain a reliable value for the scaling exponent. Theoretical work and numerical simulations based on 3-dimensional spin and gauge glasses, which are believed to be in the same universality class as the vortex glass, yield $v \approx 0.9 - 1.7$ and $z \approx 4 - 6$, which gives $s \approx 2.7 - 8.5$ (for a review see Blatter et al [12]). Our averaged field independent value of $s = 5.3 \pm 0.7$ is consistent with these values.

![Figure 34](image.png)

Figure 34. The Scaling Parameter $s$ as a Function of Applied Magnetic Field $H$. 

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Figure 35 shows the tail of the normalized resistivity as a function of the reduced temperature, \( t = T/T_c \), for both the unirradiated and the irradiated regions, at \( H = 4 \, T \parallel c \). for the unirradiated (continuous line) and the irradiated (open circles) portions.

![Figure 35. Normalized Resistivity Versus Reduced Temperature.](image)

The large square symbols represent the reduced temperatures of the I-V curves \((H = 4 \, T \parallel c)\) shown in Figure 36. Remarkably, the resistivity curves cross (at \( t = 0.90 \)), pushing the zero of resistivity to lower temperature for the irradiated region than for the unirradiated region. This result is counter-intuitive, since one expects the induced point defects to enhance pinning and consequently shift the zero resistivity temperature to higher values. We address the origin of this behavior in our discussion of the phase diagram below. In Figure 35 the reduced vortex glass temperature.
\(t_e = 0.877\), for \(H = 4\) T lies below the vortex lattice melting temperature \(t_m\) of the unirradiated crystal.

Figure 36. Current-Voltage Characteristics for the Unirradiated (Left, Top Panel) and the Irradiated (Left, Bottom Panel) Regions of the Crystal.

Figure 36 shows the current-voltage curves (I-V's), taken at temperatures corresponding to the large symbols in Figure 35. In the unirradiated region we observe ohmic behavior in the vortex liquid state above \(t_m\), shown by the linear I-V curve, and non-ohmic behavior in the vortex solid phase below \(t_m\). In contrast, the I-V's for the proton-irradiated portion show only ohmic behavior over the same temperature range. We are able to probe the I-V's only at temperatures above \(t_e\), due to the limited sensitivity of our voltmeter and due to heating from the contact resistance at higher currents. The predicted behavior for the I-V curves [12], is indicated in Figure 36. Indeed, the I-V characteristics at low current above \(T_s\) (shaded region of the inset) are expected to be ohmic, consistent with our results.
Discussion

Earlier studies on untwinned crystals of YBa$_2$Cu$_3$O$_{6.5}$ irradiated with 3 MeV protons to a dose of $5 \times 10^{15}$ p/cm$^2$ did not show vortex glass behavior [22]. Similarly, no vortex glass behavior was found in systematic studies [36] of the evolution of first order melting with successive proton irradiations in doses ranging from $2.5 \times 10^{14}$ to $2 \times 10^{15}$ p/cm$^2$. Our observation of vortex glass behavior at high proton doses suggests a threshold of point defect pinning strength below which glassy behavior is not observed. In a related study, Fendrich et al. [23], found no evidence for a vortex glass phase in untwinned YBa$_2$Cu$_3$O$_{6.5}$ crystals irradiated with 1 MeV electrons to $1 \times 10^{19}$ e/cm$^2$, a dose much higher than in the present work. The absence of the vortex glass in that study may be related to the difference in pinning efficiency of the defects produced by protons and electrons at these energies. To check this possibility, we extracted the pinning energy of the proton induced defects in the present experiment using the same interpolation formula and Arrhenius fit used by Fendrich et al. [23] for their electron irradiated sample. We obtained a pinning energy of $\approx 1870$ K at $H = 4$ T and $T = 83$ K, while for the electron irradiated sample the pinning energy was $\approx 850$ K. This higher pinning energy is obtained despite a proton dose that is $\approx 300$ times lower than the electron dose. This confirms the report by Giapintzakis et al. [37], that proton irradiation induces more efficient pinning defects than does electron irradiation. Taken together, the previous electron and proton irradiation experiments support our conclusion that the vortex glass scaling is observed only above a threshold of point disorder pinning strength.
The first order vortex melting line for the unirradiated region and the second order vortex glass transition line for the irradiated region are shown in Figure 37 for $H \parallel c$ and $H \parallel ab$.

![Figure 37. Melting and Vortex Glass Lines.](image)

The plot depicts the melting lines for $H \parallel c$ (solid triangles) and $H \parallel ab$ (open triangles) and the vortex glass transition lines for $H \parallel c$ (solid circles) and $H \parallel ab$ (open circles). Solid lines represent fits with $H = H_0 (1-T/T_{c0})^{\alpha_c}$, where $H_0 = 81 \pm 5$ T, $\alpha_c = 1.30 \pm 0.03$ before and $H_0 = 64 \pm 2$ T, $\alpha_c = 1.30 \pm 0.01$ after the irradiation, for $H \parallel c$, consistent with previously reported results [1]. The vortex-glass lines lie below the first order melting transition lines. This is consistent with systematic studies at lower proton dose which show that increasing point disorder pushes first order melting to lower temperature before suppressing it altogether [36]. We interpret this behavior as a signature of enhanced entanglement of the liquid due to point disorder.

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as found in previous experiments [23, 38]. The enhanced vortex entanglement of the liquid frustrates the formation of the unentangled lattice, depressing the first order melting temperature. The temperature and discontinuity of the first order melting are continuously depressed, until the second order glass transition emerges at a critical threshold of disorder pinning strength.

Figure 38 shows the angular dependence of the melting and vortex glass temperatures. The plot shows the first order vortex melting temperature, $T_m$ (triangles), and the second order glass transition temperature, $T_g$ (circles), as functions of the angle $\theta$ between $H$ and the $c$-axis of the crystal for $H = 4$ T.

![Figure 38. Angular Dependence of the Melting Temperature and of the Vortex Glass Temperature.](image)
The vortex glass transition temperature is extracted from scaling the resistivity versus temperature data. We find the resistivity scaling predicted for a second order phase transition over the whole range of angles investigated. Scaling over a wide angular range is consistent with a vortex glass transition, but not with a Bose glass transition, where the scaling is found only within a few degrees of the twin boundary plane [20]. Furthermore, the angular dependence of $T_g$ is consistent with a vortex glass: $T_g$ has a minimum at $\theta = 0^\circ$ and a smooth angular dependence. In contrast, the Bose glass temperature displays a sharp cusp for $H$ parallel to the twin boundary [20]. Thus, the angular dependence of our data further confirms the existence of the vortex glass and rules out the presence of a Bose glass in our sample.

Conclusions

In summary, we present the first evidence for a vortex glass transition in an untwinned single crystal of YBa$_2$Cu$_{3-x}$O$_{6.8}$ with induced point-like disorder. The first order melting transition is suppressed by proton irradiation and a second order vortex glass transition appeared at lower temperatures. The results of the scaling analysis of the resistivity yield a field-independent parameter $s$, as predicted for the vortex glass transition. The angular dependence of the vortex glass transition temperature is consistent with a vortex glass. Our results suggest that the pinning energy of the disorder must be sufficiently large in order for the vortex glass to be observed.


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CHAPTER IV

FLUX PINNING ENHANCEMENT THROUGH HEAVY ION IRRADIATION IN SINGLE CRYSTALS OF YBa_{2}Cu_{3}O_{x}

Introduction

Amorphous linear tracks or columnar defects in high temperature superconductors, created by high energy heavy ion irradiation have so far yielded the most noteworthy vortex pinning enhancement by shifting the irreversibility line to higher temperatures and increasing the critical current at both high temperatures and high fields [1-3]. The advantage of this method of introducing correlated defects is that vortex pinning can be controlled by the irradiation dose, which can be related to a dose equivalent 'matching field' $B_{p}$, where the number of columnar defects equals the number of vortices. Thus the magnetic field range over which pinning is enhanced can be tailored.

Irradiation Parallel to the c-axis

Experimental Details

Most previous experiments on the effect of heavy ion irradiation in YBCO have been performed on twinned single crystals. Twin boundaries can also act as correlated pinning sites [4], and therefore they can interact with the vortices. This interaction can compete with the interaction between vortices and the induced columnar defects, complicating the separation of the two contributions to vortex pinning. Using untwinned crystals eliminates the interfering effects from twin boundaries.
We carried out a systematic investigation of the effect of irradiation dose on a large untwinned YBa$_2$Cu$_4$O$_{1-x}$ crystal that was cleaved into five pieces, as shown in Figure 39.

This method guarantees that all crystals have the same underlying reference state and allows us to compare the effects of different doses and different irradiation ions. All the pieces were irradiated such that the heavy ions were parallel to the c-axis of the crystals. Three of the pieces were irradiated with 1.4 GeV $^{238}$U$^{64+}$ ions to doses of $n=B/\Phi_u=4.8 \times 10^{14}$ m$^{-2}$, $9.6 \times 10^{14}$ m$^{-2}$, and $1.9 \times 10^{15}$ m$^{-2}$, respectively, where $\Phi_u=2.07 \times 10^{-15}$ T-m$^{-2}$ is the flux quantum. These doses correspond to the matching fields of $B_\phi=1T$, $2T$, and $4T$, respectively. Two of the pieces were irradiated with 3.9 GeV $^{197}$Au$^{29+}$ ions to a matching field of $B_\phi=1T$ and $4T$. We will first present the

Figure 39. Sample Geometry and Incoming Heavy Ions.
effects of increasing the defect density for the samples irradiated with 1.4 GeV Uranium ions to doses of $B_\phi = 1T$, 2T, and 4T. We will then compare with the results for Gold irradiation.

Irradiation with Uranium ions

Effects of the Irradiation on the Zero-Field Transition

The zero field resistive transitions of the four crystals, unirradiated $B_\phi = 0$ and Uranium irradiated at $B_\phi = 1$, 2, and 4T, are shown in Figure 40.

![Graph](image)

Figure 40. Zero Field Resistivity Versus Temperature for $B_\phi = 0$, 1, 2, and 4 T.

As seen in the figure, as a result of the irradiation, the zero field transition temperature, $T_{\phi}$, determined from the peak in the temperature derivative of the
resistivity, $\frac{d\rho}{dT}$, is shifted to lower temperatures. We also see an increase in both the width of the transition, $\Delta T_{\text{c}}$, and in the normal state resistivity, $\rho_n(100K)$. Table 4 summarizes the values for $T_{\text{c}}$, $\Delta T_{\text{c}}$, $\rho_n(100K)$, as well as the slope of the normal state resistivity ($\frac{d\rho}{dT}$) and the linearly extrapolated normal state resistivity intercept ($\rho_n$). Before irradiation, the zero field transition temperature of the crystal was $T_{\text{c}}=92.66K$ and as a result of irradiation it decreases down to 88.57K for $B_\phi=4T$. An increase in the normal state resistivity near $T=100K$ of about 16% is observed between the unirradiated and the $B_\phi=1T$ irradiated crystal.

On the other hand, crystals irradiated with $B_\phi=2T$ and $4T$ showed increases in the normal state resistivity of 80% and 170%, respectively. Also, at these high doses, the width of the zero field resistive transition increases up to 5 times ($B_\phi=2T$) and as high as 10 times ($B_\phi=4T$) as compared to the pre-irradiation value. This is attributed to the large damage inflicted upon the sample at these high doses. For example, a dose of $B_\phi=4T$ produces a columnar defect spacing of about 220Å apart. Since the columnar defect cores are about 100Å in diameter, this gives a separation between the perimeter of the columnar defects of only 120Å.

Figure 41 shows the resistive transitions for the unirradiated sample. The dashed line represents extrapolation of the normal state resistivity above 120 K to zero temperature, depicting the negative intercept. A negative intercept is usually indicative of high quality crystals.

Table 4 shows the pre and post-irradiation characteristics in zero magnetic field. As seen in the table, the intercept increases monotonically as the irradiation dose is increased, consistent with an increase in the damage present in the sample. Figure 42 displays the dependence of the intercept $\rho_n$ and of the slope of the normal state resistivity ($\frac{d\rho}{dT}$) on the irradiation dose.
Table 4
Pre and Post-irradiation Characteristics in Zero Magnetic Field

<table>
<thead>
<tr>
<th>$B_\Phi$ (T)</th>
<th>$T_{cl}$ (K)</th>
<th>$\Delta T_{cl}$ (K)</th>
<th>$\rho_n(100K)$ (µΩ-cm)</th>
<th>$d\rho/dT$ (µΩ-cm/K)</th>
<th>$\rho_o$ (µΩ-cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>92.660</td>
<td>0.29000</td>
<td>69.590</td>
<td>0.92970</td>
<td>-16.800</td>
</tr>
<tr>
<td>1</td>
<td>90.840</td>
<td>0.97000</td>
<td>78.360</td>
<td>0.96069</td>
<td>-12.532</td>
</tr>
<tr>
<td>2</td>
<td>89.710</td>
<td>1.71000</td>
<td>125.73</td>
<td>1.2680</td>
<td>-9.1683</td>
</tr>
<tr>
<td>4</td>
<td>88.570</td>
<td>3.03000</td>
<td>187.72</td>
<td>1.7325</td>
<td>24.880</td>
</tr>
</tbody>
</table>

Figure 41. Resisitive Transition for the Unirradiated Sample. Depicting $\rho_o$, the Normal State Resistivity Intercept at $T = 0$ K.
Figure 42. Linearly Extrapolated Normal State Resistivity Intercept at T=0K ($\rho_0$) and the Slope of the Normal State Resistivity ($d\rho/dT$), as a Function of the Matching Field $B_\Phi$.

Magnetic Field Dependence of the Resistive Transition

The normalized resistivity, $\rho_n(T) = \rho(T) / \rho(T_c)$, as a function of temperature for magnetic fields of $H=0, 0.5, 1, 2, 3, 4, 5, 6, 7$ and $8$ T, applied parallel to the crystallographic c-axis and to the ab-plane is shown in Figure 43, for the pre-irradiation sample (top panel) and the post-irradiation samples (lower panels). As explained in Chapter I, the broadening of the transition is more severe when the magnetic field is applied parallel to the c-axis than when is applied parallel to the ab-plane, due to the anisotropy of the material.
Figure 43. Normalized Resistivity Versus Temperature for $H=0, 1, 2, 3, 4, 5, 6, 7, 8$ Tesla Parallel to $c$-axis and Parallel to $b$-axis.

The pre-irradiation resistivity curves show a sharp 'kink' in the resistive transition associated with the first order vortex melting transition for both $H \parallel c$ and $H$
The arrows depict the melting temperature for $H=8T$. At each field the melting occurs at a different temperature, and this dependence defines the melting line in the phase diagram.

After irradiation with $B_\phi=1$, 2, and 4T, the kink in the resistivity curve is completely suppressed and replaced with a smooth curve. Note that different trends are apparent for the two orientations of the magnetic field. For $H \parallel c$, the zero resistance temperature of the irradiated crystals shifts to higher temperature as the irradiation dose is increased. The resistivity curves shrink closer together as the dose is increased, and at the highest dose the slopes of the resistivity curves in different fields are nearly parallel. In contrast, for $H \parallel ab$, the zero resistance temperature of the irradiated crystals shifts to lower temperature as the irradiation dose is increased.

The temperature at which the resistivity goes to zero represents the onset of a true critical current and of irreversible behavior in the magnetization, and it defines the irreversibility line. We determined the irreversibility line by using a criterion of $\rho=0.01\mu\Omega\cdot\text{cm}$, the resolution of our equipment.

**Phase Diagram**

The vortex lattice melting line for the unirradiated crystal for $H \parallel c$ is shown in the top panel of Figure 44. The pre-irradiation melting line can be fitted to $H_m=88.4(1-T/T_a)^{1.26}$, consistent with other high quality single crystals of YBCO. After irradiation, the first order vortex melting is completely suppressed. The irreversibility line for $H \parallel c$ for the irradiated crystals, $H_{irr}(T)$, is shown in the lower panels of Figure 44. Notice the distinct change in slope at the matching field $B_\phi$. For $B_\phi=1T$, the irreversibility line for $H \parallel c$ follows a linear behavior above $H=1T$ and a sharp upward curvature below 1T.
Figure 44. Melting Line for the Pre-Irradiated Crystal (Top Panel) and Irreversibility Lines for Post-Irradiated Crystals (Lower Panels) for $H$ Parallel to the c-axis.

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Similarly, for $B_\theta = 2T$ and $4T$, a change in slope of the $H_{irr}(T) \parallel c$ is observed near $H = 2T$ and $4T$ respectively. This change in slope indicates a change in vortex pinning behavior. For magnetic fields below $B_\theta$, there are more columnar defects than vortices, and each vortex can be pinned by a columnar defect. The irreversibility line is represented by a rapid concave upward rise. However, for magnetic fields above $B_\theta$, the number of vortices exceeds the number of defects and the defects are not as efficient at pinning the vortices. Consequently, the sharp rise in the irreversibility line is replaced by a linear behavior with a smaller slope. The fact that we observe this change in behavior very close to the dose matching field attests to the high control over the density of induced columnar defects.

The top panel of Figure 45 shows the vortex lattice melting line for the unirradiated crystal for $H \parallel c$ and $H \parallel ab$. The lower panels of Figure 45 show the irreversibility line determined from the $\rho = 0.01\mu\Omega\text{-cm}$ criterion, for both $H \parallel c$ and $H \parallel b$. The remarkable result is the crossing of the $H_{irr}(T) \parallel c$ and $H_{irr}(T) \parallel ab$ lines at magnetic fields about three times the matching field. For $B_\theta = 1T$ and $2T$, the crossing occurs near $3.3T$ and $6.8T$ respectively. For $B_\theta = 4T$, a linear extrapolation yields a crossing near $12.5T$. The irreversibility line for $H \parallel b$ lies below the $H_{irr} \parallel c$ curve below about $\approx 3.3B_\theta$. This indicates that the pinning due to the defects is very strong, and the anisotropy is reversed. The fact that the irreversibility line is lowered for $H \parallel b$ suggests that columnar defects along the $c$-axis may act as conduits for easy vortex motion when the magnetic field lines are aligned perpendicular to the columnar defects. In our measurements, the transport current $J \parallel a$, is perpendicular to the applied magnetic field. Thus for $H \parallel b$, the Lorentz force exerted on the vortices by the magnetic field and current is on average, along the $c$-axis.
Figure 45. Melting Line for The Pre-Irradiated Crystal (Top Panel) and Irreversisbility Lines for Post-Irradiated Crystals (Lower Panels) for H parallel to c and b-axes.

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Previous work by Kwok et al. [5] has shown that intrinsic pinning due to the layered structure is responsible for localizing the vortices in the ab-plane. The induced columnar defects drilled through the layered structure of the crystal may suppress intrinsic pinning by introducing a new channel for vortex creep motion between the layers.

The crossing of the irreversibility lines is the result of the interplay between the intrinsic anisotropy of YBa$_2$Cu$_4$O$_{8-x}$ and the anisotropic character of the defects. Due to the intrinsic anisotropy of the material, in the unirradiated crystal, the melting line for $H \parallel ab$ lies above the one for $H \parallel c$. However, in the irradiated crystal, the defects provide strong pinning for $H \parallel c$ when the vortices are aligned with the defects. The columnar defects have proven strong enough to actually reverse the anisotropy of the material. In order to gain a better understanding of the interplay between the intrinsic anisotropy of the material and the anisotropic character of the defects, we investigate the angular dependence of the irreversibility line.

The data was taken by applying a magnetic field with respect to the c-axis and then measuring the resistivity as a function of temperature at each angle $\theta$. Here $\theta$ is the angle between the c-axis and the applied magnetic field $H$. Using the same $\rho=0.01\mu\Omega\cdot\text{cm}$ criterion, we determined the angular dependence of the irreversibility line.

Figure 46 shows the irreversibility line as a function of temperature at different angles $\theta$ for the $B_0=2T$. The irreversibility line is shown for angles between $0^\circ$ and $90^\circ$ in the top panel and for angles between $50^\circ$ and $90^\circ$ in the bottom panel. From the top panel of the figure it can be seen that the irreversibility line is highest for $H \parallel c \parallel$ defects. This result is consistent with enhanced pinning of the vortices when they are aligned with the columnar defects.

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Figure 46. Angular Dependence of the Irreversibility Line.

In the top panel of the figure it can also be seen that, for $0^\circ < \theta < 70^\circ$, the irreversibility line shifts down when $\theta$ increases. As the angle $\theta$ between the vortices
and the defects is increased, the cross-section between the vortices and the defects decreases, and so does the pinning strength. The vortex mobility increases with $\theta$, and the irreversibility line shifts at lower temperatures.

However, as seen in the lower panel of Figure 46 at angles above 70° the trend is reversed: the irreversibility line shifts up with increasing $\theta$. This trend reflects the intrinsic anisotropy of the material. Therefore the minimum at $\theta \approx 70^\circ$ reflects the interplay between the anisotropic character of the defect structure and the intrinsic anisotropy of the material. This minimum corresponds to the highest vortex mobility, and it must occur when the vortices are in a certain orientation: they are tilted far enough from the defects to escape their pinning potential; at the same time, they are tilted far enough from the ab planes to escape intrinsic pinning. Later in this chapter it will be shown that the minimum at $\theta \approx 70^\circ$ is consistent with the value found for the accommodation angle when $B_d=2T$ and $H=2T$.

**Angular Dependence of the Resistivity**

The behavior of the normalized resistivity for the unirradiated sample is shown as a function of the angle of the applied field with respect to the c-axis of the crystal for $H=0.5T$ in Figure 47. The magnetic field was tilted from the 90° ($H \parallel ab$-plane) to 0° ($H \parallel c$-axis) for a reduced temperature $t=T/T_c$ of 0.979, 0.977, 0.974, 0.972, and 0.969. In the case of untwinned samples, the resistivity is largest when the field is aligned parallel to the c-axis and is a minimum when the field is parallel to the ab-plane, due to the intrinsic anisotropy of the material (see for example Figure 1.8). In twinned crystals there is a dip in the resistivity as H becomes aligned with the twin boundaries at $\theta=0^\circ$. In our crystal, the resistivity is a smooth function of the angle, which is typical for a clean, untwinned crystal.
Figure 47. Angular Dependence of the Resistivity for an Unirradiated Crystal.

The angular dependence of the resistivity in the irradiated samples shows a dramatically different behavior as shown in Figure 48. Here we show the data for the sample for $B_0=2T$ in an applied field $H=0.5T$. The data was taken at the reduced temperature $t=1.010, 1.008, 1.006, 1.001, 0.994, 0.992, 0.990, 0.983, \text{ and } 0.980$. In contrast to the unirradiated crystal, the resistivity in the irradiated crystal shows a pronounced dip near $\theta=0^\circ$. This dip arises from the enhanced pinning of the vortices as they become aligned with the columnar defects, which are parallel to the c axis. One remarkable feature of the $\rho(\theta)$ curves is that the dip in the resistivity occurs even for the temperatures in the fluctuation regime, i.e., for temperatures above the midpoint of the superconducting transition. The onset of the dip occurs at $T/T_c \approx 1$ and very close to $\rho_n \approx 0.72$. Similar studies in which crystals were irradiated with Au
and Xe at a matching field $B_\phi=2T$ found the onset of pinning to be $\rho_\alpha \approx 0.28$ (Au ions) and $\rho_\alpha \approx 0.17$ (Xe ions). This indicates that the columnar defects created by Uranium irradiation are effective pinners up to very high temperatures, and points to the high vortex liquid pinning capability of these columnar defects.

![Angular Dependence of the Resistivity for an Irradiated Crystal](image)

**Figure 48.** Angular Dependence of the Resistivity for an Irradiated Crystal.

**Depinning Angle**

Another extraordinary feature of Uranium irradiation is the strength of the columnar defects at large angles. This is seen in the shape of the $\rho_\alpha(\theta)$ curves where $\rho_\alpha(\theta)$ first increases with decreasing $\theta$ (similar to the unirradiated crystal) and then begins decreasing as sections of the vortices become pinned by the columnar defects. We can define the depinning angle $\theta_{\text{dep}}$ as half the angular distance between the
maxima in $\rho_n(\theta)$. The value of the depinning angle is shown in Figure 48 by the solid triangles. The dashed line is a guide to the eye. The depinning angle represents the angle beyond which the columnar defects are no longer effective in pinning the vortex line. Also, the resistivity, and therefore the vortex mobility is maximum at the depinning angle and it represents the angle where the interplay between the intrinsic anisotropy and the defect anisotropy yields the least pinning.

We extracted the depinning angle for the crystals irradiated at $B_0=1T, 2T,$ and $4T$ and for measurements taken in applied magnetic fields $H=0.5T, 1T$ and $2T$. Figure 49 shows the depinning angle versus temperature. Each plot shows the data at a given applied magnetic field, $H=1T$ (top panel), $H=2T$ (middle panel) and $H=4T$ (bottom panel) for the three different irradiation doses used. At first glance a few features can be noted. First of all, for all the applied fields, the depinning angle curve shifts towards higher angles as the irradiation dose is increased. Thus at a given temperature and applied field $H,$ a higher irradiation dose yields a larger depinning angle. This can be explained in the following manner: at a given applied magnetic field $H,$ the number of the vortices is the same, while a higher irradiation dose means more columnar defects. Consequently, better pinning will be obtained when the same number of vortices has a larger number of defects with which they can interact. We expect this effect to saturate for applied fields much less than the matching field. It is unclear why, for $H=0.5$ T, the depinning angle is still higher for the highest dose ($B_0=4T$) as long for all the irradiation doses, there are more defects than vortices, and we would therefore expect the pinning to be equally strong for all the doses.

Secondly, we notice that the curves have strong temperature dependence at high temperatures, and they tend to level off as the temperature is decreased. At high temperatures, the thermal energies are considerable, and there is a strong dependence
Figure 49. The Depinning Angle for the Crystals Irradiated With $B_\phi=0, 1, 2,$ and 4 T.
of the depinning angle on the temperature. At low temperatures, where the thermal
energies can be neglected, the depinning angle coincides with the accommodation
angle, which is the angle at which the trapped length goes to zero. Figure 50 shows
the interaction between the vortex and the columnar defects, and provides a more
intuitive understanding of the accommodation angle.

The dashed lines in Figure 49 represent the extrapolated values of the
depinning angle, towards the low temperature region, where the depinning angle
coincides with the accommodation angle. We should note first that the three plots
describe quite different situations: for $H=0.5T$ (upper panel), there are fewer vortices
than defects for all three irradiation doses $B_o=1T$, 2T and 4T. We expect that at all
three doses the pinning will be quite strong, as the number of defects greatly exceeds
the number of vortices. Indeed, we see that the accommodation angle is large, lying
between $\approx 63'$ ($B_o=1T$) and $\approx 75'$ ($B_o=4T$). In contrast, for $H=2T$ (lower panel), the
number of vortices is half the number of defects for $B_o=4T$, equal to the number of
defects for $B_o=2T$, and double the number of defects for $B_o=1T$. In this situation we
expect stronger pinning for $B_o=4T$ than for $B_o=1T$. Our data confirms this
interpretation: $\theta_{\text{dipm}}$ reaches values as high as $\approx 71'$ for $B_o=4T$ while it levels off at $\approx 42'$ for $B_o=1T$.

Figure 51 depicts the accommodation angle, $\theta_{\text{acc}}$, versus the applied magnetic
field $H$, for the three irradiation doses, $B_o=1.2$, and 4T. Notably, for both $B_o=2T$
and $B_o=4T$, the accommodation angle is fairly constant when $0<H<2T$. Indeed, over
this range, the number of vortices is less or equal to that of the defects. On the
contrary, for $B_o=1T$, the accommodation angle drops fast with the increase in the
field $H$, reflecting the inability of the defects to pin the vortices that outnumber them.
Figure 50. Schematic Diagram of the Accommodation Angle.
Remarkably, the accommodation angle for $B_\Phi=2T$ and $H=2T$ is $\approx 70^\circ$, consistent with the angle at which the minimum in the irreversibility line was observed.

![Graph](image)

Figure 51. The Accommodation Angle as a Function of the Applied Magnetic Field $H$ for the Crystals With $B_\Phi=0$, 1, 2, and 4 T.

**Pinning Energy**

The observed angular depinning of the vortex system can yield information about the microscopic pinning characteristics [3]. Let us first consider the angular behavior of the low temperature pinning. The strength of the pinning energy per unit length $U_p$ can be estimated from the depinning angle. When the field is applied at angle $\theta$ with respect to the columnar defect, a vortex line trapped in the columnar
defect consists of three regions: a trapped piece of length \( l_{tr} = 2z \cos \theta \) and two "healing" regions (see Figure 52). The energy of the trapped region is given by

\[
E_i = \int \left[ \frac{\varepsilon_i}{2} \left( \frac{du}{dz} \right)^2 + \frac{K}{2} u^2 \right] dz - U_{pin}
\]

where \( \varepsilon_i \approx [\Phi_{\phi}/(4\pi\lambda)^2] \ln(a_n/\xi) \) is the linear tension energy. \( a_n \approx \sqrt{\Phi_n/B} \) is the distance between vortices. \( (K/2)u^2 \) represents the "cage potential" arising from the interaction with the other vortices. where \( K = \Phi_n B/(4\pi\lambda)^2 \), and \( U_{pin} \) is the pinning potential per unit length of the defect.

Figure 52. The Vortex Pinning Configuration in the Presence of a Columnar Defect

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The pinning energy can be ultimately related to the pinning angle through the following formula:

$$U_p \approx \varepsilon_0 \left[ \frac{2k_BT \tan \theta_{dep}}{\varepsilon_0\mu_0} \right]^2 \tag{4.2}$$

In these equations, $k_B$ is the Boltzmann constant, $T$ is the temperature, $\Phi_0=2.1 \times 10^{-7}$ G cm is the flux quantum, $B$ is the applied magnetic field, and $\varepsilon_0=\Phi_0/(4\pi\lambda_{ab})^2$. In our calculations we assumed that the temperature-dependent penetration depth was expressed by $\lambda_{ab}=1400\lambda/(2(1-T/T_{ct})^{1/2})$. Figure 53 shows the pinning energy, as obtained from the depinning angle.

![Figure 53. Pinning Energy for Crystals Irradiated With Uranium Ions at Matching Fields $B_0=1$, 2, and 4 T.](image-url)
We are showing the pinning energy $U_p/k_B$ versus the applied magnetic field $H$ at a reduced temperature $t=0.9865$ for the three irradiation doses. A few important features can be immediately distinguished. First of all, we notice that the pinning energy is higher for the higher irradiation dose ($U_p(B_\phi=4T)$ is $\approx 20\%$ higher than $U_p(B_\phi=1T)$ at $H=0.5T$). This is consistent with the fact that better pinning is obtained when there are more defects. It can be also noted that $U_p$ is rather constant as long as the number of vortices is less than the number of defects ($H<B_\phi$), and it drops abruptly when the number of vortices matches or exceeds the number of defects ($H\geq B_\phi$). This behavior can be explained by the inability of the defects to pin the vortices that outnumber them.

Comparison Between Uranium and Gold Ions

Defect Structure

Heavy ions in the GeV range impinging on the samples create amorphous columnar defects with diameters in the 100Å range. The exact size and shape of the defects depends strongly on the type and energy of the ions. Also, the energy loss of the ions varies with the depth in YBCO, which also affects the type of defects formed [6].

Figure 54 shows TEM micrographs of a tilted ($=15^\circ$ away from the c-axis) view of the damage formed by irradiation with 1.4GeV $^{238}$U$^{7+}$ ions. Figure 55 shows similar TEM views of the damage formed by irradiation with 3.9 GeV $^{197}$Au$^{7+}$ ions. As seen when comparing the two figures, while no marked difference is apparent in the size of the defects produced by the two different kinds of ions, the shape of the defects is rather different. Uranium ions produce columnar defects, which are nearly uniform in diameter and are continuous throughout the thickness of the TEM sample.
Figure 54. TEM Micrographs of the Tilted View of the Columnar Damage Formed by Irradiation with 1.4 GeV $^{197}$Au<sup>2+</sup>.

On the contrary, the cross section of the Gold-induced defects is not uniform along the length of the defect. The cross section oscillates quasi-periodically and the tracks consist of large spheres (90-180 Å diameter) connected by smaller cylinders (diameter ≈ 40 Å). The shape of the damage track is very similar to the "string of pearls" structure discussed by Hensel et al. [7].

Figure 55 shows TEM Micrographs of the columnar damage formed by irradiation with 3.9 GeV $^{197}$Au<sup>2+</sup> ions. The "string of pearls" structure is visible in the micrograph.
Figure 55. TEM Micrographs of the Tilted View of the Columnar Damage Formed by Irradiation with 1.4 GeV $^{238}$U.$^{238}$

Phase Diagram

Figure 56 compares the irreversibility line for $H \parallel c$ for two crystals, one irradiated with Uranium and the other irradiated with Gold, at the same dose $B_{\text{irr}}=4T$. Notably, the irreversibility line displays a bigger shift towards higher temperatures for the Uranium-irradiated samples, as compared to the Gold-irradiated one. This implies that the Uranium-induced defects are better pinners than the Gold-induced defects. We attribute the lesser pinning efficiency of the Gold-induced defects to their non-uniform character. The intersection volume between the defect and the vortex determines the pinning strength of the defects, as discussed in Chapter I. The "string of pearls" structure of the Gold-induced defects may provide a smaller intersection
volume with the vortices, and therefore yield less strong pinning than continuous, uniform defects would.

![Graph showing melting and irreversibility lines for Uranium and Gold irradiated crystals for \( \mathbf{H} \parallel \mathbf{c} \).](image-url)

**Figure 56.** Melting and Irreversibility Lines for Uranium and Gold Irradiated Crystals for \( \mathbf{H} \parallel \mathbf{c} \).

**Pinning Energy**

The angular dependence of the resistivity for the Gold-irradiated crystals also shows the characteristic dip associated with the enhanced pinning of the vortices as they become aligned with the columnar defects. We extracted the depinning angle and calculated the corresponding pinning energies.

Figure 57 compares the pinning energy at \( t=0.979 \), as obtained from the depinning angle for two crystals, one irradiated with Uranium and one irradiated with Gold, at the same dose \( B_{ir} = 4\, \text{T} \).
The remarkable feature here is the very different field dependence of the pinning energy for the two crystals. In contrast to the Uranium irradiated crystal, where the pinning energy is constant for $H < B_{\phi}$ and it drops abruptly as $H$ approaches $B_{\phi}$, the pinning energy for the Gold irradiated crystal drops monotonically as $H$ is increased. This points to the lower pinning efficiency of Gold-induced defects as opposed to the Uranium-induced ones. These results are consistent with the smaller shift to high temperatures of the irreversibility line, described previously in this chapter.
Irradiation Parallel to the ab-Plane

Introduction

In the previous sections it was shown that the columnar defects, when introduced parallel to the c-axis, reverse the anisotropy of the material. The columnar defects inhibit vortex motion for \( H \parallel c \), and they promote vortex motion for \( H \parallel ab \). These results reflect the interplay between the intrinsic anisotropy of the material and the anisotropic character of the defects. In order to gain a better understanding of this interplay, we performed this experiment, in which the columnar defects are introduced parallel to the ab plane of the crystal.

Previous experimental work has been focused on columnar defects introduced parallel to the c axis [1-3, 8, 9], or in a splayed defect geometry [10-13], where the sample is irradiated with heavy ions at two angles defining the splay plane. However, no previous work addresses columnar defects introduced parallel to the ab-plane, and our current work is a first effort in studying this defect configuration. We have preliminary results on the effects of columnar defects introduced with 1.4 GeV \(^{208}\text{Pb}^{85-}\) ions at a dose of \( B_0=1\text{T} \).

Experimental Details

In order to carry out the irradiation, the sample had to be thinned to a thickness of less than 30 \( \mu\text{m} \), in order to ensure that the heavy ions pass through the sample. Figure 58 shows the sample configuration. The figure shows the sample geometry, and it indicates the orientation of the incoming heavy ions with respect to the sample. It can be seen that in this geometry, the ions will strike the crystal parallel to the ab plane. As explained in Chapter I, YBa\(_2\)Cu\(_4\)O\(_{8.5}\) has a layered structure, which makes polishing the sample parallel to the ab plane, in order to
prepare it for irradiation parallel to the c-axis, rather feasible. However, in order to irradiate the sample parallel to the ab plane, one has to polish perpendicularly to the ab plane (see Figure 58).

![Diagram of YBa$_2$Cu$_3$O$_{7-\delta}$ Single Crystal](image)

Figure 58. Sample Geometry and Incoming Heavy Ions.

The internal structure of the material makes this very difficult, and most samples break when they become thin. After polishing, the samples are small, and putting the leads in order to create electrical contact to the sample was quite difficult. It was a real challenge to obtain the low resistivity electrical contacts needed for transport measurements. The electrical contacts deteriorated rather fast, and we were able only to obtain preliminary resistivity results. Our results proved successful in determining the effects of the columnar tracks on the vortices for $H \parallel ab$. However, further work is needed in order to track the changes on the phase diagram.
Results and Discussion

The pre-irradiation normalized resistivity as a function of temperature for various magnetic fields parallel to the crystallographic ab-plane is shown in the top panel of Figure 59. The applied magnetic fields are 0, 0.5, 1, 2, 3, 4, 5, 6, 7, 8 T | ab with an applied current of 0.5 mA. The resistivity curves display the 'kink' associated with the first order vortex melting transition.

The zero field transition temperature $T_c$ has shifted from 92.90 K to 92.51 K as a result of the irradiation. The decrease in $T_c$ is not as dramatic as the one observed.
in samples irradiated with Uranium ions at the same dose, suggesting that ion size may play a role in the severity of the damage.

The bottom panel of Figure 59 shows the post-irradiation normalized resistivity as a function of temperature for \( H = 0, 0.5, 1, 2, 3, 4, 5, 6, 7, 8 \, \text{T} \parallel \text{ab} \). After irradiation the kink in the resistivity curve is completely suppressed and replaced with a smooth curve at all the magnetic fields investigated, indicating a complete suppression of the first order melting transition.

Figure 60 shows the normalized resistivity as a function of temperature for various magnetic fields parallel to the crystallographic \( c \)-axis and the \( \text{ab} \)-plane. Each plot shows the normalized resistivity versus the normalized temperature for both the pre-irradiation (open circles for \( I = 0.5 \, \text{mA} \) and thin dashed line for \( I = 0.1 \, \text{mA} \)) and the post-irradiation crystal (solid circles for \( I = 0.5 \, \text{mA} \) and thin line for \( I = 0.1 \, \text{mA} \)), at a given magnetic field. Notably, for all the fields, the normalized zero resistance temperature shifts up as a result of the irradiation. Although both the \( I = 0.5 \, \text{mA} \) (solid circles), and the \( I = 0.1 \, \text{mA} \) (thin line) data are plotted, it is very hard to distinguish the \( 0.1 \, \text{mA} \) on this plot, as it follows the \( 0.5 \, \text{mA} \) data very closely. However, if the data is plotted on a semi-log scale, as in Figure 61, it can be seen that the curves at the two different currents split as the temperature is decreased.

The onset of non-ohmic behavior is used in order to determine the irreversibility line. Figure 62 shows the phase diagram for the pre-irradiated (open circles) and the post-irradiated (solid circles) crystal, for \( H \parallel \text{ab} \). The irreversibility line shifts to higher temperatures as a result of the irradiation. Therefore the columnar defects \textit{inhibit} vortex motion for \( H \parallel \text{ab} \). The shift at higher temperatures of the irreversibility line suggests that the anisotropy of the material is increased by the columnar tracks.
Figure 60. Normalized Resistivity as a Function of Temperature for Various Magnetic Fields $H$ Parallel to the $ab$-plane.
Figure 61. Normalized Resistivity as a Function of Temperature on a Semi-Log Scale for Various Magnetic Fields $H$ Parallel to the $ab$-plane.

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The behavior of the irreversibility line for $H \parallel c$ would completely clarify this issue. Unfortunately, we were unable to take the resistivity data for $H \parallel c$ after the irradiation, due to the fact that the electrical contacts degraded and could not be fixed. Future experiments will focus on the phase diagram for both $H \parallel c$ and $H \parallel ab$. These experiments will bring more insight into the interplay between the intrinsic anisotropy of the material and the anisotropic character of the columnar tracks.

![Figure 62. Phase Diagram for the Pre-Irradiation and the Post-Irradiation Crystals for $H \parallel ab$.](image)

Conclusions

We found that by irradiating the crystals at the high doses used in the experiment, the first order vortex melting line is completely suppressed. For the irradiation parallel to the c-axis, we found a consistent change in the slope of the
irreversibility line at the matching field for $H \parallel c$ and a remarkable reversal of the
pinning anisotropy. The columnar defects *inhibit* vortex motion when vortices are
parallel to the defects ($H \parallel c$), and they *promote* vortex motion for vortices
perpendicular defects ($H \parallel ab$).

Anisotropic pinning from columnar defects induced by uranium heavy ions
can extend up to $T_c$, suggesting that even in the superconducting fluctuation regime
these defects are capable of pinning the vortices. We have calculated the pinning
energy from the depinning angle and found that it is constant while the number of
vortices is less than the number of defects and that it drops sharply when the number
of vortices approaches and exceeds the number of defects. We have also found that
Uranium-induced defects are more efficient pinning sites than Gold-induced defects.

For the irradiation parallel to the $ab$-plane, we found an increase in pinning for
$H \parallel ab$. Therefore the columnar defects *inhibit* vortex motion for vortices parallel to
the defects ($H \parallel ab$). The results for $H \parallel ab$ suggest that the anisotropy is increased.


CHAPTER V

THERMAL STABILITY OF CORRELATED DEFECTS INTRODUCED BY HEAVY ION IRRADIATION IN YBa$_2$Cu$_3$O$_{7-\delta}$

Introduction

The correlated defects introduced by heavy ion irradiation are highly effective pinning sites for vortices in high temperature superconductors [1-5]. These defects greatly increase the critical currents and shift the irreversibility line to higher temperatures, decreasing the region of the vortex liquid state in the superconducting phase diagram and making these materials more suitable for technological applications. Many applications require post irradiation processing at elevated temperatures, for example to join segments of superconducting tapes and wires, or to attach electrical contacts. Some previous work has been done on the thermal stability of heavy ion irradiation induced defects in Bi$_2$Sr$_2$CaCu$_2$O$_{8}$ (BSCCO) [6]. However, the thermal stability of these defects in YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO), which is perhaps the most promising of the high T$_c$ superconductors for applications, has not yet been addressed. In this work we study the durability of the tracks created by 3.9 GeV $^{197}$Au$^{97}$ irradiation in YBCO single crystals. We performed both magnetization and Transmission Electron Microscopy (TEM) measurements to correlate changes in the critical current density with changes in the microstructure of the defects. Surprisingly, we find that the defects induced by the heavy ion irradiation can withstand thermal annealing up to temperatures of 600 °C. Even more remarkable is the discovery that the critical current density at temperatures above 45 K can actually increase with thermal annealing.

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Experimental Details

The samples used in this study were grown by a self-flux method using powders of Y$_2$O$_3$, BaCO$_3$, and CuO with a purity of 99.99% or better as described elsewhere [7]. The crystals were subsequently annealed for 10 days in flowing oxygen at 420 °C, yielding optimally doped twinned single crystals with a transition temperature $T_c \approx 93.5$ K. The samples were irradiated parallel to the crystallographic c-axis, with 3.9 GeV $^{197}$Au$^{2+}$ ions at the National Superconducting Cyclotron Laboratory (NSCL). The Monte Carlo simulation program TRIM [8] yields a range of $\approx 100$ μm, which is greater than the $\approx 30$ μm thickness of our crystals, ensuring that the heavy ions pass entirely through the samples. The irradiation dose was $\approx 5 \times 10^{10}$ ions/cm$^2$, as indicated by TEM micrographs. At this dose, the number of the vortices equals the number of ion tracks in an applied magnetic field of 1 T, which is referred to as the matching field $B_m$ [1].

The annealing process can induce changes in the defect microstructure and in the oxygen concentration, both of which can affect the critical current density $J_c$. To distinguish between the two effects, we measured $J_c$ in an irradiated and an unirradiated crystal as they underwent identical anneals. Each annealing treatment consisted of heating the sample for 1 hour in flowing oxygen. The treatments were done at 400 °C, 450 °C, 500 °C, 550 °C, 575 °C, and 600 °C, with magnetization measurements performed after each anneal. The irradiated crystal further underwent two additional anneals at 420 °C lasting 5 days each, to fully reoxygenate the crystal.

Changes in the microstructure were determined by TEM measurements, which is a destructive process. Thus we used a third irradiated crystal (TEM-crystal), which we broke in half. One piece was characterized with TEM immediately after irradiation and the second piece was characterized after a 20-minute anneal at 420°C.
in flowing oxygen. Finally, the irradiated crystal on which the magnetization measurements were performed was characterized with TEM only after the last anneal. All the crystals used in this study were grown concurrently in the same batch to minimize differences among the samples.

Magnetization measurements were taken before and after each thermal treatment with a Quantum Design superconducting quantum interference device (SQUID) using a 3-cm scan length to minimize variations in the magnetic field \( H (\delta H \leq 0.05\%) \). The sample was mounted with the crystallographic c-axis and the heavy ion tracks parallel to the field. The samples were cooled in zero-applied magnetic field. The superconducting transition temperature, \( T_c \), was determined from measurements of the magnetization \( M \) as a function of temperature in an applied field of 10 G, where \( T_c \) was defined at the temperature of the onset of diamagnetism.

The magnetic hysteresis was measured in fields between -5 T and 5 T after cooling in zero-applied field to the target temperature. In determining the critical current density \( J_c \) from magnetic hysteresis measurements using the Bean critical state model [9], one has to measure the magnetization of the sample as a function of the applied magnetic field. Due to magnetic flux pinning by defects in the crystal structure, hysteresis loops are obtained when the magnetic field is increased first and then decreased. From the magnetic hysteresis the induced critical currents \( J_c \) can be determined from the expression given by the Bean model [9] by

\[
J_c = 20 \frac{\Delta M}{w \left( 1 - \frac{w}{3l} \right)}
\]

for a rectangular shaped sample with \( w < l \), where \( w \) (width) and \( l \) (length) are the
sides of the rectangle, and $\Delta M = M^{+} - M^{-}$ is the difference in magnetization between the upper ($M^{+}$) and lower ($M^{-}$) branches of the hysteresis loops.

**Effects of the Irradiation**

After irradiation the superconducting transition temperature $T_{c0}$ decreased from $\sim 93.5 \text{ K}$ to $\sim 92.0 \text{ K}$ while the width of the transition, $\Delta T_{c0}$, increased from 2.5 K to 2.7 K. Figure 63 shows the magnetic hysteresis loops for the unirradiated (top panel) and the irradiated (bottom panel) samples. Note the increased value of the magnetization at all the temperatures after the irradiation. The critical current density increased roughly by a factor of two ($J_{c} \approx 1 \times 10^{4} \text{ A/cm}^{2}$ at 77 K in an applied field of 1T), consistent with similar studies [10].

Figure 64 shows a TEM micrograph of an end-on view of the heavy ion tracks in the irradiated TEM-crystal immediately after the irradiation. The most apparent feature is the alternating bright and dark stripes running at approximately 45° with respect to the frame. These regions correspond to domains on opposite sides of the twin boundaries. The defect structure due to the heavy ion irradiation is visible as an approximately circular projection of the amorphized region induced by the passage of the Au ion. Around this region, a lobed "bow-tie" contrast is apparent (see bottom panel of figure 64). Previous work by Y. Yan and M. Kirk [11] shows that this lobed "bow-tie" contrast occurs along the b axis and is due to the oxygen reordering (ORO) in the ab planes around the damaged tracks as the Au ions traverse through an anisotropic matrix.

Figure 65 shows a TEM micrograph of a tilted (≈ 15° away from the c-axis) view of the defect structure. Note that the cross section of the defect tracks is not uniform along the length of the defect. Instead, the cross section oscillates quasi-
periodically and the tracks consist of large spheres (90-180 Å diameter) connected by smaller cylinders (diameter ≈ 40 Å).

Figure 63. The Magnetic Hysteresis Loops Measured at 10K, 45K, and 77K for the Unirradiated Crystal (Top Panel) and the Irradiated Crystal (Bottom Panel).
Figure 64. TEM Micrographs of an End-On View of the Columnar Damage Formed by Irradiation With 3.9 GeV $^{197}$Au$^{29}$. 

The shape of the damage track is very similar to the "string of pearls" structure discussed by Hensel et al. [12]. In addition to the visible damage, indirect evidence was found in previous TEM measurements for a higher concentration of point defects in the matrix between the amorphous regions of irradiated samples as compared to unirradiated samples [13].

Figure 65. TEM Micrographs of the Tilted View of the Columnar Damage Formed by Irradiation With 3.9 GeV $^{197}$Au$^{2+}$.

Effects of the Anneals

Critical Currents

The magnetic hysteresis loops were measured after each anneal. In figure 66 we show the results obtained for the 400 °C, 450 °C, 500 °C and 600 °C anneals. The magnetic hysteresis loops are measured at $T=10$ K (top panel), $T=45$ K (middle panel)
and $T=77$ K (bottom panel). It is apparent that the magnetization at $T=10$ K behaves very differently from that at $45$ K and $77$ K. At $T=10$ K the magnetization loops show a decrease in hysteresis after each subsequent anneal. In contrast, the magnetization loops at $45$ K and $77$ K show a large increase in the hysteresis after the $400$ °C and $450$ °C anneals, followed by a decrease in hysteresis with annealing at higher temperatures.

Figure 67 displays the critical current density dependence on the annealing temperature, as extracted from the hysteresis loops. To make the changes in the critical current density with annealing more apparent, $J_c(H)$ was normalized by dividing the value after each anneal, $J_{cA}(H)$, by the value after the irradiation $J_{cI}$ (11). The figure shows the critical current density as a function of anneals for $T = 10$ K (top panel), $T = 45$ K (middle panel) and $T = 77$ K (bottom panel). The complete annealing sequence consisted of 1 hour anneals at each of the following temperatures: $400$ °C, $450$ °C, $500$ °C, $550$ °C, $575$ °C, and $600$ °C, and two additional anneals at $420$ °C lasting 5 days each. The second 5-day anneal at $420$ °C produces no significant change in the critical currents, as compared to the first 5-day anneal. Therefore we show only the effects of the first 5-day anneal.

The critical current density, $J_c$, of the irradiated crystal shows marked changes after annealing at $400$ °C and $450$ °C, as shown in Fig. 67. It is apparent that the $10$ K data behaves very differently than the $45$ K and $77$ K data. At $10$ K, $J_c$ decreases monotonically with each anneal. At $45$ K and $77$ K $J_c$ shows a large increase after the $400$ °C and $450$ °C anneals, followed by a decrease with annealing at higher temperatures. The critical current density at $77$ K increases as high as a factor of 5 over the as irradiated crystal.
Figure 66. The Magnetic Hysteresis Loops Measured as a Function of Anneals at 10 K (Top Panel) and 77 K (Bottom Panel).
Figure 67. Critical Current Density as a Function of the Anneals for 10 K (Top Panel), T=45 K (Middle Panel), and 77K (Bottom Panel).
The effect of annealing on $J_c$ (10 K) for the *unirradiated* crystal is shown in Figure 68 for $H=0$, 0.5 and 1T. There are no appreciable changes in $J_c$ for anneals up to 450 °C. For higher annealing temperatures $J_c$ decreases monotonically with each anneal. Similar results were obtained for $T=45K$ and $T=77K$.

![Figure 68](image)

**Figure 68.** Critical Current Density as a Function of the Anneals at $T=10K$ for the Unirradiated Crystal.

**Low Temperature Anneals**

The strong dependence of the critical current density on the annealing in the 400-450° C range suggests a change in the pinning mechanism. However, as it can be seen in Figure 69, TEM micrographs of the irradiated crystal indicate that a 20 minute anneal at 420 °C produces no visible difference in the morphology of the defects. Figure 69 shows the TEM micrographs of end-on view (images on the left) and tilted ($\approx 15^\circ$ away from the c-axis) view (images on the right side) of the columnar damage.
The micrographs show the damage immediately after the irradiation (top images) and after a subsequent 20-minute anneal at 420 °C (bottom images).

Figure 69. TEM Micrographs of End-On Views (Left Images) and Tilted Views (Right Images) of the Columnar Damage. Immediately After Irradiation With 3.9 GeV \(^{197}\text{Au}^{29+}\) (Top Images) and After a 20 Minute Anneal (Bottom Images).

The changes in \(J_c\) in the irradiated crystal for anneals up to 450 °C must be due to changes in the point defect concentration since the correlated damage tracks, including the ORO or "bow tie" region surrounding the track show no signs of change.
in the TEM micrograph. Previous studies on irradiated samples, containing only point-like disorder have shown that annealing temperatures as low at 100 °C produce significant changes in the point defect concentration [14]. We speculate that the fact that the critical current density decreases at low temperatures and increases at high temperatures may be related to a change in the role of the point defects. At temperatures as low as 10 K, the vortices are rigid and can be pinned effectively by either point-like or columnar defects [1]. At higher temperatures, the dynamics can change. Point defects have been shown to promote flux-line wandering and entanglement, whereas correlated defects inhibit wandering and promote localization [15-19]. As the temperature is increased, the vortex line tension decreases and loops or kinks nucleate more easily in the vortices. If point defects are present near the columnar defect, they might act to help stabilize the loop and reduce the pinning efficiency of the correlated defects. Anneals up to 450 °C would be effective for reducing the number of point defects, thereby recovering the pinning effectiveness of the correlated tracks which are unchanged after these anneals.

At higher temperatures, this would lead to an increase in the critical currents as seen in our data. We would expect this effect to be more pronounced at higher temperatures, where the vortices are more flexible. Notably, after annealing the sample at 450 °C. \( J_c(77K,1T) \) shows a 5 times increase, higher than the 3.5 times increase in \( J_c(45K,1T) \).

**High Temperature Anneals**

For the irradiated crystal, annealing at temperatures higher than 450 °C leads to a decrease in the critical current density at all temperatures measured. A drop in \( T_{c\theta} \) and a broadening of the transition accompanies this decrease in \( J_c \). After the 600
C anneal, $J_c$ dropped to less than 2% of the as irradiated value, while the midpoint of the superconducting transition dropped to $\approx 25$ K and the transition broadened to $\approx 35$ K. The unirradiated crystal also showed signs consistent with a loss of oxygen as can be seen in Figure 68, where $J_c(10$ K) dropped to $\approx 10$ % of its initial value following the 600°C anneal. Changes in the superconducting transition are shown in Figure 70.

![Figure 70. The Normalized Magnetization as a Function of the Temperature.](image)

The normalized magnetization is plotted as a function of temperature, for the as irradiated crystal (solid line), the unirradiated crystal (dashed line), and for both the irradiated (solid squares) and unirradiated (open circles) crystal after the 600°C anneal. Both crystals show drops in $T_c$ and a broadening of the transition consistent with oxygen loss, however the larger changes in the irradiated crystal seem to indicate that the oxygen is more mobile in the irradiated crystal. Previous work by Tetenbaum et al. [20] shows that the equilibrium oxygen concentration decreases as the annealing temperature increases over the 400-750 °C range, consistent with our results.
Thus the critical current will therefore reflect not only a possible change in the morphology of the columnar defects, but also the decrease in the oxygen concentration for the high temperature anneals. In order to isolate the two different effects, we re-annealed the irradiated crystal at 420 °C for two consecutive sets of 5 days each, in order to restore the initial oxygen concentration. After re-oxygenation, the critical currents show an appreciable increase at all temperatures measured. For example, $J_c(10 \text{ K})$ recovers to $\approx 70\%$ of the as-irradiated value. At 45 K and 77 K the recovery of the critical current density is even more pronounced, with final values of $J_c$ as high as three times that of the as irradiated crystal.

The final TEM micrographs for the irradiated crystal after it went through the complete annealing sequence are shown in the bottom panel of Figure 71 for the end-on view (image on the left) and for the tilt at 15° off the c-axis (image on the right). In order to make the changes in the defect morphology more apparent, the figure also contains the TEM images with the damage immediately after the irradiation (top panel) and after a subsequent 20 minute anneal at 420 °C (middle panel).

The TEM results show an unusual change in the defect morphology upon annealing. A removal of the large fluctuations in the diameter of the ion tracks is found, leaving behind a narrow continuous columnar defect. The diameter of the columnar defects after annealing is $\approx 40 \text{ Å}$, similar to the diameter of the thin rods that connected the large spherical defects in the as irradiated sample. The end-on view also shows no evidence of the bow-tie feature, or oxygen reordered regions (ORO) in the ab planes around the damaged tracks. After the high temperature anneals, it is evident that the defects are smaller in size. During the annealing process, the defect area shrinks as it presumably exerts a smaller stress on the surrounding area.
Figure 71. TEM Micrographs of End-On Views (Left Images) and Tilted Views (Right Images) of the Columnar Damage as a Function of Anneals.
The results of our annealing study on the thermal stability of heavy ion irradiation induced defects in YBCO are quite different than what was previously observed in BSCCO. In the BSCCO samples irradiated with 580 MeV Ag ions, Thompson et al. [6], found that $J_c$ decreased monotonically in two stages upon annealing. For anneals between room temperature and 300°C, $J_c$ dropped by ≈ 28% to 38%, and stayed fairly constant for further anneals up to 500°C. A second drop in $J_c$ occurred for anneals higher than 500 °C, with $J_c$ falling to pre-irradiation levels for annealing at 850 °C. Visible changes in the defects were observed with TEM after a 15-minute anneal at 300 °C. The dark strain field surrounding each defect in the as irradiated sample had mostly disappeared and the radius of the columnar defect decreased roughly 14%. After a 15-minute anneal at 700°C, no signs of the columnar defects were visible in TEM. They attributed the two-stage drop in $J_c$ to the annealing of the oxygen defects at lower T (≤ 300 °C) and the loss of the columnar defects at higher T (≥ 500 °C).

In contrast, in YBCO we find no TEM visible changes for annealing up to 420 °C, while anneals above 450 °C alter the morphology of the defects. For anneals at 450 °C < T ≤ 600 °C, the large diameter spherical regions on the "string of pearls" type of defect present in the as irradiated sample are reduced, leaving only a uniform diameter "string, or columnar defect behind. The continuous nature of the defects is preserved even after annealing at 600 °C.

Conclusions

In summary, we have studied the thermal stability of columnar defects in YBa$_2$CuO$_{x}$ single crystals introduced by irradiation with 3.9 GeV $^{197}$Au$^{29+}$. We find that upon annealing up to 450 °C, $J_c$(10 K) decreases by less than 10 %, while $J_c$(45
K) and $J_c(77$ K) increase by as much as a factor of 5. We speculate that this change in the critical currents might be attributed to a decrease in the point defect density in the sample. We find that the defects induced by irradiation are stable upon thermal annealing up to temperatures of 600 °C. Similar increases in $J_c$ upon annealing were recently observed in 6 GeV Pb-irradiated YBCO [21]. The increase in the critical currents at 77K with annealing emphasizes the high suitability of irradiated samples in applications that operate at liquid nitrogen temperatures.


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