## Diminishing Sign Anomaly and Scaling Behavior of the Mixed-State Hall Resistivity in Tl<sub>2</sub>Ba<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10</sub> Films Containing Columnar Defects

R. C. Budhani,<sup>1</sup> S. H. Liou,<sup>2</sup> and Z. X. Cai<sup>1</sup>

<sup>1</sup>Materials Science Division, Brookhaven National Laboratory, Upton, New York 11973

<sup>2</sup>Department of Physics, University of Nebraska, Lincoln, Nebraska 68588

(Received 25 November 1992)

The issues of sign reversal of the Hall voltage and scaling between longitudinal  $(\rho_{xx})$  and Hall  $(\rho_{xy})$  resistivities are studied in Tl<sub>2</sub>Ba<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10</sub> films in which the vortex dynamics is drastically changed by flux pinning at heavy-ion-irradiation-induced linear defects. While the sign anomaly diminishes with increasing defect concentration, the power law  $\rho_{xy} \sim \rho_{xx}^{\beta}$ ,  $\beta = 1.85 \pm 0.1$ , holds even after irradiation. This result shows that the scaling is a universal feature of the mixed state in this system. The sign anomaly, on the other hand, is not consistent with a model that invokes pinning-induced backflow in the vortex core as the mechanism for this effect.

PACS numbers: 74.60.Ge, 72.15.Gd, 74.62.Bf, 74.76.-w

Understanding the Hall coefficient in the mixed state of high temperature superconductors has become a rich area of research due to its two puzzling features which do not find satisfactory explanation in the confines of the classical hydrodynamic theories of vortex motion in charged superfluids. These are the following: (a) The Hall coefficient over a range of temperatures and magnetic fields below  $T_c$  reverses its sign [1-8]. (b) A striking scaling behavior of the type  $\rho_{xy} \sim \rho_{xx}^{\beta}$ , where  $\rho_{xx}$  and  $\rho_{xy}$  are the longitudinal and Hall resistivities, respectively, and  $\beta = 1.7$ , has been observed in the case of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO) in a temperature range close to the onset of the Hall signal [8]. The sign anomaly has found various explanations such as (1) a two band model for the condensate in which the gap parameter for the minority band is much smaller [9], (2) effects of superconducting fluctuations [10], (3) a combination of Nernst-Ettinghausen and Seebeck effects that lead to a voltage opposite in sign to that of the Hall voltage due to the majority carriers [11], (4) phenomenological [6] and microscopic [12] modifications of the damping forces in order to establish a steady state vortex flow with a velocity component  $(v_x)$ antiparallel to the transport current, and finally (5) a theory [13] that includes pinning in the standard Bardeen-Stephen model [14] for flux flow and predicts a sign change with the increasing pinning strength.

The scaling relation between  $\rho_{xy}$  and  $\rho_{xx}$  as reported by Luo *et al.* [8] in YBCO films has been argued to be a consequence of vortex-glass transition in a 3D vortex system with a weak, random disorder [15]. Recently, however, Vinokur and co-workers [16] have proposed that the relation  $\rho_{xy} \sim \rho_{xx}^{\beta}$ ,  $\beta = 2$ , is a general feature of any vortex state with disorder dominated dynamics. The experimental results of Samoilov on Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> [17] in the temperature and field range where the mixed state has a quasi-2D character [18,19] and is not likely to form a vortex-glass state [20] support this proposal.

In this paper we address the issues of sign anomaly and scaling in the  $Tl_2Ba_2Ca_2Cu_3O_{10}$  (Tl-2223) system in

which the vortex dynamics is drastically altered by heavy-ion-irradiation-induced linear defects. In the defect-free state, and the temperature range where dissipation is dominated by the thermally activated motion of pancake vortices,  $\rho_{xy} \sim \rho_{xx}^{\beta}$ ,  $\beta = 1.85 \pm 0.1$ . Interestingly, however, this relationship holds even after inclusion of the linear defects that act as strong pinning centers for vortices in the system. At low fields,  $\sim 1$  T, the Hall voltage in the unirradiated samples shows the characteristic sign anomaly in the mixed state. This anomaly diminishes as the concentration of defects and hence the pinning force is increased. We consider these results novel as they show for the first time that (1) the sign anomaly and scaling are unrelated, (2) the scaling between  $\rho_{xy}$  and  $\rho_{xx}$  is a universal feature of the vortex dynamics in this system and does not depend on the nature of the pinning force, and (3) the behavior of sign anomaly is not consistent with the concept of a pinning-induced backflow in the core as the mechanism for the sign reversal.

Thin films of TI-2223 were grown on LaAlO<sub>3</sub> 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 TI-2212 phase. The  $T_c$  in the TI-2223 system is highly dependent on Ba/Ca ratio and Tl-2212 intergrowth [21,22]. 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 [22]. A film of thickness  $\sim 1.2$  $\mu m$  was patterned in a six probe geometry, with a  $300 \times 1000 \ \mu m^2$  active area, for the measurement of transverse  $(\rho_{xy})$  and longitudinal  $(\rho_{xx})$  resistivities in magnetic field directed parallel to the c axis of the crystal lattice (parallel to the film normal). The standard ac lock-in technique was employed to measure the longitudinal and transverse voltages  $V_{xx}$  and  $V_{xy}$ , respectively, and  $\rho_{xy}$  was deduced from the asymmetric part of  $V_{xy}$  under field reversal. The current density used in these measurements was  $\sim 40 \text{ A/cm}^2$ .

Linear defects consisting of columns (50-60 nm diameter) of amorphized material were created by irradiation of the samples with 276 MeV silver ions. Details of irradiation experiments [23] and changes in critical current density  $J_c$ , irreversibility temperature, and thermally activated flux flow (TAFF) behavior of the material as a result of flux pinning by these defects have been described elsewhere [24].

In Figs. 1(a) and 1(b), respectively, we show the temperature dependence of the Hall and longitudinal resistivities measured at 1 T field before and after irradiation at fluences  $\phi = 0.5$ , 1, and 2 (×10<sup>11</sup>) ions/cm<sup>2</sup>. The Hall resistivity in the normal state of the unirradiated sample is positive and increases linearly with the applied field. On cooling below  $T_c$ ,  $\rho_{xy}$  becomes negative, with a peak negative value  $\sim 0.011 \ \mu \Omega$  cm at temperature  $T^* \sim 103.0$ K. By putting in the value of  $\rho_{xx}$  at this temperature, we obtain a Hall tangent of  $\tan \Theta_H \sim -0.0038$ . On further cooling, the Hall signal undergoes another sign change and then gradually becomes zero at  $\sim$ 75 K. The broad features of these data, including the double sign reversal which is not seen in YBCO, are similar to the earlier reports on Tl-2212 [6] and Bi-2212 [4] samples. The remaining three curves in Fig. 1(a) show  $\rho_{xy}$  after irradiation at several fluences. Two striking effects of the ioninduced defects are seen on  $\rho_{xy}$ . First, the onset of  $\rho_{xy}$ shifts to higher temperatures, and second, while the temperature at which it has the peak negative value does not change, the amplitude of the negative swing diminishes with the increasing fluence. The shift of the onset of  $\rho_{xy}$ to higher temperatures after irradiation coincides with the behavior of  $\rho_{xx}$ . The onset of longitudinal resistivity

[Fig. 1(b)] shows a large and progressive shift to higher temperatures on increasing the fluence to  $1 \times 10^{11}$  ions/  $cm^2$ , at which an optimum defect concentration is reached for maximum flux pinning. The localization of vortices at the linear defects leads to interesting phases and crossovers in the H-T plane as discussed by Nelson and Vinokur [25]. We have shown previously [24] that in the temperature range above the irreversibility temperature of the unirradiated material, the linear defects must outnumber flux lines by a margin of 2 to 1 for effective pinning. For 1 T field, this criterion is satisfied at  $\phi \sim 1 \times 10^{11}$  ions/cm<sup>2</sup>. A further increase in the fluence to  $2 \times 10^{11}$  ions/cm<sup>2</sup> does not shift the  $\rho_{xx}$  curve. In fact it moves to lower temperatures after irradiation at a cumulative fluence  $\sim 4 \times 10^{11}$  ions/cm<sup>2</sup> (data not shown). This reverse trend is a consequence of a global depression of the order parameter that also manifests as  $\sim 6$  K reduction in  $T_c$ .

Similar effects of vortex pinning by linear defects are also seen in  $\rho_{xy}$  and  $\rho_{xx}$  data taken at 5 T [Figs. 2(a) and 2(b)]. In the unirradiated state, the Hall resistivity at 5 T remains positive over the entire temperature range below  $T_c$ . The negative swing of  $\rho_{xy}$  in the 1 T data now appears as a dip in the positive quadrant located nearly at the same temperature as  $T^*$  in Fig. 1(a). As for the data at 1 T, here also the onset of  $\rho_{xy}$  shifts to higher temperatures and the dip diminishes with the increasing fluence. A comparison of  $\rho_{xy}$  and  $\rho_{xx}$  data in Figs. 1 and 2 also shows that the Hall resistivity falls below the detection limit at a much higher temperature as compared to  $\rho_{xx}$ .

In Fig. 3 we establish the scaling between  $\rho_{xy}$  and  $\rho_{xx}$  for the unirradiated samples. These measurements were



5 (a) 4 2 5 Tesla ρ ( μΩ cm ) H//c 3 2 2 0 đ 1 0 φ 0 40 50 60 70 80 90 100 110 Temperature ( K )  $10^{2}$ (b)  $10^{1}$ 5 Tesla  $\rho_{_{X\,X}}(\;\mu\Omega\;\text{cm}\;)$ H //c  $10^{0}$ Fluence (ions/cm<sup>2</sup>)  $\phi_0 = 0.0$ 10  $= 1 \times 10^{11}$ 10.3  $= 2 \times 10^{11}$ φ, 10 70 50 40 60 80 90 100 110 Temperature ( K )

FIG. 1. Hall (a) (note separate y axis for each curve) and longitudinal resistivity (b) measured at 1 T before and after irradiation. Symbols  $\phi_0$ ,  $\phi_1$ ,  $\phi_2$ , and  $\phi_3$  correspond to fluence 0.0, 0.5, 1.0, and 2.0 (×10<sup>11</sup>) ions/cm<sup>2</sup>, respectively.

FIG. 2. Hall (a) (note separate y axis for each curve) and longitudinal resistivity (b) measured at 5 T before and after irradiation. Symbols ( $\phi$ 's) used to denote the fluence are the same as in Fig. 1.



FIG. 3. Plots of  $\log |\rho_{xy}|$  vs  $\log \rho_{xx}$  for data taken at 1, 3, 5, and 7 T (curves *d* through *a*, respectively). Solid lines highlight the scaling  $\rho_{xy} \propto \rho_{xx}^{\theta}$ ,  $\beta = 1.85 \pm 0.1$ . The *y* axis for each curve is drawn separately for the sake of clarity.

performed at 1, 3, 5, and 7 T dc fields directed parallel to the c axis of the crystal lattice. Since the  $\rho_{xy}$  at 1 T changes sign, we have plotted the absolute value of  $|\rho_{xy}|$ vs  $\rho_{xx}$  on a log-log plot. Also, the data for 3, 5, and 7 T have been displaced vertically by  $\log |\rho_{xy}| = 1.0$ , 2.0, and 3.0, respectively, for the sake of clarity. The solid lines in the figure are power laws,  $\rho_{xy} \sim \rho_{xx}^{\beta}$ ,  $\beta = 1.85 \pm 0.1$ . At 1 T field, the power law is barely discernible due to a rapid drop of  $\rho_{xx}$  and  $\rho_{xy}$  over a narrow temperature range. However, the range of  $\rho_{xx}$  over which the power law persists grows with the increasing applied field. This behavior is similar to that of YBCO films [8]. But, unlike the data on YBCO, where the power law relation is observed in the regime where  $\rho_{xy}$  is negative, in the present case the  $\rho_{xy} \sim \rho_{xx}^{\beta}$  relation breaks off before the negative regime is reached on scanning from lower temperatures. The power law dependence observed in Fig. 3 persists even in the case where the vortex dynamics is significantly modified by the ion-induced linear defects. In Fig. 4 we investigate how the scaling between  $\rho_{xy}$  and  $\rho_{xx}$  is affected in the presence of linear pins. These data, taken at 5 T before and after irradiation, show that the  $\rho_{xy} = A \rho_{xx}^{\beta}$ ,  $\beta = 1.85 \pm 0.1$ , dependence persists even in the presence of linear pins. The intercept of the solid lines in Fig. 4 on the  $\log |\rho_{xx}|$  axis gives the power law constant A. The values of A are  $\sim 0.025$ , 0.01, and 0.0045 for  $\phi_0$ ,  $\phi_2$ , and  $\phi_3$ , respectively. This observation has serious implications for the physics of scaling as discussed below.

Vinokur *et al.* [16] have argued that the correct equation of motion for a steady state vortex flow with velocity  $\mathbf{v}$  ( $iv_x + jv_y$ ) in the presence of pinning is obtained by balancing the Lorentz force  $(e/cn_s\phi_0)$  ( $\mathbf{v}_s - \mathbf{v}$ )×**n** acting on *individual* flux lines by a damping force of the type  $[\eta + \gamma(v)]\mathbf{v} + \eta'\mathbf{n} \times \mathbf{v}$ . The Lorentz force is generated by the charged superfluid moving at a relative velocity ( $\mathbf{v}_s - \mathbf{v}$ ) in the vortex frame of reference, and has the same form as the Magnus force found in liquid helium



FIG. 4. Plots of  $\log \rho_{xy}$  vs  $\log \rho_{xx}$  for data taken at 5 T before and after irradiation. Solid lines highlight the power law behavior of  $\rho_{xy}$  and  $\rho_{xx}$ .

[26,27]. In these expressions,  $\mathbf{v}_s$  and  $n_s$  are the superfluid velocity and number density, respectively,  $\mathbf{n}$  the unit vector along the flux line, and  $\phi_0$  the flux quantum. The friction term  $\eta$  comes from the dissipation in the normal core, and it has the Bardeen-Stephen type form  $\eta = \phi_0 H_{c2}/c^2 \rho_n$ , where  $\rho_n$  is the normal state resistivity. The damping term  $\gamma(v)$  originates from the average pinning force on a flux line. The equation of motion can be written as

$$\overline{\gamma}(v)\mathbf{v} + \alpha \mathbf{v} \times \mathbf{n} = \phi_0/c \,\mathbf{J} \times \mathbf{n} \,, \tag{1}$$

where  $\overline{\gamma}(v) = \eta + \gamma(v)$ ,  $\mathbf{J} = en_s \mathbf{v}_s$  is the current density, and the damping term  $\alpha \mathbf{v} \times \mathbf{n}$  comes from the Lorentz force combined with the drag force  $\eta' \mathbf{n} \times \mathbf{v}$  acting orthogonal to the vortex velocity v [28]. By solving this equation for v, Vinokur and co-workers [16] have extracted the electric field  $\mathbf{E} = (\mathbf{B} \times \mathbf{v})/c$  induced by the vortex motion. Use of the Ohms law then allows the following relationship between  $\rho_{xy}$  and  $\rho_{xx}$ ,  $\rho_{xy} = \rho_{xx}^2 (c\alpha/B\phi_0)$ . Focusing on the TAFF regime of dissipation, where  $\alpha$  is a slowly varying function of temperature, we have  $\rho_{xy} \propto \rho_{xx}^{\beta}$ with  $\beta \sim 2$ . The data shown in Fig. 3 indeed show that this power law dependence develops at lower temperatures where thermally activated flux flow behavior is dominant. Since the derivation of this formula does not depend on the detailed form of the pinning potential as long as it is invariant under time reversal, one would expect a similar behavior even in samples containing the linear defects. The data shown in Fig. 4 are also consistent with this picture.

At this point we switch to a discussion on the behavior of the sign anomaly. If we tacitly assume that the sign reversal is a consequence of the vortex velocity component antiparallel to transport current and the resultant Josephson field  $E_y = (Bv_x)/c$ , the origin of such an upstream motion of vortices must be identified. Wang and Ting [13] have discussed how a pinning-induced backflow of normal carriers in the core may result in a sign reversal. However, our data show a diminishing anomaly as more and more vortices are pinned by the linear defects. This is opposite to what is expected from this theory. Hagen *et al.* [6] have given an *ad hoc* argument for an equation of motion where the Magnus force is balanced by the damping terms  $\eta \mathbf{v}$  and  $\delta \mathbf{v}_s$  for a steady state flow. Such a formalism indeed predicts a sign reversal below  $T_c$  at low magnetic fields. Microscopic arguments for a damping term proportional to the superfluid velocity  $\mathbf{v}_s$  arising from opposing drifts of thermally excited quasiparticles far outside the core have been given by Ferrell [12].

Vinokur *et al.* [16] have argued against the existence of a damping force which is proportional to the superfluid velocity  $\mathbf{v}_s$  on the grounds that it would violate the dissipationless flow properties of a superfluid. In their picture, the Hall tangent is  $\tan \Theta_H = \alpha/[\eta + \gamma(v)]$ , and its sign is decided by the sign of  $\alpha$ . If we assume that the temperature dependence of  $\alpha$  remains the same before and after irradiation, although its magnitude decreases as evident from the intercepts in Fig. 4, the singular effect of pinning [higher  $\gamma(v)$ ] would be to reduce  $\tan \Theta_H$ . This is consistent with the observation of a shift of  $\rho_{xy}$  and decrease in the negative swing as the density of linear defects is increased.

Although our data, both scaling and sign anomaly, are consistent with the arguments of Vinokur *et al.*, the equation of motion proposed by them [Eq. (1)] is purely phenomenological. Dorsey [29] has used a time dependent Ginzburg-Landau approach to solve Eq. (1) in the limit of zero disorder  $[\gamma(v)=0]$  and low flux density. The sign of the Hall voltage in this picture also depends on  $\alpha$  which has two contributions, one coming from the normal Hall effect in the core of the vortex and the other from the order parameter relaxation time. The second term can be negative depending on the detailed electronic structure of the material.

In summary, we have measured the sign anomaly of the Hall resistivity and scaling between the Hall and longitudinal resistivities in the mixed state of TI-2223 films. In the temperature range where  $\rho_{xx}$  is controlled by TAFF processes, we observe a power law relationship of the type  $\rho_{xy} \sim \rho_{xx}^{\beta}$ , with  $\beta = 1.85 \pm 0.1$ . This behavior persists even after incorporation of linear pinning centers and thus provides compelling evidence for a theory which identified the scaling as a general feature of the disorder dominated vortex dynamics. The behavior of the sign anomaly with increasing pinning strength rules out the possibility of a pinning-induced backflow in the vortex core as the source of the sign reversal.

We thank V. M. Vinokur, G. Blatter, M. Suenaga, and D. O. Welch for helpful discussions. We also thank V.M.V. and G.B. for providing a copy of Ref. [16] before publication. 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] M. Galffy and E. Zirgiebl, Solid State Commun. 68, 929 (1988).
- [2] Y. Iye, S. Nakamura, and T. Tamegai, Physica (Amsterdam) 159C, 616 (1989).
- [3] K. C. Woo, K. E. Gray, R. T. Kampwirth, and J. H. Kang, Physica (Amsterdam) 162-164C, 1011 (1989).
- [4] S. M. Artemenko, I. E. Gorlova, and Y. I. Latyshev, Phys. Lett. A 138, 428 (1989).
- [5] T. R. Chien, T. W. Jing, N. P. Ong, and Z. Z. Wang, Phys. Rev. Lett. 66, 3075 (1991).
- [6] S. J. Hagen, C. J. Lobb, R. L. Greene, and M. Eddy, Phys. Rev. B 43, 6246 (1991).
- [7] J. P. Rice, N. Rigakis, D. M. Ginsberg, and J. M. Mochel, Phys. Rev. B 46, 11050 (1992).
- [8] J. Luo, T. P. Orlando, J. M. Graybeal, X. D. Wu, and R. Muenchausen, Phys. Rev. Lett. 68, 690 (1992).
- [9] J. E. Hirsch and F. Marsiglio, Phys. Rev. B 43, 424 (1991).
- [10] A. G. Aronov and S. Hikami, Phys. Rev. B 41, 9548 (1990).
- [11] A. Freimuth, C. Hohn, and M. Galffy, Phys. Rev. B 44, 10396 (1991).
- [12] R. A. Ferrell, Phys. Rev. Lett. 68, 2524 (1992).
- [13] Z. I. Wang and C. S. Ting, Phys. Rev. Lett. 67, 3618 (1991).
- [14] J. Bardeen and M. J. Stephen, Phys. Rev. 140, A1197 (1965).
- [15] A. T. Dorsey and M. P. A. Fisher, Phys. Rev. Lett. 68, 694 (1992).
- [16] V. M. Vinokur, V. B. Geshkenbein, M. V. Feigel'man, and G. Blatter (to be published).
- [17] A. V. Samoilov, preceding Letter, Phys. Rev. Lett. 71, 617 (1993).
- [18] J. R. Clem, Phys. Rev. B 43, 7847 (1991).
- [19] 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).
- [20] M. P. A. Fisher, Phys. Rev. Lett. 62, 1415 (1989); D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B 43, 130 (1991).
- [21] S. S. P. Parkin, V. Y. Lee, E. M. Engler, A. I. Nazzal, T. C. Huang, G. Gorman, R. Savoy, and R. Beyers, Phys. Rev. Lett. 60, 2539 (1988).
- [22] S. H. Liou and C. Y. Wu, Appl. Phys. Lett. 60, 2803 (1992).
- [23] R. C. Budhani, Y. Zhu, and M. Suenaga, IEEE Trans. Appl. Supercond. 3, 1675 (1993); R. C. Budhani, Y. Zhu, and M. Suenaga, Appl. Phys. Lett. 61, 985 (1992).
- [24] R. C. Budhani, M. Suenaga, and S. H. Liou, Phys. Rev. Lett. 69, 3816 (1992).
- [25] D. R. Nelson and V. M. Vinokur, Phys. Rev. Lett. 68, 2398 (1992).
- [26] P. G. de Gennes and J. Matricon, Rev. Mod. Phys. 36, 45 (1964).
- [27] P. Nozières and W. F. Vinen, Philos. Mag. 14, 667 (1966).
- [28] W. F. Vinen, *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), Vol. 2, p. 1167; see also M. C. Marchetti and D. R. Nelson, Physica (Amsterdam) **174C**, 40 (1991).
- [29] A. T. Dorsey, Phys. Rev. B 46, 8376 (1992).