PHYSICAL REVIEW B VOLUME 47, NUMBER 18 1 MAY 1993-II

Flux flop in Y-Ba-Cu-O crystals irradiated with 5.3-GeV Pb ions

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(Received 27 January 1993)

Y-Ba-Cu-O crystals were irradiated with 5.3-GeV Pb ions. The induced columnar defects are either perpendicular or at 45° to the Cu-O planes. Magnetization curves confirm an anisotropic, unidirectional enhancement of the critical current J_c. However, a sharp crossover to *isotropic* enhancement is observed in the low-field limit. We interpret these results in terms of flux flop from a direction determined by the field to the direction of the defect. From this feature we determine the pinning energy of a flux line in a columnar defect.

Heavy-ion irradiation[1–4] has been used extensively to introduce pinning centers, in a controlled way, in high-temperature superconductors (HTS). Irradiation with ions such as Pb, Sn, and I produces defects in the form of amorphous columnar tracks embedded in essentially undamaged superconducting matrix. As expected from the size and shape of these defects, they yield strong enhancement of flux trapping with a unidirectional anisotropy, namely, stronger trapping is observed for fields parallel to the direction of the defect.[2]

In this paper we report on magnetic measurements in Y-Ba-Cu-O crystals irradiated with 5.3-GeV Pb ions[4] which produce *continuous* cylindrical amorphous tracks along their paths, with a diameter of 7 nm. We confirm previous observations of unidirectional flux trapping enhancement.[2] However, we report on a different feature: When the external field is reduced, the flux trapping enhancement becomes independent of the direction of the field, indicating reorientation of the flux lines along the direction of the defects. This reorientation is somewhat analogous to the well-known phenomenon of spin flop in magnetic systems; hence, we refer to it as "flux flop." The flop yields an experimental verification of the large pinning energy of the cylindrical defect. However, this energy may still be considerably increased if columnar defects with a diameter of the order of the penetration depth are used.

We describe here measurements on a $0.6\times0.3\times0.02$ mm^3 crystal which was irradiated along the **c** direction and a $0.6\times0.6\times0.02$ mm^3 crystal irradiated in 45° relative to this direction. We refer to these crystals as IR0 and IR45, respectively. As a reference, we also describe measurements on an unirradiated (UIR) $1.4\times0.7\times0.03$ mm^3 sample from the same batch. Sample preparation is described in Ref. 5. The transition temperature $T_c=92.5$ K of the UIR samples is reduced by 0.5 K after irradiation. Irradiation was done at the Grand Accelerateur National d'Ions Lourds (GANIL, Caen, France), with a beam of 5.3-GeV Pb ions at room temperature. The total fluence was 10^{11} ions/cm^2. All magnetic measurements were performed on an Oxford vibrating sample magnetometer (VSM). The VSM sensors define a spatial direction $\bar{x}$, and *only the component of the magnetization along* $\bar{x}$ *is measured.*

Figure 1 shows the magnetization curves **M**(**H**) at $T=60$ K for the crystal IR45, for the applied field oriented at angles $\phi=45°$ and $-45°$ relative to the **c** direction. For a schematic description of the relevant directions, see the inset to Fig. 2. The width of the magnetization curves reflects the efficiency of flux trapping and the magnitude of the critical current.[6] It is apparent from the figure that the width depends on ϕ; it is larger for $\phi=45°$ where **H** is parallel to the defects. However, the magnetization curve for $\phi=-45°$ exhibits a strong upturn at low fields and a pronounced peak around **H**=0 where the magnitude of the magnetizations of these two curves coincide. Magnetization curves for other field orientations (0 and ±30°) show a similar upturn at low fields and *all* magnetization curves coincide at **H**=0. In other words, the width of the magnetization curves in the low-field limit is *independent* of the orientation of the field relative to the direction of the defect.

It is important to point out that magnetization curves were measured for angles which are "symmetric" with respect to the **c** axis (45° and −45°). It is thus obvious that demagnetization effects *cannot* be the origin of this phenomenon. We also note that the same phenomenon was observed for IR0; we find for this sample enhanced width of the magnetization curves for **H** along the defects at high fields and isotropic behavior near **H**=0. This crossover to an isotropic trapping is our main observation in this work.

Figure 2 exhibits the width of the magnetization curves for IR45 for several field orientations and for the unirradiated sample. The increase in the width, after irradiation, is apparent. Also, the figure emphasizes the fact

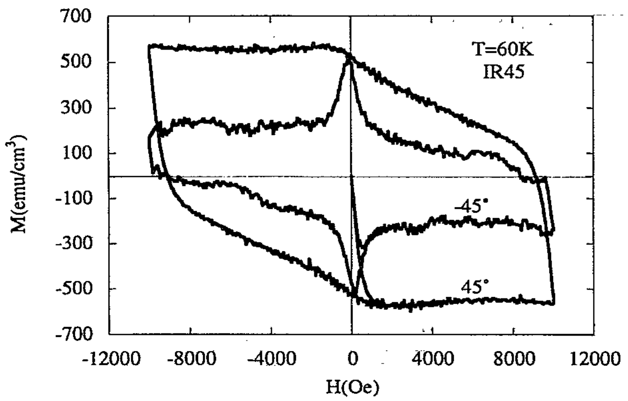

FIG. 1. The full magnetization curves of IR45 for $\phi=\pm45°$ at $T=60$ K.

that enhancement of flux trapping at low fields is independent of the orientation of the field. Another interesting point of this figure is the observation that at high fields the width of the magnetization curves of the irradiated samples, when the field is *not* along the defect, is actually the same as the width of the loop of the unirradiated sample. This implies that in this case (field not along the defects), the columnar defects act like point defects.

Figure 3 shows the angular dependence of the remanent magnetization $\mathbf{M}_{rem}(\theta)$ for IR45 for $\phi=\pm45°$ at 60 K. In these measurements the sample is cooled from above T_c in a field of $H=1.6$ T which forms an angle ϕ relative to the crystalline **c** axis. At the measurement temperature the field is turned off, the sample is rotated (the angle of rotation θ is measured to relative to **c** and for $\theta=0$ $c \| \bar{x}$) and the component of $\mathbf{M}_{rem}$ along $\bar{x}$ is measured.[7] We find that the remanent magnetization is independent of ϕ in the range of our measurements ($-45° \leq \phi \leq 45°$) and the maximum is always at $\theta=0$, implying that $\mathbf{M}_{rem}$ is pointing along the **c** direction.

Our surprising result that $\mathbf{M}_{rem}$ points along **c** even

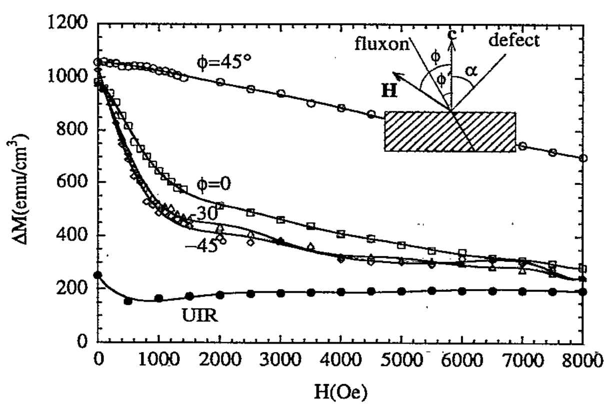

FIG. 2. Open symbols: Width of magnetization curves at $T=60$ K of IR45 for $\phi=0$, $-30°$, and $\pm45°$. Full circles: Width for the unirradiated sample for $\phi=0$. Inset: Schematic description of the relevant parameters. α, ϕ, and ϕ' denote the direction of the defect, the field, and the fluxons, respectively, relative to the c axis.

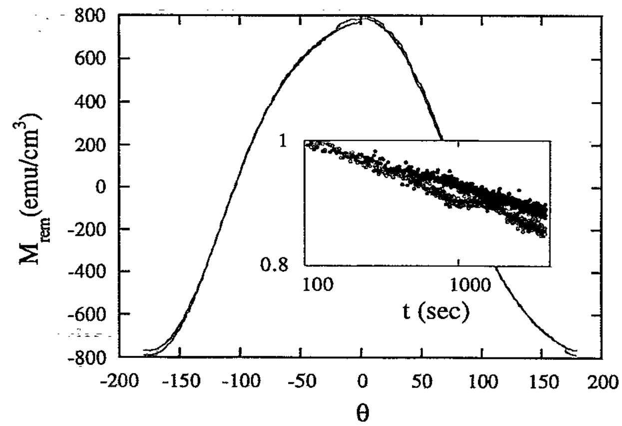

FIG. 3. The remanent magnetization of IR45 at $T=60$ K after cooling in a field of $H=1.6$ T (applied at $\phi=\pm45°$) vs the angle θ which is measured relative to the **c** direction. The two curves are almost indistinguishable. Inset: Relaxation of the remanent magnetizations at 60 K for $\phi=0$ and 45°. The relaxation data is normalized to a data point taken 100 sec after reducing the field to zero. The open and full symbols refer to data taken for $\phi=0$ and 45°, respectively.

when the vortices are trapped along the tilted defects, may be understood in view of a recent report by Hellman, Gyorgy, and Dynes.[8] They demonstrated that in flat samples such as ours, the critical currents in the bulk are forced to flow mainly perpendicular to the short distance. Thus, even when the vortices inside the sample are aligned along the defects, the *irreversible* magnetization points by preference in a direction perpendicular to the flat surface. Since the irreversible magnetization dominates the magnetic signal of our samples, we conclude that the magnetization vector points along **c** not only at $\mathbf{H}=0$ [where we have measured $\mathbf{M}_{rem}(\theta)$], but along the entire magnetization curves. We recall that in the VSM only the component of **M** along **H** is measured. Therefore, for a meaningful comparison of data taken at different angles ϕ, we divide the measured component of the magnetization by $\cos\phi$.

The sharp upturn and the sudden increase of the magnetization curves at low fields, for fields not along the defects, suggests that for fields not along the defect the fluxons flop towards the defects as soon as the field is decreased below some threshold. The flop leads to a magnetic "structure" near $\mathbf{H}=0$ which is independent of ϕ. This claim is supported by the convergence of all the magnetization curves at $\mathbf{H}=0$. Relaxation data provide a further support to this claim. We find that the effective pinning potential $U_0=k_BT/(\partial \ln \mathbf{M}_{rem}/\partial \ln t)$ of $\mathbf{M}_{rem}$ is *independent* of the direction of the original field. To demonstrate this feature we show, in the inset to Fig. 3, the almost identical decay of the maximum value of $\mathbf{M}_{rem}$ of IR45 at 60 K for $\phi=0$ and 45°. At this temperature $U_0 \cong 0.23$ eV for IR0 and IR45 as compared to 0.07 eV for the UIR sample.

The observed flux flop towards the columnar defects is similar to the predicted "lock-in" transition towards the *planes* in layered superconductors,[9] though it should be noted that the prediction is for lock-in of the *reversible* magnetization and the transition is expected for small angles (10°–20°) relative to the pinning planes. Transport measurements[10] of twinned Y-Ba-Cu-O crystals are also interpreted in terms of planar trapping of fluxons in *twin boundaries* at even smaller angles (1°–5°) of the field relative to the twin planes. Our data exhibit experimental evidence for such a transition in one-dimensional defects. The effect is much more dramatic (more than 45° jump of flux lines) due to the different energy scale for pinning along *columnar* defects; we use the term flux flop to emphasize the fact that we measure an *irreversible* process.

One may suggest that an alternative interpretation to our data could be that the large self-fields, due to the geometry of the sample,[11] blur the difference in $\mathbf{M}_{rem}$ for fluxons which maintain their original direction. As we argue below, self-fields are indeed important in the quantitative analysis of our results. However, the qualitative behavior cannot be explained by self-fields. To demonstrate this point we show in Fig. 2 the width of the magnetization loops for $\phi=0$ for both the irradiated and unirradiated samples. These two samples exhibit similar ΔM values in the high-field limit, suggesting that the irradiation did not contribute significantly to pinning efficiency for fields not along the columnar defects. Nevertheless, when the field is decreased, $\mathbf{M}_{rem}$ for $\phi=0$ for the irradiated sample is six times larger than the remanent magnetization for $\phi=0$ for the unirradiated sample. If self-fields play a major role in this experiment they should have the same effect on these two curves. Furthermore, the angular independence of the relaxation of $\mathbf{M}_{rem}$ that we discussed above indicate that the fluxons are subjected to the same pinning and this excludes the possibility that the fluxons maintain their original directions. We therefore conclude that the most plausible explanation for our data is flux flop towards the columnar defects.

The physics of flux lines pinned by columnar defects was treated by Nelson and Vinokur.[12] Within their model we estimate the pinning energy as follows: The energy of an Abrikosov vortex[13,14] is $\varepsilon_0=(\Phi_0/4\pi\lambda)^2\ln(\lambda/\xi)$ whereas the energy of a fluxon in a columnar insulating defect with a diameter $d(\lambda\gg d>\xi)$ is approximately $\varepsilon_P=(\Phi_0/4\pi\lambda)^2\ln(\lambda/d)$, because the defect diameter now plays the role of the vortex core. Substituting reasonable values for d (70 Å), ξ (13 Å), and λ (1400 Å) we find $u_P=(\varepsilon_0-\varepsilon_P)/\varepsilon_0\approx 0.4$. For comparison, for point defects the gain in the core condensation energy $\varepsilon_c=H_c^2\xi^2/8\pi$ is only a few percent of ε_0 when $\lambda\gg\xi$.[13]

In our theoretical treatment of the flux flop, we assume that the only significant pinning is due to columnar defects, and we consider fluxons not along the defect as being in a thermodynamic equilibrium state. Therefore, we determine the field at which the flux flop occurs by minimization of the total Gibbs free energy in the presence of applied field $\mathbf{H}$,[15]

$$G=F-\mathbf{H}_i\cdot\mathbf{B}/8\pi-\mathbf{M}\cdot\mathbf{H}/2\ , \qquad (1)$$

where $\mathbf{H}_i$ is the magnetic field in the sample, $\mathbf{B}$ is the magnetic induction, and F is the combined contributions of the vortex energy (ε_0 or ε_P) and of the mutual interactions between the fluxons. Since the measured samples are platelike, we use a simplified model and treat the sample as an infinite plane. Using the conventional boundary conditions for $\mathbf{H}_i$ and $\mathbf{B}$ we calculate the Gibbs energy for *unpinned* fluxons and we rewrite Eq. (1),

$$G_0=(B_Z/\phi_0)[\varepsilon_0/\cos\phi'-\phi_0 H\sin\phi\tan\phi'/4\pi +\phi_0 H\cos\phi/8\pi\cos^2\phi']\ , \qquad (2)$$

where ϕ' is the actual direction of the vortex (see inset to Fig. 2). This direction is determined by minimization of G_0 which yields

$$H_0\sin\phi'+H\cos\phi\tan\phi'=H\sin\phi\ , \qquad (3)$$

where $H_0=4\pi\varepsilon_0/\phi_0$ (for isotropic superconductor $H_0=H_{C1}$, the lower critical field). The minimum of G_0 reflects the competition between two factors: (a) alignment along the direction of $\mathbf{H}_i$ decreases the Gibbs energy by a factor which is proportional to the intensity of the field. (b) The total vortex energy is proportional to its length; in a platelet sample this length is minimized by alignment perpendicular to the flat surface. The minimum of G_0 can now be compared with the Gibbs energy for fluxon along a defect, G_P, which is at an angle α with the **c** direction and is obtained by substituting in Eq. (2) ε_P instead of ε_0 and α instead of ϕ'.

The discussion in the previous paragraphs leads us to the following scenario for the behavior of the vortices in the presence of columnar defects. In the high-field limit the vortices are aligned along the field; i.e., $\phi'=\phi$, independent of the direction of the defect. When the field is reduced, ϕ' *gradually* decreases in order to minimize the length of the flux line. Finally, at low enough field, $G_P\le G_0$ and an abrupt change (flop) occurs in the orientation of the flux line. An estimation of the field for flux flop yields $H_{fl}=0.3H_0$ for $\phi=0$ and $H_{fl}=0.15H_0$ for $\phi=-45°$. In reality, finite-size effects[11] lead to nonhomogeneous field distribution in the sample and, in particular, to a reduction in the effective field near the surface. As a result, the actual field at which flux flop occurs is sample dependent and is larger than the calculated H_{fl}. We estimate a lower bound of the pinning energy by taking the limit of $H_{fl}=0$. In this limit the flux flop towards a tilted columnar defect occurs only when the energy gain in the defect $(1-u_p)$ is smaller than the energy loss due to the increase (by a factor $1/\cos\alpha$) in the length of the fluxon. The fact that flux flop occurs at IR45 implies that $u_p\ge 1-\cos 45°\approx 0.3$. This gives an experimental estimation of the pinning energy in columnar defects. It is important to note that this pinning energy is conceptually different from that extracted from magnetic relaxation measurements; the latter is related to the energy cost of nucleus formation.[16]

All the energetic considerations above depend only weakly on temperature through the characteristic length scales. Nevertheless, $\mathbf{M}_{rem}$ at high temperature exhibits isotropic behavior at low fields only above 60 K. At lower temperatures we still see the upturn in $\mathbf{M(H)}$ which characterize the flop but $\mathbf{M}_{rem}$ depends on ϕ, being the largest for field along the defects. This observation may be explained by the use of the Bean model.[6] For fields parallel to the defects, $\mathbf{M}_{rem}$ is determined only by the lowest field which penetrates the whole sample, H^*. For fields in other directions, $\mathbf{M}_{rem}$ is determined by both H_{fl} and H^*. To clarify this point we assume, for simplification, that there is no bulk pinning from point defects. Therefore, for fields $H > H_{fl}$ which are not along the defect, $\mathbf{B}$ is constant throughout the sample. In this case, the Bean critical state starts to develop only after the fluxons become trapped by the columnar defects, i.e., below H_{fl}. In Figs. 4(a) and 4(b) we describe the flux profiles for $H_{fl} > H^*$ and for $H_{fl} < H^*$, respectively. It is apparent from Fig. 4(a) that for $H_{fl} > H^*$, $\mathbf{M}_{rem}$ reaches its maximum value independent of ϕ, whereas for $H_{fl} < H^*$, $\mathbf{M}_{rem}$ after a flop is smaller than the maximum possible value and it is ϕ dependent. Since H^* increases as temperature is decreased, we may understand the ϕ dependence of $\mathbf{M}_{rem}$ at $T < 60$ K.

Finally, we point out that despite the large pinning energy which we have measured here, magnetic relaxations are still observed even in the irradiated samples (see inset to Fig. 3). This disappointing result calls for a novel approach to flux trapping enhancement. We suggest that this may be achieved by producing columnar defects with larger diameter d. This large diameter may add a new factor to our energy consideration, namely, the contribution of surface barriers. The importance of Bean-Livingston surface barriers to irreversible magnetic features in HTS has recently been demonstrated.[17] The surface introduces potential barriers for flux penetration into the sample. Analogous with this effect, the surface of the columnar defect is a barrier for flux expulsion from the defect into the sample. However, for this surface to be most effective, it should be of a diameter larger than the penetration depth λ. Thus, if we increase d to be of order λ, we may add another energy scale which may be extremely efficient. Similar arguments were presented in the past by Campbell *et al.*[18] for increased bulk pinning by large precipitates.

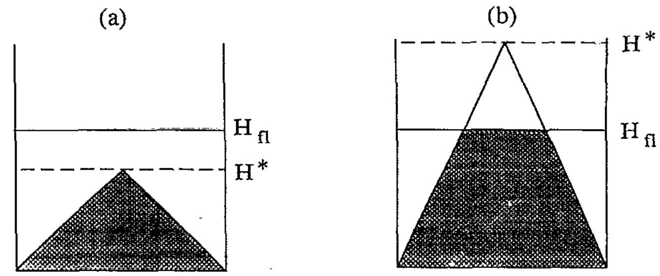

FIG. 4. The Bean profile for (a) $H_{fl} > H^*$ (high temperature) and (b) $H_{fl} < H^*$ (low temperature).

Valuable comments from V. Vinokur, M. Feigel'man, R. Goldfarb, and D. Feinberg are acknowledged. The work in Israel is partially supported by the U.S.-Israel BSF, and in part by the Ministry of Science and Technology. L.B. acknowledges support from the Raschi Foundation and from the Israel Academy of Sciences.

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