Scaling behavior and mixed-state Hall effect in epitaxial HgBa₂CaCu₂O₆₊₆ thin films

W. N. Kang,* S. H. Yun, and J. Z. Wu

Department of Physics and Astronomy, University of Kansas, Lawrence, Kansas 66045

D. H. Kim

Department of Physics, Yeungnam University, Kyungsan 712-749, Korea (Received 24 June 1996; revised manuscript received 13 September 1996)

We have measured the mixed-state Hall effect in superconducting $HgBa_2CaCu_2O_{6+\delta}$ thin films. We found that the Hall resistivity ρ_{xy} shows a double sign reversal in low fields, then shifts to positive without changing the general shape in higher fields. At the higher field region, the Hall conductivity ρ_{xy} is observed to be the sum of two terms, C_1/H and C_2H , where C_1 and C_2 are field independent. The scaling behavior between σ_{xy} and ρ_{xx} (longitudinal resistivity) shows a strong dependence on fields. The scaling exponent β in $\rho_{xy} = A \rho_{xx}^{\beta}$ increases from 1.5 ± 0.1 to 1.9 ± 0.1 as field increases from 1 to 5.5 T. The field dependence of tangent of the Hall angle is linear only in the flux-flow regime, different from Bi- and Tl-based cuprates. $[$ S0163-1829(97)01201-0]

INTRODUCTION

Since the discovery of a family of Hg-based high- T_c superconductors, $1,2$ a number of studies have been reported on these materials including the temperature dependence of the thermoelectric power, 3 the normal-state Hall effect, 4 the irreversibility line, $5,6$ and the critical current density.^{7,8} However, the Hall effect in the mixed state of Hg-based superconductors remains to be one of the unsolved problems in understanding of flux motion of type-II superconductors.

The sign reversal of the mixed-state Hall effect has been observed in most of the high- T_c superconductors^{9–15} as well as in some conventional superconductors.^{9,16} While only a single sign reversal is observed in $YBa_2Cu_3O_7$ (YBCO) crystals¹⁵ and films,¹⁰ the double sign change has been observed in $Bi_2Sr_2CaCu_2O_8$ (Bi-2212) crystals,¹¹ $Tl_2Ba_2CaCu_2O_8$ (Tl-2212) films,^{12,13} and $Tl_2Ba_2Ca_2Cu_3O_{10}$ $(Tl-2223)$ films.¹⁴ Furthermore, an interesting scaling behavior between the Hall resistivity ρ_{xy} and the longitudinal resistivity ρ_{xx} , $\rho_{xy} = A \rho_{xx}^{\beta}$, has also been observed with the scaling exponent $\beta \sim 2$ in Bi-2212 crystals¹¹ and Tl-2212 films,¹³ β =1.5–2.0 in YBCO films¹⁰ and crystals.¹⁵

It is now generally accepted that the mixed-state Hall effect in type-II superconductors in magnetic fields consists of two contributions, quasiparticle and hydrodynamic.^{17–21} The sign of the quasiparticle term remains the same as that in the normal state, but the hydrodynamic term can be negative in the mixed state so that the sign reversal takes place. Dorsey and Fisher²² showed that the scaling behavior $\rho_{xy} = A \rho_{xx}^{\beta}$, is a general picture near the vortex-glass transition but were not be able to obtain the exact estimation of the scaling exponent. A model proposed by Vinokur *et al.*²³ suggests that the scaling exponent should be 2 in the thermally assisted fluxflow (TAFF) region regardless of the pinning. Their results were in fair agreement with scaling exponents of both weakly pinned systems of Bi-2212 crystals $(\beta=2.0\pm0.1),^{11}$ and rather strongly pinned systems of heavy-ion irradiated Tl-2223 films $(\beta=1.85\pm0.1)$,¹⁴ but not with the recent results of heavy-ion irradiated YBCO $(\beta=1.5\pm0.1).$ ¹⁵ Wang, Dong, and $Ting²⁴$ explained the scaling behavior and the anomalous sign reversal of the Hall effect by taking into account the backflow current due to pinning. In their model the scaling exponent β may change from 2 to 1.5 as the pinning strength increases, and this is found to be consistent with the result of Ref. 21.

Hg-based cuprates are a very interesting system for understanding the role of pinning. For example, $HgBa_2CaCu_2O_{6+\delta}$ (Hg-1212) has a similar crystalline structure as TlBa₂CaCu₂O_{6+ δ} but higher T_c (~125 K). In particular, the irreversible line of the Hg-based system is ranged between that of YBCO and Bi-2212/Tl-2212 (Ref. 6) and the pinning strength strongly depends on applied field. The unpurterbed pinning potential U_0 of the Hg-based system was observed as $U_0(H) \propto 1/H$, ²⁶ which is similar to the YBCO, ²⁹ while $U_0(H)$ is proportional to $1/\sqrt{H}$ (Ref. 30) for Bi-2212 and Tl-2212 systems.

In this paper, we report a clear observation of the double sign reversal in the mixed-state Hall effect of high-quality Hg-1212 films and the magnetic-field dependence of the Hall scaling behavior.

EXPERIMENT

The fabrication process of Hg-1212 films used in this study is similar to that described in detail by Yun and Wu.⁷ For films on $SrTiO₃$ grown from the same batch, the midtransition temperature (T_c) ranges 123–124 K with the transition width of 2–3 K in a zero field. The critical current density at 77 K is \sim 10⁶ A/cm². The x-ray-diffraction analysis shows a highly *c*-axis-oriented Hg-1212 phase normal to the plan of substrate. The films with ~ 0.8 μ m thickness have typical dimensions of 6×4 mm². The electrical contacts $(<1$ Ω) were made by sputtering Ag followed by annealing at 350 °C for 20 min under O_2 atmosphere.

The Hall resistivity ρ_{xy} and the longitudinal resistivity ρ_{xx} were measured in a 5.5 T superconducting magnet system using the standard five-probe dc method. The magnetic fields were applied parallel to the *c* axis of Hg-1212 films, and ρ_{xy}

FIG. 1. Temperature dependence of the longitudinal resistivity in Hg-1212 films. Fields from top to bottom are 5.5, 4, 2, 1, and 0 T.

was obtained from the antisymmetric part of the Hall voltage under the magnetic-field reversal. The current density used for these measurements was \sim 250 A/cm². Both ρ_{xy} and ρ_{xx} were observed to be linear at those currents.

RESULTS AND DISCUSSION

The temperature dependence of ρ_{xx} near the transition temperature for the magnetic fields up to 5.5 T are shown in Fig. 1. T_c is 123.7 K with the transition width of 2.6 K. The zero-resistance temperatures $(T_{c,\text{zero}})$ in the limit of our instrument resolution in magnetic fields of 1 and 5.5 T were 90 and 48 K, respectively. It is interesting to compare the relative transition temperature with Bi-2212, Tl-2223, and YBCO compounds at the same field. The reduced zeroresistance temperatures $(T_{c,\text{zero}}/T_c)$ at 1 T are 0.40, 0.58, 0.73, and 0.94 for Bi-2212 crystals,²⁸ Tl-2223 films,¹⁴ Hg-1212 films, and YBCO crystals,¹⁵ respectively. These results show that the pinning strength of Hg-1212 ranges between Bi-2212/Tl-2223 and YBCO.

In Fig. 2, the corresponding Hall resistivities ρ_{xy} are shown. ρ_{xy} in the normal state is positive, linearly increases

FIG. 2. Temperature dependence of the Hall resistivity in Hg-1212 films. The double sign reversal is clearly observed below $H=2$ T.

FIG. 3. Tangent of Hall angle $(tan Θ)$ in Hg-1212 films. Peak locations shift to the lower temperature while the dip is located at the constant temperature \sim 116 K. The linear field dependence of $\tan \Theta$ was observed in the region above T^* which is marked by an arrow. Inset shows the field dependence of tan Θ as a function of temperature, in which the data point (open circle) at $106 K$ and $1 T$ indicates nonlinear behavior below *T**.

with the applied field, and only weakly depends on temperature. For the magnetic field $<$ 2 T, along with the temperature decrease below T_c , ρ_{xy} becomes negative showing a negative dip. Higher fields drive the dip to positive. On further cooling, ρ_{xy} becomes positive with a peak at a low temperature and then gradually decreases to $T_{c,\text{zero}}$. The general trends of these results, including the double sign reversal, are similar to the earlier reports on Bi-2212 $(Ref. 11)$ and Tl- 2212 (Refs. 12 and 13) samples. However, the field dependence of the location of positive peak shows a different behavior. As the field increases, the peak position shifts to lower temperature while it is located at a constant temperature for Bi-2212 and Tl-2212 systems. Furthermore the dip is deeper than Tl-2212 and Bi-2212 systems.

The temperature dependence of the tangent of the Hall angle (tan Θ) is shown in Fig. 3. As the temperature decreases, a dip at higher temperature (T_d) and a peak at lower temperature (T_p) are observed. Even though T_d is located near the constant temperature of 116 K, T_p significantly shifts to lower temperature with increasing fields, which is different from the reported data in highly anisotropic Bi- and Tl-based superconductors^{11,12} where T_p is located at a constant temperature as fields increase. In the flux-creep region $(T \leq T_n)$, tan Θ shows a complicated field and temperature dependence. In the region of flux flow $(T>T_d)$, however, the regular displacement between curves of tan Θ reflects the linear dependence of tan Θ on field and their broad features are similar to Bi-2212 (Ref. 11) and Tl-2212.¹²

According to Samoilov¹¹ and Hagen, Lobb, and Greene,¹² in the strongly anisotropic systems, tan Θ (*H*,*T*) consists of two terms as follows;

$$
\tan \Theta(H, T) = b(T)H + a(T),\tag{1}
$$

where $b(T)$ is weakly dependent on the temperature and related to the normal-state Hall effect in the vortex core, while $a(T)$ is strongly temperature dependent. The inset of Fig. 3 is clearly in agreement with this feature. However, $\tan \Theta$

FIG. 4. Temperature dependence of the Hall conductivity σ_{xy} of Hg-1212 films. Inset is magnified at the temperature region of 100– 130 K for the sake of clarity.

cannot be linearly fitted below the temperature *T** indicated as an arrow in Fig. 3. For example, the data point (open circle) at 106 K and 1 T in the inset is no longer linear with the applied field. Above T^* , the normal-state Hall effect in the vortex core is still dominant, this fact implies that the pinning is negligibly weak compared to the driving Lorentz force. In other words, pinning seems to play an important role below *T**. In case of the highly anisotropic systems such as Bi-2212 (Ref. 11) and Tl-2212,¹² tan Θ deviates from the linear field dependence below T_p which is much lower than *T** as indicated in Fig. 3.

Based upon the time-dependent Ginzburg-Landau theory, Dorsey and co-workers $17-19$ and Kopnin, Ivlev, and Kalatsky20 proposed theoretical results of the mixed-state Hall effects. The Hall conductivity σ_{xy} can be described by the sum of two contributions:

$$
\sigma_{xy} = C_1/H + C_2H,\tag{2}
$$

where the coefficients C_1 and C_2 are related to the motion of vortices and quasiparticles, respectively. Both C_1 and C_2 are independent of fields, but are expected to depend on the temperature. If C_1 and C_2 have opposite signs, then the Hall effects can change sign as H is varied. Figure 4 shows the plot of the temperature dependence of the Hall conductivity as a function of magnetic field. The inset in Fig. 4 is magnified at the temperature region of 100–130 K for the sake of clarity. To test the validity of Eq. (2) , we fit our data using the equation $\sigma_{xy}H = C_1 + C_2H^2$. The plot of $\sigma_{xy}H$ vs H^2 is shown in the inset of Fig. 5. In making these fits, we have excluded the 1 T data since these data are far from a linear behavior. Higher field data show a good fit to the linear behavior. The temperature dependence of the intercept C_1 and the slope C_2 obtained from this linear fit is shown in Fig. 5. As the temperature decreases from T_c , C_1 becomes negative, and then reaches a minimum at 115 K. On further cooling, C_1 abruptly increases toward positive, while C_2 monotonously increases with decreasing temperature. According to Eq. (2) , the sign of mixed-state Hall effect is decided by the competition between C_1 and C_2 . In the region of negative

FIG. 5. Temperature dependence of intercept C_1 (solid square) and slope C_2 (open circle) of Hg-1212 films. Inset shows $\sigma_{xy}H$ vs H^2 , higher field data show a good fit to the linear behavior.

 C_1 , and if C_1 dominates C_2 , the negative Hall sign can take place. However, the reason that the negative Hall behavior due to the vortex motion can happen with varying temperature still remains as an unsolved problem. We expect that the theory^{18–20} can be extended to yield the temperature dependence of the coefficients C_1 and C_2 .

A plot of the scaling behavior between ρ_{xy} and ρ_{xx} is shown in Fig. 6, for the fields of 1, 2, 4, and 5.5 T. Since ρ_{xy} data below 2 T have a negative value in a certain temperature region, we have plotted using the absolute value of ρ_{xy} . The exponent β in the power law, $\rho_{xy} = A \rho_{xx}^{\beta}$, can be deduced from the slope of the solid lines in Fig. 6. The field dependence of the scaling behavior, which is different from the reported data on Bi-2212 (Ref. 11) and Tl-2212,¹² was clearly observed. The scaling exponent changes from 1.5 ± 0.1 to 1.9 \pm 0.1, as the fields increase from 1 to 5.5 T. This result is in good agreement with our earlier data¹⁵ on the twinned YBCO crystals without columnar defects, in which β changes from $\beta \sim 1.5 \pm 0.1$ to $\beta \sim 2.0 \pm 0.1$ as fields increase. This observation implies an important physical meaning of the Hall scaling behavior as discussed below.

Considering the force balance equation for a stationary moving vortex, Vinokur *et al.*²³ proposed a universal scaling law of $\rho_{xy} \propto \alpha \rho_{xx}^2$, where $\alpha = \eta \tan \Theta$ and η , the usual viscous coefficient, is independent of any vortex state in the presence of the quenched disorder and thermal noises in the mixed-state of a type-II superconductor. They showed that the scaling exponent should be 2 regardless of the pinning in TAFF region, where α is a slowly varying function of temperature. On the other hand, Wang, Dong, and $\text{Ting}^{24,25}$ (WDT) recently developed a general theory of the flux motion including both pinning-induced backflow and thermal fluctuations in the force balance equation. The additional transverse term $\mathbf{F}_p \times \mathbf{n}$ with \mathbf{F}_p being the pinning force and **n** being a unit vector in the direction of magnetic field, is produced by backflow current inside the normal core. Their main result is summarized by

$$
\rho_{xy} = \frac{\beta_0 \rho_{xx}^2}{\Phi_0 B} \left\{ \eta (1 - \overline{\gamma}) - 2 \overline{\gamma} \Gamma(\nu_L) \right\},\tag{3}
$$

where $\beta_0 = \mu_m H_{c2}$ with μ_m being the mobility of the charge carrier and H_{c2} being the upper critical field,

FIG. 6. Scaling behavior, $\rho_{xy} = A \rho_{xx}^{\beta}$, between ρ_{xy} and ρ_{xx} in Hg-1212 films. The scaling exponent β significantly increases from β =1.5±0.1 to 1.9±0.1 with increasing field. The solid lines are power laws, $\beta=1.5$ and $\beta=1.9$, for the sake of clarity.

 $\overline{\gamma} = \gamma (1 - \overline{H}/H_{C2})$ with \overline{H} being the average field over the core and γ as the parameter describing the contact force on the surface of core, which depends on temperature in the following way: $\gamma \sim 0$ for $\zeta / \zeta \ll 1$ and $\gamma \sim 1$ for $\zeta / \zeta \ll 1$ with *l* as the mean free path of the carrier, and $\Gamma(v_l)$ is the coefficient of the time average of pinning force $\langle \mathbf{F}_p \rangle = -\Gamma(v_L) \mathbf{v}_L$. At the region of relatively high temperatures $(\gamma \sim 1)$, Eq. (3) predicts a negative dip in ρ_{xy} if the field is low enough and the pinning is relatively strong, that is, a double sign reversal can be observed. As the field increases, the negative dip of can be observed. As the field increases, the negative dip of ρ_{xy} changes to positive since $\overline{\gamma}$ decreases with increasing field. In the relatively strong pinning system like Hg-1212, the depth of the dip is deeper than weak pinning systems such as Bi-2212 (Ref. 11) and Tl-2212.¹² The data shown in Fig. 2 are in good agreement with this picture.

According to Eq. (3) , there are two distinct regimes for the Hall scaling behavior in the region of γ -1. For the weak pinning regime, $\Gamma(v_L) \ll \eta H/H_{c2}$ in relatively high fields, Eq. (3) becomes $\rho_{xy} \sim A \rho_{xx}^2$, having the same scaling exponent β =2 as the model by Vinokur *et al.*²³ But for the strong nent β =2 as the model by Vinokur *et al.*²³ But for the strong pinning regime, where $\Gamma(v_L)$ $\gg \eta \overline{H}/H_{c2}$ in low fields, then β is no longer 2. Since $\Gamma(v_L) \sim v_L^{-1/2}$ in the strong pinning case,²⁷ the scaling behavior modifies to $\rho_{xy} \sim A \rho_{xx}^{1.5}$. In the intermediate case between two limiting regimes, β can vary from 1.5 to 2.0. Within the context of the WDT model, the result of $\beta=1.5\pm0.1$ for the field of 1 T corresponds to the theoretical estimation of 1.5 of the strong pinning case, while the result of $\beta=1.9\pm0.1$ for the field of 5.5 T corresponds to the weak pinning case. The present results suggest that the

pinning strength decreases with increasing field, which is consistent with the observed field dependence of the pinning energy.²⁶ This result is also consistent with our earlier work on the twinned YBCO crystals.¹⁵

At this point, we discuss the previous work on the Hall scaling behavior. Budhani, Liou, and Cai^{14} reported the Hall scaling in Tl-2223 films. If we get the scaling exponent β from Fig. 3 in Ref. 14, a log-log plot of ρ_{xy} vs ρ_{xx} for the sample without columnar defects, the 1 T data can be better fitted with the scaling exponent of 1.5 rather than 1.85 as the authors claimed, while the 7 T data are well fitted with 1.85. Thus we argue that their results are consistent with the present data as well as our earlier work in YBCO crystals. However, the present result seems to be inconsistent with the earlier work on Bi-2212 crystals reported by Samoilov.¹¹ Based on the Hall scaling result in which β is independent of the field up to 5 T, Samoilov concluded that the Hall scaling behavior is universal in the TAFF regime. But the Bi-2212 crystal may not be an appropriate material to investigate the role of pinning in different fields, because its pinning is the weakest among Hg, Tl, and Bi compounds and it is weakly field dependent as reported by Zavaritsky, Samoilov, and Yurgens,²⁸ in which the pinning potential decreases slightly from 35 to 56 meV with increasing fields from 0.3 to 5 T.

CONCLUSIONS

We have shown the double sign reversal in the Hall resistivity ρ_{xy} and strong pinning-strength dependence of its scaling behavior in the mixed state of the high-quality Hg-1212 films. The magnetic-field dependence of the tangent of the Hall angle was observed to be nonlinear in the flux-creep region, different from the results observed in Bi- and Tlbased compounds. At the higher field region, the Hall conductivity σ_{xy} has been well fitted to $\sigma_{xy}H = C_1 + C_2H^2$, where C_1 and C_2 are field independent. The scaling exponent β in $\rho_{xy} = A \rho_{xx}^{\beta}$ increased from 1.5±0.1 to 1.9±0.1 as fields increased from 1 to 5.5 T. These results are consistent with the recent theory²⁴ including both the backflow effects due to pinning and thermal fluctuations.

ACKNOWLEDGMENTS

The authors are very grateful to K. W. Wong for useful discussions and to the Midwest Superconductivity Inc. for the support of materials and various experimental facilities. This work is supported partially by the University of Kansas GRF fund and NSF EPSCoR fund. D.H.K. is supported by KOSEF 951-0209-044-2 and Korea MOE through BSRI-95- 2437.

- *Present address: Texas Center for Superconductivity at the University of Houston, Houston, TX 77204. Electronic address: wnkang@bayou.uh.edu
- ¹S. N. Putilin, E. V. Antipov, O. Chmaissen, and M. Marezio, Nature (London) 362, 226 (1993).
- $2A$. Schilling, M. Cantoni, J. D. Guo, and H. R. Ott, Nature (London) 363, 56 (1993).
- 3 C. K. Subramanian, M. Paranthman, and A. B. Kaiser, Phys. Rev. B 51, 1330 (1995).
- ⁴ J. M. Harris, H. Wu, N. P. Ong, R. L. Meng, and C. W. Chu, Phys. Rev. B 50, 3246 (1994).
- ⁵Z. J. Huang, Y. Y. Xue, R. L. Meng, and C. W. Chu, Phys. Rev. B 49, 4218 (1994).
- 6A. Schilling, O. Jeandupeux, J. D. Guo, and H. R. Ott, Physica C **216**, 6 (1993).
- 7 S. H. Yun and J. Z. Wu, Appl. Phys. Lett. **68**, 862 (1996).
- 8 L. Krusin-Elbaum, C. C. Tsuel, and A. Gupta, Nature (London) **373**, 679 (1995).
- ⁹ S. J. Hagen, C. J. Lobb, R. L. Greene, M. G. Forrester, and J. H. Kang, Phys. Rev. B 41, 11 630 (1990).
- ¹⁰ J. Luo, T. P. Orlando, J. M. Graybeal, X. D. Wu, and R. Muenchausen, Phys. Rev. Lett. **68**, 690 (1992).
- 11 A. V. Samoilov, Phys. Rev. Lett. **71**, 617 (1993).
- ¹²S. J. Hagen, C. J. Lobb, and R. L. Greene, and M. Eddy, Phys. Rev. B 43, 6246 (1991).
- 13A. V. Samoilov, Z. G. Ivanov, and L.-G. Johansson, Phys. Rev. B 49, 3667 (1994).
- 14R. C. Budhani, S. H. Liou, and Z. X. Cai, Phys. Rev. Lett. **71**, 621 $(1993).$
- 15W. N. Kang, D. H. Kim, S. Y. Shim, J. H. Park, T. S. Hahn, S. S. Choi, W. C. Lee, J. D. Hettinger, K. E. Gray, and B. Glagola, Phys. Rev. Lett. **76**, 2993 (1996).
- 16A. W. Smith, T. W. Clinton, C. C. Tsuei, and C. J. Lobb, Phys. Rev. B 49, 12 927 (1994).
- 17 A. T. Dorsey, Phys. Rev. B 51, 8376 (1992).
- ¹⁸ S. Ullah and A. T. Dorsey, Phys. Rev. B 44, 262 (1991).
- 19 R. J. Troy and A. T. Dorsey, Phys. Rev. B 47, 2715 (1993).
- 20 N. B. Kopnin, B. I. Ivlev, and V. A. Kalatsky, J. Low Temp. Phys. 90, 1 (1993).
- 21V. B. Geshkenbein and A. I. Larkin, Phys. Rev. Lett. **73**, 609 ~1994!; D. M. Ginsgerg and J. P. Manson, Phys. Rev. B **51**, 515 $(1995).$
- 22A. T. Dorsey and M. P. A. Fisher, Phys. Rev. Lett. **68**, 694 $(1992).$
- 23V. M. Vinokur, V. B. Geshkenbein, M. V. Feigel'man, and G. Blatter, Phys. Rev. Lett. **71**, 1242 (1993).
- 24Z. D. Wang, J. Dong, and C. S. Ting, Phys. Rev. Lett. **72**, 3875 $(1994).$
- ²⁵ Z. D. Wang and C. S. Ting, Phys. Rev. Lett. **67**, 3618 (1991); Phys. Rev. B 46, 284 (1992).
- 26 B. W. Kang, W. N. Kang, S. H. Yun, and J. Z. Wu (unpublished); S. Y. Ding, J. Li, H. M. Shao, J. W. Lin, C. Ren, and X. X. Yao, Phys. Rev. B 53, 900 (1996).
- 27V. M. Vinokur, M. V. Feigel'man, V. B. Geshkenbein, and A. I. Larkin, Phys. Rev. Lett. **65**, 259 (1990).
- 28N. V. Zavaritsky, A. V. Samoilov, and A. A. Turgens, Physica C **180**, 417 (1991).
- ²⁹T. Matsuura and Itozaki, Appl. Phys. Lett. **59**, 1236 (1991).
- 30 A. Nishida and K. Horai, Solid State Commun. **86**, 447 (1993).