Defect Independence of the Irreversibility Line in Proton-Irradiated Y-Ba-Cu-O Crystals

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Flux pinning in Y-Ba-Cu-O crystals is studied as a function of fluence of 3-MeV protons, which create random local defects. Order-of-magnitude increases in the critical current density are deduced from magnetic hysteresis loops, with values up to 2×10^5 A/cm^2 observed at 77 K and 1 T. However, the irreversibility line in the field-temperature plane and the pinning potentials deduced from flux-creep studies are hardly changed. These results are compared to melting and pinning models.

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One of the major surprises in the high-temperature superconductors has been the discovery of a more complex magnetic phase diagram than the behavior observed in conventional type-II superconductors. The main new feature[1] is the appearance of an "irreversibility line" in the field-temperature (H-T) plane which separates regions of reversible from irreversible magnetic behavior within the Abrikosov mixed phase region. The onset of irreversible magnetic behavior across this line correlates closely with the onset of critical currents and nonlinear I-V transport characteristics,[2] and it occurs well below[3] the conventional diamagnetic upper critical field H_{c2}. The interpretation of this line has been the subject of intense study,[1] with theoretical proposals ranging from a granular superconducting model,[4] to thermally activated depinning of vortices out of defect potential wells,[5-7] to vortex-lattice melting,[8-10] to vortex-glass freezing.[11]

While such theories are still at an early state of development, it is obviously essential to establish the basic experimental features of the irreversibility line. Its upward curvature in the H-T plane[1] and its approximately logarithmic or weak power-law dependence on frequency[6,8] are well established, but little is known about its dependence on the nature and density of defects in the material. Since the irreversible behavior below the line, which is closely related to the critical current density, depends directly on the defect pinning, it is natural to suppose that the irreversibility line will also.

The central result of this paper is, however, that the irreversibility line is found to be largely *independent* of the defect density in well-characterized Y-Ba-Cu-O crystals, even at defect levels which enhance the irreversible magnetization by more than an order of magnitude. As will be discussed below, this result is difficult to reconcile with most existing theories. It is a basic feature of the irreversibility line in Y-Ba-Cu-O to be considered in the further development of any theory. We also report a closely related and novel observation, namely, that the effective activation energy determined in flux-creep measurements is almost independent of defect density. A further implication, at least in the perspective of the pinning theory, would be that the operative defects determining the critical current density in the as-grown crystals are the same as those introduced by the proton irradiation, which we use to controllably increase the defect concentration. This suggests the likelihood of the local defects being oxygen vacancies or interstitials, since oxygen defects are the ones most likely to exist in the unirradiated crystals.

Five well-characterized fully oxygenated Y-Ba-Cu-O twinned crystals have been used in these experiments. Initial T_c's were near 93.5 K, and transition widths $\Delta T_c < 0.5$ K. Typical dimensions were $1\times1\times0.03$ mm^3 with the c axis parallel to the shorter dimension. Details of sample preparation have been reported previously.[12]

Radiation damage is a standard procedure to controllably introduce defects into a material, and there have been many studies in high-T_c materials using mostly neutrons, but also ions and electrons.[13-18] However, so far these studies have concentrated on increasing critical current density rather than on the physics of the irreversibility line. We chose to use 3-MeV protons, irradiating at room temperature at a flux of 3×10^{12} cm^{-2}sec^{-1}. At the highest doses used in these experiments (2×10^{16} cm^{-2}), T_c decreases by no more than 2 K. The range of 3-MeV protons in Y-Ba-Cu-O is about 45 μm, greater than the crystal thickness. Their predominant interaction is to displace atoms in small, uncorrelated clusters of one or a few displacements. But, about 30% of the total displacements are generated in higher-energy events, also uncorrelated with one another, in which clusters of ~30 displacements are produced. Neutron irradiation produces similar clusters. Monte Carlo calculations[19] show that on average each 3-MeV proton produces ~6 displacements in penetrating 30 μm (assuming a displacement threshold of 20 eV). The calculated mean concentration of displaced atoms for a dose of $10^{16}/cm^2$ is 270 ppm varying ±30% with depth.

Since at least some irradiation-produced defects are mobile at room temperature,[20] it is to be expected that point-defect clusters and extended defects will nucleate and grow during and after irradiation. Electron microscopy has been used to characterize this post-irradiation microstructure. Initial TEM observations after irradiation to a dose of 2×10^{16} cm^{-2} show the presence of small clusters, about 30 Å in size spaced ~300 Å apart. These observations support the notion that the defects caused by proton irradiation are random, but they leave open the question of whether the dominant contributions to flux pinning are from the larger clusters or from background of defects which may not be visible in electron microscopy.

Magnetization hysteresis loop measurements $M(H)$ for applies fields H up to 5.5 T in the $\mathbf{H}\parallel\mathbf{c}$ orientation were performed in a Quantum Design SQUID magnetometer. Figures 1(a) and 1(b) show $M(H)$ of crystal No. 3 at $T=5$ and 77 K, respectively, for various irradiation doses. The most relevant feature of these figures is the systematic enhancement of the irreversible magnetization with increasing dose. The data in Fig. 1(a) emanating from the origin represent the virgin magnetization curve after zero-field cooling, and the constancy of the initial slope confirms that the amount of superconducting material is not significantly reduced at these levels of irradiation. In Fig. 1(b), which shows the data at 77 K, a new feature is evident: the presence of a reversible $M(H)$ regime at high fields, with a field onset nearly independent of dose. A more sensitive measure of the onset of reversibility is provided by the ac susceptibility measurements described below.

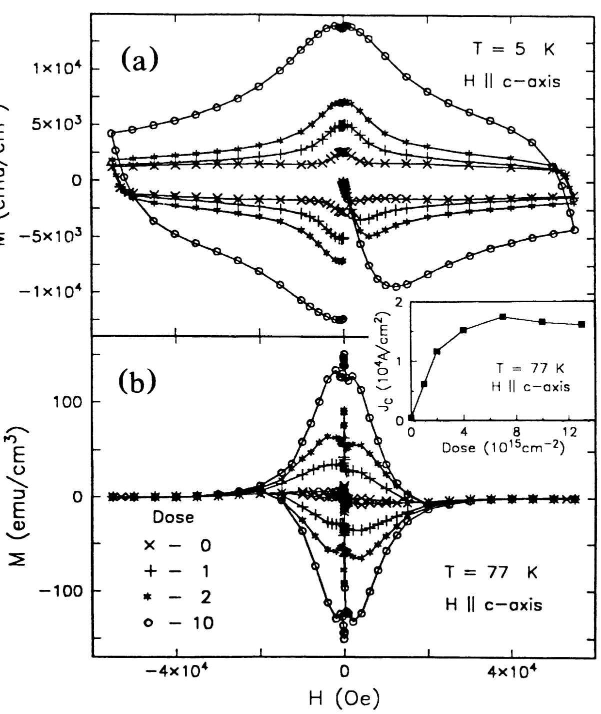

FIG. 1. Magnetization $M(H)$ vs magnetic field of Y-Ba-Cu-O crystal No. 3 for different doses of 3-MeV proton irradiation, at (a) $T=5$ K and (b) $T=77$ K. Inset: Dose dependence of the critical current at 77 K and 1 T. The doses are given in units of 10^{15} protons/cm^2.

We have used the $M(H)$ data to extract the critical current density J_c corresponding to currents flowing in the a-b plane, according to the Bean model.[21] The dose dependence of J_c at 77 K and $H=1$ T, in the inset to Fig. 1(b), shows a linear initial rise followed by a saturation and an eventual decrease at higher doses. In other crystals a peak value of close to 2×10^5 A/cm^2 was obtained,[16] comparable to values reported recently for other irradiation techniques.[14,15]

We have also studied the time relaxation (flux creep) of the magnetization of crystal No. 1 at $H=1$ T and various temperatures from 5 to 60 K using a SQUID magnetometer, both before and after irradiation to a dose of 10^{16} cm^{-2}. The sample was cooled to the desired temperature in zero field. Because of the large critical current in the irradiated crystal, it was necessary to cycle the crystal around a hysteresis loop before stopping at 1 T to ensure that the crystal was in the critical state. The magnetization $M(t)$ was then recorded over a period from 100 to 10000 sec. A logarithmic decay of M as a function of time was observed for all the temperatures measured, for both the unirradiated and the irradiated states. If we use the conventional flux-creep relation[22,23]

$$M(t)=M_0\left[1-\frac{kT}{U_{\rm eff}}\ln(t/t_0)\right], \qquad (1)$$

an effective activation energy $U_{\rm eff}$ can be obtained from the slopes of curves of M/M_0 vs $\ln(t)$. Here t_0 is the vortex-hopping attempt time. If we take M_0 to be the initial magnetization at the beginning of the experiment, we obtain the results for $kT/U_{\rm eff}$ [from Eq. (1)] as shown in Fig. 2. It is clear that irradiation changes $U_{\rm eff}$ by less than 25% over the entire temperature range where reliable measurements were possible ($T=5$–60 K), even though the critical current density at 77 K changes by more than an order of magnitude. We emphasize that at all except the lowest temperatures (e.g., above about 20 K in Fig. 2) $U_{\rm eff}$ is only an effective activation barrier because the assumptions which led to Eq. (1) break down in the limit of large relaxation (large T) and in the presence of a distribution of barriers. Its meaning in the context of recent theories of collective pinning[24] and the vortex-glass transition[11,25] will be discussed elsewhere. The apparently finite value of $kT/U_{\rm eff}$ as T approaches zero suggests a tunneling contribution.[26]

A third measurement is that of the irreversibility line, defined here by the peak in the loss component χ'' of the ac susceptibility[27] measured at 1 MHz. Shown in Fig. 3

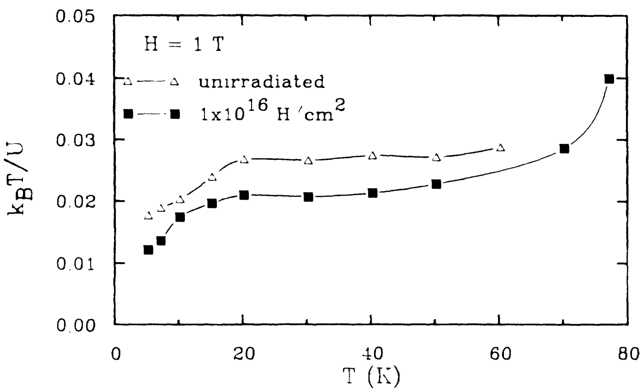

FIG. 2. kT/U_{eff} as a function of temperature for sample No. 1, before irradiation and after irradiation to a dose of 10^{16} cm^{-2}. The critical current at 77 K and 1 T for this crystal was 2.2×10^3 before irradiation and 2.0×10^5 A/cm^2 after irradiation.

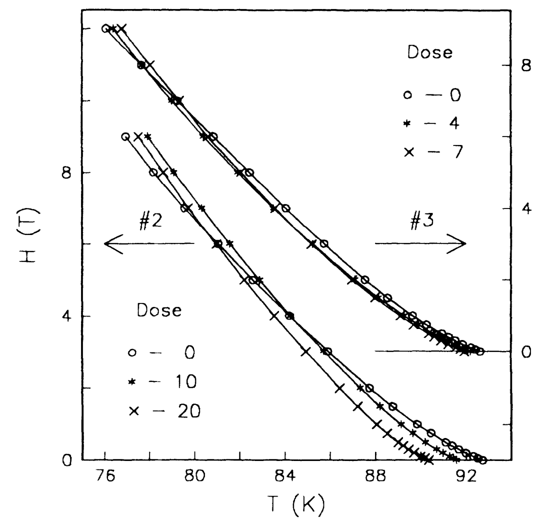

FIG. 3. Irreversibility line $H_{irr}(T)$ of samples No. 2 and No. 3 as a function of proton fluence, determined by ac susceptibility. The vertical scale has been shifted for clarity.

are the results for crystals No. 2 and No. 3, where the line is only slightly shifted by the irradiation, in spite of the large enhancement of J_c. We have also made a preliminary measurement of the frequency dependence of the irreversibility temperature at a given field (7 T). It is weak and approximately logarithmic, as reported earlier,[6] and, what is more, it is essentially the same in irradiated and unirradiated samples. This is a further confirmation of the apparent defect independence of the irreversibility line. Whether the irreversibility line as measured by ac susceptibility is an approximation of a true phase transition[11] or whether it represents a measurement-dependent crossover between fast- and slow-flux dynamics[5] is still unsolved. However, the observation that the onset of irreversibility is almost independent of the irradiation dose, when measured either by ac susceptibility or through dc magnetization (although we recognize that the last technique provides a poor determination of H_{irr}), is the central experimental result of this paper.

Next, we examine several alternative models for the irreversibility line and flux creep. The defect independence of the irreversibility line is at first glance most easily explained by the vortex-lattice-melting model,[8-10] in which the line is determined by the competition between intervortex elastic energy which keeps the lattice ordered and the thermal energy which disorders it. By assumption, defects play no role here. Such a model is reasonable when the spacing of defects is greater than several vortex-lattice spacings. As discussed above, however, even the large clusters appear at spacings of only 300 Å, while at 1 T, the vortex-lattice spacing $a_0 \equiv \sqrt{\phi_0/B}$ is still about 450 Å. Alternatively, if the pinning is weak enough, the vortices can be pinned collectively,[24] forming "Larkin domains" of ordered hexagonal structure, which are disordered only on a larger length scale. Vortex-lattice melting of the type described above can then occur *inside* the Larkin domains. However, it remains to be established theoretically how sensitive the size of the Larkin domains will be to the defect density and what effect this should have on the activation energy.

Another widely considered model for the irreversibility line is based on thermal activation of vortices out of pinning wells.[4-6,22,23] The hopping rate will be dominated by a factor $\exp(-U/kT)$, where U is the barrier energy, and thus the irreversibility line is essentially determined by the condition U/kT=const. With an appropriate H and T dependences of U, one can then explain the observed H-T and frequency dependences.[4,5] Our observed defect independence of the irreversibility line seems nicely consistent in this perspective with the defect independence of the flux-creep barriers U at low temperatures.

Nevertheless, problems in this picture arise from considering the conventional relation[23] between J_c and U, expressed in terms of a pinning force per unit volume,

$$J_cB = U/aV, \qquad (2)$$

where a is the size of the potential well and V is usually considered to be an activation volume. Thus, unless V depends inversely on defect concentration, it is hard to understand our experimental result that U could remain constant while J_c increases strongly with concentration.

If widely spaced pinning centers are strong enough to individually pin a vortex, which is elastically bound to a surrounding bundle, U becomes the pinning potential of one defect, and V in Eq. (2) can be interpreted as the volume per defect. Then we obtain the desired result that U is constant and J_c scales with dose or defect concentration. Once the distance between defects becomes smaller than a_0, V becomes limited by a_0 (at least in the a-b plane) and there is a tendency for J_c to saturate, as observed experimentally. Nevertheless, this picture is put in doubt by the likely presence of a higher density of local point pinning centers. Evidence that these, rather than the larger clusters observed in TEM, are the relevant pins comes from the fact that the activation energy barriers are essentially the same both before and after irradiation.

For a higher density of pinning centers, collective pinning theory[24] becomes relevant. In this case, the activation volume V is the Larkin-domain volume $V_c = R_c^2 L_c$, where R_c and L_c are the transverse and longitudinal dimensions, respectively. The net pinning energy is given by[7,24] $U_c = \sqrt{WV_c}$, where W is a mean-square pinning force per unit volume. For the localized defects generated by the irradiation, W will depend linearly on dose Φ. R_c and L_c are known[7,24] to scale as the square of the vortex-lattice elastic moduli, divided by W. Combining these simple relationships with Eq. (2) leads to the results that $U \propto 1/\Phi$ and $J_c \propto \Phi^2$, which disagrees with our experimental results. Thus three-dimensional collective pinning can be ruled out.

In summary, we see that existing theories have difficulty reconciling our observation of the defect independence of the irreversibility line and activation barriers with the simultaneous large increase in the critical current density of these proton-irradiated Y-Ba-Cu-O crystals. The results are also consistent with our observations of comparable irreversibility lines[28] in laser-ablated Y-Ba-Cu-O films which have even larger critical current densities compared to the crystals. It will, of course, be important to explore other high-temperature superconductors, in which results could be quite different because of the different anisotropies or coherence lengths, and to develop theories of the defect dependence in other models such as vortex-glass freezing.[11]

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