Suppression of surface barriers for flux penetration in $Bi_2Sr_2CaCu_2O_{8+\delta}$ whiskers by electron and heavy ion irradiation

J. K. Gregory, M. S. James, and S. J. Bending Department of Physics, University of Bath, Bath BA2 7AY, United Kingdom

C. J. van der Beek and M. Konczykowski

Laboratoire des Solides Irradiés, Ecole Polytechnique, 91128 Palaiseau Cedex, France (Received 4 April 2001; revised manuscript received 18 June 2001; published 12 September 2001)

We have used micron-sized linear Hall probe arrays to investigate the effects of irradiation on surface barriers for flux penetration in individual superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ whiskers. Samples were irradiated with 2.5-MeV electrons or 9-GeV heavy (Pb) ions. The magnetization was investigated in the temperature range between 5 K to above the superconducting transition temperature, in magnetic fields up to 1 T. At all temperatures, irradiation by high-energy electrons or swift heavy ions reduces the penetration field substantially. At low temperatures (T < 50 K) we attribute this to the suppression of a Bean-Livingston (BL) surface barrier for two-dimensional "pancake" vortices, and our results are in *reasonable* agreement with recent theoretical predictions. At high temperatures (T > 50 K) we tentatively propose that the reduction in $H_p(T)$ is due to suppression of a BL surface barrier for flux lines. While electron irradiation strongly reduces magnetic irreversibility at high temperatures, the moderate hysteresis measured after heavy ion radiation suggests that this creates additional bulk pinning for flux lines on columnar defects.

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The magnetic properties of the highly anisotropic hightemperature superconductors (HTS's) continue to attract considerable attention. In particular, the physical mechanism controlling the penetration field (H_p) at which flux first enters the bulk of the zero-field-cooled superconductor remains controversial. It is well known that H_p can be much larger than the lower critical field H_{c1} due to the influence of kinetic barriers at the sample edges,¹ although this effect is partially compensated by the shape-dependent demagnetization effect. While a number of distinct types of surface barriers exist, two are of particular relevance for bulk samples.² Bean-Livingston (BL) barriers arise from the competition between the attraction between a vortex and the sample surface and the repulsion between the vortex and the Meissner screening currents flowing at the very same surface.³ In addition, a barrier of geometrical origin may also be present in plateletlike single crystals of rectangular cross section, such as those found in HTS's. This arises from the increased line energy and the repulsion by the Meissner currents in the corners experienced by the necessarily curved flux lines as they attempt to enter the platelet.⁴ In contrast to the BL barrier, which is strongly suppressed by surface damage, the geometrical barrier is expected to be very robust since the Meissner currents in a flat geometry extend over the entire sample width. It is also sufficiently high that, well below T_c , there should be no appreciable thermal activation over it.

Although a geometrical barrier is often invoked to explain magnetization data in HTS's, compelling evidence exists to the contrary. The pronounced suppression of $H_p(T)$ in YB₂C₃O_{7- δ} single crystals after low-dose electron irradiation was interpreted as evidence for a disorder-sensitive BL barrier.⁵ Furthermore, we have recently demonstrated⁶ that the penetration field (T < 50 K) of single-crystal Bi₂Sr₂CaCu₂O_{8+ δ} (BSSCO) whiskers displays an exponen-

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tial temperature dependence in excellent agreement with recent estimates^{7,8} for the creep of independent twodimensional pancake vortices over a Bean-Livingston surface barrier. Additional evidence for this conclusion arises from the observed asymmetric relaxation rates for vortex entry and vortex exit⁹ and the fact that the magnetization for flux entry is well described by $M \sim H_p^2/2H$ for $H \gg H_p$.⁸ References 7 and 8 predict

$$H_p \approx 0.76 H_c \exp\left[-\frac{T}{T_0}\right] \quad (T \ll T_0), \tag{1}$$

where $H_c = \Phi_0/2\sqrt{2}\pi\mu_0\lambda\xi$ is the thermodynamic critical field, Φ_0 is the flux quantum, λ is the penetration depth, and ξ is the coherence length. kT_0 is a characteristic energy (which is itself temperature dependent) given by

$$kT_0(T) = \frac{\varepsilon_0(T)sk}{\ln(t/t_0)}.$$
(2)

Here, *s* is the separation of the copper-oxide planes, *t* is the time since application of the field *H*, t_0 is a fundamental time scale for vortex oscillations, and $\varepsilon_0(T) = \Phi_0^2/4\pi\mu_0\lambda^2 = \Phi_0 H/\sqrt{2}\kappa$ is the vortex line energy per unit length ($\kappa \equiv \lambda/\xi$ is the Ginzburg-Landau parameter). The effects on the surface barrier of the controlled modification of the bulk sample (pinning) properties by irradiation have, until now, not been considered. In Ref. 8, it was supposed that the opening of new penetration channels after surface damage by irradiation changes the prefactor of Eq. (1) but not the exponent. Here we report systematic investigations on the effect of swift electron and heavy-ion irradiation on the surface barrier in Bi₂Sr₂CaCu₂O_{8+ δ} whiskers. Whiskers have the advantage that they show very weak bulk pinning; moreover, their small cross section of $0.4 \times 12 \ \mu m^2$ means that the field

perturbation (or the magnetic moment) due to the presence of a surface barrier overwhelms that produced by the Bean critical state due to bulk pinning.

Irradiation by swift heavy ions introduces strongly pinning amorphous columnar defects into the superconducting matrix. The effect of this on the surface barrier should be threefold.¹⁰ First, the attraction between the vortex and the surface is altered by the presence of the columnar defects. This can be taken into account by summing over an infinite number of image vortices inside the columnar defects as well as outside the sample. More importantly the fact that the Meissner current will be restricted to flow between the specimen surface and the columnar defects means that the current density will be larger than that in the sample before irradiation; for the investigated defect density of 5×10^{10} cm⁻² it is larger by a factor of nearly 2.¹⁰ Moreover, the Meissner current is expected to be largest near the circumference of the columns, closest to the surface, leading to the nucleation of vortices on the first "row" of defects in the sample. Thus, the introduction of nonsuperconducting holes near the surface is effectively equivalent to the creation of preferential "channels" for penetration. This effect leads to a decrease of the prefactor in Eq. (1) by an amount proportional to the increase of the Meissner current.

The final consequence of heavy-ion irradiation is that, since the first vortices are nucleated on a column (of radius c_0), one should account for the fact that the vortex free energy per unit length ε_0 is decreased by the pinning energy $U_P = \varepsilon_0 (c_0/2\xi)^{1/2}$.¹¹ Naively this is expected to lower both T_0 and the prefactor in Eq. (1), yielding

$$H_p \approx \sqrt{2}\kappa \frac{\varepsilon_0 - U_p}{\Phi_0 s} \exp\left[\frac{T \ln(t/t_0)}{(\varepsilon_0 - U_P)s}\right].$$
 (3)

The effects of electron irradiation are more subtle. The introduction of point defects by this method increases the pinning energy as well as the vortex entropy due to pancake wandering¹² so that T_0 is again reduced, be it to a much lesser extent than after heavy-ion irradiation. However, there is no substantial modification of the image force or of the Meissner currents, so that the main effect is expected to arise from surface damage.

A number of individual BSCCO whiskers from the same growth batch have been investigated. The samples were produced by annealing a quenched melt of appropriate stoichiometry in flowing oxygen; a more detailed description of the whisker growth and characterization is given in Ref. 13. The whiskers are of high crystallographic perfection and have no extended defects, although there may be some point defects that give rise to bulk pinning at very low temperatures (T< 20 K). They do not have optimum oxygen stoichiometry as indicated by their critical temperatures, which we estimate to be 77 and 79 K for whiskers I and II, respectively. These estimates were obtained by fitting a cubic polynomial to the high-temperature $H_p(T)$ data and extrapolating to zero field. Due to their very regular surfaces and narrow widths (whisker I had dimensions $108 \times 12 \times 0.4 \,\mu \text{m}^3$; whisker II had dimensions $44 \times 12 \times 0.4 \,\mu \text{m}^3$), the magnetic properties of these whiskers are dominated by surface effects, and they represent model systems for flux penetration studies.

Measurements have been carried out using miniature GaAs/Al_xGa_{1-x}As heterostructure Hall probe arrays based on a 2- μ m-wide wire width with 4- μ m center-to-center spacing between Hall voltage contacts. The probes were operated with a 2- μ A rms 32-Hz ac current and the Hall voltage detected with a lock-in amplifier. The whiskers were positioned in the desired location on the Hall probe with a micromanipulator where they were held by their mutual electrostatic attraction (see inset of Fig. 2). Characterization of the same whisker was attempted both before and after irradiation, but this was not always possible due to damage that sometimes occurred during manipulation. The samples were then mounted on a temperature-controlled probe and inserted into a ⁴He cryostat containing a small superconducting solenoid, with the magnetic field applied along the whisker c axis, perpendicular to the (largest) a-b face.

Samples I(AG) and I(EI) represent the same whisker before (as-grown, AG) and after electron irradiation (EI). The latter was performed at 20 K using 2.5-MeV electrons from a Van de Graaf accelerator, which produces randomly distributed isolated Frenkel pairs. The damage produced by the known dose of 160 mC is estimated to be 1.7×10^{-4} displacements per atom (d.p.a.).¹⁴ Agglomeration of small defect clusters is expected when the sample is warmed to room temperature for transfer to the measuring cryostat. The residual damage, after room-temperature annealing, is in the range of $(0.6-1.2) \times 10^{-4}$ d.p.a. Whisker II(HII) (after heavy-ion irradiation, HII) has been irradiated along the caxis with heavy ions (9-GeV Pb ions) at 20 K and a total fluence of 5×10^{10} ions/cm² ($B_{\phi} \cong 1$ T). Each ion is known to produce a continuous amorphous track through the whisker with a diameter of 5-7 nm;¹⁵ the induced columnar defect serves as an oriented pinning site for vortices. In this case we do not have an unirradiated reference data set, but, since all the whiskers from this growth batch behaved in a very similar fashion, we expect the virgin $H_p(T)$ to coincide with that of whisker I(AG) to within $\pm 25\%$.

Figure 1 shows six "local" magnetization loops (defined by $\mu_0 M_l = B_m - \mu_0 H_a$, where B_m is the measured induction and H_a is the applied field) measured at the center of the whiskers at temperatures of 10, 20, 40, 50, 65, and 70 K. The symbol M_1 is used to differentiate between our "local" magnetization and the conventional bulk magnetization. The field is applied parallel to the crystallographic c axis (the thin dimension of the whisker). In each panel, H_p can be identified as the field at which M_l deviates sharply from a linear diamagnetic behavior near the origin. The magnitude of the "local" magnetization and the Meissner slope depends to some extent on the separation between whisker and Hall probe, which may vary not only when the whisker is repositioned on the array, but also as function of temperature. However, H_p is independent of such considerations. Clearly the penetration fields and the width of the hysteresis loops reduce very rapidly as the temperature is increased in all cases. At 40 K and above, the asymmetry between the increasing and decreasing branches of the local magnetization loops of all the whiskers is entirely characteristic of a sys-



FIG. 1. Local magnetization loops at various temperatures for whisker I(AG) (solid black lines), I(EI) (dark gray lines) and II(HII) (black dashed lines) measured near the sample center with the applied field parallel to the c axis.

tem dominated by a surface barrier.¹ This arises because the surface barrier hinders vortex entry much more than vortex exit.⁵ At low temperatures, e.g., at 10 K, the $M_l(H)$ loops become more symmetric and it appears that bulk pinning is beginning to play a role. Figure 1 allows a direct comparison of the effect of irradiation on the "local" magnetization loops of the whiskers. Electron irradiation [I(EI)] results in a sharp reduction in the penetration field at a given temperature as compared to the virgin state [I(AG)], with even stronger suppression evident in the heavy-ion-irradiated whisker [II(HII)].

In Fig. 2 we have plotted the temperature dependence of the penetration field for each sample on a semilogarithmic plot. At low temperatures ($T \le 50$ K) an exponential temperature dependence of the form predicted in Eq. (1) is displayed over nearly two orders of magnitude in H_p in all cases. Irradiation seems to reduce the prefactor of Eq. (1) with almost no change in the exponent. This is illustrated in Fig. 2 by the near parallel fit lines for whiskers I(AG), I(EI), and I(HII) at T < 50 K, which yield values for T_0 of 18.5, 17, and 19.4 K, respectively. These agree well with values of 18 and 14 K we have measured previously in different unirradiated whiskers⁶ and compare reasonably well with estimates of 10 ± 1 K (Ref. 7) and 27.3 K (Ref. 16) in large BSCCO single crystals. The prefactor is reduced by a factor 1.2 for



FIG. 2. Temperature dependence of the field of first flux penetration, $H_p(T)$, for whisker I(AG) (open circles), I(EI) (filled circles), and II(HII) (filled triangles). The dashed line is a fit to Eq. (1) for the parameters indicated in the text. The inset shows an optical micrograph indicating the relative orientation of whisker I(AG) and the Hall bar.

the electron-irradiated whisker, and by a factor 3.6 in the ion-irradiated whisker, in good agreement with recent calculations.¹⁰ Also shown in this figure (dashed line) is a fit to Eq. (1) for the unirradiated whisker where the temperature dependence of $\varepsilon_0(T)$ has been explicitly included, assuming $\varepsilon_0(T)s = 1000 \text{ K} \times (1 - T/T_c), \quad \kappa = 120, \text{ and } \ln(t/t_0) = 30.$ Note that we have had to scale the data by $\frac{1}{12}$ to get asymptotic agreement at low temperature, and even then the fit is relatively poor above T = 20 K. We attribute this to the fact that Eq. (1) has been calculated in the London approximation and neither treats the vortex core nor the nucleation of the instability of the order parameter at the point of vortex entry and is therefore not a full description of penetration. We also note that the approximately constant slope observed in Fig. 2, even after heavy-ion irradiation, is in contradiction to the predictions of Eq. (3). The inapplicably of Eq. (3) is perhaps to be anticipated since, in the presence of columnar defects, the maximum of the penetration barrier will lie at an intermediate position between the surface and defect, which will not have been lowered by the full pinning energy.

At higher temperatures (T > 50 K) we are unable to obtain a good fit to Eq. (1) with the same set of parameters that was used to generate the dashed line in Fig. 2. In a previous publication we have tentatively attributed the behavior in this regime to the penetration of well-correlated flux lines over a BL barrier.⁶ To gain deeper insight into this regime we have estimated the irreversibility at a given temperature via the quantity $\Delta M_l(T) = M_{l\downarrow}(2H_p,T) - M_{l\uparrow}(2H_p,T)$. This is plotted in Fig. 3 for the different samples as a function of temperature. Since the width of our whiskers is only ~5–10 times the separation between the whisker and Hall probe, which changes a little each time a sample is mounted, ΔM_l can vary by as much as 25% between manipulations, and



FIG. 3. Measured irreversibility $\mu_0 \Delta M_l$ (see definition in text) calculated at $2H_p$ for whisker I(AG) (solid black line), I(EI) (dark gray line), and II(HII) (dotted line) with the field applied parallel to the *c* axis.

only effects of greater magnitude can be considered significant. We find that the irreversibility of whisker I(AG) is reduced manyfold by electron irradiation [I(EI)], suggesting that, even at these high temperatures, its origin is predominantly due to a BL-type surface barrier. This is consistent

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with results on large BSCCO single crystals,¹⁷ where the transport critical current was shown to fall by a factor of up to 20 when surface barriers were eliminated in a Corbino measurement geometry. The situation is not so clear-cut in the heavy-ion-irradiated whisker [II(HII)], which displays quite a wide plateau in ΔM_1 with magnitude slightly larger than whisker I(EI). We propose that in this case heavy-ion irradiation both decreases the surface barrier and increases the bulk pinning for flux lines via the introduction of amorphous columnar defects. We note that the sharp drop in $\Delta M_1(T)$ near 75 K may be due to the same entropic reduction of pinning that causes the barrier to decrease in this temperature regime.

In conclusion, we present compelling evidence that flux penetration in BSCCO whiskers is governed by creep over Bean-Livingston-type surface barriers, which can be considerably weakened by high-energy electron or swift heavy-ion irradiation. At low temperatures (T < 50 K) the exponential form of $H_p(T)$ is in reasonable agreement with recent theories^{7,8} for creep of 2D pancakes over a BL barrier. At higher temperatures (T > 50 K) irradiation also suppresses the penetration field and the irreversibility, suggesting that their origin is again due to BL-type surface barriers. We tentatively propose that the creep of flux lines over BL surface barriers is important in this regime.

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