

## Sign reversal of the Hall effect below $T_c$ in untwinned single-crystal $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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Because of possible effects from (110) twin planes and  $a$ - $b$  anisotropy in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , measurements on untwinned crystals are desirable. We have measured the Hall effect and resistivity simultaneously in untwinned single-crystal  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  with a superconducting transition temperature  $T_c = 93$  K, with the magnetic field along the  $c$  axis. Below  $T_c$ , the measured Hall resistivity reverses sign, becoming negative over a range of temperature and magnetic field, and the location and magnitude of the minimum is similar to that measured previously by others on twinned samples. These results show that the negative Hall resistivity is not caused by twin planes inducing guided vortex motion or vortex pinning. In addition, the exponent characterizing a scaling relation of the longitudinal to the Hall resistivity has a value consistent with that measured on thin films.

### I. INTRODUCTION

Sample defects often interfere with attempts to understand the intrinsic solid-state properties of materials. One classic case is the Hall effect below  $T_c$  in type II superconductors, which can be complicated by effects associated with pinning and guiding of the vortices.<sup>1,2</sup> In high- $T_c$  superconductors such as  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ,<sup>3-7</sup>  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$ ,<sup>4</sup> and  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ ,<sup>8</sup> and in some samples of low- $T_c$  superconductors such as Nb,<sup>9</sup> V,<sup>10</sup> and In-Pb alloys,<sup>11</sup> the Hall effect exhibits an interesting reversal of sign in the superconducting state from that in the normal state. For example, in high- $T_c$  samples such as  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , for which the charge carriers in the CuO planes are hole-like and hence have a positive Hall coefficient above  $T_c$ , the Hall resistivity becomes negative over a range of magnetic field and temperature in the mixed state below  $T_c$ . One wonders, however, whether this sign reversal would occur in a homogeneous sample having no defects, or whether it is extrinsic, resulting entirely from the defects that exist in real samples.

We focus on (110) twinning, a common defect in samples of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . The crystallographic  $a$  and  $b$  axes interchange when passing across a twin plane, and the local crystal structure in the vicinity of the twin plane is distorted. (The CuO planes lie in the  $a$ - $b$  plane, and the  $b$  axis is the direction of the CuO chains.) A variety of experiments have shown that twin planes can greatly increase the pinning force on vortices at various magnetic fields and temperatures.<sup>12-17</sup> A recent calculation indicates that the sign reversal of the Hall resistivity may be associated with pinning forces.<sup>18</sup> We therefore measured the Hall effect and the resistivity in untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and compared the behavior with that of twinned samples.

### II. PROCEDURE

The untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystal investigated here is the same one used in our study of the Hall effect above

$T_c$ .<sup>19</sup> It was made in an yttria-stabilized zirconia crucible. The value of  $T_c$ , defined by the midpoint of the zero-field resistive transition, is 94.5 K. Above about 96.7 K the Hall resistivity was found to be a linear function of magnetic field. Below  $T_c$ , the field dependence of the Hall resistivity is nonlinear, as we shall see.

As in the previous study, we used a five-probe contact arrangement consisting of two large current contacts and three small voltage contacts. A uniform current density of 440 A/cm<sup>2</sup> rms at 37.8 Hz was directed along the crystallographic  $b$  axis from the reference channel of a lock-in amplifier. We denote the current direction as the  $x$  axis. The resulting in-phase voltages were measured with lock-in amplifiers referenced to the current. For measurements of the resistivity  $\rho_{xx}$  we used the first two voltage contacts, aligned along the crystal's  $b$  axis, to measure the voltage drop along the current direction. For measurements of the Hall resistivity  $\rho_{xy}$ , a high-resistance potentiometer was connected across those two voltage contacts, and we measured the voltage between the wiper of the potentiometer and the third voltage contact. This third contact was approximately aligned with the second voltage contact along the crystallographic  $a$  axis, which we denote as the  $y$  axis. Thus the voltage drop in the absence of an applied magnetic field, resulting from the slight misalignment of the voltage contacts, was small. This misalignment voltage was nulled by adjusting the potentiometer with the magnetic field off and the sample at a temperature a few degrees above  $T_c$ .

We performed magnetic-field sweeps ( $\mathbf{H}$  parallel to the  $c$  axis) with the sample at a fixed temperature. A given field sweep cycle consisted of incrementally changing the field from 0 to 7 to  $-7$  to 0 T, using an increment of 0.28 T throughout. The temperature, resistivity  $\rho_{xx}$ , and Hall resistivity  $\rho_{xy}$  were measured at each field. The measured standard deviation of the temperature was typically below 3 mK during each field cycle. Such tight temperature control was essential, since  $\rho_{xx}$  and  $\rho_{xy}$  are strongly temperature dependent in the temperature and field ranges of interest. Other than the negligible effects re-

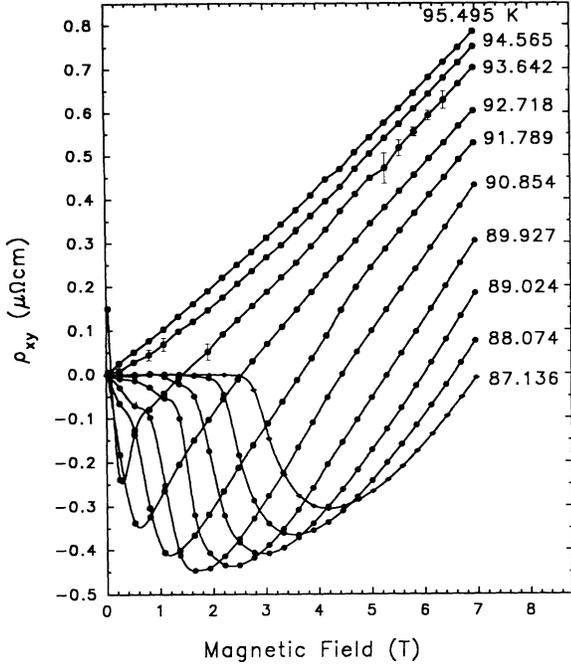


FIG. 1. Hall resistivity  $\rho_{xy}$  vs magnetic field at several fixed temperatures near  $T_c$ . The error bars represent standard deviations of voltage readings, which are significant only for a few data points.

sulting from temperature drift (within the 3 mK standard deviation), no hysteretic effects were seen in  $\rho_{xx}$  or  $\rho_{xy}$  during the field cycles.

The misalignment resistivity had been nearly nulled by adjusting the potentiometer with the sample above  $T_c$  in

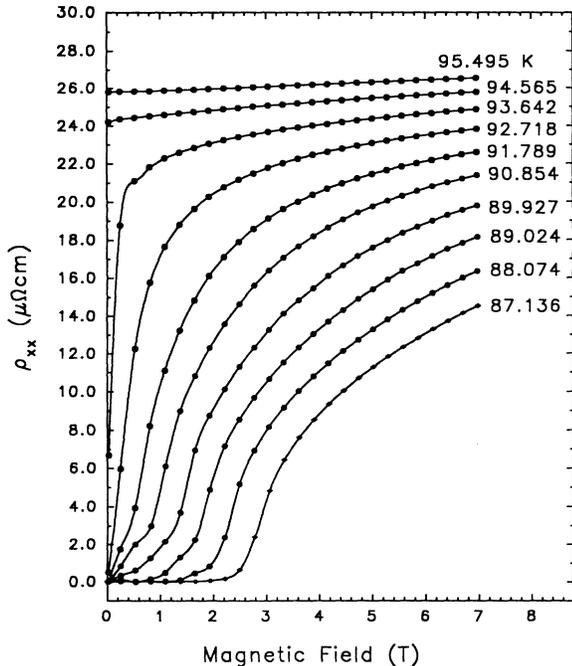


FIG. 2. Resistivity  $\rho_{xx}$  vs magnetic field, measured simultaneously with the data of Fig. 1.

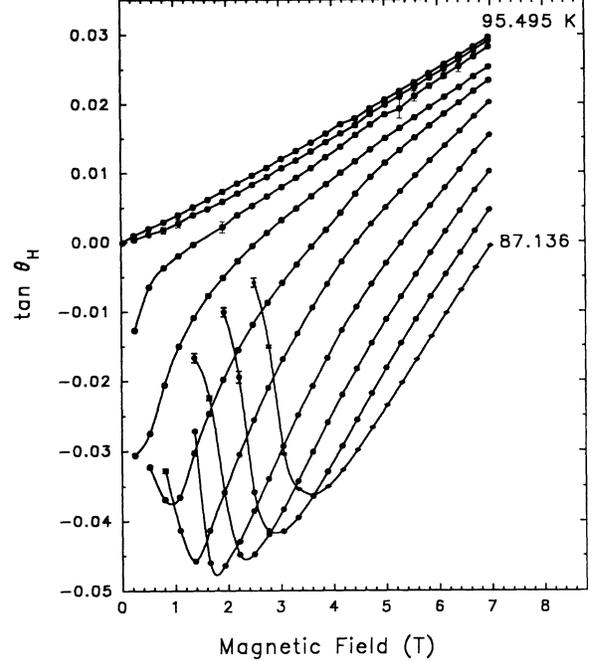


FIG. 3. Tangent of Hall angle ( $\tan\theta_H = \rho_{xy}/\rho_{xx}$ ) vs magnetic field calculated from the data in Figs. 1 and 2.

zero magnetic field. Thus our measured values of  $\rho_{xy}$  were nearly antisymmetric under field reversal. To completely remove contact misalignment effects, we took the antisymmetric part of the data by subtracting the negative-field values from the positive-field values and dividing by 2. The resulting  $\rho_{xy}$  values are shown in Fig. 1. The measured values of  $\rho_{xx}$  were symmetric under field reversal, and the data are shown in Fig. 2. Values of the tangent of the Hall angle ( $\tan\theta_H = \rho_{xy}/\rho_{xx}$ ) are plotted in Fig. 3.

Similar results were obtained in data collected using a reference frequency of 1.1 kHz. Also, using a separate untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystal, we looked for current dependence of the measured resistivities, but there was none except near the onset of nonzero  $\rho_{xy}$  and  $\rho_{xx}$ , where vortex depinning first occurs.

### III. RESULTS AND ANALYSIS

From Fig. 1 we see that reducing the temperature causes the Hall resistivity  $\rho_{xy}$  to start becoming nonlinear in  $H$  just above  $T_c$ . At temperatures below  $T_c$ ,  $\rho_{xy}$  is negative at low fields, exhibiting a minimum at a characteristic field that depends upon temperature. At higher fields  $\rho_{xy}$  becomes positive and approaches the linear field dependence found above  $T_c$ , but with a sizeable negative intercept. In our lowest-temperature field sweeps, the Hall resistivity is evidently zero below a characteristic temperature-dependent field  $B^*$ . Presumably vortices are pinned below this field. Similarly, the resistivity  $\rho_{xx}$ , shown in Fig. 2, is nearly zero below a certain field in the lowest-temperature field sweeps.

In Fig. 4 we plot contours of the  $\rho_{xy}$  data in the  $H$ - $T$

phase diagram. Shown are onset points of  $\rho_{xy}$ , points where  $\rho_{xy}$  reaches its minimum, and points where  $\rho_{xy}$  crosses through zero. For reference, we show also the vortex lattice melting line determined by a torsional oscillator technique in a separate study on a different sample.<sup>20</sup> (The sample in that study was also an untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystal grown by us by the same technique). In drawing the melting line shown in Fig. 4, we used a  $T_c$  of 94.5 K, taken, as indicated above, from the midpoint of the resistive transition of the sample used in our Hall effect experiment. Since it is not clear that this is an appropriate criterion for determining the value of  $T_c$  to be used in drawing the melting line, there is some degree of uncertainty about the exact position of the line. We include it in Fig. 4 only to show that the region where the Hall resistivity becomes nonzero, but has not reached its minimum, is approximately the same region where the vortex lattice melts.

Recently, Luo *et al.*<sup>7</sup> have discovered a striking power-law behavior between their Hall effect and resistivity data on thin film  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . In the region of the  $H$ - $T$  phase diagram where  $\rho_{xy}$  becomes nonzero but has not yet reached its minimum, they find that

$$|\rho_{xy}(T)| \propto [\rho_{xx}(T)]^\alpha \quad (1)$$

at fixed field, with  $\alpha = 1.7 \pm 0.1$ . To see if this power law fits our data, we plot  $\log|\rho_{xy}|$  versus  $\log(\rho_{xx})$  in Fig. 5 for

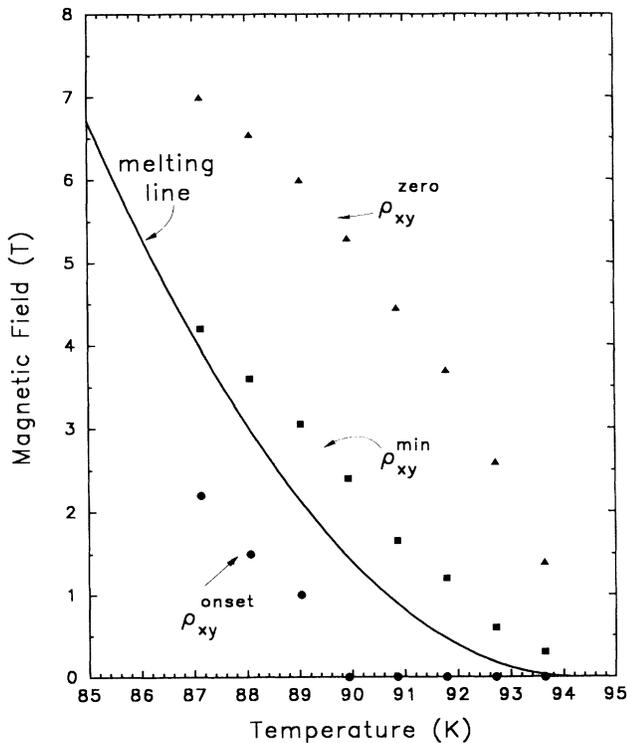


FIG. 4. Contours in the  $H$ - $T$  phase diagram:  $\rho_{xy}^{\text{onset}}$  is where the Hall resistivity first becomes nonzero,  $\rho_{xy}^{\min}$  is where it reaches its negative minimum, and  $\rho_{xy}^{\text{zero}}$  is where it crosses zero, passing from negative to positive with increasing field. For comparison, we also plot the melting line of Ref. 20 (see text).

the three different fields in this region of the phase diagram where we have enough temperature points to make the plot. While the density of our data points in this region is not as great as that of Luo *et al.*, our range of temperatures (about 3 K) is equivalent. Our data are consistent with the power-law behavior, including the exponent of 1.7.

Our data on an untwinned single crystal of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  are similar to those on thin films<sup>4,5,7</sup> and twinned crystals.<sup>6</sup> For purposes of comparison we consider the global minimum of  $\rho_{xy}$  in our data in Fig. 1, which occurs at a particular value of temperature and magnetic field. As seen in Table I, our values for the global minimum of  $\rho_{xy}$  falls among the values measured by other groups on twinned samples at similar values of temperature and magnetic field. Thus, the sign change of the Hall resistivity from positive at high fields and above  $T_c$  to negative at low fields below  $T_c$  is not a result of twinning.

It is well known that a potential difference can be induced across a type II superconductor in the mixed state by the motion of quantized flux vortices. The electric field  $\mathbf{E}$  produced by vortices directed along the  $z$  axis (i.e., the  $c$  axis of our crystal) moving relative to the superconductor at velocity  $\mathbf{v}_L$  was shown by Josephson<sup>21</sup> to be given by Faraday's law of induction:

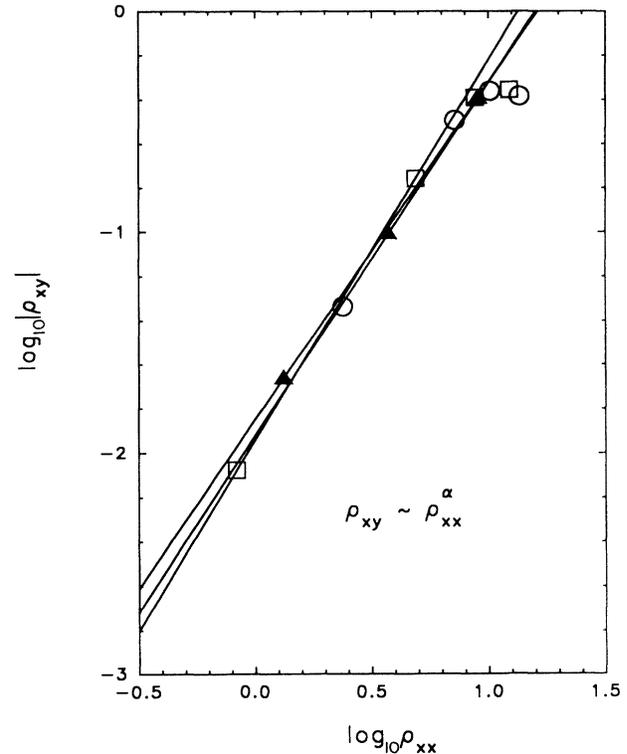


FIG. 5. Scaling behavior of longitudinal and Hall resistivities in untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Circles, squares, and triangles are data at magnetic fields of 2.2, 1.9, and 1.4 T, respectively, and temperature is the implicit variable. The lines shown have slopes  $\alpha = 1.6 \pm 0.1$ , which is consistent with the results in Ref. 7, for data over a range of field and temperature where  $\rho_{xy}$  is between  $\rho_{xy}^{\text{onset}}$  and  $\rho_{xy}^{\min}$ . The units of  $\rho_{xy}$  and  $\rho_{xx}$  are  $\mu\Omega \text{ cm}$ .

TABLE I. Comparison of the global minimum in the Hall resistivity  $\rho_{xy}$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  measured by several groups, along with the field and temperature where that minimum occurred.

Source	Sample type	Minimum $\rho_{xy}$ ( $\mu\Omega$ cm)	$B$ (T)	$T$ (K)
Fig. 1	untwinned crystal	-0.45	1.7	90.9
Ref. 6	twinned crystal	-0.2	2	87.8
Ref. 5	thin film	-0.5	1.9	88.4
Ref. 7	thin film	-0.3	3	89
Ref. 4	thin film <sup>a</sup>	-0.05	3.2	79

<sup>a</sup> $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$  film with  $T_c$  of 81 K.

$$\mathbf{E} = -\mathbf{v}_L \times \mathbf{B}. \quad (2)$$

Defining  $\phi_0$  as the flux quantum and  $n$  as the areal density of vortices,  $B = n\phi_0$ . With  $\mathbf{B}$  along the unit vector  $\hat{\mathbf{z}}$ , vortices moving at an angle  $\theta$  with respect to the  $-y$  axis induce an electric field at angle  $\theta$  with respect to the  $x$  axis. The components of this electric field are expressed in the longitudinal and Hall resistivities. The Hall angle  $\theta_H$  calculated from the measured longitudinal and Hall resistivities is the same as the angle  $\theta$ . The anomalous negative Hall effect would then imply that there is a component of vortex motion antiparallel to the transport current direction: the vortices flow upstream. Various effects, such as thermodynamic fluctuations<sup>22</sup> or a combination of Ettingshausen and Seebeck effects,<sup>23</sup> could affect the Hall angle. Some authors have attributed the anomalous negative Hall effect below  $T_c$  to these additional effects. The influence of fluctuations is difficult to estimate, since it would depend upon complicated microscopic details.<sup>22</sup> An estimate of the electric field produced by the combined Ettingshausen and Seebeck effects shows that this effect is two orders of magnitude too small to account for the observed negative Hall angle in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .<sup>23</sup>

The vortices are driven to move by the transport current  $\mathbf{J}$ . For instance, following the model of Bardeen and Stephen,<sup>24</sup> we may assume that the driving force on a vortex per unit length is the Lorentz force

$$\mathbf{F} = \phi_0 \mathbf{J} \times \hat{\mathbf{z}}. \quad (3)$$

In the model of Nozieres and Vinen,<sup>25</sup> it is argued that the driving force on a vortex is given by the Magnus force

$$\mathbf{F} = n_s e \phi_0 (\mathbf{v}_s - \mathbf{v}_L) \times \hat{\mathbf{z}}, \quad (4)$$

where  $n_s$  is the superfluid electron density and  $\mathbf{v}_s = \mathbf{J}/n_s e$  is the superfluid velocity. These forms for the driving force differ only when the vortices are depinned, where  $\mathbf{v}_L$  becomes nonzero.

In both models the driving force is balanced by a friction force  $\mathbf{f}$ ; the net force  $\mathbf{F} + \mathbf{f} = 0$  in steady state. Bardeen-Stephen assumed a friction force  $\mathbf{f} = -\eta \mathbf{v}_L$  acting in a direction opposite to the vortex velocity, but No-

zieres and Vinen<sup>25</sup> considered the form  $\mathbf{f} = -a \mathbf{v}_s$ , which acts in a direction opposite to  $\mathbf{J}$ . In each of these models, the coefficients  $\eta$  and  $a$  are temperature dependent. While both of these models predict a nonzero Hall resistivity, neither can account for the sign change of  $\rho_{xy}$  in our data. Hagen *et al.*<sup>8</sup> have pointed out that a combination of Bardeen-Stephen and Nozieres-Vinen friction forces, given by

$$\mathbf{f} = -\eta \mathbf{v}_L - a \mathbf{v}_s, \quad (5)$$

can account for a sign change of the Hall resistivity over a temperature range where  $\eta a > (n_s e \phi_0)^2$ . However, this assumption seems to be rather arbitrary.

Recently a theory by Wang and Ting<sup>18</sup> appeared, associating the anomalous sign change of the Hall effect with pinning. The theory reexamines the Nozieres-Vinen model and takes account of the backflow current due to pinning forces. The theory predicts a component of vortex velocity antiparallel to the current direction over a certain range of field and temperature, in qualitative agreement with experiment. A detailed quantitative comparison of the theory with our data would involve a six-parameter fit to Eq. (12) of Ref. 18. This fit was not attempted because the large number of adjustable parameters would render the result unconvincing. Instead, we present here a simpler comparison. At a temperature  $T_0$  where the mean-free path  $l_0$  is equal to the superconducting coherence length  $\xi$ , the minimum Hall resistivity was predicted to be

$$\rho_{xy}^{\min} \simeq -2\rho_n \mu_n B^* = -2R_H B^*, \quad (6)$$

where  $\rho_n$  and  $\mu_n$  are the normal-state resistivity and mobility, and  $R_H$  is the normal-state Hall coefficient. Using the value of  $R_H$  measured previously<sup>19</sup> for this sample ( $0.15 \mu\Omega \text{ cm/T}$ ), and the values of  $B^*$  from Fig. 1, we find that Eq. (6) correctly predicts the value of the minimum  $\rho_{xy}$  in Fig. 1 at a temperature between 88 and 89 K, which we identify as the  $T_0$  of the Wang-Ting model. Assuming  $15 \text{ \AA}$  for the zero-temperature value of  $\xi$ , the coherence length is expected to be about  $60 \text{ \AA}$  at 88.5 K, which is indeed about the same value that we estimate for the mean-free path  $l_0$ .

Thus the Wang-Ting model, which predicts a sign change of  $\rho_{xy}$  by incorporating the effect of pinning forces, is able to account for the size of the negative minimum in the Hall effect seen in our untwinned crystal. This interpretation has a problem, however. If the sign change is caused by pinning forces, it seems surprising that the most negative value of  $\rho_{xy}$ ,  $(\rho_{xy})_{\min}$ , and the field value  $B_{\min}$  at which it occurs are similar for crystals and for thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , as shown in Table I. Various crystals and films would be expected to exhibit quite different strengths of vortex pinning. The similarities of these values may therefore indicate that the Wang-Ting mechanism is not the source of the sign reversal of  $\rho_{xy}$ . On the other hand, it is not surprising that  $(\rho_{xy})_{\min}$  and  $B_{\min}$  in our untwinned sample are similar to the values measured in twinned samples, shown in Table I, since we now present a line of reasoning indicating that pinning by twin planes is not an appreciable effect near

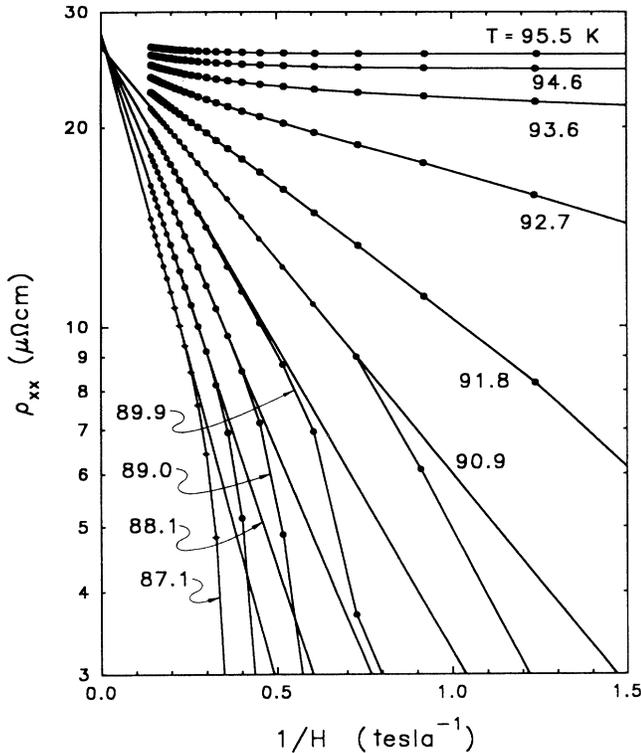


FIG. 6. Plot showing that Eq. (7) fits the high-resistance part of the  $\rho_{xx}$  data, indicating flux-flow behavior similar to that of the twinned crystal of Ref. 6.

$(\rho_{xy})_{\min}$ . Since twin planes are known to pin vortices,<sup>12-17</sup> one might expect our twinned crystal to have less pinning than untwinned crystals. The resistivity experiments of Crabtree *et al.*<sup>17</sup> in which twin planes were shown to give the dominant pinning force when the magnetic field was aligned with them, focused on the lower part of the field-induced resistive transition, where the resistivity was less than  $\frac{1}{3}$  of the normal-state value. Our data (Figs. 1 and 2), however, show that the negative minimum in the Hall resistivity occurs at a field and temperature where the resistivity is greater than  $\frac{1}{3}$  of the normal-state value. We refer to this high-resistance region as the flux-flow regime. In this regime, the data of Crabtree *et al.* imply that the pinning force from twin planes is negligible. Thus the pinning force that would cause the negative Hall resistivity within the Wang-Ting model could not come from twin planes. Thus our observation of a negative Hall resistivity in untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is consistent with the Wang-Ting model only if a sufficient pinning force is generated by some inhomogeneity other than twin planes.

Further evidence supporting this conclusion can be

found from a quantitative analysis of the resistivity (Fig. 2) in the flux-flow regime. To obtain information about the pinning force, we follow the analysis of Chien *et al.*<sup>6</sup> Replotting the resistivity data as shown in Fig. 6, we see that  $\log \rho_{xx}$  is linear in  $1/H$ . Using the notation of Chien *et al.*, we write

$$\rho_{xx}(T, H) = \rho_0 \exp \left[ \frac{-a'(1-T/T_c)^{q'}}{H} \right], \quad (7)$$

where  $\rho_0$  has a value of  $27 \mu\Omega \text{cm}$ , which is about the normal-state resistivity just above  $T_c$ . If we fix  $q' = 1.5$  (the value of Chien *et al.*), then the best fit to the temperature dependence in Eq. (7) gives the values  $T_c = 94.2 \text{ K}$  and  $a' = 234 \text{ T}$ . Following Chien *et al.* in identifying the term  $a'(1-T/T_c)^{q'}/H$  with the ratio  $U'/k_B T$ , where  $U'$  is the activation energy for vortices moving in the presence of pinning forces, we find that  $U' < k_B T$ , consistent with flux-flow behavior. Furthermore, the value of  $U'$  from our untwinned sample has the same order of magnitude as that of the twinned sample of Chien *et al.* This result provides further evidence that the pinning forces dominating in the flux-flow region, where  $\rho_{xy}$  reaches its minimum, are not generated by twin planes. [Equation (7) does not fit our data at lower resistivities, presumably because the activation energy  $U' > k_B T$ , and the situation is more accurately described as being thermally activated motion than as flux-flow (or diffusive) behavior.<sup>6</sup>]

#### IV. CONCLUSIONS

A sign reversal of the Hall effect occurs below  $T_c$  in untwinned single-crystal  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . The magnitude of the negative minimum of the Hall resistivity is the same as that which has been seen previously in twinned samples. This result indicates that the negative Hall angle is not a result of twin-plane-guided vortex motion. If, as asserted by Wang and Ting, the sign reversal is associated with pinning forces in the flux-flow region of the  $H$ - $T$  phase diagram, then these pinning forces are not exerted by twin planes. The experimental data fit the theoretical relation given in Eq. (6). On the other hand, because different types of samples, probably with very different pinning strengths, show a similarity in the magnitude of the Hall coefficient's sign reversal and the field at which the reversal is strongest, the Wang-Ting mechanism may not be important in explaining the sign reversal.

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