Vortex Confinement by Columnar Defects in $YBa_2Cu_3O_7$ Crystals: Enhanced Pinning at High Fields and Temperatures

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We report the realization of a microstructure which leads to very strong high-temperature flux pinning in $YBa_2Cu_3O_7$ crystals. Aligned discontinuous columns of damaged material, about 50 Å in diameter and more than 15 μm long, are produced by 580-MeV Sn-ion irradiation. The enhancement of flux pinning is largest when the applied magnetic field is aligned with these tracks. At high temperatures and fields the pinning is much greater than that produced by random point defects, and causes a considerable enlargement of the irreversibility region in the H-T plane.

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Deliberate introduction of defects by irradiation in $YBa_2Cu_3O_7$ has been shown [1–4] to result in large increases in magnetization for temperatures and fields in the irreversible region. However, in this material the boundary between the reversible and the irreversible region in the H-T plane (the irreversibility line) does not change noticeably upon proton irradiation [3]. This raises the question of whether the position of this line is an intrinsic feature of the material. The present work shows that the irreversibility line can be moved to higher field and temperature if a special microstructure is provided to pin the magnetic flux lines. This microstructure consists of linear, discontinuous columns of damaged material extending through the crystal, and is obtained by very-high-energy heavy-ion irradiation. Our results indicate that the pinning of flux lines is directional and is particularly strong when the magnetic field is aligned with the columns of damage.

Flux pinning from radiation damage is ascribed to the creation of local regions where the superconductivity is *weakened,* so that the energy cost of locating a flux line in this region is reduced. The system therefore *gains* [5] an amount of condensation energy $U_p = \eta(H_c^2/8\pi)v$, where $\eta \leq 1$ is the fractional suppression of superconductivity in the defect, H_c is the thermodynamic critical field, and v is the volume of the vortex core that is pinned. Along the field direction, v is limited by the greater of the coherence length ξ or the defect size. The point defects created by light-ion irradiation are not the optimum to pin the vortex array. A *dilute* random distribution of point defects will pin only a small fraction of the total length of each vortex, and the net pinning energy per unit volume will be small. If the defect distribution is *dense,* each vortex will simultaneously interact with a large number of them. In this situation, described by the collective pinning theory [6], the core energy will be almost independent of the location of the flux line, and the critical current will be significantly smaller than that expected from a simple sum of elemental pinning forces. We have indeed found [7] that the amorphous limit [6] of collective pinning provides the best description for our results in proton-irradiated $YBa_2Cu_3O_7$ crystals.

Obviously, defects that confine a longer section of vortex core should provide better pinning. The *optimum* pinning sites should consist of columns of nonsuperconducting material that completely traverse the sample *in the direction of the applied field.* To provide the maximum pinning force, the diameter of these columns should be of the order of ξ. We have produced such defects by irradiation with 580-MeV Sn ions.

Several $YBa_2Cu_3O_7$ single crystals, grown using a flux-melt technique [8], have been used in these experiments. All of them had initial T_c's near 93.5 K and transition widths of approximately 0.5 K. The crystals are platelets of typically $1\times1\times0.02$ mm^3, with the c axis parallel to the shortest dimension. Prior to irradiation crystals were characterized by dc magnetization measurements at fields up to 5.5 T, and by ac susceptibility at $\omega = 1$ MHz and fields up to 9 T, to determine the location of the irreversibility line $H_{irr}(T)$ [9].

The crystals were irradiated with 580-MeV $^{116}Sn^{30+}$ at the Holifield Facility at Oak Ridge National Laboratory. Irradiation was done with the incident beam close to the c axis (in fact 2° from it, to avoid axial channeling) and at incident angles of 30° and 45°. Different crystals were irradiated to doses of 4.8×10^{10}, 1.5×10^{11}, and 2.4×10^{11} ions/cm^2, which were chosen so that the density of damage tracks would match the vortex density at fields of 1, 3, and 5 T, respectively. Throughout the remainder of the paper we will refer to dosages in terms of this equivalent field, B_Φ. The irradiations were done at room temperature at a flux less than 2×10^8 ions/cm^{2}sec, where beam-heating effects are negligible.

It has recently been established [10,11] that ionization energy loss by fast heavy ions can produce permanent damage effects in $YBa_2Cu_3O_7$ if the linear ionization energy-loss rate exceeds 2 keV/Å, and that this damage, unlike that produced by elastic collisions with nuclei in the target, forms a linear track. We have used 580-MeV Sn ions, whose ionization energy-loss rate is 2.7 keV/Å and whose range in $YBa_2Cu_3O_7$ is 26.8 μm. The ionization energy-loss rate of these ions exceeds 2 keV/Å over the first 18 μm of their path in $YBa_2Cu_3O_7$. Thus, the damage microstructure expected from these irradiations consists of very long columns of highly defected material, aligned with the initial beam direction, but arranged randomly in the plane perpendicular to the beam.

We have confirmed this expectation in TEM observations of a crystal irradiated to a dose equivalent to $B_\Phi = 3$ T at an incident angle of 2° to the c axis. Micrographs of this sample after thinning are shown in Fig. 1. The upper part of the figure shows an end-on view of the damage tracks, whose density was measured as $(1.35 \pm 0.20) \times 10^{11}$ cm^{-2}, in excellent agreement with the nominal dose of 1.5×10^{11} cm^{-2}. The columnar nature of the damage is illustrated in the lower micrograph, which shows the same area tilted by 27°. The columns can be seen to consist of small ($\sim$50 Å) regions of damage separated by about 100 Å. The angular spread of the tracks is due to multiple scattering of the ions, and indicates that the TEM sample was made from a depth towards the end of the tracks, where also the ionization density is expected to be less than the maximum.

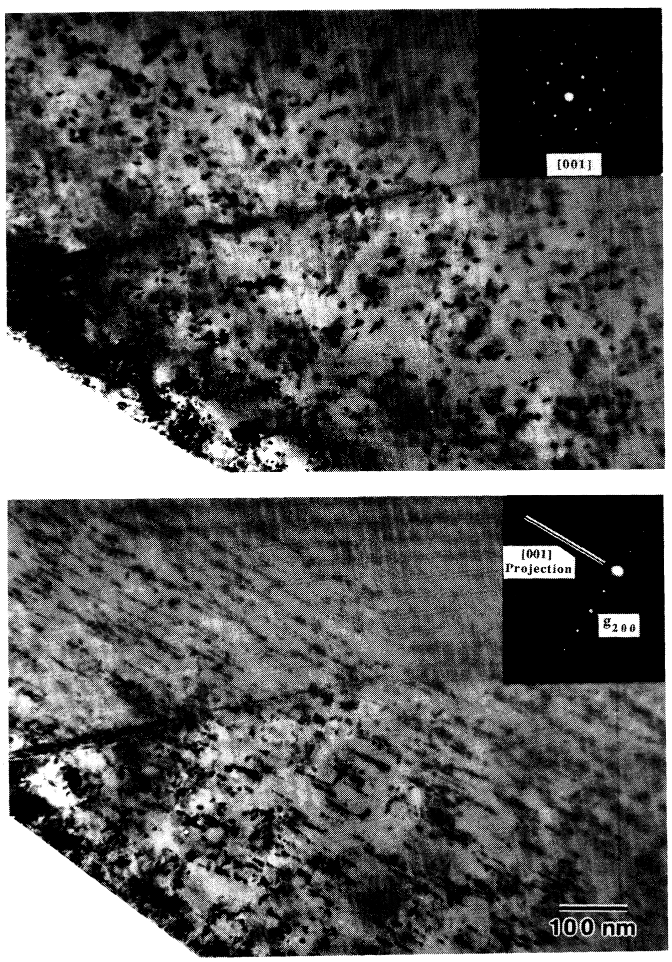

FIG. 1. Electron micrographs of the same area of an irradiated crystal imaged in multiple-beam conditions at two tilts after thinning. Upper: exact alignment with the c axis (001 pole), and showing the ion tracks end-on. Lower: tilted 27° in a (200) direction. The bar in the inset diffraction pattern shows the irradiation direction. The tracks are seen to consist of discontinuous columns of highly damaged or amorphous regions, each about 50 Å in diameter and 100 Å apart.

The critical current J_c was derived using the Bean critical-state model [12] from dc magnetization measurements. Figures 2(a) and 2(b) show J_c at $T = 5$ and 77 K, respectively, for three $YBa_2Cu_3O_7$ crystals irradiated at 2° from the c axis, with doses equivalent to $B_\Phi = 1$, 3, and 5 T. In all the cases the applied field H was parallel to the c axis, and, consequently, to the tracks of damage (within a few degrees). For comparison, we have plotted J_c data corresponding to our best result obtained in proton-irradiated crystals [3,7], and for a typical unirradiated crystal.

At high temperature and high field, heavy-ion irradiation produces much larger enhancement of J_c than proton irradiation. In fact, we see in Fig. 2(b) that at $T = 77$ K and $H = 1$ T, the crystal irradiated with the largest dose of heavy ions has $J_c \simeq 4.5 \times 10^5$ A/cm^2, similar to the best reported values [2] for irradiated $YBa_2Cu_3O_7$, but for $H > 3$ T, where the unirradiated and proton-irradiated crystals have close to zero J_c, the J_c in Sn-irradiated crys-

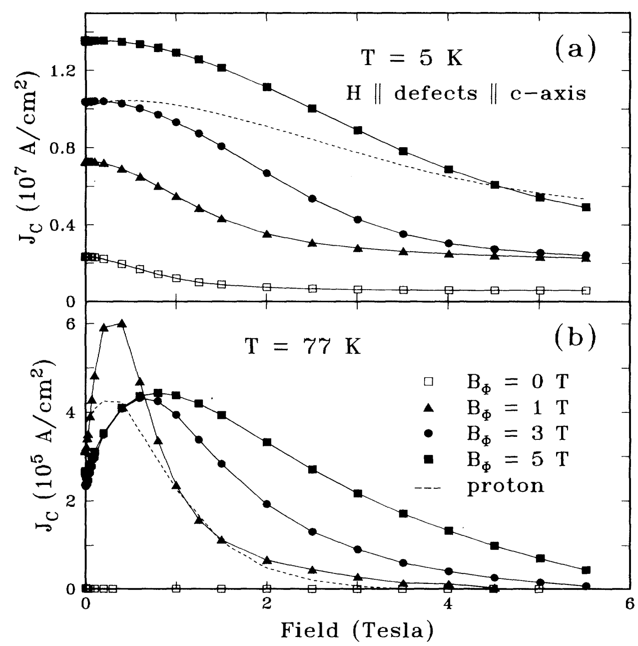

FIG. 2. The critical current J_c, in the a-b plane, at (a) $T = 5$ K and (b) $T = 77$ K for three crystals irradiated with 580-MeV Sn ions along the c axis at doses that produce densities of defects equal to the vortex density at fields of 1, 3, and 5 T. For reference, our best results for a proton-irradiated crystal and an unirradiated crystal are shown.

tals is still quite significant. At low temperatures, the results of proton and heavy-ion irradiation are similar. It is important to emphasize that, while the J_c values for proton-irradiated crystals shown in Fig. 2 are the maximum values that we have been able to obtain (further proton irradiation reduces J_c), the J_c of Sn-irradiated crystals is monotonically increasing with dose at the dose levels that we have studied so far. It is thus likely that higher heavy-ion doses will further enhance J_c.

Evidence that the strong pinning is due to the alignment of flux lines with damage columns is shown in Fig. 3. This crystal was irradiated at a dose of $B_\Phi = 3$ T at a 30° angle with respect to the c axis. Both hysteresis loops shown in the figure were taken at $T = 70$ K, with the magnetic field applied at an angle of 30° from the c axis. In one case H was parallel to the direction of irradiation, and in the other the crystal was tilted in the opposite direction so that the angle between H and the columnar defects was 60° (see Fig. 3). With this procedure we can clearly separate defect alignment effects from the intrinsic anisotropy of the material. In both orientations J_c is considerably enhanced with respect to the preirradiation result in the same conditions (in the scale of the figure, the preirradiation hysteresis loops are indistinguishable from the horizontal axis), but there is clearly a larger enhancement when H is aligned along the damage tracks. In particular, while the reversible regime is reached at fields above 4.5 T in the misaligned orientation, in the aligned case the hysteresis loop remains open up to the maximum accessible field, 5.5 T. We found that the difference in J_c between aligned and misaligned configurations is particularly pronounced at high temperatures, and becomes less at lower temperatures. In a previous work in $YBa_2Cu_3O_7$ thin films heavy-ion irradiated in the c-axis direction [13], a small enhancement of J_c for **H**∥**c** was accompanied by a reduction for **H**∥**a**-**b**, and this was attributed to an alignment effect. However, recent measurements in $YBa_2Cu_3O_7$ films irradiated with protons show similar results [14]. In contrast, the data of Fig. 3 clearly establish an alignment effect. Weak-angle-dependent pinning in $YBa_2Cu_3O_7$ is also produced by twin boundaries [15], but not by the random distribution of point defects created by proton irradiation [16].

Figure 4 shows the ac irreversibility line in the **H**∥**c** configuration for the crystals of Fig. 2. It is apparent that the location of $H_{irr}(T)$ is progressively shifted to higher temperatures with increased irradiation doses. This result is consistent with the observation that Sn irradiation is effective in extending the region of nonzero J_c to larger fields (Fig. 2). These data indicate that, at the characteristic temperature of 77 K, the potentially useful $J_c > 0$ regime of $YBa_2Cu_3O_7$ can be enlarged by several tesla. This shift should be contrasted with the results of proton [3,16] and neutron [4] irradiation on single-crystal $YBa_2Cu_3O_7$ which show almost no change in the location of the irreversibility line.

For the crystals irradiated along the c axis, when **H**∥**c** the confined volume of the core is $v \simeq \pi \xi_{ab}^2 L$, where L is the length of the track, which can approach the total thickness of the crystal. Thus $U_p \simeq \eta (H_c^2/8\pi) \pi \xi_{ab}^2 L$, which is larger by a factor L/ξ_c than for the case of random point defects. The main consequence of this larger U_p is the reduction of the thermally activated relaxation. The advantage of columnar defects over random point defects is thus expected to be larger at high temperature, where thermal relaxation is more significant, as indeed observed in Fig. 2. The larger U_p also explains the observed shift in the irreversibility line. Preliminary measurements of the time decay of J_c show a smaller relaxation rate in Sn-irradiated crystals as compared with unirradiated and proton-irradiated crystals.

Where thermal relaxation can be neglected, $J_c = cU_p/$

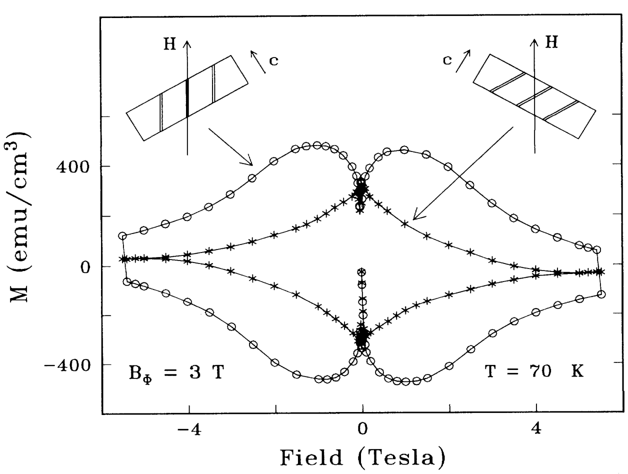

FIG. 3. Hysteresis loops taken at 30 K for a crystal irradiated at 30° off the c axis. The hysteresis loops are shown with the applied field aligned ±30° with respect to the c axis. Inset: The relationship between the radiation and field directions.

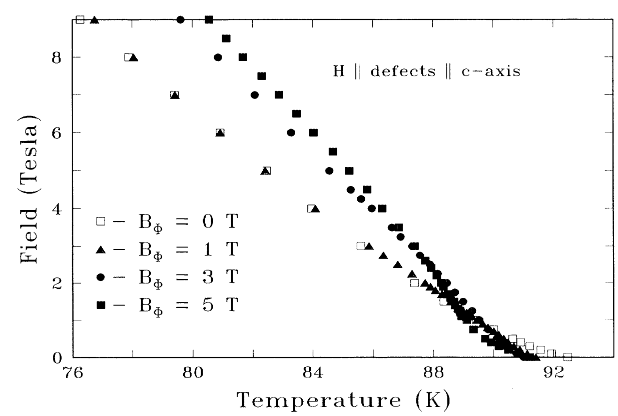

FIG. 4. The irreversibility line measured by ac susceptibility for three crystals irradiated at a dosage equivalent to one damage track for each flux line at 1, 3, and 5 T. An unirradiated crystal is shown for reference.

BlV_c, where $l \simeq \xi_{ab}$ is the size of the defect in the direction of the vortex movement, V_c is the volume of the bundle, and c is the speed of light. If $B \ll B_\Phi$, we can suppose that all the vortices are pinned, and $V_c = a^2 L$, where $a = (\phi_0/B)^{1/2}$ is the vortex spacing. Thus

$$J_c \simeq \eta c \frac{H_c^2}{8\pi} \pi \frac{\xi_{ab}}{\phi_0} = \frac{3\sqrt{3}}{16} \eta J_0 ,$$

where J_0 is the depairing current. In this regime J_c is independent of both B and B_Φ. If $B \gg B_\Phi$ all the pinning sites are occupied and $V_c \simeq d^2 L$, where $d \simeq (\phi_0/B_\Phi)^{1/2}$ is the distance between defects, then

$$J_c \simeq \eta c \frac{H_c^2}{8\pi} \pi \frac{\xi_{ab}}{\phi_0} \frac{B_\Phi}{B} .$$

At low temperature, the large magnitude of the estimated self-field [17,18] (0.7, 1.1, and 1.8 T for $B_\Phi = 1$, 3, and 5 T) and the limited field range of the magnetometer make it impossible to test either limit in detail. It is also clear from the comparison with the results of proton irradiation that point defects do almost as well as the linear tracks.

At 77 K the self-field is much smaller. As a result, the general features of our simple analysis are evident in the data of Fig. 2(b): At low field there is very little dose dependence, while at higher fields J_c decreases with field, and there is a definite J_c increase with increasing dose. The reason for the decrease in J_c at low applied field is not clear and deserves further study. Obviously, a detailed analysis of the results at this temperature must include thermal activation.

In summary, we have used heavy-ion irradiation to generate long tracks of damaged material in $YBa_2Cu_3O_7$ single crystals. We have found that J_c largely increases at all temperatures and fields, and that the enhancement is largest when the applied field is parallel to the irradiation direction. In contrast to light-ion irradiation, this microstructure significantly enlarges the irreversible regime. The results demonstrate that columnar defects are more effective than point defects at pinning flux lines at high temperatures and fields.

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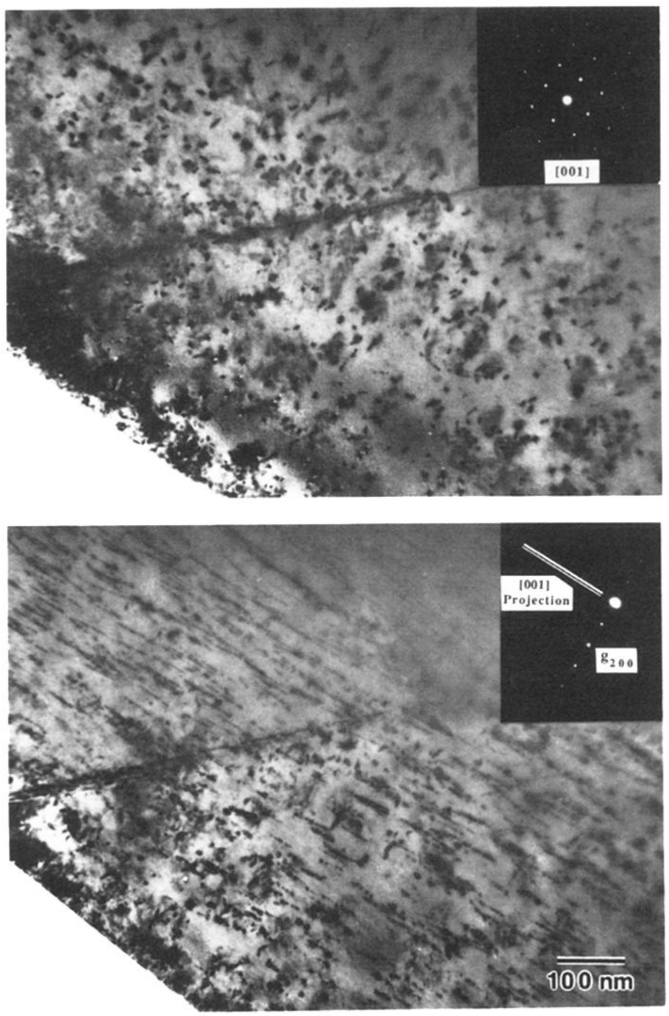

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