

## Twin-boundary effect on the Hall conductivity in high- $T_c$ superconducting thin films

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The pinning influence on the Hall conductivity in the mixed state of high-temperature superconductors is still experimentally and theoretically controversial. In this work, the effect of twin-boundary pinning on the Hall conductivity is studied, with particular emphasis on the high-field results. The longitudinal and Hall resistivities were measured in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{YbBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films, up to 18 T: a signature due to the pinning of the vortices in the twin boundaries is clearly visible in the longitudinal resistivity  $\rho_{xx}$  and in the Hall resistivity  $\rho_{xy}$ , but disappears when the Hall conductivity  $\sigma_{xy} \cong \rho_{xy}/\rho_{xx}^2$  is computed. However, at lower temperatures, a minimum of  $d\sigma_{xy}/dT$  is proved to be a weak signature of the twin-boundary pinning. These results are confirmed by similar measurements done on one sample rotated  $16^\circ$  away from the magnetic field direction. [S0163-1829(97)04133-7]

The Hall effect in the mixed state of high-temperature superconductors remains one of the most intriguing and challenging problems exhibited by these new materials, and it probably contains the key for a better understanding of vortex dynamics. The issues in discussion can be summarized as follows: (i) the sign reversal presented by the Hall resistivity with respect to its normal-state value, for temperatures near  $T_c$  and for moderate fields;<sup>1</sup> (ii) the scaling relation between the Hall and longitudinal resistivities, for low resistivity values, which takes the form  $\rho_{xy} \propto \rho_{xx}^\beta$  with  $\beta$  between 1.5 and 2 for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ,  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ , and  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ ;<sup>2-4</sup> (iii) the role of disorder (pinning) on the Hall effect<sup>5</sup> and, in particular, on the Hall conductivity.<sup>3,4,6-9</sup>

Many different approaches have been developed to explain the Hall effect, invoking magnetic scattering,<sup>10</sup> two carrier types,<sup>11</sup> motion of vacancies in a pinned vortex lattice,<sup>12</sup> Andreev reflection,<sup>13</sup> quasiparticle-vortex scattering,<sup>14</sup> charged vortices,<sup>15</sup> time-dependent Ginzburg-Landau theories<sup>16,17</sup> or fluctuation effects.<sup>18-20</sup> This long but not exhaustive list illustrates the fact that the Hall effect is far from being well understood.<sup>21</sup>

However, since the transport properties of the mixed state are dominated by the presence of vortices, several attempts have been made in order to explain the Hall effect in the frame of vortex dynamics. Vinokur *et al.*<sup>7</sup> proposed a model which incorporates the effect of pinning on vortex dynamics in an averaged way, leading to the result  $\rho_{xy} = \alpha(T, B)\rho_{xx}^2/\Phi_0 B$ , where  $\alpha$  is a microscopic parameter which imposes the  $\rho_{xy}$  signal. According to this model,  $\alpha(T, B)$  is not affected by the disorder present in the samples, which means that the Hall conductivity,  $\sigma_{xy} = \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2) \approx \alpha(T, B)/\Phi_0 B$ , is also disorder independent.

In the low-temperature region, the rapid variation of  $\rho_{xx}$  due to pinning effects hides the temperature dependence of  $\alpha(T, B)$ , leading to the simple result  $\rho_{xy} \propto \rho_{xx}^2$ . Recently, Liu *et al.*<sup>9</sup> developed a model based on a perturbation approach which considers collective pinning effects; their main conclusion is that the Hall conductivity is independent of pinning to the first nonvanishing order of the perturbation correction term, thus supporting the conclusion of Vinokur *et al.*

On the other hand, Wang, Dong, and Ting (WDT),<sup>8</sup> considering explicitly the effect of backflow current due to pinning, arrived at the conclusion that pinning not only is responsible for the sign reversal observed near  $T_c$  but also affects the Hall conductivity in a direct way. According to this model, the relation  $\rho_{xy} = \alpha(T, B)\rho_{xx}^2/\Phi_0 B$  still holds but an explicit dependence on the pinning appears in  $\alpha(T, B)$  leading to  $\rho_{xy} \propto \rho_{xx}^\beta$ , with  $\beta = 1.5$  for strong pinning and  $\beta = 2$  for weak pinning.

Experimental results on heavy-ion-irradiated samples, showing the same temperature dependence of  $\sigma_{xy}$  before and after the irradiation,<sup>3</sup> have been interpreted as presenting evidence for the Vinokur *et al.* model. However, Kang *et al.*<sup>4</sup> showed with similar experiments that an explicit dependence of  $\sigma_{xy}$  on pinning appears if  $\sigma_{xy}$  is plotted as a function of  $T/T_c$  instead of  $T$  as in the previous experiments. According to Kang *et al.*, irradiation changes the critical temperature of the samples, a fact disregarded in the previous work. However, this interpretation may be oversimplified: the modification in  $T_c$  is induced by a change of microscopic parameters which can lead to a different temperature dependence of  $\sigma_{xy}$ , not fully accounted for by a simple rescaling in  $T/T_c$ . It is thus our opinion that to clarify these matters and

TABLE I. Characteristics of the samples: The critical temperature  $T_c$  is determined by the middle point of the resistive transition, and the width of the transition at zero field,  $\Delta T_c$ , by the usual 10–90 % criterion.

Sample	Composition	Thickness (nm)	$T_c$ (K)	$\Delta T_c$ (K)	$\rho_{xx}(100\text{ K})$ ( $\mu\Omega\text{ cm}$ )	$R_H(100\text{ K})$ $10^{-9}$ S.I.
No. 1	$\text{YbBa}_2\text{Cu}_3\text{O}_7$	600	89.2	.9	115	2.05
No. 2	$\text{YbBa}_2\text{Cu}_3\text{O}_7$	600	89.4	.8	120	2.55
No. 3	$\text{YBa}_2\text{Cu}_3\text{O}_7$	375	90	.4	103	2.01

to establish the influence of pinning on the Hall conductivity, experiments must be performed on identical samples, without changing any microscopic parameters. To this purpose, we present in this paper measurements of the Hall effect up to 18 T in  $\text{YbBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films, which present at high fields a clear signature of pinning by the twin boundaries (TB's). This pinning signature provides a simple way to check the effects of pinning on the Hall conductivity and our results show that pinning influences the Hall conductivity, though this effect is weak and occurs at low temperature.

The  $\text{YbBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films were grown on  $\text{LaAlO}_3$  by the planar high-oxygen-pressure sputtering technique from stoichiometric targets;<sup>22</sup> the sample characteristics are summarized in Table I. The longitudinal resistance was measured by the standard dc four-point method. To measure the Hall resistance, the direction of the magnetic field was inverted to avoid spurious effects due to the Hall contacts misalignments or due to an inhomogeneous current distribution. The magnetic field was applied perpendicularly to the substrate, i.e., parallel to the  $c$  axis of the film. We will focus on the high-field results which display clear pinning effects.

Figure 1 shows the temperature dependence of the longitudinal resistivity  $\rho_{xx}$  and of the Hall constant  $R_H$  for sample No. 1 in fields of 14, 16, and 18 T. The longitudinal resistivity curves exhibit a characteristic ‘‘shoulder’’ separating a high-temperature regime, where  $\rho_{xx}$  is almost linear with temperature, from a rapid decrease of  $\rho_{xx}$  at lower temperatures. For all the magnetic fields, the shoulder occurs for the same  $\rho_{xx}$  value ( $\approx 46\ \mu\Omega\text{ cm}$ , for sample No. 1). Such a feature was proved to be the onset of vortex pinning by the twin boundaries<sup>23</sup> and will be of central importance in our analysis. Remarkably, the  $R_H(T)$  curves show a similar crossover between two regimes and for the same temperature values of the  $\rho_{xx}(T)$  curves (cf. Fig. 1), leading us to attribute this effect also to the TB. In order to emphasize the similarity between the longitudinal and Hall resistivities, the derivatives  $d\rho_{xx}/dT$  and  $d\rho_{xy}/dT$  were calculated from the experimental results and are shown in Fig. 2 for sample No. 1, for  $B=16$  and 18 T. The onset of the pinning by the twin boundaries is identified by a simultaneous and rapid increase of these two quantities below a characteristic temperature, defined in the following as  $T_{\text{TB}}(B)$ .<sup>24</sup> An effect of the TB on the negative minimum of the Hall resistivity was shown by Harris *et al.*<sup>25</sup> in twinned single crystals and, more recently, Morgoon *et al.*<sup>6</sup> showed that, in unidirectional twinned single crystals, the Hall conductivity depends on the angle between the current and the TB. However, to our knowledge, it is the first time that the TB characteristic ‘‘shoulder’’ present in the  $\rho_{xx}$  curves is simultaneously observed in the

$\rho_{xy}$  curves. The existence of such a clear pinning signature in the longitudinal and in the Hall resistivities provides a good way to investigate the effect of pinning on the Hall conductivity, without any modification of the samples like the ones occurring in irradiation experiments: according to the model of Vinokur *et al.*, the behavior of the Hall conductivity  $\sigma_{xy}$  should not reveal any anomaly at the pinning temperature  $T_{\text{TB}}$  whereas, according to WDT, some signature due to the pinning should be found.

$\sigma_{xy}$  was calculated as  $\sigma_{xy} \approx \rho_{xy}/\rho_{xx}^2$  using the values of  $\rho_{xy}$  and  $\rho_{xx}$  obtained experimentally. The results obtained for the Hall conductivity  $\sigma_{xy}$  in sample No. 1 (corresponding to the curves of Figs. 1 and 2) are displayed in Fig. 3; the derivative  $d\sigma_{xy}/dT$  is shown in the inset to this figure. Figure 3 shows that, contrary to the  $\rho_{xx}(T)$  or  $\rho_{xy}(T)$  curves, no clear anomaly or change of regime occurs in  $\sigma_{xy}(T)$  at the

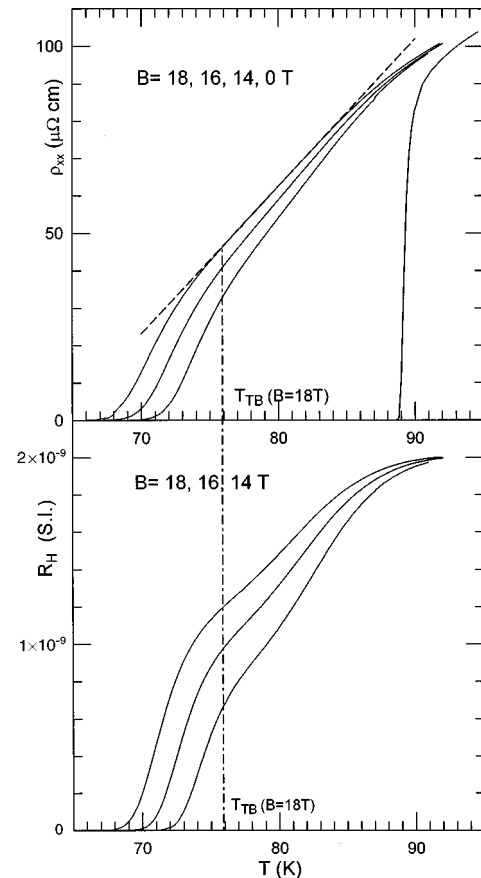


FIG. 1. Longitudinal resistivity and Hall constant at high fields for sample No. 1. The dashed line is displayed to emphasize the linear regime of  $\rho_{xx}$ . The dot-dashed line signals the onset of the pinning by the twin boundaries for  $B=18$  T, according to its definition in Fig. 2.

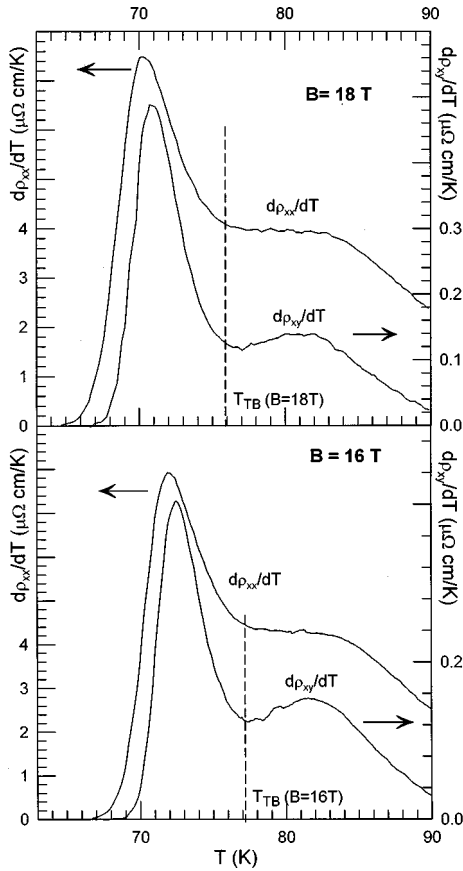


FIG. 2. Derivative of the longitudinal and Hall resistivities with respect to the temperature for sample No. 1 at  $B = 18$  and  $16$  T. The dashed line shows our determination of  $T_{TB}(B)$ .

pinning temperature  $T_{TB}(B)$  (signalled by arrows): the pinning signature is not visible in the  $\sigma_{xy}(T)$  curves at  $T_{TB}$ . Also in the  $d\sigma_{xy}(T)/dT$  curves, no clear feature is observed at  $T_{TB}$ . These results are in agreement with the Vinokur *et al.* model which predicts the independence of the Hall conductivity on pinning.

However, the analysis of the  $d\sigma_{xy}/dT$  curves at lower temperature must moderate this statement: while the feature appearing at  $T_{TB}$  on the  $d\rho_{xx}(T)/dT$  or  $d\sigma_{xy}(T)/dT$  does not subsist on the conductivity derivative, a pronounced minimum appears in  $d\sigma_{xy}/dT$  at a temperature lower than  $T_{TB}$ , for all fields. To check if this minimum is due to pinning effects or to an intrinsic (i.e., pinning independent) thermal behavior of the Hall conductivity, its derivative  $d\sigma_{xy}/dT$  is plotted versus  $\rho_{xx}$  (Fig. 4). As shown by the  $d\rho_{xx}/dT$  curve in this figure, such a plot allows us to separate easily the regime where  $\rho_{xx}$  and  $\rho_{xy}$  are dominated by the pinning in the TB ( $\rho_{xx} \leq 46 \mu\Omega \text{ cm}$  for sample No. 1,  $\rho_{xx} \leq 53 \mu\Omega \text{ cm}$  for sample No. 2) from the flux-flow regime.<sup>26</sup> Two features are displayed by this plot: at  $\rho_{xx} = 46 \mu\Omega \text{ cm}$  (continuous vertical line in Fig. 4),  $d\sigma_{xy}/dT$  does not display any anomaly or regime change as it occurs for  $d\rho_{xx}/dT$  for all magnetic fields, confirming our previous analysis; at  $\rho_{xx} \approx 25 \mu\Omega \text{ cm}$  for all magnetic fields (dashed vertical line in Fig. 4),  $d\sigma_{xy}/dT$  goes through a minimum, in close analogy with the  $d\rho_{xx}/dT$  curves whose maxima also occurs at constant  $\rho_{xx}$  values ( $\rho_{xx} \approx 16 \mu\Omega \text{ cm}$ ) for all fields. Though our results are not very accurate for the low  $\rho_{xx}$

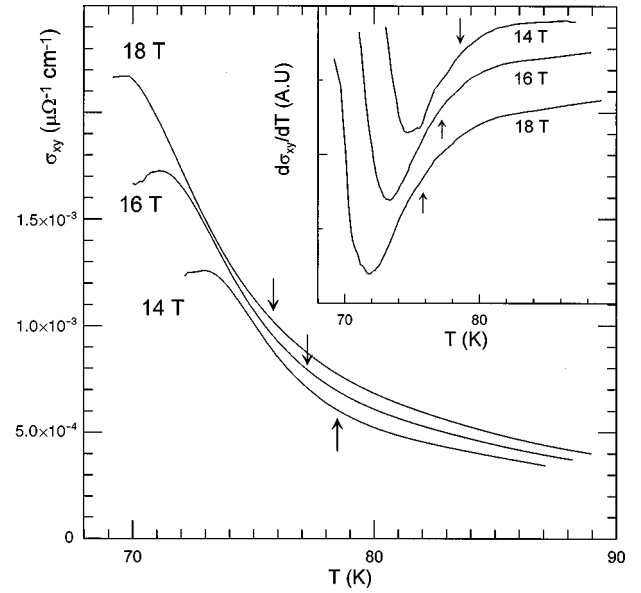


FIG. 3. Temperature dependence of the Hall conductivity for  $B = 14, 16, 18$  T. Inset: derivative of the Hall conductivity (for the sake of clarity, the curves have been vertically shifted). The arrows indicate  $T_{TB}(B)$ . No pinning signature is visible at  $T_{TB}$ , for  $\sigma_{xy}$  or  $d\sigma_{xy}/dT$ .

values,  $d\sigma_{xy}/dT$  goes to zero (corresponding to the maximum of the  $\sigma_{xy}$  curves of Fig. 3) for a common value  $\rho_{xx} \approx 12 \mu\Omega \text{ cm}$  for all fields. Similar results are obtained for all the samples where the TB signature is clearly detected (see, e.g., inset to Fig. 4).

This last feature (phenomenon occurring at constant  $\rho_{xx}$  value) is typical of two cases related to vortex motion: the melting of the Abrikosov lattice, occurring at  $\rho_{xx}/\rho_{xx}(T_c) \approx 20\%$  (Ref. 27) and the onset of vortex pinning by the TB.<sup>23</sup> Thus, we are led to the conclusion that the minimum of  $d\sigma_{xy}/dT$  at  $\rho_{xx} \approx 25 \mu\Omega \text{ cm}$  (and probably, the low-temperature part of the  $\sigma_{xy}$  curve) is caused by the pinning of the vortices, contradicting the Vinokur *et al.* prediction, but in qualitative agreement with the WDT work. One question remains: Why is the pinning signature visible in the Hall conductivity for resistivity values significantly lower than that at which appears the signature in  $\rho_{xx}$  and in  $\rho_{xy}$ ? The answer probably lies in the previously mentioned work of Liu *et al.*:<sup>9</sup> the effect of pinning on the Hall conductivity (if any) is a second- or higher-order one, thus being very weak and, consequently, only visible at a temperature lower than  $T_{TB}$ , i.e., at resistivity values lower than the one characterizing the pinning by the TB.

Another way to check the TB pinning effects is through the rotation of the samples. As shown in Ref. 23, the angle  $\theta$  between the  $\mathbf{c}$  axis and the external magnetic field plays a crucial role in the pinning process of the vortices by the TB, the resistivity curves being strongly affected below  $T_{TB}$  by a change of this angle. We performed a similar experiment with one of our samples: sample No. 3 was rotated away from the direction of the magnetic field by an angle  $\theta = 16^\circ$ , keeping the current perpendicular to the magnetic field to maintain a maximum Lorentz force configuration. Due to the geometrical and *intrinsic* anisotropy effects resulting from the rotation, the longitudinal resistivity, the Hall

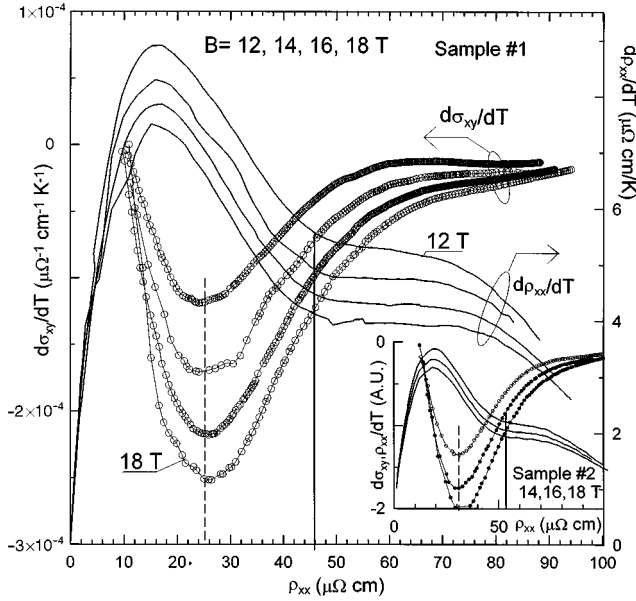


FIG. 4. Derivative of the Hall conductivity (open circles) and of the longitudinal resistivity (solid lines) vs the longitudinal resistivity for sample No. 1 (inset: the same for sample No. 2). For all fields, the pinning effects appear on  $d\rho_{xx}/dT$  for the same  $\rho_{xx}$  value (solid line), at which no anomaly appears on  $d\sigma_{xy}/dT$ . For all fields, the minimum of  $d\sigma_{xy}/dT$  (dashed line) and the maximum of  $d\rho_{xx}/dT$  occur for a constant  $\rho_{xx}$  value: this feature strongly suggests that the minimum in  $d\sigma_{xy}/dT$  must be ascribed to a pinning effect. The inset shows these same features for sample No. 2. Note that  $d\sigma_{xy}/dT$  seems to go to zero for a common value of  $\rho_{xx}$  for all magnetic fields.

resistivity, and the Hall conductivity should scale as<sup>28</sup>  $\rho_{xx}(H/\varepsilon, \theta) = \rho_{xx}(H, 0)$ ,  $\rho_{xy}(H/\varepsilon, \theta) = \rho_{xy}(H, 0)\cos(\theta)/\varepsilon$ ,  $\sigma_{xy}(H/\varepsilon, \theta) = \sigma_{xy}(H, 0)\cos(\theta)/\varepsilon$ , where  $\varepsilon^2 = \cos^2(\theta) + \sin^2(\theta)/\gamma^2$  ( $\gamma^2 = m_c/m_{ab}$ ). For  $\theta = 16^\circ$  and using  $\gamma = 7$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , we obtain  $\varepsilon \cong \cos(16^\circ)$  and hence, the factor  $\cos(16^\circ)/\varepsilon$  (present in  $\rho_{xy}$  and  $\sigma_{xy}$ ) is  $\cong 1$ , leading to  $\rho_{xy}(H/\varepsilon, 16^\circ) \cong \rho_{xy}(H, 0)$  and  $\sigma_{xy}(H/\varepsilon, 16^\circ) \cong \sigma_{xy}(H, 0)$ ; therefore, increasing the field by a  $1/\varepsilon$  factor in the rotated configuration must lead to  $\rho_{xx}(T)$ ,  $\rho_{xy}(T)$ , and  $\sigma_{xy}(T)$  curves unchanged with respect to the initial configuration ( $\theta = 0^\circ$ ), as far as the effect of anisotropic pinning centers, like the TB, is neglectable. Following this result, the measurements in the  $\theta = 16^\circ$  configuration were performed under magnetic fields given by  $H(16^\circ) = H(0^\circ)/\varepsilon = H(0^\circ)/0.96$  and compared with the results obtained in the  $\theta = 0^\circ$  configuration.

For temperatures above  $T_{TB}$ , our experimental results show that the curves  $\rho_{xx}(T)$ ,  $\rho_{xy}(T)$ , and  $\sigma_{xy}(T)$  are identical in both configurations ( $\theta = 0^\circ$  and  $\theta = 16^\circ$ ); for greater clarity and accuracy, the values of  $d\rho_{xx}/dT$ ,  $d\rho_{xy}/dT$ , and  $d\sigma_{xy}/dT$  for  $H(0^\circ) = 16$  T and  $H(16^\circ) = H(0^\circ)/0.96 = 16.64$  T are displayed in Fig. 5. These results clearly show that, for  $T > T_{TB}$ , the scaling law is thoroughly verified and, therefore, the pinning by the TB is neglectable in this temperature range. In agreement with the results of Ref. 23, the longitudinal resistivity is modified only for temperatures lower than  $T_{TB} \approx 78$  K ( $\rho_{xx} \approx 40 \mu\Omega \text{ cm}$ ) where the pinning by the TB becomes dominant, the same occurring for the Hall resistivity: the drop in  $\rho_{xx}$  and  $\rho_{xy}$  below  $T_{TB}$  becomes

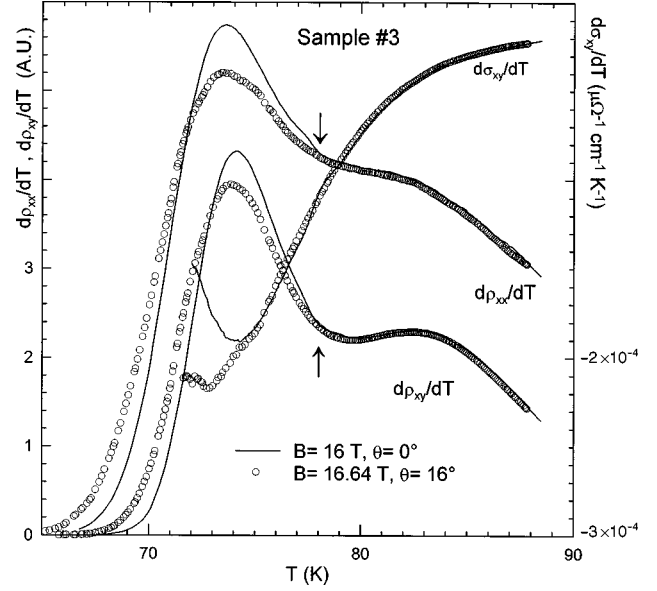


FIG. 5. Derivative of  $\rho_{xx}$ ,  $\rho_{xy}$ , and  $\sigma_{xy}$  vs temperature for  $\theta = 0^\circ$  (solid lines,  $B = 16$  T) and  $\theta = 16^\circ$  (circles,  $B = 16/\varepsilon = 16.64$  T) for sample No. 3.  $d\rho_{xx}/dT$  and  $d\rho_{xy}/dT$  differ only for  $T < T_{TB} \approx 78$  K (arrows) whereas  $d\sigma_{xy}/dT$  remains unchanged down to a lower value ( $\approx 75$  K).

less pronounced for  $\theta = 16^\circ$  (as seen by a lower maximum in the derivative), as expected in a TB pinning situation since the pinning force decreases when  $\theta$  increases.<sup>23</sup> For  $d\sigma_{xy}/dT$ , the curves for both configurations ( $\theta = 0^\circ$  and  $\theta = 16^\circ$ ) become different only at a temperature ( $\approx 75$  K) significantly lower than  $T_{TB}$ ; the well-defined minimum present for  $\theta = 0^\circ$  disappears (or is shifted to lower temperatures) for  $\theta = 16^\circ$ , showing that the minimum seen for  $\theta = 0^\circ$  is intimately related to the pinning of the vortices in the TB. We

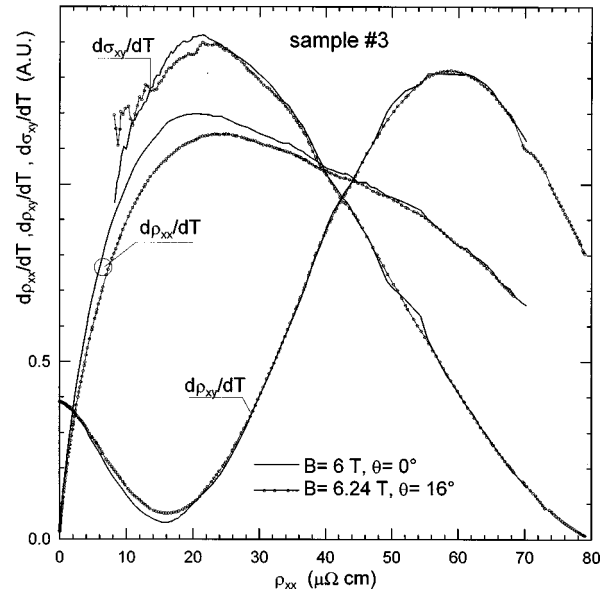


FIG. 6.  $d\rho_{xx}/dT$ ,  $d\rho_{xy}/dT$ , and  $d\sigma_{xy}/dT$  vs  $\rho_{xx}$  for  $\theta = 0^\circ$  (solid lines,  $B = 6$  T) and  $\theta = 16^\circ$  (circles,  $B = 6/\varepsilon = 6.24$  T) for sample No. 3.  $d\rho_{xx}/dT$  differs between the two configurations for  $\rho_{xx} \leq 44 \mu\Omega \text{ cm}$  whereas  $d\sigma_{xy}/dT$  remains unchanged down to a lower value.

take this result as a strong proof that the Hall conductivity is sensitive to the vortex pinning in the TB, supporting our previous analysis and recent results on single crystals with unidirectional TB.<sup>6</sup>

Let us note that these results are in agreement with the two-component model<sup>17,29</sup> which predicts that the Hall conductivity is the sum of a quasiparticle contribution, insensitive to macroscopic defects and proportional to  $H$  and a contribution due to the vortex motion, proportional to  $1/H$ . In the framework of this model, the Hall conductivity would not change abruptly at  $T_{TB}$  because the quasiparticle contribution is dominant at high temperatures and has a soft temperature dependence. On the other hand, at lower temperature, the Hall conductivity due to the vortex motion becomes more important and the effects of pinning can be seen.

At low fields, when the Hall conductivity is negative in the superconducting state, our results are less clear due to the lower experimental accuracy on  $\rho_{xy}$ . However, they show (Fig. 6) that the longitudinal and Hall resistivities in the  $\theta = 0^\circ$  and  $\theta = 16^\circ$  configurations differ for  $\rho_{xx} < 40 \mu\Omega \text{ cm}$  due to TB pinning, whereas the Hall conductivity is significantly modified only at a lower value, confirming the high-field results. As already pointed out in Ref. 17, let us note that at still lower fields (not shown) the Hall conductivity becomes negative for a longitudinal resistivity region in which the resistivity was shown to be mainly described by fluctuation effects<sup>30</sup> instead of pinning effects: for instance, at 2 T,  $\sigma_{xy}$  is negative for  $\rho_{xx} < 65 \mu\Omega \text{ cm}$  (75% of the resistivity in the normal state at  $T_c$ ). So, our results show that the Hall conductivity is sensitive to the pinning of the vortices but the negative part of the Hall conductivity is probably better explained by fluctuation effects as already proposed<sup>19,17,31</sup> than by the WDT model in which the Hall

conductivity becomes negative only when pinning is important.

In summary, our high-field results allow us to identify a clear pinning signature due to the TB, both in the longitudinal and Hall resistivities, supporting an interpretation of the Hall effect in the frame of vortex dynamics. This clear pinning signature at  $T_{TB}$  provided us an excellent tool to check the pinning effects on the Hall conductivity, without any modification of sample properties or morphology as it occurs in irradiation experiments. Our results show consistently that the Hall conductivity is affected by the pinning in the twin boundaries, though this effect is weak and occurs for temperatures lower than  $T_{TB}$ . The Vinokur *et al.* model appears then as a good first approximation as showed by the Liu *et al.* but an explicit dependence of the Hall conductivity on pinning must be considered for extended and anisotropic defects as the TB. To compare our high-field results with the WDT model, more theoretical details on the explicit dependence of the Hall conductivity are needed. The procedure described in this work to test the pinning dependence of the Hall conductivity can also be used for samples containing other type of defects leading to a signature in the resistivity curves as electron-irradiated samples<sup>32</sup> in order to check the effect of the morphology of the defects on the Hall conductivity. These high-field Hall results, added to other types of transport measurements,<sup>23,33</sup> may help to determine more precisely the modification of the vortex motion induced by the TB.<sup>34</sup>

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