# Vortex Liquid State in an Electron Irradiated Untwinned $YBa_2Cu_3O_{7-\delta}$ Crystal

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We report on the temperature and field dependencies of the resistivity of an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal before and after 1 MeV electron irradiation in fields up to 8 T. We show, for the first time, direct suppression of the first order vortex liquid-to-solid phase transition due to the controlled introduction of point defects, and that transport in the vortex liquid is dominated by viscosity rather than by single vortex pinning. Furthermore, we demonstrate that the first order vortex transition can be recovered by reducing the density of point defects through subsequent annealing of the sample.

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Recent investigations of the magnetic phase diagram of the mixed state in $YBa_2Cu_3O_{7-\delta}$ have revealed a large region in which the vortex lattice has melted into a vortex liquid state. In clean untwinned crystals, the melting line which separates the solid and liquid states is apparently of first order, characterized by a sharp "kink," hysteresis, and superheating in the temperature and the field dependence of the resistivity [1–4]. In addition, a "peak effect" in the critical current $J_c$ as a precursor to melting has recently been reported, indicating the softening of the lattice shear modulus prior to melting [5]. Quenched random disorder added to clean systems has been suggested to round first order phase transitions [6]. For instance, features of the first order melting transition have not been observed in thin films with high critical currents [7]. Instead, a second order vortex glass transition has been predicted [8] and observed [7]. While pinning in the vortex solid state has been extensively studied both theoretically and experimentally, pinning of the vortex liquid has not been well characterized. In the liquid state, thermal disorder has been predicted to lead to vortex entanglement [9]. Recently, correlated disorder in the form of columnar defects [10] and twin boundaries [11] has been shown to localize the vortices in the liquid state. A pinning-depinning transition has been reported in very dilutely twinned crystals, suggesting that the vortex may be highly correlated [5].

In this Letter we investigate the nature of the vortex liquid state and the first order vortex solid-to-liquid melting transition by the controlled introduction of *point defects* into a clean untwinned $YBa_2Cu_3O_{7-\delta}$ crystal via electron irradiation. We demonstrate, for the first time, the disappearance of the sharp melting transition with controlled induced disorder. We observe pinning in the liquid state which indicates that the vortex liquid is highly viscous, a signature of possible topological entanglement of the vortices [9]. We also demonstrate that the sharp melting transition and the associated hysteresis in the temperature and field dependencies of the resistivity for both $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{H} \parallel \mathbf{ab}$ can be recovered by subsequent annealing of the sample. This lends further support to the contention that the discontinuity in the magnetoresistivity is indeed an intrinsic feature in untwinned $YBa_2Cu_3O_{7-\delta}$ associated with a first order phase transition.

The single crystal used in our study was grown at Argonne National Laboratory using the self-flux method [12]. The as-grown crystal of dimensions $1200(l) \times 490(w) \times 60(t)\ \mu m^3$ was annealed, detwinned, and had contacts applied for standard four-probe ac resistivity measurements as reported elsewhere [3]. The zero field resistive transition temperature $T_{c0}$ was 93.0 K and width $\Delta T_{c0(10\%-90\%)} \sim 400$ mK with a typical measuring current density of 1 $A/cm^2$.

Electron irradiation of the detwinned crystal was performed at the high-voltage electron microscope (HVEM) accelerator facility at Argonne National Laboratory. The detwinned single crystal was cooled to $T \approx 40$ K to prevent crystalline changes due to beam heating and irradiated with 1 MeV electrons in a vacuum of $10^{-6}$ Torr. An irradiation dose of $1 \times 10^{23}$ $m^{-2}$ created approximately $3.4 \times 10^{15}$ and $2.7 \times 10^{14}$ $m^{-2}$ Cu and O displacements, respectively, in each Cu-O plane of the sample [13]. Subsequent annealing of the sample was performed in air at various temperatures between 100 and 240 °C.

Figure 1 shows the resistive transition in magnetic fields up to 8 T for $\mathbf{H} \parallel \mathbf{c}$ *before* and *after* electron irradiation. The figure is presented in reduced resistivity $\rho(T)/\rho(T_c)$ and temperature $t = T/T_c$, in order to account for the depression of $T_c$ by about 1 K and the 12% increase in the normal state resistivity after electron irradiation as shown in the inset. The thick lines depict the untwinned crystal *before* irradiation and show a smooth decrease in resistivity down to a sharp kink at $T_m$ near the tail of the transition. The temperature $T_m$ where the resistive discontinuity occurs is associated with the first order vortex liquid-to-solid phase transition reported earlier [1–4]. A similar kink in the temperature dependence of the resistivity is also observed for $\mathbf{H} \parallel \mathbf{ab}$ [see Fig. 4(b)] and the resistive height of the kink for both field orientations is $\rho(T_m)/\rho(T_c) \sim 0.18$.

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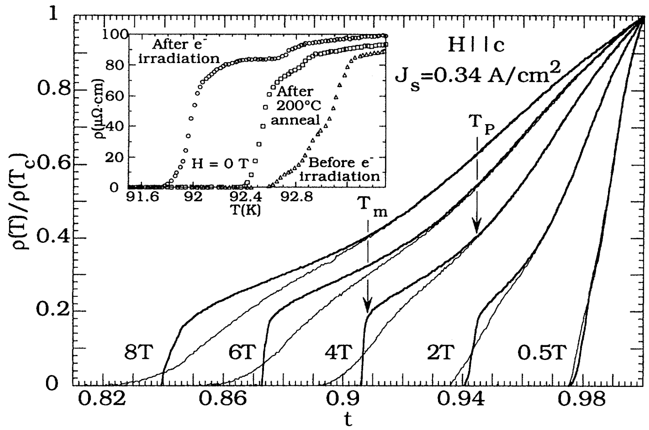


FIG. 1. Resistivity of an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal versus temperature in reduced values for **H** || **c**. Thick lines are *before* the crystal was irradiated with 1 MeV electrons. The thin lines are *after* the electron irradiation. Inset shows the superconducting transition in zero field before (triangles), after (circles) electron irradiation, and after an annealing at 200 °C (squares).

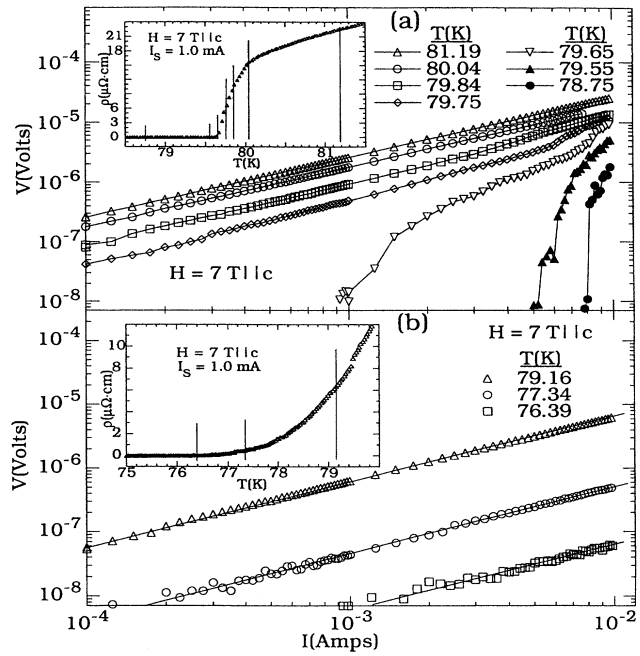


FIG. 2. (a) Voltage-current characteristics for $H = 7$ T || **c** *before* electron irradiation at several temperatures. (b) Voltage-current characteristics *after* electron irradiation at $T = 79.16$, 77.34, and 76.39 K for $H = 7$ T || **c**. Insets show the resistivity versus temperature data with vertical lines indicating the temperatures where the $V$-$I$ curves were obtained.

The thin lines in Fig. 1 show the normalized resistivity *after* electron irradiation for **H** || **c**. Whereas at high temperatures the postirradiation curves follow the preirradiation curves very closely, they begin to deviate downwards at lower temperatures. The dramatic difference between the preirradiation and postirradiation curves is that the sharp kink associated with the first order phase transition completely disappears and the resistivity becomes a smooth, continuous curve. The temperature at which the postirradiation resistivity begins to fall below the preirradiation resistivity corresponds to $\rho/\rho(T_c) \approx 0.4$ and will be referred to as $T_P$. The curves for $H = 2$ and 8 T clearly demonstrate that the deviation between the preirradiation and postirradiation curves increases with magnetic field. If the irreversibility line is defined as the temperature of the first occurrence of a critical current, our results indicate that the irreversibility line will be depressed after electron irradiation. We contend that the electron irradiation induced point disorder prevents the vortex liquid from forming an Abrikosov lattice at the preirradiation freezing temperature $T_m$ and shifts vortex solidification to lower temperatures.

This interpretation is supported by the voltage-current curves shown in Fig. 2. The *preirradiation* untwinned crystal is characterized by Ohmic behavior above and non-Ohmic behavior below the melting temperature, as shown in Fig. 2(a). The S shape in the $V$-$I$ curve before irradiation has been reported earlier [3,14] and has been related to the thermally activated flux flow [15] of the vortex solid at low excitation current and free flux flow of the vortex structure at high excitation currents. *After* irradiation the voltage-current characteristic shows Ohmic behavior at all temperatures investigated as shown in Fig. 2(b) for $T = 79.16$, 77.34, and 76.39 K, indicating that the vortices are in the liquid state at and above these temperatures. This is quite surprising, since, in the presence of weak random point disorder, a second order phase transition from the vortex liquid to a vortex glass state has been predicted [8]. A fit of the electron irradiated resistivity data near the tail of the transition by $\rho \propto (T - T_g)^s$ yields a field-dependent value for the scaling exponent in the range of $1.2 < s < 3.0$ for $1 < H < 8$ T in clear disagreement with the vortex glass prediction of a field-independent scaling exponent closer to 6.5 [7,16].

We now address the question of the dissipation mechanism in the liquid state. The preirradiation resistivity data in the temperature range $T_m < T < T_P$ show linearity in field with a zero intercept, and extrapolate to $\rho_n$ at the mean field $H_{c2}$. Furthermore, the resistivity determined from the high current portion of the S-shaped $V$-$I$ curves and from the curves above $T_m$ shown in Fig. 2(a) agrees quantitatively with that measured in the ac resistivity experiment. Thus, we identify the preirradiation resistivity in the temperature range $T_m < T < T_P$ with the Bardeen-Stephen [17] free flux flow resistivity $\rho_f$. This establishes a reference dissipation where neither pinning by defects nor the vortex viscosity due to shear processes plays a role. Comparison of $\rho_f$ with the postirradiation resistivity allows us to extract the decrease in dissipation due only to the induced point defects. We will show, by examining the activation energy of the point defect resistivity, that it is dominated by the viscosity rather than single vortex pinning interactions. This is consistent with the theory of viscous dissipation in the vortex liquid state near the melting transition [18]. In this theory, the resistivity is given by $\rho \approx \rho_0 e^{-U(T,H)/T}$, where $U$ is a "plastic energy" related to vortex-vortex interactions and not directly to vortex-pin site interactions.

The resistivity due to point defects, $\rho_P$, is extracted from the postirradiation resistivity $\rho$ using the addition formula $\rho = (1/\rho_f + 1/\rho_P)^{-1}$ [19]. As shown in Fig. 3, $\rho_P$ follows an exponential behavior given by $\rho_P = \rho_0 \exp[-U(1 - t)/t]$, where the activation energy $U(T) = U_0(1 - t)$ has the temperature dependence expected for the Ginzburg-Landau condensation energy. The field dependence of $U(T)$ at $T = 83$ K is shown in the inset to Fig. 3 and can be fitted by a power law, $H^{-0.7\pm0.1}$.

The lower resistance value of the postirradiation curve compared to the preirradiation curve in the temperature range $T_m < T < T_P$ in Fig. 1 implies that point defects partially pin the liquid, as has previously been observed for columnar defects [10] and twin boundaries [11]. We can rule out pinning of the individual vortices in the liquid state as the cause of the point defect resistivity. From the volume density of defects, $n$, and the approximation for the elementary pinning force $f_P$ of a small void [20], we obtain a field-independent pinning energy

$$\frac{U_P}{k_B}(T = 83\text{ K}) \approx \frac{\pi}{k_B \Gamma^{1/3}} \varepsilon_0 (n D_\nu^4 \xi_0^2)^{1/3} \sim 0.11\text{ K}.$$

Here $\varepsilon_0 = \phi_0^2/4\pi\mu_0\lambda^2$ is a characteristic vortex energy scale, $D_\nu$ is the defect diameter for Cu and O ions, $\Gamma$ is the effective mass anisotropy, $\phi_0$ is the flux quanta, and $\xi_0$ and $\lambda$ are the superconducting coherence length and penetration depth, respectively. This pinning energy due to the induced point defects is far too small to account for the change in dissipation. Point defect clustering, which has been suggested to occur in other electron irradiations [13] and seen in proton irradiations [21], has to be considered. Although their larger size gives a larger pinning potential, it is still small compared to $T_m$. However, the point defects and defect clusters can create modulations in the vortex velocity. We will demonstrate that the dissipation is dominated by viscous processes in the liquid state, which have much larger activation energies [18,19].

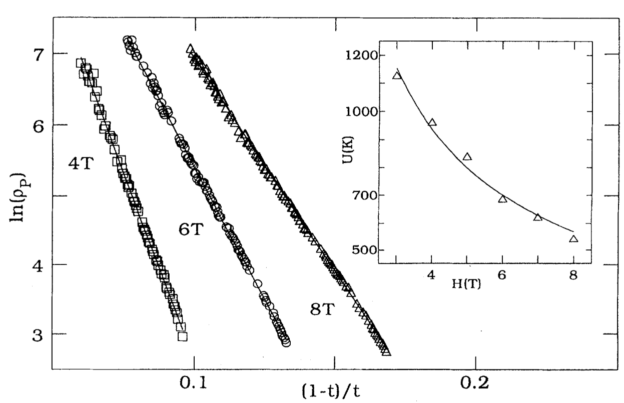


FIG. 3. $\ln(\rho_P)$ vs $(1 - t)/t$ for $H = 4$, 6, and 8 T. Solid lines indicate linear fits to the data. Inset shows magnetic field dependence of the activation energy at $T = 83$ K. Solid line is a fit with $U \sim H^{-0.7}$.

The viscous shear processes that take place in liquid flow and govern the viscosity of the vortex liquid can be related to a "plastic energy" $U_{\rm pl}$ proportional to $T_m$ describing the energy involved in vortex cutting and recombination in an entangled liquid [18,19]. Taking $U_{\rm pl} \propto (1 - t)/H^{0.7}$ as measured in the experiment, we obtain the form of the melting curve from the condition $U_{\rm pl} \sim T_m$, leading to $H_m \sim (1 - t)^{1.4}$. This is in good agreement with recent transport measurements of the melting curve in twinned and untwinned single crystals [1,2]. The experimentally determined activation energy is $U/k_B \approx 800$ K at $T = 83$ K and $H = 5$ T. A recent numerical calculation [22] finds the energy required for vortex crossing near the melting line to be $U_x = 7.5 k_B T_m$. The numerical value of this estimate at $T = 83$ K and $H = 5$ T gives $U_x/k_B \approx 600$ K, the same order as our experimentally obtained value for the same temperature and field. Thus, while direct single vortex pinning in an unentangled liquid is not able to account for the large activation energy or its field dependence, viscous processes associated with vortex entanglement are consistent with both observations.

Thus our data indicate that the reduced dissipation arising from electron irradiation induced point defects is primarily due to viscous processes. The difference in dissipation is attributed to the highly viscous nature of the vortex liquid which becomes apparent only with the introduction of point defects. The probable origin of the viscosity is the entanglement of the vortices arising from thermal disorder [9]. Adhering to this interpretation, our data indicate that vortex entanglement grows with increasing field as shown by $\rho_f - \rho$ in Fig. 1. It is also consistent with recent high field measurements on clean untwinned crystals where the sharp kink in the magnetoresistance associated with the first order phase transition becomes smeared at higher fields, and the finite resistivity curve displays Ohmic behavior [1–3,16]. At high fields, the high density vortices entangle, preventing a first order phase transition at $T_m$, and instead leads to a polymerlike glass state due to the quenching of the entangled vortex liquid at lower temperatures.

Finally, we demonstrate that the first order vortex liquid-to-solid phase transition may be recovered by reducing the density of point defects through subsequent annealing of the crystal. Fig. 4(a) shows the resistive tail of the transition at $H = 4$ T $\parallel$ **c** before irradiation, directly after irradiation, and after subsequent annealing at 200 and at 240 °C. After the first annealing shown,

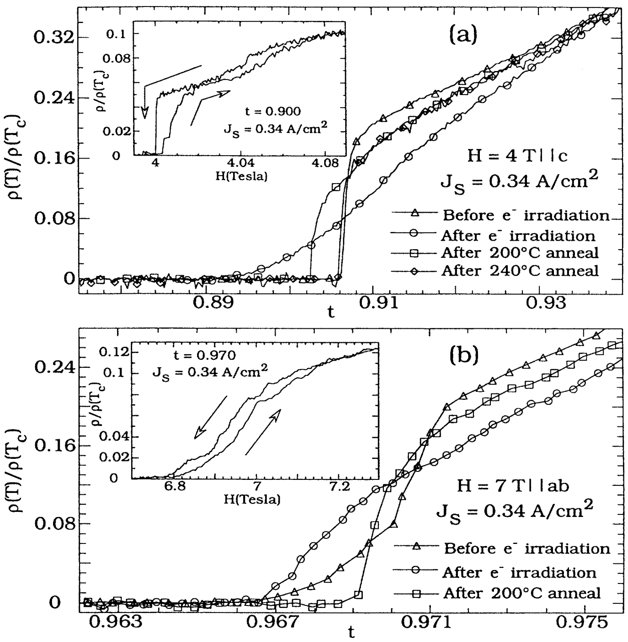


FIG. 4. (a) Resistivity versus temperature in reduced values near the resistive tail for $H = 4$ T || **c** *before* (triangles) and immediately *after* (circles) electron irradiation, subsequent annealing at 200 °C (squares), and further annealing at 240 °C (diamonds). (b) Resistivity versus temperature in reduced values near the resistive tail for $H = 7$ T || **ab** *before* (triangles), immediately *after* (circles) electron irradiation, and after subsequent annealing at 200 °C (squares). Insets show the hysteresis in the field dependence of the resistivity after the 200 °C anneal.

the resistive kink associated with the first order phase transition reappears, and the resistivity in the liquid state increases towards the preirradiation value. The melting temperature $T_m$ occurs at a slightly lower temperature than the preirradiation value. In addition, the hysteresis associated with this kink transition is recovered as shown in the inset to Fig. 4(a). After further annealing, $T_m$ returns to its original preirradiation value, however, the resistivity just above $T_m$ is still slightly depressed. The incomplete recovery of the resistivity is possibly due to some remaining disorder. A similar behavior is observed for **H** || **ab** as shown in Fig. 4(b). The preirradiation curve for $H = 7$ T || **ab** displays a sharp kink which disappears after irradiation. With a subsequent anneal at 200 °C the kink is recovered and is actually sharper than the preirradiation behavior. Furthermore, in the inset of Fig. 4(b) we show, for the first time, hysteresis in field for **H** || **ab** after annealing the sample. The annealing results show that reducing the number of point defects (by Frenkel pair annihilation, defect clustering, or diffusion to the surface or into other preexisting defects) reduces the disorder in the vortex liquid, and enables freezing of the vortex liquid at $T_m$.

In conclusion, we have shown that the first order vortex liquid-to-solid phase transition in clean untwinned crystals of $YBa_2Cu_3O_{7-\delta}$ can be suppressed by the controlled introduction of point defects via electron irradiation. The induced disorder allows observation of the viscosity of the liquid state characterized by an exponential temperature dependence. The source of the viscosity is probably vortex entanglement arising from thermal fluctuations which increase with magnetic field and may give rise to formation of a polymerlike glass. Furthermore, we have shown for the first time that the hysteresis and discontinuity in the magnetoresistance associated with the first order transition for **H** || **c** and **H** || **ab** in clean untwinned $YBa_2Cu_3O_{7-\delta}$ crystals can be recovered by subsequent annealing of the crystal.

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