Magnetic-field dependence of critical currents in proton-irradiated YBa₂Cu₃O_{7- δ} films: Conventional behavior of the pinning-force density

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We have measured the magnetic-field dependence (with the field applied perpendicular to the a-b plane) at 77 K of the critical current density (J_c) of a proton-irradiated YBa₂Cu₃O_{7- δ} thin film. When fluences of 2-MeV protons were varied from 0 to 2×10^{16} protons/cm², the transition temperature (T_c) and the field at which J_c vanishes were diminished slightly compared with the suppression of J_c . The curves of pinning-force density versus field can be scaled to follow a common functional dependence, which indicates that the distribution of pinning energies in the film is not altered by increasing proton irradiation. The pinning-force data indicate that the critical current is limited by thermally activated flux creep between sites on the flux lattice, and the reduction in $J_c(H)$ is explained by the suppression of T_c with increasing proton damage.

A major obstacle to the application of high- T_c superconductors for use in high magnetic fields is the suppression of the critical current density (J_c) at high temperatures (77 K) by magnetic fields much less than the upper critical field, H_{c2} . YBa₂Cu₃O_{7- δ} has an extremely high upper critical field, but the transport critical current vanishes at a much lower field (H^*) closely related to the irreversibility field H_{irr} .^{1,2} The irreversibility field is essential in defining a boundary in the H-T plane, the irreversibility line. Above this line, the field cooled and zero-field cooled magnetization curves coincide because the effects of flux pinning become negligible.

While several models have been proposed to explain the existence of the irreversibility line, no consensus has been reached as to whether H_{irr} depends on just extrinsic factors (e.g., the underlying defect structure) or also on the intrinsic parameters of the superconducting state (e.g., T_c or H_{c2}). Recent magnetization data by Civale *et al.*² show that the irreversibility line and T_c remain fixed even while the critical current density of a single crystal of YBa₂Cu₃O_{7- δ} is enhanced dramatically after irradiation by protons. We present data measured on a YBa₂Cu₃- $O_{7-\delta}$ thin film which show that changes in $H^*(77 \text{ K})$ after proton irradiation are due to reductions in T_c by the proton-induced damage.

Studies of the pinning-force density (F_p) in conventional superconductors have shown that when the number of defects is changed but the distribution of pinning energies remains fixed, plots of F_p vs H_a can be scaled to lie on a universal curve.^{3,4} To examine the effect of changing the defect structure on the pinning energy distribution, we have measured the pinning-force density of a protonirradiated thin film of YBa₂Cu₃O_{7- δ}. F_p is derived from inductive measurements of the transport critical current as a function of applied field (H_a) at 77 K. After irradiating the film with 2-MeV protons, changes in $J_c(77 \text{ K}, H_a)$, H^* , and T_c are correlated to determine the effect of the proton-induced damage on the film.

Two films were grown on (100) $SrTiO_3$ substrates by pulsed-laser deposition.⁵ The films were analyzed by x-ray diffraction and found to be *c*-axis aligned, and pole-

figure analysis indicated that only 90° misorientations were present in the *a-b* plane.⁶ The better film was selected for the irradiation study based on the sharpness of its superconducting transition ($T_c = 88 \pm 0.25$ K) and its larger J_c at 77 K, 3.29×10^6 A/cm². The values for T_c and J_c are consistent with the results attained for highquality, *in situ* films grown at other labs. A beam of 2-MeV protons was rastered across the film at room temperature to produce four steps of equal proton dose (5×10^{15} protons/cm²). The protons were prevented from channeling by positioning the film's normal axis at a 7° angle with respect to the beam direction. The 2-MeV protons did not lose a significant fraction of their energy traversing the 3000-Å film, so the depth profile of the damage was essentially uniform.

The technique used to measure T_c and J_c of the films is described completely in Ref. 7. Briefly, the transition temperature (T_c) and critical current density (J_c) are measured using an ac technique which induces transport currents in the film. Currents at frequencies of 10 kHz $(T_c$ measurement) or 1 kHz $(J_c$ measurement) are passed through a coil coupled to the film. As the temperature or the amplitude of the ac drive current is varied, changes in the harmonic content of the coil voltage are correlated to T_c or J_c of the film being measured.

The critical current was measured at 77 K in magnetic fields generated by a superconducting solenoid held in the persistent-current mode. The field was applied normal to the film's surface (parallel to the *c* axis) for each measurement so that the least favorable pinning direction was sampled. After each field change, the sample was warmed to 120 K and then cooled in the applied field to reduce ambiguities arising from stray shielding currents in the film. This allowed measurements of J_c to be reproduced to within 5%.

We generally see that the film's superconducting and normal-state properties are degraded by the proton irradiation. The room-temperature resistivity increases from 285 to 354 $\mu\Omega$ cm over the full course of the irradiation, and J_c is depressed sharply with field and with increasing proton dose. The pinning-force density, calculated from the critical current density $(\mathbf{F}_p = \mathbf{J}_c \times \mathbf{B}_a)$, is plotted in Fig. 1. The plot shows that $\mu_0 H^*$, the field at which F_p approaches zero, decreases little with increasing proton fluence, compared with the decrease in height of each curve, F_p^{max} .

Scaling F_p by F_p^{\max} and H_a by H^* and plotting $f(F_p/F_p^{\max})$ vs $h(H_a/H^*)$ reveals the common functional form for the pinning-force density plotted in Fig. 2. This functional form is similar to the measurements of Kerchner et al.⁸ on YBa₂Cu₃O_{7- δ} thin films, and to results that we have obtained in YBa₂Cu₃O_{7- δ} thin films deposited on MgO and LaAlO₃ substrates. The functional dependence of the scaling is similar to that observed in many conventional superconductors, ^{3,4} namely, $F_p/F_p^{\text{max}} \propto (H_a/H_{c2})^{1/2}(1-H_a/H_{c2})^2$. However, the field dependence seen in Fig. 1 differs from that observed in conventional superconductors in that the scaling field H^* (~5) T) is much lower than the upper critical field (≈ 26 T).⁹ At low temperatures, the appropriate scaling field should be H_{c2} because the ratio, H_a/H_{c2} , partially defines the elastic moduli of the flux lattice. This apparent inconsistency between data measured on YBa₂Cu₃O_{7- δ} and classical superconductivity theory is resolved by considering the thermally activated hopping of fluxoids between flux-lattice sites (i.e., thermally activated flux creep).¹⁰

Specifically, Tinkham¹⁰ has shown that the fieldinduced broadening of the resistive transition can be explained by thermally activated flux creep. This model has been extended by Hettinger *et al.*¹¹ to describe the scaling of the pinning-force density in thin films of YBa₂Cu₃- $O_{7-\delta}$. They assume that the critical current at low magnetic fields is characterized by single-particle pinning, and that at some crossover field, the single-particle pinning is then limited by flux creep. The measurements that we report here extend to low fields where the field dependence of f_p is found to be $h^{1/2}$, as shown by the inset of Fig. 2.

In the flux creep model, macroscopic currents in the su-



FIG. 1. Pinning-force density vs applied field at 77 K.



FIG. 2. The scaled pinning-force density, where the scaling field is defined from the point at which J_c approaches zero. The solid line is proportional to $h^{0.46}(1-h)^{2.26}$ and the dotted line represents a fit to Eq. (2).

perconductor tip the potential wells that trap the flux bundles.¹² As the current density is increased, the flux bundles are accelerated to a drift velocity v by the Lorentz force. Once the vortex drift velocity reaches a critical value, v_c , dissipation in the film is detected. Thus, a voltage or resistance criterion can be defined in terms of v_c . In our experiment, the current-dependent dissipation results in the appearance of components of the coil voltage at odd harmonics of the drive frequency. Then, J_c is defined by extrapolating, to zero voltage, a line tangent to the third-harmonic voltage versus current curve at the point corresponding to an electric field of 10 μ V/cm.^{7,13}

In the limit of strongly interacting vortices, the fluxoids form an ordered lattice. The lattice may be distorted by the pinning forces so that only short-range order is obtained; nevertheless, thermally activated motion of fluxoids dissipates energy in the superconducting state. To describe the thermally activated hopping of flux bundles between sites on the flux lattice, the pinning-force density can be written as 10-12

$$J_{c}B = (k_{B}T/V_{d}L)\sinh^{-1}\{\exp[U_{0}/k_{B}T - \ln(v_{0}/v_{c})]\}.$$
(1)

 (v_c/v_0) is the ratio of the minimum detectable vortex drift velocity to that with the vortices completely depinned, U_0 is the pinning energy, observed to be proportional to 1/B, $^{10,14}V_d$ is the volume over which the driving force on a moving flux bundle is determined, and L is the distance over which a bundle is moved.) Tinkham argues¹⁰ that an energy of the order of the difference between a square and a triangular Abrikosov lattice is needed to move a flux bundle from one lattice site to the next. The hopping energy can then be written as $U_0 = 0.02(4\pi\mu_0)H_c^2\xi\phi_0/B$. (H_c is the thermodynamic critical field, ξ is the Ginzburg-Landau coherence length, and ϕ_0 is the flux quantum.) Hettinger *et al.*¹¹ argue that V_d can be represented as a parallelepiped with dimensions N_4 coherence lengths in the field direction, N_3 magnetic penetration depths (λ) in the direction of motion, and N_2 flux-lattice spacings $[a_0 = (\phi_0/B)^{1/2}]$ wide. Then, V_dL can be written as $V_dL = N\lambda\xi\phi_0/B$ where $N = N_2N_3N_4$. We rewrite Eq. (1), by substituting the above relations for U_0 and V_dL , the depairing critical current $(J_{c0} = 4H_c/3\sqrt{6}\lambda)$, and the Ginzburg-Landau relation ($\phi_0 = 2\sqrt{2}\pi\mu_0H_c\xi\lambda$),

$$F_p = F_p^0(B/B_0) \sinh^{-1} \{ \exp[\ln(v_0/v_c)(B_0/B - 1)] \}, \qquad (2)$$

where $B_0 = (0.03\sqrt{3}\phi_0^2)J_{c0}/k_BT\ln(v_0/v_c)$ and $F_p^0 = 0.06 \times \sqrt{6\pi\mu_0H_cJ_{c0}/N\ln(v_0/v_c)}$. Thus, the pinning energy can be scaled in terms of the parameters B_0 and F_p^0 , if $\ln(v_0/v_c)$ is constant. The last assumption is reasonable for the data presented here because v_0/v_c should be relatively insensitive to irradiation.

The pinning-force density data of Fig. 2 are scaled to unity by F_p^{max} and H^* . Since the scaling which would be appropriate to Eq. (2) is different, the parameters F_p^{max} and H^* are converted to values corresponding to F_p^0 and B_0 by fitting the data of Fig. 2 to Eq. (2) using F_p^0 , B_0 , and $\ln(v_0/v_c)$ as adjustable parameters. We fit the data using a nonlinear-least-squares routine and then calculate the values of B_c/N and J_{c0} from F_p^0 and B_0 , respectively. The constant, $\ln(v_0/v_c)$, is found to be 6.6; B_0 and J_{c0} are plotted in Fig. 3 as a function of their expected temperature dependences¹² $[J_{c0}^{2/3}=1-(77 \text{ K})/T_c$ and $(1-B_0/N)^{1/2}=(77 \text{ K})/T_c]$. The linear correlation seen in these plots indicates that the decrease in the critical current with proton irradiation can be explained by the reduction of T_c in the bulk of the film. We estimate that $E_0 \approx 0.74$ mV/cm based on the experimental criterion for J_c , $E_c = 10$



FIG. 3. J_{c0}^{23} vs T/T_c (solid squares) and $(1-B_0)^{1/2}$ vs T/T_c (open circles). The linear correlation of the plots indicates that the radiation is changing the field dependence of the critical current by reducing T_c . Note that $J_{c0}=11$ MA/cm² corresponds to $U_0B=U^0=0.10$ eVT, in agreement with the values reported in Ref. 12.

 μ V/cm, and the value of $\ln(v_0/v_c)$, 6.6, found by fitting the scaled F_p vs *B* data. In a 2-T field, the corresponding drift velocity is ≈ 0.3 m/s, consistent with the predictions of the Bardeen-Stephen theory.¹⁵

This picture changes at low fields where the fluxoids are spaced far enough apart that they probably interact individually with the pinning sites rather than with their nearest neighbors in the Abrikosov lattice. The \sqrt{B} dependence of F_p that we observe in low applied fields (see Fig. 2) is similar to that predicted by Kramer³ for plastic shearing of the vortex lattice and by Dobrosavljević-Grujić¹⁶ for pinning at parallel grain boundaries. However, since the fluxoids are pinned individually, the functional dependence of F_p on B_a depends on the details of the microstructure of the pinning sites rather than on the dynamical properties of the flux lattice.^{17,18} The change in the field dependence of F_p may indicate that $B_a = 1$ T marks a crossover from a region where the pinning volume (V_p) is determined by the inter-vortex spacing to a region where V_p is fixed by the spacing between pinning sites.

Pinning-force data have been obtained from magnetization data measured on single crystals of YBa₂Cu₃O_{7- δ} by Li *et al.*¹⁹ They observe a functional dependence for F_p , $F_p \propto h^2(1-h)^4$, that is different from the one we observe in thin films. One explanation for this difference is that J_c is several orders of magnitude smaller in single crystals than in thin films of YBa₂Cu₃O_{7- δ}. This indicates that the defect structure responsible for the flux pinning is different in thin films from that in single crystals of YBa₂Cu₃O_{7- δ}. Also, there can be problems with the determination of J_c from magnetization measurements. Daeumling, Seuntjens, and Larbalestier²⁰ point out the difficulties in inferring J_c values from magnetization versus applied field data, especially in oxygen-deficient YBa₂Cu₃O_{7- δ} single crystals.

Our result that $J_c(77 \text{ K})$ is monotonically reduced by radiation damage is consistent with the observations of other investigators. Roas *et al.*²¹ see similar reductions for thin films of YBa₂Cu₃O_{7- δ} irradiated by ¹⁶O ions. However, our observations differ from the data of Schindler *et al.*²² where the J_c of a neutron-irradiated thin film of YBa₂Cu₃O_{7- δ} is enhanced at small fluence levels. One explanation for this difference is that the neutrons do less damage per unit fluence than charged particles and so produce a different type of defect.²¹

The effect of irradiation on bulk materials is quite different from that on thin films. Experiments by van Dover *et al.*²³ show that flux pinning in single crystals of YBa₂Cu₃O_{7- δ} is enhanced by proton irradiation. In another study,²⁴ not only J_c , but also the irreversibility line, is shown to increase upon irradiation of bulk YBa₂Cu₃O_{7- δ} by fast neutrons. This contrasts with the data of Civale *et al.*²⁵ which indicate that while $J_c(H,77$ K) is enhanced, the irreversibility line remains fixed in single crystals of YBa₂Cu₃O_{7- δ} irradiated by protons.

The measurements of Ref. 25 probe the effect of temperature changes and irradiation on the scaling of the pinning-force density in single crystals of YBa₂Cu₃O_{7- δ}. The data show that the field dependence of the pinningforce density, measured at different temperatures, can be scaled to follow a universal function. The same is not demonstrated for pinning-force density data measured at different levels of radiation damage (for fluences similar in magnitude to those reported here in thin films) in YBa₂Cu₃O_{7- δ} single crystals. Rather, for radiation doses small enough so that T_c remains unchanged, the value of F_p^{max} in single crystals increases by a factor of 10 while the value for B_0 does not change. At the highest dose (8×10¹⁶ cm⁻²), T_c is reduced by 4 K and B_0 (77 K) is decreased roughly by $\frac{1}{2}$. In contrast, the J_c data presented here show that F_p^{max} , B_0 , and T_c decrease monotonically over the full range of applied irradiation doses. Moreover, the scaling of the pinning-force density with irradiation dose is shown to be consistent with the changes in T_c .

The test for scaling indicates that radiation changes the

- ¹A. P. Malozemov, in *Physical Properties of High Temperature Superconductors*, edited by D. M. Ginsberg (World Scientific, Singapore, 1989), p. 71, and references therein.
- ²L. Civale *et al.*, Phys. Rev. Lett. **65**, 1164 (1990).
- ³Edward J. Kramer, J. Appl. Phys. 44, 1360 (1973).
- ⁴W. A. Fietz and W. W. Webb, Phys. Rev. **178**, 657 (1969).
- ⁵D. B. Chrisey *et al.*, J. Superconductivity **4**, 57 (1991); K. S. Grabowski *et al.* (unpublished).
- ⁶K. S. Grabowski (private communication).
- ⁷J. H. Claassen, IEEE Trans. Magn. **25**, 2233 (1989); J. H. Claassen, M. E. Reeves, and R. J. Soulen, Jr., Rev. Sci. Instrum. **62**, 996 (1991).
- ⁸H. R. Kerchner et al., in High Temperature Superconductors: Fundamental Properties and Novel Materials Processing, edited by David Christen, Jagdish Narayan, and Lynn Schneemeyer, MRS Symposia Proceedings No. 169 (Materials Research Society, Pittsburgh, 1990), p. 903.
- ⁹U. Welp et al., Phys. Rev. Lett. 62, 1908 (1989).
- ¹⁰M. Tinkham, Phys. Rev. Lett. 61, 1658 (1988).
- ¹¹J. Hettinger et al., Phys. Rev. Lett. 62, 2044 (1989).

distribution of pinning energies in single crystals but not in thin films. The as-grown single crystals are relatively free of defects so that radiation enhances flux pinning as long as the doses are low enough that T_c is not affected. In contrast, the as-deposited films may have a high enough density of defects that the pinning is already optimized. Further damage reduces T_c and the pinning energy, in a manner consistent with predictions of the Anderson-Kim flux creep theory.¹²

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- ¹²M. Tinkham, Introduction to Superconductivity (Krieger, Malabar, FL, 1980); P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. 36, 39 (1964).
- ¹³J. W. Ekin, Appl. Phys. Lett. 55, 905 (1989).
- ¹⁴Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
- ¹⁵J. Bardeen and M. J. Stephen, Phys. Rev. 140, A1197 (1965).
- ¹⁶Lj. Dobrosavljević-Grujić, Phys. Rev. B 36, 1899 (1987).
- ¹⁷H. R. Kerchner, J. Low Temp. Phys. 50, 337 (1983).
- ¹⁸E. H. Brandt, Phys. Rev. Lett. **57**, 1347 (1986).
- ¹⁹J. N. Li et al., Physica C 169, 81 (1990).
- ²⁰M. Daeumling, J. M. Seuntjens, and D. C. Larbalestier, Nature (London) **346**, 332 (1990).
- ²¹B. Roas et al., Appl. Phys. Lett. 54, 1051 (1989).
- ²²W. Schindler et al., Physica C 169, 117 (1990).
- ²³R. B. van Dover *et al.*, Nature (London) **342**, 55 (1989), E.
 M. Gyorgy *et al.*, Appl. Phys. Lett. **56**, 2465 (1990).
- ²⁴H. Küpfer *et al.*, IEEE Trans. Magn. **27**, 1369 (1991).
- ²⁵L. Civale *et al.*, Phys. Rev. B **43**, 13732 (1991).