## Hall Effect in  $YBa_2Cu_3O_7 - \delta$  in the Limit of Free Flux Flow

Milind N. Kunchur, David K. Christen, and Charles E. Klabunde Solid State Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6057

Julia M. Phillips

AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, New Jersey 07974

(Received 29 November l993)

The Hall effect of an YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>-<sub>6</sub> epitaxial film was studied using high pulsed current densities *J* to suppress flux pinning and reveal intrinsic behavior in the free-flux-flow limit. With increasing  $J$  there is an enhancement of the anomalous sign reversal, inconsistent with mechanisms based on pinning and inhomogeneity. At high H and J the Hall angle  $\alpha$  appears to decompose into  $\alpha = \alpha_M + \alpha_n$ ;  $\alpha_M(T)$  saturates to a negative value independent of H and J, and  $\alpha_n$  ( $\alpha$ H) has the normal-state value. The results are consistent with time-dependent Ginzburg-Landau calculations.

PACS numbers: 74.25.Fy, 74.60.Ec, 74.60.6e

The Hall effect in the mixed state of high- $T_c$  superconductors is a topic of great current interest and one surrounded by considerable controversy and confusion. Central to the controversy is the observed sign reversal of the Hall angle  $\alpha = \rho_{xy}/\rho_{xx}$  as the system enters the superconducting state [1-3]. Details of the sign reversal tend to be sample and field dependent, which has led to the proposal of models based on disorder. In particular it has been shown that a negative  $\alpha$  can result from the backflow current caused by pinning [4], the guiding of vortices [5], and from inhomogeneous percolating current flow [5,6]. The present work represents the first measurement of the Hall effect over an extended current-density range, where high current densities overcome pinning and bring the behavior closer to free flux flow (FFF) [7]. The high-J measurement provides direct information on the evolution of a transport quantity (in this case  $\alpha$ ) as pinning is systematically suppressed and the current flow pattern becomes more uniform. As demonstrated here, under these conditions the observations are inconsistent with the above three mechanisms. In addition the results seem to argue against a model based on spontaneous basal-plane components of vortex lines due to thermal fluctuations [8]. Instead the findings confirm an intrinsic mechanism, and are in good agreement with timedependent Ginzburg-Landau (TDGL) calculations that account for the Magnus force on the body of the vortex [9-11].

The sample was a  $c$ -axis oriented epitaxial film of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  on a (100) LaAlO<sub>3</sub> substrate. The film was deposited and postannealed by means of the  $BaF<sub>2</sub>$ process, described elsewhere [12]. The precise stoichiometry and postannealing conditions were chosen to produce films that were relatively defect-free and had single-crystal-like quality [7,13]. Measurements were made on a patterned bridge of dimensions  $(\ell \times \ell \times w)$ : 100 nm  $\times$ 3 mm $\times$ 100  $\mu$ m, with narrow Hall leads connected to the two sides of the bridge at the center. Current leads had contact resistances of about 50  $\mu \Omega$ .

The sample resistance was measured by applying rectangular current pulses 6  $\mu$ s wide with a duty cycle of 1 part in 10<sup>5</sup>. Longitudinal  $(V_{xx})$  and transverse  $(V_{xy})$ voltages were preamplified and then displayed and measured on a digital storage oscilloscope. The preamplification stage had a gain of 10000 and a common-mode rejection (CMR) of 107 dB at  $\sim$  1 MHz. By measuring the sample well above  $T_c$  (where there is no intrinsic current dependence) with both the high pulsed currents (PC) and low-value continuous dc (CDC) currents, it was verified that the PC measurement had an accuracy of (1-2)%. Similarly the apparatus was tested by measuring a network of resistors simulating the Hall signal (10 m $\Omega$  sandwiched between two 50  $\Omega$  resistors). The slight rise in temperature that occurs at the highest currents can be accurately determined and subtracted to obtain the corrected temperatures:  $T = T_{sink} + pR_{th}$ , where  $T_{sink}$ is the temperature of the sample block,  $p$  is the power dissipation density  $(p = \rho J^2)$ , and  $R_{th}$  is the total thermal resistance between film and sample block. Details about this procedure, the measurement and calculation of  $R_{th}$ , and other information about the technique are published elsewhere [7,14,15].

The procedure for taking data was to fix the magnetic field  $[H]$  axis (z direction) to within 0.5°] and current  $[along bridge length (x direction)]$  and slowly sweep the temperature down from above  $T_c$  (typically over 8 h). The field direction was then reversed and the sweep repeated. Data were measured at 50-150 mK intervals and were found to be completely reproducible at other sweep rates (3-16 <sup>h</sup> per sweep) and also for the opposite temperature-sweep direction (i.e., while warming up). Similarly no hysteresis was observed with respect to changes in  $H$  and  $J$ . The Hall resistivity is obtained from the transverse electric field  $E_y^{\pm}$  measured for the two magnetic-field directions by  $\rho_{xy} = (E_y^+ - E_y^-)/2J$ . As commonly observed by others there is a nonzero offset resistivity  $\rho_{\text{off}} = (E_y^+ + E_y^-)/2J$  associated with Hall lead mismatch.  $\rho_{off}$  roughly scales with the longitudinal resis-

tivity  $\rho_{xx}$  [Fig. 1(a)] with a factor ( $\sim$ 62) that is independent of  $H$ ,  $T$ , and  $J$ . This is uncharacteristic of conventional Hall measurements done at low J where  $\rho_{\text{off}}$  and  $\rho_{xx}$  often have very different dependencies on T, presumably due to temperature-sensitive percolating current paths [5,6]. Figure 1(a) also compares  $\rho_{xx}$  measured with CDC  $(J=5\times10^3 \text{ A/cm}^2)$  and PC  $(J=3\times10^5 \text{ A/m})$  $\text{cm}^2$ ). The overlap above the transition serves to show the consistency between the two methods and that there is negligible heating, despite very different dissipation levels  $[3 \times 10^3 \text{ W/cm}^3$  (CDC) and  $1 \times 10^7 \text{ W/cm}^3$  (PC) at  $T = 100$  K]. The departure at low temperatures is mainly because pinning is more influential in restricting Aux motion at lower J. Current-induced pair breaking also modifies the behavior at high  $J$ , but that effect is only a fraction of a kelvin. Figure  $1(b)$  shows the Hall angle  $\alpha$  at three current densities: 5 (CDC), 300 (PC), and 712 (PC) kA/cm<sup>2</sup>. Again the overlap above  $T_c$  shows that the high-pulsed-current measurement is in good agreement with the conventional low-current one. Spurious transverse voltages that can arise from the Nernst and Seebeck thermoelectric effects due to possible temperature gradients were calculated to be negligible compared with the actual Hall signals.

Figure 2(a) shows  $\alpha/H$  plotted against T for different H, measured at low current (CDC). Since  $\alpha_n \propto H$ , the



FIG. l. (a) Comparison between longitudinal resistivity (H  $=$  4 T) measured by the PC  $(J=3\times10^5 \text{ A/cm}^2)$  and CDC  $(J = 5 \times 10^3$  A/cm<sup>2</sup>) methods (upper curves) and between the longitudinal and Hall-offset resistivities  $(J=3 \times 10^5 \text{ A/cm}^2)$ (lower curves). (b) Hall angle measured at three different current densities. The method of measurement and the value of  $J$  (in MA/cm<sup>2</sup>) are indicated for each curve.

quantity  $\alpha/H$  is plotted in favor of  $\alpha$  so that the curves scale together in the normal state. The data at low fields show the controversial and much discussed sign reversal. Notice that the behavior undergoes both qualitative and quantitative changes as the field is increased. The negative minimum is absent for  $H = 6$  and 8 T. Also the position of the minimum shifts with field. It was these types of observations, in part, that led to early speculations that the sign reversal may be caused by pinning or inhomogeneities. (The decrease in magnitude as  $H$  is increased is partly an artifact of the way the data are plotted, i.e.,  $\alpha/H$  rather than just  $\alpha$ .) Figure 2(b) shows a similar set of curves measured at much higher (PC) currents. The behavior is dramatically altered. The negative minimum is considerably enhanced [the systematics of which can be seen for  $H = 4$  T in Fig. 1(b)] and its position has become less field dependent. Also a minimum is now seen to be a



FIG. 2. (a) Temperature dependence of  $\alpha/H$  for indicated fixed fields, measured at  $J=5$  kA/cm<sup>2</sup> (CDC). (b) Similar data measured with pulsed current at  $J=0.7$  MA/cm<sup>2</sup> (H) =0.5, 1, 2, and 4 T),  $J=1.1$  MA/cm<sup>2</sup> ( $H=6$  T), and  $J=1.5$  $MA/cm<sup>2</sup>$  ( $H=8$  T). (c) Magnus-force component of the Hall angle  $\alpha_M = \alpha - \alpha_n$ , for the same data shown in (b). Here,  $\alpha_n = 21.9 \times H/T^2$  is the observed T- and H-dependent normalstate contribution. The dashed line is a fit by Eq. (1) for  $B=8$ T.

universal feature—present even at 8 T—superimposed on a positive background that appears to be a continuation of the normal-state Hall angle. These observations all point towards an intrinsic mechanism. Figure 2(c) shows the same data as in (b) with the normal-state background subtracted, i.e.,  $\alpha_M = \alpha - \alpha_n$ , where  $\alpha_n(T) = 21.9 \times H/T^2$ is a fit to the normal-state data  $(H$  is in tesla and  $T$  in kelvin). Displayed in this way, the data show more clearly the sign-reversing field-independent component  $\alpha_M$ , and elucidate its universal nature. Data at all fields show a common positive slope just below  $T_c$ . As the temperature is lowered, pinning sets in, freezing out both the Hall and longitudinal signals. This effect occurs at higher temperatures for lower fields. In the limit of high  $H$  and  $J$ , the  $\alpha_M(T)$  curves measured at different H and J appear to converge to a common behavior with weak temperature dependence; note that the 4, 6, and 8 T curves are at different currents: 0.71, 1.1, and 1.5  $MA/cm<sup>2</sup>$ , respectively. This qualitative behavior, where there is an apparent decomposition of  $\alpha$  into two components, one field independent and the other proportional to  $H$ , has also been observed in the  $Tl_2Ba_2CaCu_2O_8$  superconductor [2]. However, in that work the field-independent component had a complicated and nonmonotonic temperature dependence (similar to what we observe at low fields) which has been speculated [9,10] to be due to an unusual temperature dependence of the complex order-parameter relaxation time. In our data the nonmonotonic nature of  $\alpha_M(T)$  disappears at high fields and currents, showing that it is likely a pinning effect.

As mentioned above, there is a model that attempts to explain the sign reversal based on the mechanism of thermally induced vortex fluctuations [8]. The above model is specific to quasi-2D layered superconductors, while the present measurements in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$  are in a temperature regime that is essentially 3D. Moreover, if fluctuations played a principal role, the effect should depend strongly on  $T$ ,  $H$ , and  $J$ . In the present studies, however, we find that the sign-reversing component converges to a roughly  $T<sub>z</sub>$ ,  $H<sub>z</sub>$ , and J-independent behavior [Fig. 2(c)]. It seems more likely that previous experiments, all conducted at low current densities, can be explained in terms of a suppression of flux pinning by the applied field, without invoking the fluctuation argument.

Having demonstrated inconsistency with the first four models (based on pinning, flux guiding, percolation, and thermal vortex fluctuations), we now interpret the results based on recent theoretical considerations that lead to a component of flux flow that is parallel to the current [9-11]. <sup>A</sup> transverse electric field can arise from two distinct mechanisms. First, since the vortex core behaves like a cylinder of radius  $\xi$  containing normal fluid, it will give rise to a transverse electric field by the customary Hall mechanism, i.e., due to the force experienced by normal carriers in motion in a magnetic field. This Hall signal will not depend on the details of the hydrodynamics governing vortex motion but will be proportional to the

magnetic field  $H_{\text{core}}$ , within the vortex core. For  $H \gg H_{c1}$ (the case here),  $H_{\text{core}} \approx B \approx H$ , so that for free flux flow we expect this component to have the same magnitude  $(a_n)$  and sign as in the normal state. The second origin of  $E_{xy}$  is due to the Magnus force on the "body" of the vortex (as opposed to the normal fluid within the core). This will force vortex motion which will in general subtend a finite angle to  $B \times J$ , giving rise to a transverse electric field. Note, however, that this mechanism is not a "Hall effect" in the usual sense of moving charges being forced by a magnetic field. This Magnus force also exists in neutral superfluids and does not have an a priori dependence on  $H$ . Thus it is possible to have a contribution to the Hall effect that is actually field independent. This can explain the observed decomposition of  $\alpha(T)$  into a normal-state-like field-dependent component  $\alpha_n(T)$  and a field-independent hydrodynamic component  $\alpha_M(T)$ . One difficulty with earlier phenomenological hydrodynamic models, such as by Bardeen and Stephen (BS) [16] and Nozieres and Vinen (NV) [17], is that they always predicted the mixed-state Hall effect to have the same sign as the normal state. Hagen et al. [2] generalized the phenomenological hydrodynamic approach by combining the driving force and damping terms of the BS and NV models in an *ad hoc* manner. In this way they were able to show that it is possible to obtain a negative sign.

Recent calculations  $[9-11]$  of the Magnus force within the TDGL framework can lead to a negative sign for  $\alpha_M$ without any additional assumptions. Furthermore the complete expression [10] for  $\alpha$  valid for our range of T and  $H$ ,

$$
a = a_n(T) + g\left[\frac{H_{c2}(T) - B}{H_{c2}(T)}\right],
$$
 (1)

contains the decomposition that we have observed and contains an " $\alpha_M(T)$ " term that saturates to a T- and Hindependent value, when  $H_{c2}(T)$  becomes large compared to  $B$ , qualitatively similar to what we have observed. The dashed line in Fig. 2(c) is a fit by Eq.  $(1)$ . In Eq.  $(1)$ , g is proportional to the ratio of the real and imaginary parts of the order-parameter relaxation time and depends on the energy derivative of the density of states averaged over the Fermi surface. At present a calculation of the complex relaxation time based on microscopic theory does not exist, so it is not possible to make a quantitative comparison with the data. It should be noted that because g depends on band structure [10,18] and other microscopic details, there can be sample-to-sample variability in both the magnitude and sign of the Hall effect—as in fact found in the literature. One must therefore be cautious in interpreting experiments that study the effects of artificially introduced defects [3] or oxygen concentration [19]. In addition to altering pinning, these experiments can alter the electronic structure and influence the intrinsic mechanism itself. In this respect the use of high J to suppress pinning is a more controlled experiment for

isolating the role of pinning. Similarly the high-J experiment has certain advantages over transport measurements that have been done in intrinsically weak-pinning materials such as  $T_2Ba_2CaCu_2O_8$  [2]. Because of the role played by electronic structure, just discussed, it is not possible to apply the results in one material to another. Furthermore, these weak-pinning materials have a more pronounced 2D layered structure which introduces another factor and adds complexity (e.g., vortex fluctuations) to the interpretation.

In conclusion, the Hall effect in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> -  $\delta$  was measured at high-pulsed-current densities to reveal the intrinsic behavior in the FFF limit. The results show that traditional measurements made at low CDC current densities can depart dramatically from intrinsic behavior as a result of pinning. This work also contains important information about the nature of the vortex core. We found that the normal-state field-dependent component of  $\alpha$ (i.e.,  $\alpha_n$ ) is consistent with a direct extrapolation of the behavior above  $T_c$ . In previous work [7] we found similar characteristics for the longitudinal resistivity under FFF conditions. There the Bardeen-Stephen result was found to be satisfied if  $\rho_n$  within the core was taken as the extrapolation from above  $T_c$ . Thus the vortex core in high- $T_c$  superconductors seems to be conventional (with respect to both  $\rho_{xx}$  and  $\rho_{xy}$ ) in the temperature and magnetic field ranges studied, unaffected by energy-level quantization [20] or changes in the quasiparticle damping rate upon entry into the superconducting state [21,22].

The authors acknowledge useful discussions with E. C. Jones, C. J. Lobb, A. T. Dorsey, N. P. Qng, R. C. Dynes, G. Giuliani, and V. G. Kogan, and would like to thank S. Y. Hou for help with the sample preparation. Research was sponsored by the Division of Materials Sciences, U.S. Department of Energy under Contract No. DE-AC05- \$4OR21400, with Martin Marietta Energy Systems, Inc. Partial support for the program was administered by the Oak Ridge Institute for Science and Education.

- [1] M. Galffy and E. Zirgiebl, Solid State Commun. 68, 929 (1988); Y. lye, S. Nakamura, and T. Tamegai, Physica (Amsterdam) 159C, 616 (1989); K. C. Woo et al., Physica (Amsterdam) 162-164C, 1011 (1989); S. M. Artemenko er a/. , Phys. Lett. A 13\$, 428 (1989); T. R. Chien er al., Phys. Rev. Lett. 66, 3075 (1991); J. P. Rice et al., Phys. Rev. B 46, 11050 (1992).
- [2] S. J. Hagen, C. J. Lobb, R. L. Greene, and M. Eddy Phys. Rev. B43, 6246 (1991).
- [3] R. C. Budhani, S. H. Liou, and Z. X. Cai, Phys. Rev. Lett. 71, 621 (1993}.
- [4] Z. D. Wang and C. S. Ting, Phys. Rev. B 46, 284 (1992); Phys. Rev. Lett. 67, 3618 (1991).
- [5] E. C. Jones et al. (to be published).
- [6] R. C. Dynes (private communication).
- [71 M. N. Kunchur, D. K. Christen, and J. M. Phillips, Phys. Rev. Lett. 70, 998 (1993).
- [8] J. M. Harris et al., Phys. Rev. Lett. 71, 1455 (1993).
- [9] A. T. Dorsey, Phys. Rev. B 46, 8376 (1992).
- [10] N. B. Kopnin et al., J. Low Temp. Phys. 90, 1 (1993).
- [11] R. J. Troy and A. T. Dorsey, Phys. Rev. B 47, 2715 (1993).
- [12] M. P. Siegal et al., J. Appl. Phys. 68, 6353 (1990); D. J. Carlson et al., J. Mater. Res. 5, 2797 (1990).
- [13] M. P. Siegal et al., Appl. Phys. Lett. 60, 2932 (1992).
- [14] M. N. Kunchur et al., Phys. Rev. Lett. 72, 752 (1994).
- [15] M. N. Kunchur, Bull. Am. Phys. Soc. 38, 459 (1993); in Proceedings of the Workshop on the Statics and Dynamics of Vortices, LT20 satellite conference, 1993 (to be published).
- [16]J. Bardeen and M. J. Stephen, Phys. Rev. 140, A1197 (1965).
- [17] P. Nozieres and W. F. Vinen, Philos. Mag. 14, 667 (1966).
- [18] H. Fukuyama et al., Prog. Theor. Phys. 46, 1028 (1971).
- [19] E. C. Jones et al., Phys. Rev. B 47, 8986 (1993).
- [20] C. Caroli et al., Phys. Lett. 9, 307 (1964); A. L. Fetter and P. C. Hohenberg, in Superconductivity, edited by R. D. Parks (Marcel Dekker, New York, 1969), p. 889.
- [21] W. N. Hardy et al., Phys. Rev. Lett. 70, 3999 (1993).
- [22] Y. Matsuda (to be published).