## Vortex pinning by splayed columnar defects in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>: Influence of large crossing angles

V. Hardy, S. Hébert, C. Goupil, Ch. Simon, J. Provost, and M. Hervieu

Laboratoire Cristallographie et Sciences des Matériaux, Unité Mixte de Recherches 6508, Institut des Sciences de la Matière et du *Rayonnement et Universite´ de Caen, 6 Boulevard du Mare´chal Juin, 14050 Caen-Cedex, France*

## P. Lejay

*Laboratoire Centre de Recherches sur les Tre`s Basses Tempe´ratures, Centre National de la Recherche Scientifique, 25 Avenue des Martyrs, 38042 Grenoble Cedex 9, France*

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Various configurations of columnar defects have been installed by heavy-ion irradiation in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> crystals: a standard configuration of tracks parallel to *c*, a bimodal splay consisting of crossed tracks at  $\pm$  5° to the *c* axis, and a "fan" configuration consisting of crossed tracks at  $\pm 5^{\circ}$  and  $\pm 45^{\circ}$ , with a fraction of large angles equal to 1/5. The aim is to directly investigate the influence of a small proportion of tracks at large angle on the vortex pinning. This study leads us to conclude that the large-angle tail of a Gaussian angular distribution should not, in itself, impede the occurrence of a beneficial splay effect, i.e., a further pinning enhancement relative to a standard irradiation along  $c$ .  $[$0163-1829(99)00113-7]$ 

Columnar defects can be introduced in high- $T_c$  superconducting oxides by heavy-ion irradiation.<sup>1,2</sup> These defects consist of continuous, amorphous latent tracks, with a core diameter of the order of 10 nm. According to their size and shape, such linear defects can act as very efficient pinning centers for the vortices. Many studies have reported huge improvements of pinning properties when the magnetic field and the track setting are both oriented along the  $c$  axis.<sup>3,4</sup>

It has been proposed by Hwa *et al.*<sup>5</sup> and Le Doussal *et al.*<sup>6</sup> that pinning (for *H*||c) can be further improved by a dispersion in track directions (around the  $c$  axis). The expected beneficial effects are related, first, to the existence of an entangled vortex ground state, and, second, to a modification of the variable-range-hopping regime at low currents. In  $YBa_2Cu_3O_7$  (YBCO) and other compounds of moderate anisotropy, several results have provided evidence for enhancements of pinning efficiency in splayed defect settings compared to parallel defect settings.<sup>7-11</sup> For instance, it is now well established that a characteristic splay angle of 10° relative to the *c* axis yields an improvement of transport properties in YBCO.<sup>7,10–12</sup> For the widely used bimodal configuration (two track directions at  $\pm \theta_i$  relative to the *c* axis), an optimal value of  $\theta_i$  equal to 5<sup>°</sup> was even reported.<sup>11</sup>

There is less consensus in the literature about the efficiency of large splay angles, e.g., 45 °, in YBCO or closely related compounds. In  $DyBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>$  single crystals, Schuster *et al.*<sup>9</sup> found a larger pinning enhancement for tracks at  $\pm 45$ ° to the *c* axis than for tracks oriented along *c*, in an observation of flux penetration by magneto-optics. We previously carried out magnetization measurements in YBCO crystals irradiated with three symmetrically distributed incident directions at  $\theta_i$  to the *c* axis.<sup>10</sup> It was found that such a splay arrangement for  $\theta_i = 45^\circ$  leads to an increased pinning efficiency compared to  $\theta_i = 0^\circ$ , at high temperatures and low fields. There is even a range of low fields where the pinning enhancement is larger for  $\theta_i = 45^\circ$  than for  $\theta_i = 10^\circ$ . Prozorov *et al.*<sup>13</sup> measured, by Hall probe magnetometry, the irreversibility temperature  $T_{irr}$  versus the angle  $\theta$  of the field with respect to the *c* axis, in irradiated YBCO thin films. In a narrow angular range around  $\theta=0$ ,  $T_{irr}(\theta)$  has a cusp shape for tracks at  $\pm 45$ °, whereas it exhibits a dip for parallel tracks along *c*. This difference was ascribed to a suppression of the relaxation rate when the field is applied along the mid-direction of crossed defects. On the contrary, there are several results which support the absence of (beneficial) splay effects for large angles like 45°. Numerical simulations of  $E(J)$  curves performed by Krusin-Elbaum *et al.*<sup>11</sup> display regimes where the vortex velocity for  $\pm 45$  ° exceeds that for the parallel configuration. On the basis of transport measurements in YBCO thin films, Park *et al.*<sup>12</sup> found that the activation energy for crossed tracks at  $\pm 40^{\circ}$  is lower than for parallel tracks, in all fields. In YBCO crystals irradiated at  $\pm \theta_i$  to the *c* axis, Lopez *et al.*<sup>14</sup> recorded resistive transition curves with the current flowing either parallel or perpendicular to the irradiation plane. The absence of resistivity anisotropy, in the case  $\theta_i = 45^\circ$ , made them claim the ineffectiveness of the splay for such a large angle.

From a theoretical point of view, it was early specified that too large angles should reduce the irreversible domain in the  $H-T$  plane.<sup>5,6</sup> In addition to the large value of  $\theta_i$ , which would merely impede vortex accommodation to the columns, another negative effect for pinning might derive from the high density of track crossings generated by large inclination angles. This latter idea is related to an antagonist role of crossings pointed out by Krusin-Elbaum *et al*. 11,15 The crossings impede the motion of vortex kinks between tracks, but they also favor zigzag vortex configurations which may stimulate creep.<sup>16</sup> The competition between these antagonist influences could result in regimes where splay enhances vortex motion. This competition, and thus the possible occurrence of a detrimental effect, is promoted by large angles.

Recently, this influence of track crossings has been invoked in the analysis of a Gaussian splay of full width at half maximum (FWHM) equal to  $\pm$ 4.47°, achieved by defocusing the incident ion beam through a metallic foil.<sup>11</sup> Despite the apparent similarity to the bimodal configuration  $\pm 5$ °,



FIG. 1. Schematic representation of the three investigated angular distributions of columnar defects (see text):  $P(\theta_i)$  is the fraction of tracks tilted by  $\theta_i$  relative to the *c* axis. The parallel configuration  $(a)$  is a standard irradiation along  $c$ . For the splayed configurations  $(b)$  and  $(c)$ , there is only one irradiation plane, containing the  $c$  axis.

the persistent current densities *J* were observed to be much lower for the Gaussian splay than for the planar splay. The Gaussian splay was even found to be less efficient than the standard defect setting parallel to *c*, in the major part of the temperature range. This behavior has been ascribed to the large-angle tail of the Gaussian which generates numerous intersections of tracks.

However, it must be realized that a Gaussian splay with  $\pm \theta_G$  FWHM differs from a planar splay  $\pm \theta_G$  in many other respects (numerous tracks at  $\theta_i < \theta_G$ , continuous nature of the distribution, etc.). From a practical point of view, Gaussian splays are attractive because they can be easily achieved, just by placing a foil on the samples. Note also that this type of splay actually corresponds to the case which is most often addressed in theoretical works.<sup>5,17</sup> For all these reasons, the influence of a low density of highly angled tracks deserves to be directly investigated. This is the aim of the present study.

In YBCO single crystals, we will compare the pinning efficiency of three configurations of linear defects, which are sketched in Fig. 1: (a) all the tracks are parallel to the  $c$  axis (referred to as the "parallel" configuration); (b) crossed tracks, inclined at  $\pm 5$  ° with respect to the *c* axis (referred to as the "cross" configuration); (c) crossed tracks at  $\pm 5$  ° or  $\pm$  45 $\degree$ , with one track by five being at 45 $\degree$  (referred to as the "fan" configuration). The fan configuration allows us to distinguish the specific role of the low density of large angles in the Gaussian splay, discarding all the other features of this defect setting. Note also that this ''fan'' configuration enhances the possible detrimental influence of large angles, since their relative abundance is larger than in a Gaussian distribution of FWHM close to  $\pm$  5 ° (only 5% of the angles are higher than  $10^{\circ}$ , in this latter case).

The samples are 30  $\mu$ m thick YBCO single crystals of  $T_c$ equal to  $91.9\pm0.3$  K. The crystals were grown by a pseudoflux method at 980 °C in gold crucibles. Selected crystals were submitted to post-oxygenation in pure oxygen at  $420\degree$ C for 20 days. They have been irradiated with 6 GeV Pb ions at GANIL (Caen, France), on a rotating sample holder permitting the achievement of configurations described in Fig. 1. In all cases, the total ion fluence was



FIG. 2. Persistent current density versus magnetic field, at 85 K, in a virgin YBCO single crystal  $(\nabla)$ , and in samples irradiated with  $10^{11}$  Pb cm<sup>-2</sup> according to the angular distributions displayed in Fig. 1: parallel configuration ( $\bigcirc$ ); cross configuration at  $\pm 5^{\circ}(\square)$ ; fan configuration at  $\pm 5$ ° and  $\pm 45$ °( $\blacklozenge$ ).

 $10^{11}$  cm<sup>-2</sup>. For the fan configuration, the sample was irradiated with  $4 \times 10^{10}$  Pb cm<sup>-2</sup> in both directions at 5<sup>o</sup>, and with  $10^{10}$  Pb cm<sup>-2</sup> in both directions at 45 $^{\circ}$ . The 6 GeV Pb ions are known to induce, in YBCO, the creation of continuous, amorphous tracks with a core diameter close to 7 nm.<sup>18</sup> The pinning properties of each sample have been investigated by superconducting quantum interference device magnetometry, before and after irradiation, with the magnetic field *H* applied along the *c* axis. Hysteresis loops were recorded with a waiting time of 1 min after each field change. The field and temperature dependences of the persistent current *J*, for this characteristic time of 1 min, were derived from the Bean model.

Figure 2 shows  $J(H)$  curves, at 85 K, in the virgin state and in the three defect configurations sketched in Fig. 1. Firstly, one observes that the introduction of columnar defects strongly increases the persistent current density for all configurations. Secondly, it is also exhibited that the cross  $(\pm 5^{\circ})$  configuration has a much better pinning efficiency than the parallel one. This behavior supports the occurrence of a splay effect in YBCO for such a small angle, in accordance with the literature. In the present work, the most important point is the location of the  $J(H)$  curves in the fan configuration ( $\pm$  5°; $\pm$ 45°), with respect to the parallel and cross ( $\pm$ 5 $\degree$ ) configurations. Figure 3 displays *J(H)* curves for these three configurations, at several temperatures from 40 up to 80 K. Two main features can be observed. First, the *J*(*H*) curves of the cross and fan configurations are close to each other. Secondly, they systematically lie above those of the parallel configuration, for all values of field and temperature.

In a closer examination, one observes, however, that the persistent current densities in the fan configuration drop more rapidly than those in the cross configuration, when approaching the irreversibility field. At this stage of the comparison, it must be pointed out that the effective density of linear defects is the lowest in the fan configuration, owing to the presence of large irradiation angles ( $\theta_i = \pm 45^{\circ}$ ). Indeed, several geometrical effects are associated with the inclination of the ion beam with respect to the short direction of a platelike sample (i.e., the  $c$  axis in our case). For a given fluence (which is always measured perpendicular to the ion beam), an irradiation angle  $\theta_i$  with respect to the *c* axis induces not only an increase of the individual track length (by  $1/\cos \theta_i$ ),



FIG. 3. Curves of persistent current density versus magnetic field at various temperatures  $[40 \text{ K (a)}; 55 \text{ K (b)}; 70 \text{ K (c)}; 80 \text{ K}$ (d)], in the three configurations of Fig. 1: parallel ( $\circlearrowright$ ); cross ( $\Box$ ); fan  $(\bullet)$ .

but also a decrease (by cos  $\theta_i$ ) of the *number of tracks per unit area of the sample surface*. Although the total damaged volume is the same for all values of  $\theta_i$ , such a decrease of the areal density of columnar defects per *ab* plane can affect the pinning ability in high fields applied parallel to *c*. In addition to the standard fluence equivalent field  $B_{\Phi t} = \Phi t$  $\times\Phi_0$ , where  $\Phi t$  is the ion fluence and  $\Phi_0$  is the flux quantum, let us introduce the notion of field equivalence to the track density per  $ab$  plane, denoted  $B_{cd}$ . For the cross configuration at  $\pm 5^\circ$ , $B_{cd} \approx B_{\Phi t} \times \cos 5^\circ \approx B_{\Phi t} = 2$  T. For the fan configuration,  $B_{cd} \approx [(4/5) \times \cos 5^\circ + (1/5) \times \cos 45^\circ]$  $\times$ 2*T* $\approx$  1.88 T. For symmetric track arrangements, it was shown<sup>19</sup> that the location of the irreversibility line in  $Bi_2Sr_2CaCu_2O_8$  crystals is directly driven by  $B_{cd}$ . The present difference in  $B_{cd}$  between the cross and fan configurations could account for the greatest part of the observed variation between their  $J(H)$  curves (see Fig. 2 for instance).

Figure 2 also shows that the *J* values are slightly larger in the ''fan'' than in the ''cross,'' in a restricted range of low fields. This feature is in qualitative agreement with a previous study<sup>10</sup> comparing the pinning efficiency of various splay angles  $\theta_i$ . In YBCO single crystals irradiated along three directions tilted by  $\theta_i$  from the *c* axis, it was shown that, as  $\theta_i$  increases (e.g., from 10 $\degree$  to 45 $\degree$ ), the peak of  $J(H)$  increases while shifting towards lower fields. The presence of a small proportion of tracks at  $45^{\circ}$  in the "fan" could be at the origin of the slight enhancement of *J* which is observed at low fields.

Beyond these above-mentioned effects, the important qualitative result, when comparing the fan  $(\pm 5^{\circ}; \pm 45^{\circ})$ and cross ( $\pm$ 5 $\degree$ ) configurations, is that the introduction of a low density of highly angled tracks does not drastically affect the strong pinning efficiency of a moderate splay  $(e.g.,)$  $\pm$  5 $\degree$ ). The comparison to parallel configurations exhibits still more clearly the very different behaviors of the fan and Gaussian splays. In Ref. 11, for a fluence equivalent field  $B_{\Phi t}$ =3 T, the persistent current (measured at 5 K; 1 T) was found to be smaller for a Gaussian splay  $(\pm 4.47^{\circ}$ FWHM)



FIG. 4. Time relaxation of persistent current, normalized to its value at 1 min, for the three configurations of Fig. 1 [parallel  $(\bigcirc)$ ; cross ( $\Box$ ); fan ( $\blacklozenge$ )], under an applied field of 0.5 T, at 40 K (a) and  $80 \text{ K}$  (b).

than for a parallel configuration, in the widest part of the temperature range, i.e., from 16 K up to  $T_c$ . On the contrary, the persistent currents of the fan configuration ( $\pm 5$ °;  $\pm$ 45<sup>°</sup>) are systematically larger than those of the parallel configuration, in the whole investigated ranges of field  $(0-4)$ T) and temperature  $(40-85 \text{ K})$ . Vortex dynamics has been also investigated by recording magnetic relaxation. Figure 4 shows the variation in time of *J* at low and high temperatures  $(40$  and  $80$  K), under a moderate field of  $0.5$  T, in order to minimize the influence of the different  $B_{cd}$  values among the three configurations. At 40 K [Fig. 4(a)], the relaxation data confirm the measurements of  $J(1 \text{ min})$ . The relaxation rate is clearly lower in the fan than in the parallel configuration. At 80 K [Fig. 4(b)], the relaxation is less pronounced than at 40 K, and it is not so directly connected to  $J(1 \text{ min})$ , since very similar relaxation rates are found in the three configurations. Actually, this indicates that *J*'s in both the fan and the cross configurations remain larger than in the parallel one, at any time.

Our results, about persistent current and relaxation, show that the peculiar behavior of the Gaussian splay in Ref. 11 is not simply due to the presence of large angles in the distribution of track orientations. Let us specify that there are many other features which should be considered in a Gaussian splay resulting from beam defocusing. First of all, in a Gaussian splay of FWHM equal to  $\pm \theta_G$ , a large proportion of tracks are inclined at very low angles  $|\theta_i| \ll \theta_G$ . For  $\theta_G$  $\approx$  5 $\degree$ , these angles are likely below the optimal misorientation value, which results in reducing the overall pinning efficiency with respect to a cross configuration ( $\pm \theta_G$ ). The Gaussian splay is also characterized by the continuous nature of the angular distribution. This feature could influence the vortex dynamics, owing to the determinant role of angle selection in a regime of variable-range hopping.<sup>5</sup> Moreover, to obtain a large enough angular dispersion, the ion energy is strongly reduced by the foil, hence before penetrating the sample. As a consequence, the individual nature of the tracks may be different from that obtained in a direct, high-energy irradiation. This can impede a straightforward comparison to the parallel configuration. Finally, with low incident energy, the natural splay inside the target is not negligible, $<sup>7</sup>$  and leads</sup>

to a drastic variation of the net angular dispersion along the sample thickness.

In summary, we have introduced, in YBCO single crystals, a small amount (about  $20\%$ ) of tracks at large angle  $(\pm 45^{\circ})$  in a configuration of crossed tracks at low angle  $(\pm 5^{\circ})$ . Magnetization measurements have been carried out in this fan configuration, as well as in a planar splay  $\pm 5$  ° and a standard configuration of tracks parallel to *c*. It was found that the persistent current densities of the fan always exceed by far those of the parallel configuration, in the whole range of temperature and field. The fan stays even close to the splay  $\pm 5$ °, except at very high fields. These results

- <sup>1</sup>D. Bourgault, M. Hervieu, S. Bouffard, D. Groult, and B. Raveau, Nucl. Instrum. Methods Phys. Res. B 42, 61 (1989).
- <sup>2</sup>B. Roas, B. Hensel, S. Henke, S. Klaumünzer, B. Kabius, W. Watanabe, G. Saemann-Ischenko, L. Schultz, and K. Urban, Europhys. Lett. **11**, 669 (1990).
- 3L. Civale, A. D. Marwick, T. K. Worthington, M. A. Kirk, J. R. Thompson, L. Krusin-Elbaum, Y. R. Sun, J. R. Clem, and F. Holtzberg, Phys. Rev. Lett. **67**, 648 (1991).
- <sup>4</sup>M. Konczykowski, F. Rullier-Albenque, E. R. Yacoby, A. Shaulov, Y. Yeshurun, and P. Lejay, Phys. Rev. B 44, 7167 (1991).
- 5T. Hwa, P. Le Doussal, D. R. Nelson, and V. M. Vinokur, Phys. Rev. Lett. **71**, 3545 (1993).
- ${}^{6}P$ . Le Doussal and D. R. Nelson, Physica C 232, 69 (1994).
- ${}^{7}$ L. Civale, L. Krusin-Elbaum, J. R. Thompson, R. Wheeler, A. D. Marwick, M. A. Kirk, Y. R. Sun, F. Holtzberg, and C. Feild, Phys. Rev. B 50, 4102 (1994).
- 8T. Schuster, H. Kuhn, M. Indenbom, M. Leghissa, M. Kraus, and M. Konczykowski, Phys. Rev. B 51, 16 358 (1995).
- <sup>9</sup>T. Schuster, H. Kuhn, M. Indenbom, G. Kreiselmeyer, M. Leghissa, and S. Klaumünzer, Phys. Rev. B 53, 2257 (1996).
- 10V. Hardy, A. Ruyter, A. Wahl, A. Maignan, D. Groult, J. Provost, Ch. Simon, and H. Noël, Physica C 257, 16 (1996).
- <sup>11</sup>L. Krusin-Elbaum, A.D. Marwick, R. Wheeler, C. Feild, V.M.

show that the small proportion of tracks at large angles, which results from a beam defocusing, should not be necessarily detrimental to the splay effect related to the smaller angles. This encourages us to proceed with the study of Gaussian splays resulting from beam defocusing, taking the various effects associated with this method into account.

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Vinokur, G.K. Leaf, and M. Palumbo, Phys. Rev. Lett. **76**, 2563  $(1996).$ 

- <sup>12</sup> J.H. Park, D.H. Kim, S.Y. Shim, Y.H. Kim, J.M. Lee, T.S. Hahn, J.D. Hettinger, D.G. Steel, K.E. Gray, B. Glagola, J. Lee, and Z.G. Khim, Physica C 281, 310 (1997).
- 13R. Prozorov, M. Konczykowski, B. Schmidt, Y. Yeshurun, A. Shaulov, C. Villard, and G. Koren, Phys. Rev. B **54**, 15 530  $(1996).$
- 14D. Lopez, L. Krusin-Elbaum, H. Safar, V.M. Vinokur, A.D. Marwick, J.Z. Sun, and C. Feild, Phys. Rev. Lett. **79**, 4258 (1997).
- 15L. Krusin-Elbaum, A.D. Marwick, R. Wheeler, C. Feild, V.M. Vinokur, G.K. Leaf, and M. Palumbo, Proceedings of the 21st ICLTP, Prague 1996 [Czech. J. Phys. 46, 1799 (1996)].
- <sup>16</sup>Note that zigzag configurations have been contrariwise assumed to strongly reduce vortex creep in Ref. 13, because of the required nucleation of multiple half-loops.
- 17T.P. Devereaux, R.T. Scalettar, G.T. Zimanyi, K. Moon, and E. Loh, Phys. Rev. Lett. **75**, 4768 (1995).
- 18V. Hardy, D. Groult, M. Hervieu, J. Provost, B. Raveau, and S. Bouffard, Nucl. Instrum. Methods Phys. Res. B 54, 472 (1991).
- <sup>19</sup>S. Hébert, V. Hardy, G. Villard, M. Hervieu, Ch. Simon, and J. Provost, Physica C 299, 259 (1998).