Lorentz-Force Independence of Resistance Tails for High-Temperature Superconductors in Magnetic Fields near T_c

K. C. Woo,[a] K. E. Gray, R. T. Kampwirth, J. H. Kang,[b] S. J. Stein, R. East, and D. M. McKay
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

(Received 5 July 1989)

An unusual loss mechanism is commonly observed in the high-temperature superconductors in a magnetic field near the zero-field transition temperature T_c. Resistive transitions in highly c-axis-oriented $Tl_2Ba_2CaCu_2O_x$ thin films are presented as a function of field perpendicular to the c axis for the current *both perpendicular and parallel to the field.* The data are essentially the same over a span of 5 orders of magnitude below the normal-state resistance. Since the *macroscopic* Lorentz force is zero for the current parallel to the magnetic field, it plays, at most, a significantly reduced role in this loss mechanism. Our results seriously question any explanation of these losses based on magnetic-flux motion.

PACS numbers: 74.60.Ge, 74.70.Ya

An unusual loss mechanism has been commonly observed in the high-temperature superconductors (HTS) in a magnetic field near the zero-field transition temperature T_c. This loss manifests itself as a current-independent resistance[1] (for sufficiently low currents) which broadens the transition in an uncharacteristic manner. In conventional superconductors, magnetic fields shift the resistive transitions to lower temperatures, with relatively small broadening, so the shift can be regarded as a measure of the upper critical field, H_{c2}. For the HTS in a magnetic field, there is relatively little change in temperature of the initial drop of resistance, at T_c(onset), whereas after an initial decrease, the resistance appears to be activated; i.e., it increases approximately exponentially with inverse temperature. These effects are more dramatic in[1] Bi-Sr-Ca-Cu-O and[2] Tl-Ba-Ca-Cu-O materials but are also found in[3] $YBa_2Cu_3O_7$. Additionally, magnetization measurements in $YBa_2Cu_3O_7$ single crystals indicate that H_{c2} is larger than that determined from any extrapolation to zero resistance or even by using the resistive midpoints.[4]

This unusual behavior has led authors to suggest the existence of thermally activated magnetic-flux creep,[1,5] flux-line melting,[6] or another unknown loss mechanism.[1] In this paper, resistive transitions of highly c-axis-oriented $Tl_2Ba_2CaCu_2O_x$ thin films are presented as a function of field perpendicular to the c axis, for the current *both perpendicular and parallel to the field.* The data for these orientations are essentially the same over a span of 5 orders of magnitude below the normal-state resistance (just above T_c). Since the *macroscopic* Lorentz force is zero for the current parallel to the magnetic field, it can play, at most, a significantly reduced role in this loss mechanism, and our results seriously question any explanation of these losses based on magnetic-flux motion. This new insight has important implications for understanding discrepancies between magnetic and transport measurements in HTS.

Sputtered films of $Tl_2Ba_2CaCu_2O_x$ were prepared[2,7] in a three-gun dc magnetron sputtering system which used a turbomolecular pump to provide a typical base pressure of $(2-3)\times10^{-8}$ Torr. The three guns are aimed at a common point about 6 in. above the sources which provides compositional uniformity to $\pm1\%$ over a 2-cm^2 substrate area. Targets of Tl, Cu, and a 1:1 BaCa mixture are simultaneously sputtered in a 20-mTorr argon atmosphere with an oxygen partial pressure of 0.1 mTorr being introduced directly adjacent to the substrates. The films were deposited onto (100)-oriented single-crystal or polycrystalline ZrO_2-9%Y_2O_3 substrates, which were kept at ambient temperature during the deposition. Film thicknesses are ~1900 nm. The films were annealed in a closed Au crucible, which was placed in a flowing-oxygen tube furnace at 850°-890°C for about 5-30 min. It was also found necessary to include a small pellet of target material in the Au crucible in order to adequately reduce the loss of the highly volatile Tl. Electrical contact was made by evaporating patterned Ag films and applying silver epoxy.

Scanning electron micrographs of such films indicate an interconnected backbone structure, which looks topologically similar to that found in epitaxial $YBa_2Cu_3O_7$ films on $SrTiO_3$ substrates,[8] except that the grains are not oriented along the principal crystal axes of the substrates in our $Tl_2Ba_2CaCu_2O_x$ films. The composition was measured by using energy dispersive x rays, and x-ray diffraction analysis indicates a high degree of orientation with the c axis perpendicular to the substrate. Samples were mounted in a gas-flow variable-temperature insert of a 13.5-T superconducting solenoid. Resistance measurements (ac) were taken with a standard lock-in technique as a heater surrounding the sample chamber increased the temperature slowly through the transition. Strictly identical thermal conditions (starting temperature and heating rate) were used for all runs. Standard magnetoresistance corrections were used for the carbon-glass thermometer, but these do not affect the relative measurements between current parallel and per-

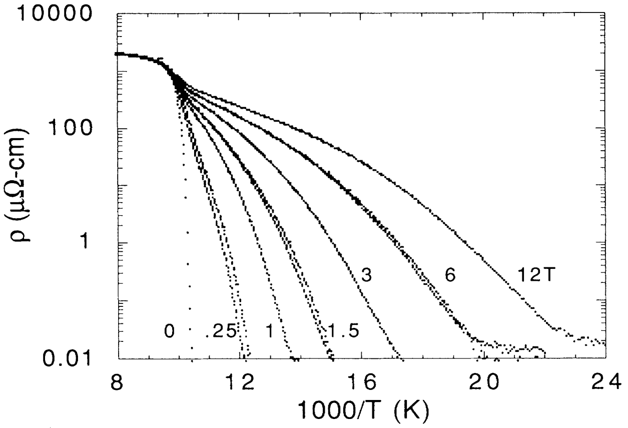

FIG. 1. The resistive transitions for various indicated fields which are perpendicular to the c axis. Both field orientations with respect to the current are displayed for fields of 0.25, 1.5, and 6 T. The current density was 7 A/cm^2.

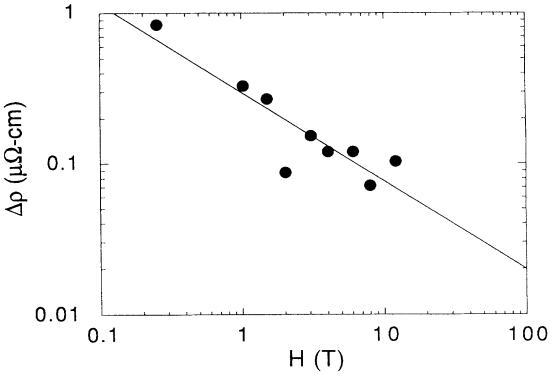

FIG. 2. The differences in resistivity, $\Delta\rho$, between H perpendicular and parallel to the current. They *decrease* with H, and therefore are unlikely to be a weak manifestation of the Lorentz force which is proportional to H. Comparisons were made at a fixed value of $\rho = 0.57$ $\mu\Omega$ cm for parallel field.

pendicular to the field.

Evidence for adequate alignment of the polycrystalline grains in the film and of the film in the magnet come from resistive measurements of the upper critical field[2] in the same apparatus which gave a giant anisotropy of ~70. Further evidence for the orientational uniformity of the film come from the torque magnetization anisotropy[9] (~94) in our films.

Some of the data are shown in Fig. 1 for various fields perpendicular to the c axis; both field orientations with respect to the current, I, which corresponds to 7 A/cm^2, are shown for fields of 0.25, 1.5, and 6 T, and they display minor differences compared to the overall effect. The resistance for H perpendicular to I is higher in these cases as well as all others, but this effect is seen in Fig. 2 to *decrease* with the field strength as $H^{-0.59}$. It is therefore unlikely to be a weak manifestation of the Lorentz force which is proportional to H, but more likely a result of warming the sample to room temperature and remounting it between measurements in the parallel and perpendicular orientations. Such small variations have been noted previously after thermal cycling when the field orientation was unchanged.

The behavior of the resistive transition versus field follows that obtained previously,[2] with the midpoints of the transition exhibiting a very large upper-critical-field slope. The linear portions of the curves shown in Fig. 1 occur for the lowest resistances above the noise level and can be fitted using a simple thermal activation model.[1] However, we do not find a single field-independent prefactor, and hence cannot reduce all our data to a single curve as claimed in Ref. 1. Figure 3 shows that the activation energies, U_0, follow an $H^{-0.47}$ scaling law: They are ***indistinguishable*** from measurements in $Tl_2Ba_2CaCu_2O_x$ single crystals,[10] are much smaller than for[3] $YBa_2Cu_3O_7$, but are about a factor of 2 higher than for Bi-Sr-Ca-Cu-O single crystals.[1] The latter reflects a somewhat smaller effect of the field on these $Tl_2Ba_2CaCu_2O_x$ films; e.g., in a 12-T field, a reduced resistance of 10^{-5} is found at about 44 K in our films and at 32 K in the Bi-Sr-Ca-Cu-O single crystal. At low fields, the prefactor increases significantly, by a factor of 10^{23} from 12 to 0.25 T. However, it should be noted that the natural logarithm of the prefactor divided by U_0 varies by no more than 40% over all the field values *including zero field.*

In principle, these resistive tails could be caused by film defects like grain boundaries since, in spite of the high degree of c-axis orientation, the films are not epitaxially grown. However, the overall behavior of the excess resistance found here is so similar to that of single crystals[10] of $Tl_2Ba_2CaCu_2O_x$ (our U_0 values, over the

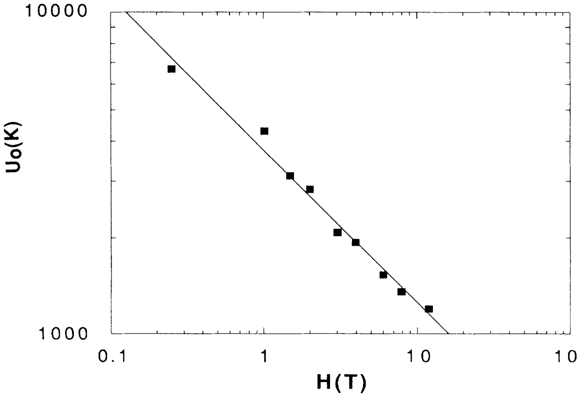

FIG. 3. The values of U_0 follow approximately an $H^{-0.47}$ scaling law. They are about a factor of 2 higher than for the Bi-Sr-Ca-Cu-O single crystal, but much smaller than found in $YBa_2Cu_3O_7$.

entire field range, are indistinguishable from Ref. 10) and the other HTS[1] that this possibility seems very remote. Additionally, although one can consider that the current I and/or flux-line direction may deviate from the macroscopic average and identify flux-flow dissipation mechanisms for each field orientation, we cannot envision *any* flux-flow scenario giving identical dissipations. Wanderings of current and field from their macroscopic directions would lead to regions with oppositely directed Lorentz forces, which would, at least, partially cancel resulting in a smaller net Lorentz force with $\mathbf{I} \parallel \mathbf{H}$. Although conventional superconductors in the "force-free configuration," with $\mathbf{I} \parallel \mathbf{H}$, indicate that the critical current density increases by factors of 20 or more,[11] numerous studies, including the novel idea of flux cutting,[12] indicate that flux motion can occur. However, such effects must be even smaller in our study because the ratio of current density to field is significantly reduced (down to 10^{-6}) in the data of Fig. 1, and remain *unchanged* as the current is further decreased (until the voltage becomes smaller than the noise). The fact that the excess resistance is largely unaffected by the orientation of current and field implies very strongly that the Lorentz force is not the origin of the resistive loss in the HTS. Recent measurements[13] of the excess resistance in single crystals of $YBa_2Cu_3O_7$ indicate two components, one showing the $\sin^2\theta$ dependence of the Lorentz force, where θ is the angle between $\mathbf{I}$ and $\mathbf{H}$, and the other being independent of θ as in our $Tl_2Ba_2CaCu_2O_x$ films. It will be important to repeat these measurements in single crystals and films of the other HTS exhibiting such lossy behavior.

The authors thank John Mattson for the data acquisition software. This work was supported by the U.S. Department of Energy, Division of Basic Energy Sciences-Materials Sciences and Office of Energy Storage and Distribution, Conservation and Renewable Energy, under Contract No. W-31-109-ENG-38 and the National Science Foundation—Office of Science and Technology Centers under Contract No. STC-8809854 (D.M.M., Science and Technology Center for Superconductivity, University of Illinois, Urbana-Champaign). K.C.W. acknowledges support from the Division of Educational Programs, Argonne National Laboratory.

Note added.—Similar studies by others have come to our attention. Iye, Nakamura, and Tamegai[14] have performed the valuable contribution of measuring this effect as a function of angle, to rule out misalignment with the field as its cause. Kitazawa *et al.*[15] show that the width of the transition, defined over about 1 order of magnitude in resistance, depends on the field orientation *with the c axis and not the current.*

---

[a] Also at University of Illinois at Chicago, Chicago, IL 60680.

[b] Present address: Westinghouse Research Laboratory, Pittsburgh, PA 15235.

[1] T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1662 (1988).

[2] J. H. Kang, K. E. Gray, R. T. Kampwirth, and D. W. Day, Appl. Phys. Lett. **53**, 2560 (1988).

[3] T. T. M. Palstra, B. Batlogg, R. B. van Dover, L. F. Schneemeyer, and J. V. Waszczak, Appl. Phys. Lett. **54**, 763 (1989).

[4] U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).

[5] Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988); M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988); P. H. Kes, J. Aarts, J. van den Berg, C. J. van der Beek, and J. A. Mydosh, Supercond. Sci. Technol. **1**, 242 (1989).

[6] D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988); P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, Phys. Rev. Lett. **61**, 1666 (1988).

[7] J. H. Kang, R. T. Kampwirth, and K. E. Gray, Phys. Lett. A **131**, 208 (1988).

[8] S. H. Liou, M. Hong, B. A. Davidson, R. C. Farrow, J. Kwo, T. C. Hsieh, R. M. Fleming, H. S. Chen, L. C. Feldman, A. R. Kortan, and R. J. Felder, in *Proceedings of the American Vacuum Society Topical Conference on Thin Film Processing and Characterization of High Temperature Superconductors,* edited by J. M. E. Harper, R. J. Colton, and L. C. Feldman, AIP Conference Proceedings No. 165 (American Institute of Physics, New York, 1988).

[9] K. E. Gray, R. T. Kampwirth, and D. E. Farrell (unpublished).

[10] H. Iwasaki, N. Kobayashi, M. Kikuchi, S. Nakajima, T. Kajitani, Y. Syono, and Y. Muto (private communication).

[11] G. D. Cody and G. W. Cullen, RCA Rev. (USA) **25**, 466 (1964); J. W. Heaton and A. C. Rose-Innes, Cryogenics **4**, 85 (1964); V. R. Karasik and V. G. Veresshchagin, Zh. Eksp. Teor. Fiz. **59**, 36 (1970) [Sov. Phys. JETP **32**, 20 (1971)].

[12] J. R. Clem, Phys. Rev. B **26**, 2463 (1982).

[13] W. K. Kwok, U. Welp, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu (unpublished).

[14] Y. Iye, S. Nakamura, and T. Tamegai, Physica (Amsterdam) **159C**, 433 (1989).

[15] K. Kitazawa, S. Kambe, M. Naito, I. Tanaka, and H. Kojima, Jpn. J. Appl. Phys. **28**, L555 (1989).