## Hall-conductivity sign reversal and fluctuations in $YBa_2Cu_3O_{7-\delta}$ films

Wu Liu, T. W. Clinton, A. W. Smith, and C. J. Lobb

Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, Maryland 20742

(Received 3 January 1997)

We measured the longitudinal and Hall resistive transitions of  $YBa_2Cu_3O_{7-\delta}$  films in applied fields up to 9 T. The longitudinal fluctuation conductivities obey the scaling behavior of the three-dimensional Hartree fluctuation theory of Ullah and Dorsey. The fluctuation Hall conductivities do not scale. Our analysis shows that there are substantial contributions from the Maki-Thompson (MT) process as well as the Aslamazov-Larkin (AL) one. Using an *ansatz* suggested by the microscopic theory of Fukuyama *et al.*, we separated the AL term from the total fluctuation conductivity. The AL Hall conductivity follows a scaling law, and the AL and MT terms have opposite sign. The sign of the AL term is consistent with the sign change in the flux-flow regime, as predicted by theory. At low fields and temperatures just above  $T_c(H)$ , the AL term dominates the MT term, therefore causing the total Hall-conductivity sign change. [S0163-1829(97)11117-1]

The sign reversal of the Hall conductivity in the mixed state of both high- $T_c$  and low- $T_c$  superconductors has been one of the most intriguing and controversial transport phenomena.<sup>1</sup> Despite many efforts, the mechanism responsible for the anomalous Hall effect is still unclear. Even the basic question of whether the sign change is due to the intrinsic property of vortex dynamics or some extrinsic property like pinning is unsolved. Wang et al.<sup>2</sup> propose that the backflow current caused by vortex pinning leads to the sign change. However, based on a general argument of pinning theory, Vinokur et al.,<sup>3</sup> and later Liu et al.<sup>4</sup> on a rigorous calculation argue otherwise, concluding that the sign change is not due to pinning and Hall conductivity is independent of pinning. Some experiments show Hall conductivity independent of pinning,<sup>5</sup> implying that sign change is not due to pinning. Other experiments have shown some quantitative agreement with the pinning mechanism.<sup>6</sup> There are also models based on the carrier density difference between inside and outside the vortex core,<sup>7,8</sup> as well as a microscopic calculation by van Otterlo *et al.*<sup>9</sup> which supports Ref. 8. A general approach which accommodates a sign change is the time-dependent Ginzburg-Landau (TDGL) theory,<sup>10,11</sup> but it needs microscopic information to determine certain parameters.

In this paper, we report Hall-effect measurements which are primarily in the fluctuation regime, where vortices are not present. We interpret our data in terms of fluctuation theory by Ullah and Dorsey.<sup>12</sup> We will present data from one YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (YBCO) film made by pulsed-laser deposition with thickness of 3000 Å, and  $T_c$  of 90 K [midpoint of transition region, Fig. 1(a) inset] with a transition width less than 1 K (10–90 %). We have measured several other samples, and they show similar results.

In Fig. 1 we plot longitudinal and Hall resistivities vs field for temperatures near and above the zero field  $T_c$ , where fluctuations make significant contributions to the conductivity.<sup>13–15</sup> It is clear from Fig. 1(b) that there is a sign change around 90 K, even though the longitudinal resistivities *never go to zero* as shown in Fig. 1(a). Notice also that for temperatures around 90 K and a few degrees above, the high-field Hall resistivities do not extrapolate to zero, but have negative intercepts. The importance of the fluctuation contribution to Hall-conductivity sign change has been addressed recently by Jin and Ott.<sup>16</sup>

The longitudinal and Hall conductivities can be written as  $\sigma_{xx} = \sigma_{xx}^n + \Delta \sigma_{xx}$  and  $\sigma_{xy} = \sigma_{xy}^n + \Delta \sigma_{xy}$ , where  $\sigma_{xx}^n$  and  $\sigma_{xy}^n$ 



FIG. 1. Longitudinal and Hall resistivities near and above  $T_c$ . For both plots, the temperatures corresponding to each curve at far right are listed in kelvin. Inset in (a) shows the zero-field resistive transition.

© 1997 The American Physical Society

are normal-state contributions. According to Ullah and Dorsey,<sup>12</sup> in the three-dimensional (3D) case, the fluctuation conductivities have the following behavior:

$$\Delta \sigma_{xx} = c_1 \Gamma_0^{-1} \left( \frac{T^{2/3}}{H^{1/3}} \right) \mathcal{F} \left( c_2 \left( \frac{T - T_c(H)}{(HT)^{2/3}} \right) \right), \tag{1}$$

$$\Delta \sigma_{xy} = c_1 \lambda_0^{-1} \left( \frac{T^{2/3}}{H^{1/3}} \right) \mathcal{F} \left( c_2 \left( \frac{T - T_c(H)}{(HT)^{2/3}} \right) \right), \tag{2}$$

where  $c_1$  and  $c_2$  are two constants,  $\mathcal{F}$  is a universal function,  $T_c(H)$  is the mean-field transition temperature, while  $\Gamma_0^{-1}$  and  $\lambda_0^{-1}$  are real and imaginary parts of the order-parameter relaxation rate of TDGL theory. Equations (1) and (2) are only true in the high-field (lowest-Landau-level) limit. The original derivation only included the Aslamazov-Larkin (AL) contribution, and did not include the Maki-Thompson (MT) contribution.<sup>17</sup> From Eqs. (1) and (2), we have the theoretical prediction  $\Delta \sigma_{xy} / \Delta \sigma_{xx} = \lambda_0^{-1} / \Gamma_0^{-1} = \text{const}$ , since  $\lambda_0^{-1}$  and  $\Gamma_0^{-1}$  are both constants within TDGL theory.

We obtained fluctuation conductivities by subtracting the normal-state contributions. The normal-state longitudinal resistivity can be described by  $\rho_{xx}^n(T) = [0.81T(K)]$ +8.67]  $\mu\Omega$  cm, which was obtained by fitting resistivity from 150 to 250 K. The Hall coefficient was obtained in the  $R_H(T) = [1/(0.045T(K))]$ same temperature range, +1.2)]  $\mu\Omega$  cm/T. The normal-state Hall resistivity is given by  $\rho_{yx}^n = R_H(T)B$ . Normal-state conductivities were obtained by  $\sigma_{xx}^n = \rho_{xx}^n / [(\rho_{xx}^n)^2 + (\rho_{yx}^n)^2]$  and  $\sigma_{xy}^n = \rho_{yx}^n / [(\rho_{xx}^n)^2 + (\rho_{yx}^n)^2]$ . The mean-field transition temperature  $T_c(H)$  is chosen to be  $T_c(H) = [-0.387H(T) + 88.2]$  K. This corresponds to  $-dH_{c2}(T)/dT = 2.58$  T/K. The choice of  $T_c(H)$ is based on achieving the best scaling plot for  $\Delta \sigma_{xx}$  and a linear relationship between  $T_c(H)$  and H.<sup>15,18</sup> The interpolated  $T_c(0)$  is known to not match experimental zero-field transition temperature exactly.<sup>15,18</sup>

After subtracting out the normal-state contributions, we scale the data based on Eqs. (1) and (2). As shown in Fig. 2, the longitudinal fluctuation conductivity follows the Ullah and Dorsey scaling law as observed by many groups,<sup>15,18</sup> while the fluctuation Hall conductivity does not scale. Also, we plot in Fig. 2(c)  $\Delta \sigma_{xy} / \Delta \sigma_{xx}$  vs temperature. It is strongly temperature and field dependent, as opposed to what the theory predicts. We point out that in order to make  $\Delta \sigma_{xy}$  scale, we have tried many different choices of  $T_c(H)$ , including some which sacrifice scaling of  $\Delta \sigma_{xx}$ , but all attempts failed.

This failure to scale may be due to the fact that the TDGL theory only includes AL contributions, but fails to include MT contributions. As we can see from the insets of Figs. 2(a) and 2(b), the fluctuation longitudinal conductivities monotonically increase with decreasing temperature. The fluctuation Hall conductivities first increase with decreasing temperature, then around  $T_c(H)$  start to change sign, becoming increasingly negative as temperature is lowered. This crossover might be due to competition between the two terms in fluctuation Hall conductivity.



FIG. 2. Scaling of fluctuation conductivities with field strengths of 2 to 9 T with increments of 1 T. (a) Longitudinal conductivity, which scales. (b) Hall conductivity, which does not scale. Insets show the corresponding fluctuation conductivities. (c) Ratio of  $\Delta \sigma_{xx} / \Delta \sigma_{xx}$  vs temperature.

We now turn to the microscopic theory for guidance, which includes both AL and MT terms in a Gaussian approximation. From microscopic theory by Fukuyama *et al.*,<sup>19</sup> in the 3D low-field limit,

$$\sigma_{xx} = \sigma_{xx}^{n} + \frac{e^{2}}{32\hbar\xi(0)} \left(\frac{1}{\eta^{1/2}} + \frac{4}{\eta^{1/2}}\right), \qquad (3)$$

$$\sigma_{xy} = \sigma_{xy}^{n} + \frac{e^2}{32\hbar\xi(0)} \frac{\sigma_{xy}^{n}}{\sigma_{xx}^{n}} \left(\frac{\pi\alpha}{36} \frac{1}{\eta^{3/2}} + \frac{4}{\eta^{1/2}}\right), \quad (4)$$



FIG. 3. The ratio of dimensionless fluctuation conductivity vs temperature for magnetic field from 2 to 9 T with increments of 1 T. Upper inset shows  $(\sigma_{xx} - \sigma_{xx}^n)/\sigma_{xx}^n$  and lower inset shows  $(\sigma_{xy} - \sigma_{xy}^n)/\sigma_{xy}^n$  in the same temperature range.

where  $\eta = \ln(T/T_c(H)) \approx [T - T_c(H)]/T_c(H)$ , and  $\alpha \propto N'(\epsilon_F)$ , the derivative of the density of states at the Fermi energy. In both Eqs. (3) and (4), the first fluctuation term corresponds to the AL and the second to the MT contributions. Near  $T_c(H)$ ,  $\eta^{-3/2}$  dominates  $\eta^{-1/2}$  terms, while away from  $T_c(H)$ ,  $\eta^{-3/2}$  is much smaller than  $\eta^{-1/2}$ . If  $\alpha < 0[N'(\epsilon_F) < 0]$ , it is conceivable that near  $T_c(H)$ , the AL Hall term can overpower the MT Hall term and normal-state contribution, therefore leading to a sign change.

At higher temperatures, we have

$$[(\sigma_{xy} - \sigma_{xy}^{n})/\sigma_{xy}^{n}]/[(\sigma_{xx} - \sigma_{xx}^{n})/\sigma_{xx}^{n}]$$
  
=  $(\pi \alpha/36 \eta^{3/2} + 4/\eta^{1/2})/(1/\eta^{1/2} + 4/\eta^{1/2}) \approx 4/5 = 0.8.$ 

In Fig. 3 we plot  $[(\sigma_{xy} - \sigma_{xy}^n)/\sigma_{xy}^n]/[(\sigma_{xx} - \sigma_{xx}^n)/\sigma_{xx}^n]$  vs temperature, where around 100 K the ratio is about 1.5.

As pointed out by Fukuyama *et al.*, the AL term and MT term in longitudinal conductivity, and apart from a factor of  $\sigma_{xy}^n/\sigma_{xx}^n$  the Hall MT term, all have the same temperature and field dependence. We now propose an *ansatz*: even as fluctuations become large, and the low-field Gaussian approximation must be replaced by the high-field Hartree approximation, the functionality between longitudinal AL and MT terms, as well as the Hall MT term (apart from the factor  $\sigma_{xy}^n/\sigma_{xx}^n$ ) may still remain the same, but the relative strength between them might be different. We propose that we can separate the AL Hall contribution from the total fluctuation Hall conductivity by using

$$\Delta \sigma_{xy}^{\text{AL}} \approx \sigma_{xy}^{n} \left[ \left( \frac{\sigma_{xy} - \sigma_{xy}^{n}}{\sigma_{xy}^{n}} \right) - g \left( \frac{\sigma_{xx} - \sigma_{xx}^{n}}{\sigma_{xx}^{n}} \right) \right], \quad (5)$$

where g is a numerical factor. We found that with g = 1.6and using the *same* mean-field transition temperatures  $T_c(H)$  used for  $\Delta \sigma_{xx}$  scaling, we can achieve a nice scaling for the AL Hall conductivity. This factor g = 1.6 is actually very close to what Fig. 3 suggested, 1.5, and is *twice* the low-field theoretical limit.

In Fig. 4 we perform the scaling plot of the AL and MT



FIG. 4. Scaling of AL Hall and MT Hall conductivities with field strengths of 2 to 9 T with increments of 1 T. (a) AL Hall conductivity. (b) MT Hall conductivity. Insets show the corresponding conductivities. (c) Ratio of  $\Delta \sigma_{xy}^{AL}/\Delta \sigma_{xx}$  vs temperature.

Hall conductivities obtained from Eq. (5). The AL Hall conductivity scales nicely, while the MT Hall conductivity does not. Scaling of the total fluctuation Hall conductivity fails because of the importance of the MT term. (The fact that the fluctuation longitudinal conductivity scales without separation of AL and MT contributions agrees with the microscopic calculation and our *ansatz*.) The AL and MT Hall terms shown in Fig. 4 insets are similar to those obtained by fitting low-field data in Fig. 8 of Lang *et al.*<sup>14</sup> Other experiments with low-field data have shown disagreement with MT contributions.<sup>20,21</sup> Our observation that the ratio of dimensionless fluctuation conductivity approaches 1.5 around 100 K implies that the strength of the longitudinal MT term in high field is substantially smaller than the low-field theoretical result has suggested.

As we can see from Figs. 4(a) and 4(b), insets, the AL Hall conductivity has sign opposite to the MT Hall conductivity. At certain fields and temperatures close to  $T_c(H)$ , the AL Hall term starts to dominate the MT Hall term, and  $\Delta \sigma_{xy}$  becomes negative. When this negative  $\Delta \sigma_{xy}$  is larger

in magnitude than the normal-state contribution  $\sigma_{xy}^n$ , the Hall conductivity changes sign, as shown in Fig. 3, lower inset.

At still lower temperatures, fluctuation effects will be dominated by flux-flow effects, and the sign change may be due to another mechanism. This is reflected in Fig. 4(c) where we plot  $\Delta \sigma_{xy}^{AL}/\Delta \sigma_{xx}$  vs temperature at several fields. Even though both  $\Delta \sigma_{xx}$  and  $\Delta \sigma_{xy}^{AL}$  scale using the same mean-field transition temperature  $T_c(H)$ , their ratio is *not* temperature and field independent as suggested by the theory. This ratio, however, is clearly an improvement on Fig. 2(c), where the AL and MT terms were not separated. We note that the main deviations from temperature independence come from lower temperatures and fields, where data are obviously in the flux-flow regime. The relatively weak temperature dependence of  $\Delta \sigma_{xy}^{AL}/\Delta \sigma_{xx}$  may indicate that the TDGL relaxation rates  $\Gamma_0^{-1}$  and  $\lambda_0^{-1}$ , which should be temperature independent according TDGL, are actually weakly temperature dependent.

We want to emphasize that our approach to separate the AL and MT Hall conductivity relies on our *ansatz*. Though this lacks rigorous theoretical justification, it is however a very simple approach, and there is no other reliable method available. The fact that after separation the AL Hall term

- <sup>1</sup>C. J. Lobb *et al.*, Appl. Supercond. **2**, 631 (1994), and references cited therein.
- <sup>2</sup>Z. D. Wang et al., Phys. Rev. Lett. 72, 3875 (1994).
- <sup>3</sup>V. M. Vinokur *et al.*, Phys. Rev. Lett. **71**, 1242 (1993).

<sup>4</sup>Wu Liu *et al.*, Phys. Rev. B **52**, 7482 (1995); for detailed calculations, see Wu Liu, Ph.D. thesis, University of Maryland, 1996.

- <sup>5</sup>A. V. Samoilov *et al.*, Phys. Rev. Lett. **74**, 2351 (1995). <sup>6</sup>W. N. Kang *et al.*, Phys. Rev. Lett. **76**, 2993 (1996).
- w. N. Kalig *et al.*, Fliys. Kev. Lett. **70**, 2995 (1990)
- <sup>7</sup>D. I. Khomskii and A. Freimuth, Phys. Rev. Lett. **75**, 1384 (1995).
- <sup>8</sup>M. V. Feigel'man *et al.*, Physica C **235-240**, 3127 (1994); JETP Lett. **62**, 834 (1995).
- <sup>9</sup>A. van Otterlo et al., Phys. Rev. Lett. 75, 3736 (1995).
- <sup>10</sup>A. T. Dorsey, Phys. Rev. B 46, 8376 (1992).
- <sup>11</sup>N. B. Kopnin et al., J. Low Temp. Phys. 90, 1 (1993).

scales is consistent with Ullah and Dorsey's prediction and is an indirect indication of the correctness of our *ansatz*. By separating AL and MT Hall contributions and demonstrating the scaling behavior of the AL Hall term, we have clearly identified the *cause* of the Hall conductivity sign change in the fluctuation regime. We further note that the clear occurrence of sign-reversing terms in the fluctuation regime, where there are no vortices, indicates that pinning cannot fully account for the sign reversal.<sup>2</sup>

In summary, we have analyzed our data in terms of Ullah and Dorsey's fluctuation scaling theory. The longitudinal fluctuation conductivity scales well but the Hall conductivity does not. After separating the AL and MT terms from fluctuation Hall conductivity, the AL Hall term follows the scaling law, while the MT Hall term does not. Furthermore, the AL Hall term is opposite in sign to the normal-state Hall effect. Near  $T_c$  in the fluctuation-dominated regime, the sign change is due to the AL Hall term if  $N'(\epsilon_F) < 0$ .

We are very grateful for extensive discussions with and comments by Professor A. T. Dorsey, Professor D. M. Ginsberg, and Professor W. Lang. We thank Dr. W. Jiang, Dr. C. Kwon, Dr. Q. Li, and Dr. I. Takeuchi for providing the YBCO films. This research was supported in part by the NSF under Grant No. DMR-9510464.

- <sup>12</sup>S. Ullah and A. T. Dorsey, Phys. Rev. B 44, 262 (1991).
- <sup>13</sup>J. P. Rice et al., Phys. Rev. B 44, 10158 (1991).
- <sup>14</sup>W. Lang et al., Phys. Rev. B **49**, 4209 (1994).
- <sup>15</sup>A. V. Samoilov, Phys. Rev. B 49, 1246 (1994).
- <sup>16</sup>R. Jin and H. R. Ott, Phys. Rev. B 53, 9406 (1996).
- <sup>17</sup>W. J. Skocpol and M. Tinkham, Rep. Prog. Phys. **38**, 1049 (1975).
- <sup>18</sup>U. Welp et al., Phys. Rev. Lett. 67, 3180 (1991).
- <sup>19</sup>H. Fukuyama *et al.*, Prog. Theor. Phys. **46**, 1028 (1971). The original formula given by Fukuyama *et al.* has numerical factor errors. Equations (3) and (4) are due to Alan T. Dorsey [Alan T. Dorsey (private communication)].
- <sup>20</sup>J. P. Rice and D. M. Ginsberg, Phys. Rev. B 46, 12 049 (1992).
- <sup>21</sup>K. Semba et al., Phys. Rev. Lett. 67, 769 (1991).