

## Giant Suppression of Flux-Flow Resistivity in Heavy-Ion Irradiated $Tl_2Ba_2Ca_2Cu_3O_{10}$ Films: Influence of Linear Defects on Vortex Transport

R. C. Budhani and M. Suenaga

*Materials Science Division, Brookhaven National Laboratory, Upton, New York 11973*

S. H. Liou

*Department of Physics, University of Nebraska, Lincoln, Nebraska 68588*

(Received 7 October 1992)

A large shift of the onset of flux-flow resistivity and the irreversibility line  $H_{irr}(T)$  to higher temperatures is observed in  $Tl_2Ba_2Ca_2Cu_3O_{10}$  films containing linear defects created by  $Ag^{+21}$  ion irradiation. The  $H_{irr}(T)$ , which has a characteristic L shape in highly anisotropic Tl and Bi based cuprates, becomes more like that of  $YBa_2Cu_3O_7$  in the presence of these defects. The  $J_c$  at 77 K also shows a large increase as a result of flux localization at the defects. The transport data indicate that in the  $H$ - $T$  plane above  $H_{irr}(T)$  of the unirradiated material, an ensemble of unoccupied defects is required for effective pinning of each flux line in the system.

PACS numbers: 74.60.Ge, 74.70.Vy, 74.75.+t

Understanding the dynamics of Abrikosov vortices in the force field of structural defects has been of considerable scientific and technological interest in the case of copper-oxide based superconductors. A subclass of this broad field concerns interactions between assemblies of flux lines and linear defects of diameter  $b_0 > 2\xi_{ab}$  [1,2], where  $\xi_{ab}$  is the  $a$ - $b$ -plane coherence length. Recently, it has been shown that heavy-ion induced linear defects in  $YBa_2Cu_3O_7$  (YBCO) crystals provide strong flux pinning at high temperatures [3,4]. The enhanced pinning is manifested in a several orders of magnitude increase in the magnetically measured critical current density ( $J_c$ ) at 77 K in substantial fields and a large shift of the irreversibility line  $H_{irr}(T)$  to higher temperatures. Magnetic measurements of the  $J_c$  and  $H_{irr}(T)$  in heavy-ion irradiated Bi-2:2:1:2 crystals, on the other hand, suggest that dissipation in this highly anisotropic cuprate occurs by motion of pancake vortices which are poorly pinned by linear defects in the temperature range  $\sim 77$  K [5,6].

The  $Tl_2Ba_2Ca_2Cu_3O_{10}$  (Tl-2:2:2:3) phase of the thallium cuprates is one of the most interesting high- $T_c$  superconductors. It has the highest known critical temperature  $T_c$  [7], and some transport [8] data suggest that it may be less anisotropic as compared to the Bi based cuprates. These properties, together with its tendency for a platelet-type growth in the basal plane in which current transport is not impeded by grain boundaries [9], make it a promising cuprate for large-scale conductor applications. However, these favorable characteristics are masked by the large reversible region in the  $H$ - $T$  plane [10], which does not allow usable supercurrent transport in the temperature and field regimes of interest.

In contrast to the aforementioned magnetization data on YBCO and Bi-2:2:1:2, and some low-temperature magnetic measurements on Tl-2:2:2:3 ceramics [11], here we report detailed transport measurements of the vortex dynamics in the Tl-2:2:2:3 system with linear defects created by heavy-ion irradiation. At the optimum defect

density, we measure a thousandfold enhancement in  $J_c$  at 77 K for  $\sim 2.5$  T field aligned parallel to the defects. This change is accompanied by a large shift of  $H_{irr}(T)$  and the onset of thermally activated flux flow (TAFF) resistivity to higher temperatures. We consider these results novel as they reveal the following for the first time: (a) Flux pinning by linear defects extends the temperature and field range of usable transport critical current in this system; (b) the L-shaped irreversibility line, which is characteristic of the highly anisotropic, double Bi-O and Tl-O layer containing cuprates, becomes more like that of YBCO; and (c) thermal fluctuations of a pinned flux line transverse to its length require unoccupied defects in its vicinity for optimum pinning at temperatures above the  $H_{irr}(T)$  of the unirradiated material.

Thin films of Tl-2:2:2:3 were grown on  $LaAlO_3$  substrates by rf sputtering. X-ray  $\theta$ - $2\theta$  scans showed a highly  $c$ -axis oriented growth normal to the plane of the substrate and 96% phase purity. The remaining 4% of the material was Tl-2:2:1:2 phase. The  $T_c$  in the Tl-2:2:2:3 system is highly dependent on Ba/Ca ratio and Tl-2:2:1:2 intergrowth [7,12]. The resistively measured zero-field  $T_c$  (midpoint) of the film used here was 106.7 K and the transition width  $\sim 2$  K. Details of film preparation have been discussed elsewhere [12]. Two films of thickness  $\sim 1.2$   $\mu m$  were chosen for transport and ac screening measurements before and after irradiation. One of the films was patterned with a  $300 \times 1000$   $\mu m^2$  active area for measurements of resistivity and  $J_c$ . Standard ac and dc techniques as described elsewhere [13] were used for this purpose. The current density used for resistivity measurements was  $\sim 40$  A/cm<sup>2</sup>. The ac screening measurements were performed using a two-coil mutual inductance technique similar to the one described by Jeanneret *et al.* [14] and operated at 100 kHz.

Silver ions, accelerated to 276 MeV in a tandem Van de Graaff accelerator, were used to irradiate the films. The ion beam was incident at 2 deg off the film normal.

The sample used in transport measurements was irradiated at fluences of  $\phi=0.5, 1, 2,$  and  $4 (\times 10^{11} \text{ ions/cm}^2)$ , which correspond to the field-equivalent fluences  $B_\phi (=1.2\phi\phi_0)=1.2, 2.3, 4.6,$  and  $9.2 \text{ T}$ , respectively, where  $\phi_0$  is the flux quantum. The approximate range of these ions in Tl-2:2:2:3 is  $\sim 16 \mu\text{m}$  and the energy loss rate  $\sim 25 \text{ keV/nm}$ . It has been shown previously that irradiation of high- $T_c$  cuprates with silver and gold ions of this energy results in formation of linear tracks, 5-10 nm in diameter, of amorphized material [13,15].

Activation of vortices from their respective pinning wells at temperatures where the thermal energy becomes comparable to the pinning potential is the primary cause of dissipation in high- $T_c$  cuprates. The TAFF resistivity, which varies as  $\rho = \rho_0 \exp[-U(T)/k_B T]$  in the classical Anderson-Kim picture, is a manifestation of this process. In this simplest single-particle activation picture of dissipation which neglects many-body effects [16], any enhancement in the pinning potential  $[U(T)]$  due to the linear defects would push the onset of dissipation to higher temperatures. In Fig. 1 we show the resistivity of the film before and after irradiation at several fluences. The measurements were performed at 1 T dc field directed perpendicular to the plane of the film. In the unirradiated state, the dissipation becomes detectable at  $\sim 60 \text{ K}$ . This large broadening of the transition for  $\mathbf{H} \parallel \mathbf{c}$  is a characteristic feature of the highly anisotropic cuprates [8,17]. Even at the lowest fluence ( $5 \times 10^{10} \text{ ions/cm}^2$ ), we observe a large shift ( $\sim 21 \text{ K}$ ) of the onset to higher temperatures. This shift reaches a saturation value at  $\phi \sim 2 \times 10^{11} \text{ ions/cm}^2$  and then decreases after irradiation at  $\phi = 4 \times 10^{11} \text{ ions/cm}^2$ . A clear understanding of these data requires a knowledge of the competing factors such as pinning due to linear defects, relative concentrations of the defects and flux lines, and changes in  $T_c$  ( $\delta T_c$ ) of the material after irradiation. The  $\delta T_c$  varies as  $\sim -(9.96 \times 10^{-12} \phi + 1.27 \times 10^{-23} \phi^2)$ , with a reduction of  $\sim 6 \text{ K}$  after irradiation at  $\phi \sim 4 \times 10^{11} \text{ ions/cm}^2$ . We

can understand this behavior by mapping the flux-line lattice (FLL) at 1 T onto the 2D array of linear defects distributed with a zero translational order. Since the shear strength of the FLL is much weaker compared to the pinning force of linear defects [2], at  $\phi = 5 \times 10^{10} \text{ ions/cm}^2$  ( $B_\phi \sim 1.2 \text{ T}$ ) all flux lines will be captured by the defects. This results in a large shift of the resistivity curve. At low temperatures, where thermal fluctuations are negligible, this situation would correspond to an optimally pinned phase. Further irradiation to a cumulative fluence equivalent to  $B_\phi = 2.3 \text{ T}$  leads to a situation where the defects outnumber flux lines in the system. At sufficiently low temperatures, this would correspond to the Bose-glass phase of the mixed state as described by Nelson and Vinokur [1]. However, as thermally activated lateral displacements of a flux line in the form of loops grow with temperature, effective pinning would require a finite number of unoccupied defects as the nearest neighbors. This behavior is reflected in the data for  $B_\phi = 2.3 \text{ T}$  which shifts to still higher temperatures. No further gain in pinning for  $B_\phi = 4.6 \text{ T}$ , as revealed by the saturation of the shift in Fig. 1, suggests that at this stage the number of defects in the film is more than the critical concentration required for optimum pinning. A further increase in the defect density would lead to a global suppression of the superconducting order parameter ( $\psi$ ), as evident from the suppression of  $T_c$ , and would counteract the gains in pinning. The downward shift of the resistivity curve at  $\phi = 4 \times 10^{11} \text{ ions/cm}^2$  must be due to a severe suppression of the order parameter in the sample. This global loss in  $\psi$  is a compound effect of the amorphized area under the defects ( $\sim 11.3\%$  at  $\phi = 4 \times 10^{11} \text{ ions/cm}^2$ ) and distortion of the oxygen sublattice in the regions surrounding them.

This mapping between the defects and vortices is also seen in the  $\rho$  vs  $T$  data taken at 5 T (Fig. 2). In this case also, a large and progressive shift of the onset of dissipation is seen after irradiation. However, in spite of a

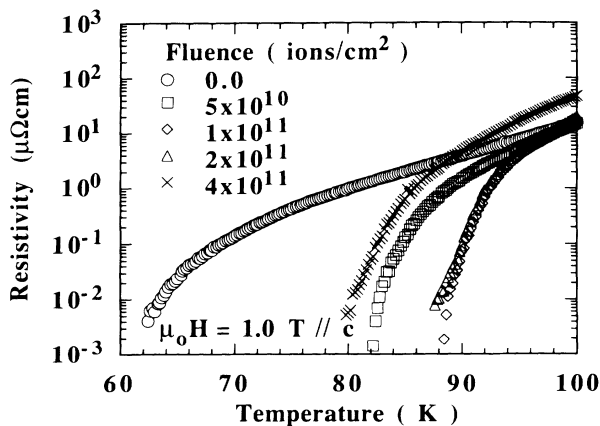


FIG. 1. Electrical resistivity of a Tl-2:2:2:3 film before and after irradiation with the Ag ions. The data were taken at 1 T ( $\mathbf{H} \parallel \mathbf{c}$ ).

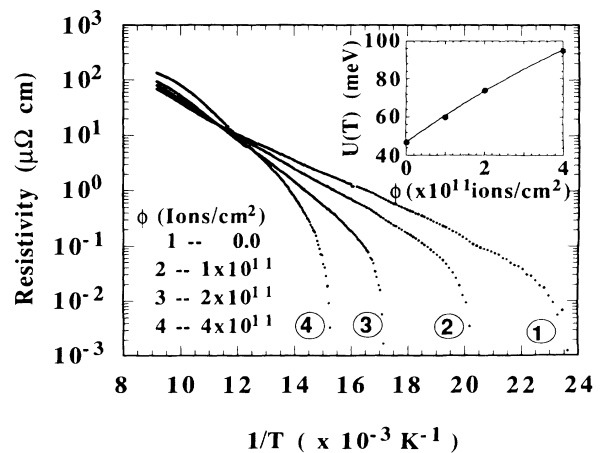


FIG. 2. The resistivity data at 5 T ( $\mathbf{H} \parallel \mathbf{c}$ ) before and after irradiation. Inset: A plot of the activation energy at 77 K vs  $\phi$ .

significant degradation in zero-field  $T_c$  at the highest fluence, the shift in this case continues in one direction. Since at 5 T only the maximum fluence ( $B_\phi=9.2$  T) provides enough defects to facilitate independent pinning of each flux line in the film, the data in Fig. 2 are a direct confirmation of the one-to-one mapping between linear defects and vortices in the system. We can quantify the enhancement in pinning by calculating the activation energy at a fixed temperature (say 77 K) from the Arrhenius plots of the form  $\ln\rho$  vs  $1/T$ . In the inset of Fig. 2, we show the activation energy  $U(77$  K and 5 T) vs  $\phi$ . To a first approximation, this  $U(T)$  can be regarded as the pinning energy of the most loosely bound lines in the system. This is expected to increase as more and more neighbors of such flux lines are trapped by the defects.

These effects of vortex confinement are seen in the behavior of  $J_c$  as well. In Fig. 3 we show the  $J_c$  data (criterion  $10 \mu\text{V}/\text{cm}$ ) taken at 77 K as a function of magnetic field. As seen in this figure,  $J_c$  of the unirradiated film is zero for  $\mu_0 H > 1$  T. This precipitous drop in  $J_c$  is a manifestation of weak pinning and a large reversible region in the mixed state of Tl-2:2:2:3. After irradiation, however, the range of measurable  $J_c$  extends to higher fields. There are some other interesting features in the data shown in Fig. 3. First of all, for  $\mu_0 H < 2.5$  T, the  $J_c$  after irradiation at the maximum fluence ( $\phi=4 \times 10^{11}$  ions/cm<sup>2</sup>) is considerably lower than the  $J_c$  at the lower fluences. This behavior is a consequence of  $\sim 6$  K reduction in  $T_c$  of the material, and is consistent with the  $\rho$  vs  $T$  curve at this fluence (Fig. 1). For  $\mu_0 H > 2.5$  T, however, the contribution of additional pinning centers becomes evident as the  $J_c$  vs  $\mu_0 H$  curve crosses the data for  $\phi=2 \times 10^{11}$  ions/cm<sup>2</sup>. Again, this crossover is in agreement with the measurement of linear resistivity at 5 T (Fig. 2).

For the values of fluence which do not cause a severe reduction in  $T_c$ , the critical current shows a weak field dependence for  $B < 0.5B_\phi$ . The drop in  $J_c$  for fields

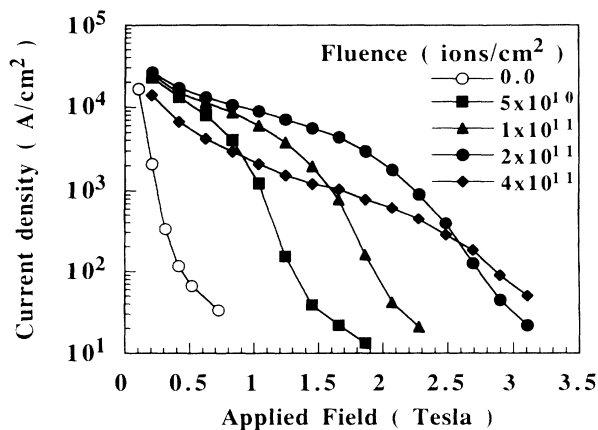


FIG. 3. Critical current density plotted as a function of applied field ( $H \parallel c$ ). The data were taken at 77 K.

$> 0.5B_\phi$  is much more rapid. This is a strikingly different behavior as compared to the low-temperature  $J_c$  data of Bi-2:2:1:2 crystals irradiated with iodine ions [5], which show a universal  $J_c \propto B^{-2}$  behavior for  $0.5 < B/B_\phi < 2.0$ . The data in Fig. 3 indicate that at high temperatures, a one-to-one correspondence between the flux lines and linear defects is not sufficient for optimum pinning. Instead, the defects must outnumber flux lines for an appreciable gain in  $J_c$  [1].

Finally, in Fig. 4 we show the irreversibility line of a Tl-2:2:2:3 film before and after irradiation at  $\phi=1$  and  $3 \times 10^{11}$  ions/cm<sup>2</sup>. The irreversibility temperature has been deduced from the position of the  $\chi''$  peak in the mutual-inductance data taken at 100 kHz with the external dc field  $H \parallel c$ . The inset of the figure shows quadrature components of the pickup voltage at 4.4 T. The strongly depressed, L-shaped,  $H_{\text{irr}}(T)$  of the unirradiated sample is a characteristic of the double Tl-O or Bi-O layer containing highly anisotropic cuprates [10,18,19]. The irreversibility line shifts to higher temperatures after irradiation. However, the shift is not the same at each field. The columnar defects tend to remove the curvature at the knee of  $H_{\text{irr}}(T)$  and make it more like the irreversibility line of the less anisotropic  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [3]. The large shift of the  $\chi''$  peak, which essentially signals the onset of linear response at the measurement frequency [20], is similar to the behavior of linear resistivity and  $J_c$  as discussed earlier. However, while correlating the position of the peak in  $\chi''$  with the dc linear response, the former must be deduced in the limit of zero frequency.

A complete understanding of the linear and nonlinear responses of vortices in the presence of columnar defects requires a knowledge of the factors such as FLL elasticity, anisotropy, and thermal fluctuations. For fields  $B \ll B_\phi$ , every flux line is trapped by the defects. At

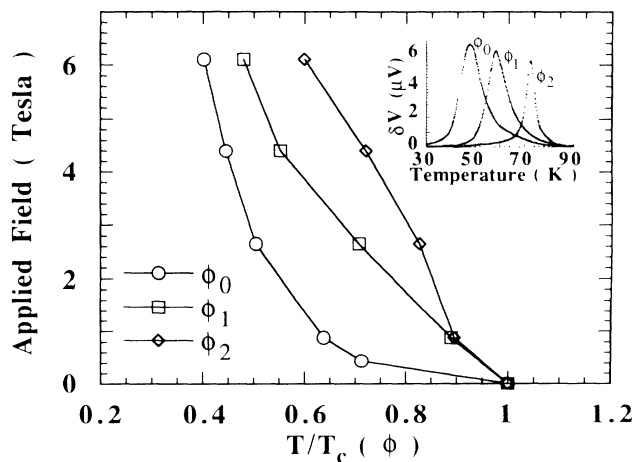


FIG. 4. Irreversibility line before ( $\phi_0$ ) and after irradiation at  $\phi_1=1 \times 10^{11}$  and  $\phi_2=3 \times 10^{11}$  ions/cm<sup>2</sup>. Inset: The quadrature ( $\chi''$ ) component of the pickup voltage at 4.4 T before and after irradiation.

sufficiently low temperatures,  $J_c$  in this limit is  $\sim cU_0/\phi_0 b_0$ , where  $U_0$  is the depth of the pinning potential for unit length of the flux line,  $\phi_0$  the flux quantum,  $c$  the velocity of light, and  $b_0$  the width of the well. However, as thermal fluctuations become stronger, vortex transport is decided by the balance of thermal, flux-line tension, and pinning energies with respect to the Lorentz force. When the line tension is significant, dissipation occurs by excitation of vortex loops of the  $z$  dimension spanning several  $\text{CuO}_2$  planes in the crystal lattice. In the limit of completely decoupled layers, motion of pancake vortices [21] would be responsible for dissipation. Although from the data in hand we can only speculate about the role of pancake vortices, our measurements of the critical current density at 77 K unambiguously show that the linear defects in this system can provide effective pinning even in a temperature range where the Josephson interlayer coupling is considerably weaker.

In summary, we have measured the TAFF resistivity,  $J_c$ , and irreversibility temperature of Tl-2:2:2:3 films containing linear defects of radius  $> \xi_{ab}$ . Trapping of vortices in these defects leads to a large shift of the onset of dissipation to higher temperatures. The linear defects shrink the reversible region on the  $T$ - $H$  plane and increase  $J_c$  in a temperature range where thermal fluctuations are important. In this regime, however, defects must outnumber vortices by a significant margin for effective pinning. Our data also show that a severe depression of the order parameter at high fluences limits extension of this criterion to arbitrarily large fields.

We thank D. O. Welch, J. R. Thompson, and V. M. Vinokur for helpful discussions. We also thank the control room staff at Brookhaven Tandem Van de Graaff for their cooperation during the irradiation experiments. This research has been supported by the U.S. Department of Energy, Division of Materials Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76CH00016. One of us (S.H.L.) was supported by NASA Lewis Grant No. NAG 3-886.

- 
- [1] D. R. Nelson and V. M. Vinokur, Phys. Rev. Lett. **68**, 2398 (1992).
  - [2] E. H. Brandt, Europhys. Lett. **18**, 635 (1992).
  - [3] L. Civale, A. D. Marwick, T. K. Worthington, M. A. Kirk, J. R. Thomson, L. Krusin-Elbaum, Y. Sun, J. R. Clem, and F. Holtzberg, Phys. Rev. Lett. **67**, 648 (1991).

- [4] M. Konczykowski, F. Rullier-Alebenque, E. R. Ycoboy, A. Shaulov, Y. Yeshurun, and P. Lejay, Phys. Rev. B **44**, 7167 (1991).
- [5] W. Gerhauer, G. Ries, H. W. Neumuller, W. Schmidt, O. Eibl, G. Saemann-Ischenko, and S. Klaumunzer, Phys. Rev. Lett. **68**, 879 (1992).
- [6] J. R. Thompson, Y. R. Sun, H. R. Kerchner, D. K. Christen, B. C. Sales, B. C. Chakoumakos, A. D. Marwick, L. Civale, and J. O. Thomson, Appl. Phys. Lett. **60**, 2306 (1992).
- [7] Z. Z. Sheng and A. M. Hermann, Nature (London) **332**, 138 (1988); S. S. P. Parkin, V. Y. Lee, E. M. Engler, A. I. Nazzari, T. C. Huang, G. Gorman, R. Savoy, and R. Beyers, Phys. Rev. Lett. **60**, 2539 (1988).
- [8] D. H. Kim, K. E. Gray, R. T. Kampwirth, J. C. Smith, D. S. Richardson, T. J. Marks, J. H. Kang, J. Talvacchio, and M. Eddy, Physica (Amsterdam) **177C**, 431 (1991).
- [9] L. N. Bulaevskii, J. R. Clem, L. I. Glazman, and A. P. Malozemoff, Phys. Rev. B **45**, 2545 (1992).
- [10] V. K. Chan and S. H. Liou, Phys. Rev. B **45**, 5547 (1992); D. N. Zheng, A. M. Campbell, R. S. Liu, and P. P. Edwards (to be published).
- [11] V. Hardy, D. Groult, J. Provost, M. Hervieu, B. Raveau, and S. Bouffard, Physica (Amsterdam) **178C**, 255 (1991).
- [12] S. H. Liou and C. Y. Wu, Appl. Phys. Lett. **60**, 2803 (1992).
- [13] R. C. Budhani, Y. Zhu, and M. Suenaga, Appl. Phys. Lett. **61**, 985 (1992).
- [14] B. Jeanneret, J. L. Gavilano, G. A. Racine, Ch. Leemann, and P. Martinoli, Appl. Phys. Lett. **55**, 2337 (1989).
- [15] Y. Zhu, R. C. Budhani, Z. X. Cai, D. O. Welch, and M. Suenaga (unpublished); R. C. Budhani, Y. Zhu, and M. Suenaga, IEEE Trans. Appl. Supercond. (to be published).
- [16] D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988); M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); M. V. Feigelman, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Phys. Rev. Lett. **63**, 2303 (1989).
- [17] K. C. Woo, K. E. Gray, R. T. Kampwirth, J. H. Kang, S. J. Stein, R. East, and D. M. McKay, Phys. Rev. Lett. **63**, 1877 (1989); T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1662 (1988).
- [18] Y. Xu, M. Suenaga, Y. Gao, J. E. Crow, and N. D. Spencer, Phys. Rev. B **42**, 8756 (1990).
- [19] V. Hardy, J. Provost, D. Groult, M. Hervieu, B. Raveau, S. Durcok, E. Pollert, J. C. Frison, J. P. Chaminade, and M. Pouchard, Physica (Amsterdam) **191C**, 85 (1992).
- [20] V. B. Geshkenbein, V. M. Vinokur, and R. Fehrenbacher, Phys. Rev. B **43**, 3748 (1991).
- [21] J. R. Clem, Phys. Rev. B **43**, 7837 (1991).