

Anisotropy and Lorentz-force dependence of the critical currents in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thick films deposited on nickel-alloy substrates

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We investigate the angular dependence of the critical currents in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thick films deposited on metallic nickel-alloy substrates, with the aid of an yttria-stabilized zirconia buffer layer. Overall, the critical current shows a clear Lorentz-force dependence and two distinctive peaks, one for fields parallel to the ab plane, due to bulk intrinsic pinning by the layered structure of the superconductor, and the other for fields parallel to the c axis, which we attribute to twin-boundary c -axis correlated pinning, as suggested by its Lorentz-force dependence.

At the forefront of using high-temperature superconductors (HTS) for bulk applications is the necessity to produce large-scale conductors with high critical currents that can be wound to produce coils or other devices. This requirement has been frustrated by the metallurgical properties of this class of materials: they are fragile and prone to contain large-angle grain boundaries which act as weak links to limit the current-carrying capabilities of the conductor.¹ These problems have been partially solved by the fabrication² of multifilamentary Bismuth-based $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-\delta}$ and $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_{8-\delta}$ (BSCCO) silver-clad HTS wires and tapes, with very good mechanical properties. The main disadvantage of these samples is that the materials of choice are among the most anisotropic in the HTS family. This pushes the vortex solid to liquid transition³ to lower temperatures and fields, narrowing the region of the field-temperature plane where these materials have a finite critical current. As an example, multifilamentary BSCCO tapes lose their critical current at a field of ~ 0.5 T at 75 K. By means of irradiation with high energy protons,⁴ one can create very strong pinning centers and move this point of zero critical current up to 1.2 T at 75 K.

It was recently shown⁵ that thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) films can be deposited on flexible metallic substrates. In this procedure an ion-beam-assisted-deposition (IBAD) process is used to deposit a biaxially textured yttria-stabilized zirconia (YSZ) buffer layer onto nickel-based substrates. Later, an YBCO thick film is deposited by pulsed laser deposition (PLD) onto the YSZ layer. This technique yields YBCO/YSZ/Ni conductors, with superior flexibility to that of multifilamentary BSCCO/Ag tapes. The biaxial texture of the YSZ buffer layer is also present in the YBCO thick film, and is crucial to inhibit the presence of large-angle grain boundaries, since it was observed⁶ that the in-plane critical current density decreases exponentially with increasing misalignment. Typical YBCO/YSZ/Ni thick films, with an average of only 6° of in-plane misalignment, have critical currents above 10^6 A/cm² at zero applied magnetic field and liquid nitrogen temperatures. Tunneling electron microscopy (TEM) shows a typical grain size of about $5 \mu\text{m}$. These grains are seen to have many twin boundaries, with a typical separation of $0.2 \mu\text{m}$ between them. High resolution electron

microscopy (HREM) reveals very clean and sharp interfaces between adjacent grains, without spurious phases.

A crucial advantage of YBCO superconductors is that they are much less anisotropic than BSCCO. The vortex solid to liquid transition is located at about 10 T at 75 K,³ almost a tenfold increase with respect to the irradiate BSCCO tapes. In addition to this, naturally occurring strong c -axis pinning centers are ubiquitous in YBCO, in the form of twin boundaries. These defects have a profound effect on the flux-flow resistance,⁷ and are expected to play a major role in determining the vortex dynamics and critical currents for fields parallel to the c axis.⁸ For fields applied parallel to the ab planes, on the other hand, it was suggested that intrinsic pinning by the layered crystal structure is possible,⁹ and indeed sharp peaks in the transport critical current were observed for fields applied parallel to the layers.¹⁰ These findings, however, were criticized¹¹ by the fact that magnetic surface pinning¹¹ or Bean-Livingston barriers¹² can produce similar results even in the case of isotropic, low- T_c superconductors.

In this paper we report on the angular dependence of the in-plane critical current density J_c in YBCO/YSZ/Ni thick films. We found that J_c has two clear peaks, when the magnetic field is applied parallel to the c axis and to the ab plane. As we will show, the peak for fields applied parallel to the c axis has a peculiar Lorentz force dependence that strongly suggests it is due to c -axis correlated pinning. In addition to this, its angular width is in agreement with the one predicted⁸ for c -axis extended disorder. We attribute this peak to pinning by twin boundaries which are present in a dense concentration in our samples.

On the other hand, the peak in J_c for fields applied parallel to the ab planes is consistent with the presence of bulk intrinsic pinning, since our samples are about 10 times thicker than the penetration depth. Thus our observation constitutes clear evidence for intrinsic pinning in the transport critical current density, as opposed to geometrical effects.

These peaks in J_c persist up to the maximum magnetic field of 9 T used in our experiment, while the relative heights are strongly field dependent. In addition to this, J_c is clearly Lorentz-force dependent for all possible configurations, indicating the absence of percolative flow in these thick films.

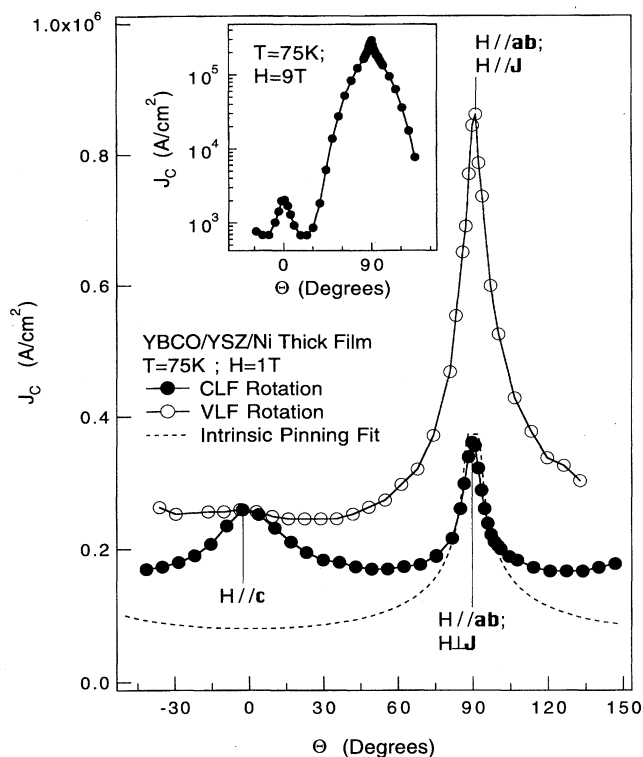


FIG. 1. Angular dependence of the in-plane critical current density J_c at $T=75$ K and $H=1$ T. Θ is the angle between the magnetic field and the sample's c axis. Full circles: constant Lorentz-force (CLF) rotation; empty circles: variable Lorentz-force rotation; dashed line: fit to the intrinsic pinning model. Inset: Angular dependence of J_c at $T=75$ K and $H=9$ T in a CLF rotation.

Our measurements were performed by the standard four-contact method on YBCO/YSZ/Ni films, of thicknesses ranging from 0.7 to 2 μm . The critical current was determined at an electric field criterion of 1 $\mu\text{V}/\text{cm}$. To avoid sample heating, all the measurements reported here were performed with the samples immersed in liquid nitrogen. The samples were mounted on a rotating sample holder that allowed for single-axis rotations with respect to the applied field, with an angular resolution of about 0.1° .

We measured several YBCO/YSZ/Ni thick films and, as a comparison, one YBCO thin film deposited by PLD on a single-crystalline YSZ substrate. The critical current of this latter sample shows similar behavior to the YBCO/YSZ/Ni films for all fields and orientations, and therefore will not be further discussed in this paper.

Shown in Fig. 1 is the angular dependence of the critical current density for one of the samples investigated. Experimentally, we found that J_c has a fourfold symmetry, and for simplicity only one portion of the data is shown in the figure. In this experiment we performed the rotation in two different ways: first we rotated the angle between the magnetic field and the c axis, keeping the magnetic field perpendicular to the transport current. In this way, for a given current, the Lorentz force is constant throughout the rotation. We will call this a constant Lorentz-force experiment, or CLF. The result of such a rotation is shown by the full circles in Fig. 1.

For this rotation procedure, we find two peaks in J_c , when the field is parallel and normal to the c axis.

In an independent experiment, we rotated the magnetic field with respect to the c axis, but now keeping the magnetic field in the plane defined by the electrical current and the c axis vectors. In this type of rotation, for a given electrical current, the Lorentz force does not remain constant during the rotation, instead it varies as $F_L(\Theta) = F_L \cos\Theta$. We will call this a variable Lorentz-force experiment, or VLF. The result of such a rotation is shown by the empty circles in Fig. 1.

As seen in Fig. 1, J_c is strongly Lorentz-force dependent for any orientation between the magnetic field and the c axis. A particularly notable feature is that the peak seen close to $H\parallel c$ in the CLF rotation is absent in the VLF experiment. This peak in the critical current is not always present, suggesting it is strongly dependent of sample morphology: for example, among the results cited in Ref. 10, it is present in the samples measured by Roas, Schultz, and Saemann-Ishenko, but not in those studied by the other groups. Recently, a similar peak in the critical currents close to $H\parallel c$ was reported,¹⁴ and attributed to flux pinning by miscut-growth-initiated columnar defects. However, the Lorentz-force dependence of this peak, such as the one we show in Fig. 1, has never been previously reported. This dependence is an essential ingredient for the complete understanding of this feature, since it suggests that the pinning force for fields close to $H\parallel c$ decreases at the same rate as the Lorentz force itself. This type of angular dependence could be attributed to extended pinning centers, i.e., correlated disorder that extends parallel to the c axis. One can expect that, in a first approximation, the strength of these correlated pinning centers will be proportional to the length of the vortex that it is trapped by them. Furthermore, our samples (both YBCO/YSZ/Ni and YBCO deposited onto single crystalline YSZ) have a high concentration of twin boundary planes, which are known to be strong pinning centers of the extended type.

In the CLF rotation, however, J_c is seen to change somewhat faster than a simple $\cos\Theta$. This may be attributed to the elastic properties of the vortex lines, an issue neglected in our simplistic model. Once the elastic properties are taken into account, for the case of irradiation-induced columnar defects, there is an "accommodation angle" (Ref. 8), predicted to be about 30° to 45° in YBCO, above which the tracks are ineffective pinning centers. Such an effect was indeed found¹⁵ in the angular dependence of the vortex glass to liquid transition temperature in the presence of columnar defects. While the former concepts were developed for the case of columnar defects, it can be argued that some general features, in particular those related to the elastic properties of the vortex line, can still be applicable for the case of twin boundaries. Indeed, our measurements show that the peak at $H\parallel c$ extends up to about $\Theta=50^\circ$, in very good agreement with the model for columnar disorder.

The peak at $H\parallel c$ in J_c is seen even at the highest fields we measured, $H=9$ T. However, since J_c for the $H\parallel c$ orientation drops much faster than for $H\parallel ab$, this central peak is not apparent in linear plots. For this reason, we show in the inset to Fig. 1 a semilogarithmic plot of the angular dependence of the critical current at $H=9$ T, in a CLF rotation. In spite of the J_c values for $H\parallel c$ being two orders of magnitude smaller

than for $H\parallel ab$, the peak at $H\parallel c$ is clearly seen, with the J_c value at the peak being about three times larger than at $\Theta=30^\circ$. The fact that correlated pinning is effective so close to the vortex solid liquid transition (estimated from linear resistance measurements to be at 10 T at this temperature) strongly suggests that this will be of the Bose-glass type.¹¹

We will concentrate now on the narrow peak in J_c seen when the magnetic field is applied parallel to the ab planes for both rotations. This peak has been observed by many other groups,¹⁰ and generally interpreted as due to intrinsic pinning⁹ of the vortex lines by the ab planes. Convincing evidence for intrinsic pinning was presented by Christen *et al.*,¹⁰ who excluded the possibility of this peak being solely due to the anisotropy of the superconducting order parameter and also showed that the peak in the critical current aligns with the crystallographic ab planes and not the film surface by studying YBCO films with the ab plane 6° off the substrate plane. However, in the samples studied in our work the ab plane lies parallel to the substrate surface. In this situation, it has been noted¹⁰ that a similar peak can be observed even in the case of isotropic, low-temperature superconductors, when the sample thickness is smaller than the London penetration depth. In this case, a peak in J_c for fields applied parallel to the sample plane was also observed,¹¹ but due to surface magnetic pinning. Our samples, however, are about 10 times thicker than the penetration depth, and thus surface magnetic pinning can be disregarded. We should also note that a surface Bean-Livingston barrier can contribute to the magnetic hysteresis and critical current, even in samples much larger than the London penetration depth.^{12,13} Such a barrier can indeed produce a peak in the critical current¹² similar to the one shown in Fig. 1 for $H\parallel ab$. However, Bean-Livingston barriers can be quantitatively and qualitatively disregarded in our experiment: it has been shown¹³ that the critical transport current due to surface barriers is $J_{cb} \approx cH_p^2/4\pi dH$, where d is the sample thickness and H_p is the field for first vortex penetration in the sample, which can be as large as the thermodynamic critical field $H_c \approx 0.1$ T at $T=75$ K. With this value we obtain an upper bound for the surface critical current of $J_{cb} < 1.6 \times 10^5$ A/cm², more than a factor of 2 lower than the experimental value obtained in the CLF measurements shown in Fig. 1. In addition to this, it should be noted that surface imperfections will further reduce the effect of the Bean-Livingston barrier. In any case, the discrepancy becomes even more severe at higher fields, since the experimentally determined J_c decreases much slower than the $1/H$ dependence expected from the surface critical current: at $H=9$ T there is a factor of 10 difference between the model prediction and the experimental value. Having disregarded surface effects, we can turn now to bulk pinning: our CLF data can be qualitatively fit to the intrinsic pinning model,⁹ shown by the dashed line in Fig. 1. While this model adequately describes the data in the 80° to 100° interval, it fails for angles farther from the $H\parallel ab$ orientation and is clearly inadequate where the peak for $H\parallel c$ in our data starts to be prominent. It should be noted that previous^{9,10} (and somewhat better) fits to this model were performed in samples with weaker twin-boundary pinning (i.e., samples not showing the peak in the critical current at $H\parallel c$) or alternatively at lower temperature, where isotropic point defects are equally effective pinning centers. The data shown in Fig. 1

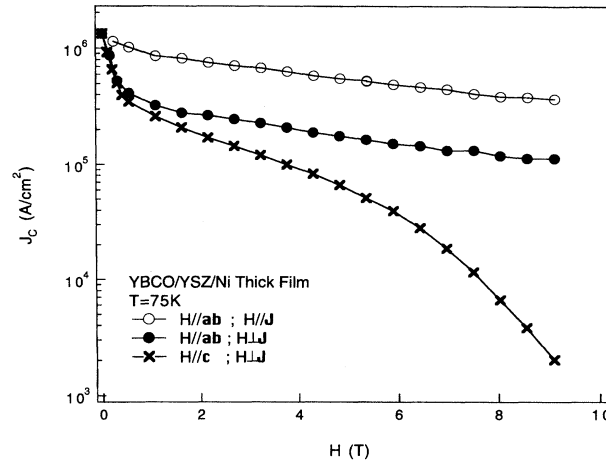


FIG. 2. Field dependence of the in-plane critical current density J_c for the three principal configurations. Empty circles: $H\parallel ab$ and $H\parallel J$; full circles: $H\parallel ab$ and $H\perp J$; crosses: $H\parallel c$ and $H\perp J$.

show that, for these samples and at these higher temperatures, the effects of c -axis correlated pinning are comparable to intrinsic pinning, and thus a more complicated angular dependence should be considered.

Shown in Fig. 2 is the field dependence of J_c for the three principal orientations at a temperature of 75 K. The critical currents for $H\parallel c$ are above the 10^3 A/cm² level at $H=9$ T, probably due to the strong twin-boundary pinning described in the paper. Due to the anisotropy of the system, the critical current for $H\parallel c$ is seen to drop to zero much faster than the critical current for $H\parallel ab$. However, as we show in Fig. 1, the peak in the critical currents for $H\parallel c$ is present over all the field range investigated. The Lorentz-force dependence for the $H\parallel ab$ configuration is clearly seen to persist up to the highest fields, with the ratio between both $H\parallel ab$ configurations being almost constant over the entire field range. The $H\parallel ab, H\parallel J$ configuration shows a remarkably flat field dependence, without the rapid falloff seen for $H < 1$ T in the other configurations, and with J_c values that remain in the vicinity of the 10^6 A/cm² range, even for fields as high as 9 T. As a consequence of this, the J_c value at $H=9$ T for this configuration is almost equal to the value at $H=0.5$ T for the other configurations.

In conclusion, we have shown that the angular dependence of the critical currents of YBCO/YSZ/Ni thick films shows clear evidence for both bulk, intrinsic pinning when the magnetic field is applied parallel to the ab plane and pinning due to a dense distribution of correlated disorder for $H\parallel c$. This correlated pinning is likely to be due to twin boundaries, and is effective at the highest fields investigated, even at the border of the vortex glass to liquid transition. This suggest that this transition will be of the Bose glass type, naturally produced by the existing twin boundaries as opposed to artificially introduced columnar defects. Overall, the critical current density shows a very strong Lorentz-force dependence. All these features, only previously seen in high-quality single crystalline samples, demonstrate the excellent degree of crystallographic alignment of the YBCO/YSZ/Ni thick films.

- ¹See D. C. Larbalestier and M. P. Maley, *Mater. Res. Bull.* **18**, 50 (1993) and references therein.
- ²G. N. Riley *et al.*, *Physica C* **235-240**, 3407 (1994).
- ³For a review see D. J. Bishop, P. L. Gammel, D. A. Huse, and C. A. Murray, *Science* **255**, 165 (1992) and references therein.
- ⁴L. Krusin-Elbaum *et al.*, *Appl. Phys. Lett.* **64**, 3331 (1995); H. Safar *et al.*, *ibid.* **67**, 130 (1995).
- ⁵X. D. Wu *et al.*, *Appl. Phys. Lett.* **65**, 1961 (1994); X. D. Wu *et al.*, *ibid.* (to be published).
- ⁶D. Dimos *et al.*, *Phys. Rev. Lett.* **61**, 219 (1988).
- ⁷W. K. Kwok *et al.*, *Phys. Rev. Lett.* **64**, 966 (1990); S. Fleshler *et al.*, *Phys. Rev. B* **47**, 14 448 (1993).
- ⁸D. R Nelson and V. M. Vinokur, *Phys. Rev. Lett.* **68**, 2398 (1992); *Phys. Rev. B* **48**, 13 060 (1993).
- ⁹M. Tachiki and S. Takahashi, *Solid State Commun.* **70**, 291 (1989); **72**, 1083 (1989).
- ¹⁰B. Roas, L. Schultz, and G. Saemann-Ishenko, *Phys. Rev. Lett.* **64**, 479 (1990); D. K. Christen *et al.*, *Physica B* **165&166**, 1415 (1990); Terukazu Nishizaki *et al.*, *Physica C* **204**, 305 (1993).
- ¹¹G. Stejic *et al.*, *Phys. Rev. B* **49**, 1274 (1994).
- ¹²Amit Das Gupta and Edward J. Kramer, *Philos. Mag.* **26**, 779 (1972).
- ¹³L. Burlachkov and V. M. Vinokur, *Physica B* **194-196**, 1819 (1994).
- ¹⁴D. H. Lowndes *et al.*, *Phys. Rev. Lett.* **74**, 2355 (1995).
- ¹⁵W. Jiang *et al.*, *Phys. Rev. Lett.* **72**, 550 (1994).