Double sign reversal of the vortex Hall effect in YBa$_2$Cu$_3$O$_{7-\delta}$ thin films in the strong pinning limit of low magnetic fields

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Measurements of the Hall effect and the resistivity in twinned YBa$_2$Cu$_3$O$_{7-\delta}$ thin films in magnetic fields B oriented parallel to the crystallographic c axis and to the twin boundaries reveal a double sign reversal of the Hall coefficient for $B \lesssim 1$ T. In high transport current densities, or with B tilted off the twin boundaries by $5°$, the second sign reversal vanishes. The power-law scaling of the Hall conductivity to the longitudinal conductivity in the mixed state is strongly modified in the regime of the second sign reversal. Our observations are interpreted as strong, disorder-type dependent vortex pinning and confirm that the Hall conductivity in high-temperature superconductors is not independent of pinning.

The unusual behavior of the Hall effect in many high-temperature and in some conventional superconductors in the mixed state attracts considerable interest. In particular, the sign reversal of the Hall angle below the critical temperature T_c, as compared to the normal state, is in contrast to traditional models for the vortex Hall effect and is regarded as a fundamental problem of vortex dynamics. Several theoretical approaches have attempted to explain this phenomenon, but no agreement has been achieved. The questions, whether the Hall anomaly is an intrinsic electronic property, determined by the trajectory of an individual vortex[1–4] if collective vortex phenomena are essential,[5,6] or if vortex pinning is indispensable for the sign reversal,[7,8] are currently not resolved. Other models are based on the general grounds of the time-dependent Ginzburg-Landau theory,[9–12] but one needs a microscopic theory to predict the sign of the vortex Hall effect.

The experiments revealed that the Hall anomaly in high-temperature superconductors (HTS's), that is only observed in moderate magnetic fields, becomes more prominent in smaller magnetic fields, and attains its maximum within the vortex liquid and thermodynamic fluctuation range.[13,14] Above T_c, a rapid drop of the Hall resistivity ρ_{yx} precedes its sign reversal.[15] The occurrence of the Hall anomaly appears to be connected with the carrier concentration,[16] being absent in heavily overdoped cuprates. In highly anisotropic HTS, like Bi$_2$Sr$_2$CaCu$_2$O$_x$, a double sign reversal is observed[17] that is attributed to weak pinning in these cuprates. This latter observation raised the question of whether such double sign reversal could also exist in YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO). In fact, a positive Hall effect has been observed in YBCO far below T_c when pinning is overpowered by intense measurement transport currents.[18]

Another issue is the scaling of the transverse (Hall) resistivity to the longitudinal resistivity $|\rho_{yx}| \propto \rho_{xx}^\beta$ that can be experimentally observed with $\beta \sim 1.7$,[19] irrespective of the sign of ρ_{yx}.[20] Theoretically, an universal scaling law was derived near a vortex-glass transition,[21] or, as a general fea-

ture of the disorder-dominated vortex dynamics, with the specific prediction of $\beta = 2$.[22,23] The latter model also concluded that the Hall conductivity $\sigma_{xy} = \rho_{yx}/(\rho_{xx}^2 + \rho_{yx}^2)$ should be independent of pinning, in sharp contrast with theories that originate the Hall anomaly on pinning.[7,8] Several groups have reported experiments on samples with artificially introduced defects, and their data have been interpreted both in favor and against the pinning independence of σ_{xy}.[24,25] Measurements on pure YBCO single crystals revealed a sharp change of σ_{xy} when crossing through the melting transition.[26] Finally, recent theoretical results have suggested that σ_{xy} can be influenced by strong pinning, eventually leading to the Hall effect with opposite sign, as compared to its value in the flux-flow regime,[27] and that the presence or absence of such effect depends on the dimensionality of the pinning centers near Bose or vortex glass transition, respectively.[28]

In this paper, we report measurements of the vortex Hall effect in YBCO thin films in a large range of magnetic fields, with particular emphasis put on small magnetic fields. Our studies allow us to investigate the Hall effect in the limit of strong pinning on twin boundaries with a dilute vortex density, as opposed to previous experiments, where pinning was enhanced by radiation damage. The latter may cause spurious effects, such as amorphous regions and local changes of the oxygen content along the heavy-ion tracks. The low-field results are augmented by pulsed high transport current density and oblique magnetic field measurements of the vortex Hall effect.

We present data collected from 100-nm-thick epitaxial YBCO films, deposited either by single-target rf sputtering on LaAlO$_3$ substrates or by pulsed-laser deposition on MgO substrates. In both cases, the films were highly epitaxial with the onset of superconducting transition at 90 K and with critical current densities $j_c > 3$ MA/cm^2 at 77 K. The experiments were performed with 17-Hz ac currents at $j = 250$ A/cm^2. The measurements from 1 to 13 T were made
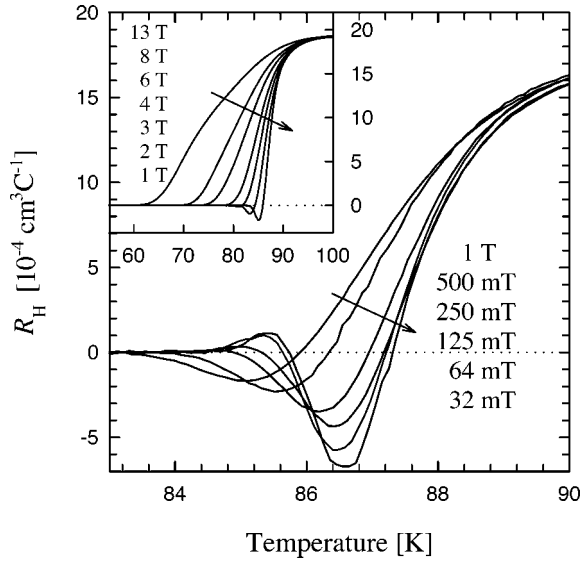
FIG. 1. Temperature dependence of the Hall coefficient of a YBCO thin film for various magnetic fields B parallel to the film c axis. The inset shows the conventional high-B field dependence of R_H on temperature.

FIG. 2. B-field-normalized Hall conductivity of YBCO dependence on temperature for various magnetic fields B parallel to the film c axis. The inset shows the conventional high-B field dependence of $\sigma_{xy}B^{-1}$ on temperature, and the broken line represents an extrapolation of the normal-state behavior below T_c.

in a superconducting solenoid using a standard cryogenic technique, while the low-field measurements from 32 mT to 1 T were performed in a closed-cycle cryocooler and with an electromagnet. The high sensitivity was achieved by fabricating thin film structures with the excellent alignment of Hall probes. Particular care was exercised to exclude spurious signals from the Earth's magnetic field and the remanence of the magnet's pole pieces. To overcome the pinning, measurements with 3-μs-long high-current density pulses were performed using a four-probe method, fast differential amplifier, and voltage detection by a boxcar averager. The temperature rise of the sample relative to the bath was smaller than 1 K and was always corrected using the YBCO film as an intrinsic thermometer.[29]

Resistivity of the YBCO thin film deep in the mixed state with B parallel to c axis is smaller than that in single crystals due to enhanced pinning at defects and does not show a first-order transition at low temperatures. At the same time, the shape of the upper part of the transition curve is common to both thin films and single crystals[30] and can be well characterized by renormalized superconducting–order-parameter fluctuations.[31] Figure 1 displays the Hall coefficient R_H of a YBCO film for a wide range of magnetic fields from 32 mT to 13 T. It demonstrates that R_H is positive at all temperatures for $B \geqslant 4$ T (inset) and reverses its sign at lower fields. The negative sign Hall anomaly increases significantly when the magnetic field is reduced below 1 T. Simultaneously, the very surprising result, observed as a second sign reversal of R_H, can be clearly identified below 0.25 T. This finding contradicts the notion[18] that the double sign reversal can only be observed in weak pinning. On the contrary, we will show that the double sign reversal of R_H presented in Fig. 1 is a result of strong pinning of vortices.

It is instructive to look at the Hall conductivity σ_{xy}, normalized to B, as shown in Fig. 2, since $\rho_{yx}=R_H B$, σ_{xy}/B is independent of B in the normal state above 90 K and roughly follows a $\sigma_{xy} \propto T^{-3}$ temperature dependence. This trend ex-
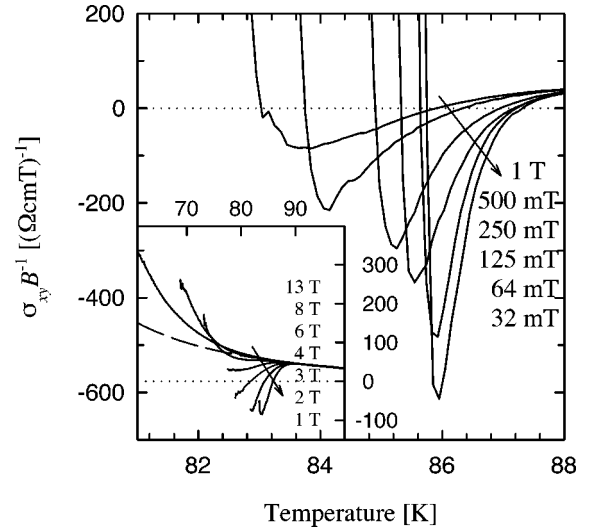
tends into the vortex-liquid region at $B=13$ T, followed by a gradual increase of the exponent as T is reduced. At lower magnetic fields, however, a negative contribution appears to gain importance over the extrapolated (broken line in Fig. 2) normal-state behavior, and at $B<3$ T leads to a sign change of σ_{xy}. With the further B decrease, a third, positive contribution sets in sharply at low fields and leads to the second sign reversal of σ_{xy} (and ρ_{yx}) at $B \lesssim 1$ T. Thus the delicate interplay of these three contributions evokes the complex features and sign reversals of the Hall effect in YBCO.

The Hall conductivity may be decomposed into

$$\sigma_{xy} = \sigma_{xy}^N + \sigma_{xy}^S + \sigma_{xy}^P, \tag{1}$$

where σ_{xy}^N represents a quasiparticle or vortex-core contribution, associated with the normal-state excitations, σ_{xy}^S a superconducting contribution, resulting from hydrodynamic vortex effects and superconducting fluctuations,[2,9–12] and σ_{xy}^P allows for a possible pinning dependence of σ_{xy}. The sign of σ_{xy}^N is the same as that of the normal-state Hall effect, but the sign of σ_{xy}^S depends on details of the Fermi surface.[9–11,16] The initially proposed pinning independence of the Hall conductivity[22,23] implies that $\sigma_{xy}^P=0$.

The results presented in Fig. 2 can be understood according to Eq. (1) as follows: $\sigma_{xy}^N \propto B$ dominates at the high B range and follows the extrapolation from the normal state (broken line in Fig. 2). Its increase above the extrapolation below $T_c(B)$ indicates reduced quasiparticle scattering in the superconducting state.[32] The contribution $\sigma_{xy}^S<0$ is roughly $\propto 1/B$ in fields of a few tesla, and is commonly associated with the hydrodynamic flux-flow effects.[4] In fields $B<1$ T, however, the $1/B$ dependence is strongly violated and rather approaches a B-linear behavior. Only the fluctuation models[12] allow for natural explanation of the observed B dependence of σ_{xy}^S, shown in Fig. 2 (excluding the second sign reversal).[33]
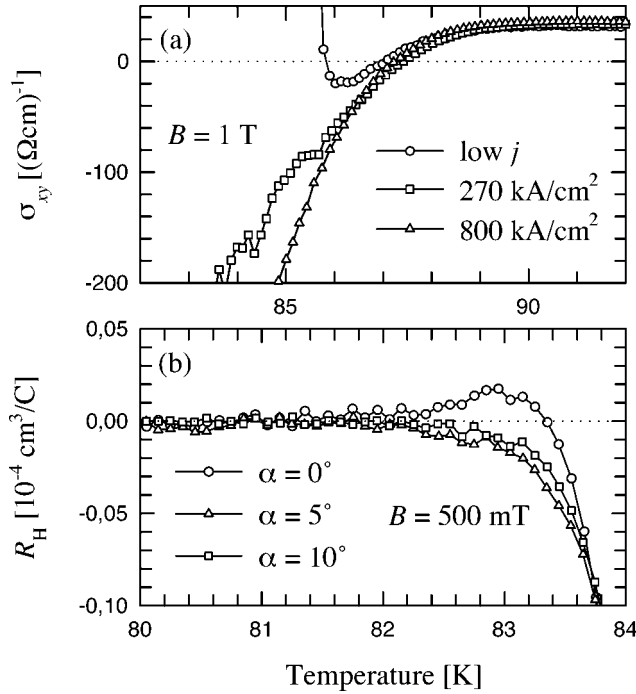
FIG. 3. Disappearance of the Hall effect's second sign reversal in YBCO films. (a) Hall conductivity versus temperature for various current densities with $B=1$ T, parallel to the film c axis. (b) Hall coefficient ($j=250$ A/cm^2) in oblique magnetic fields oriented at the angle α relative to the film c axis and to the twin boundaries.

The third (positive) contribution that is responsible for the second sign reversal of R_H (see Fig. 1), and that is evident from Fig. 2, is attributed to σ_{xy}^P. Kopnin *et al.*[27] have proposed that $|\sigma_{xy}^P|$ can exceed $|\sigma_{xy}^S|$, leading eventually to an additional sign reversal of the vortex Hall effect due to strong pinning. Ikeda[28] has stressed that in the scenario of vortex-glass fluctuations, the sign of σ_{xy}^P does depend on the dimensionality of the pinning, namely $\text{sgn}(\sigma_{xy}^P)=\text{sgn}(\sigma_{xy}^S)$ for nearly three-dimensional systems with disordered point-like pinning sites, and $\text{sgn}(\sigma_{xy}^P)\neq\text{sgn}(\sigma_{xy}^S)$ when linelike pinning disorder dominates (Bose glass). The latter situation corresponds to our YBCO films with B oriented parallel to the twin-boundary planes.

Two additional experiments allow us to test the above notion on the origin of σ_{xy}^P. The results are shown in Fig. 3. Figure 3(a) presents σ_{xy} dependence on temperature for $B=1$ T and various transport current densities. In high current densities, the pinning force is overcome by large Lorentz forces on the vortices and the sharp upturn of σ_{xy}, seen for low j (see also Fig. 2), is canceled. Thus σ_{xy} remains negative below T_c and rapidly decreases at lower temperatures. In this case, σ_{xy} strongly resembles the behavior seen in single crystals.[26] Tilting B field off the c axis at a small angle $\alpha \leqslant 10°$, as shown in Fig. 3(b), also leads to the disappearance of the Hall effect's second sign reversal. In this case, pinning is changed from linelike pinning along the twin boundary planes ($\alpha=0°$) to a reduced dimensionality pinning at an oblique field. At $\alpha<10°$ we do not expect to reach pointlike pinning, but rather pinning along short segments, where the vortices run parallel to the twin boundaries. Simultaneously, the longitudinal conductivity σ_{xx} is not changed in oblique
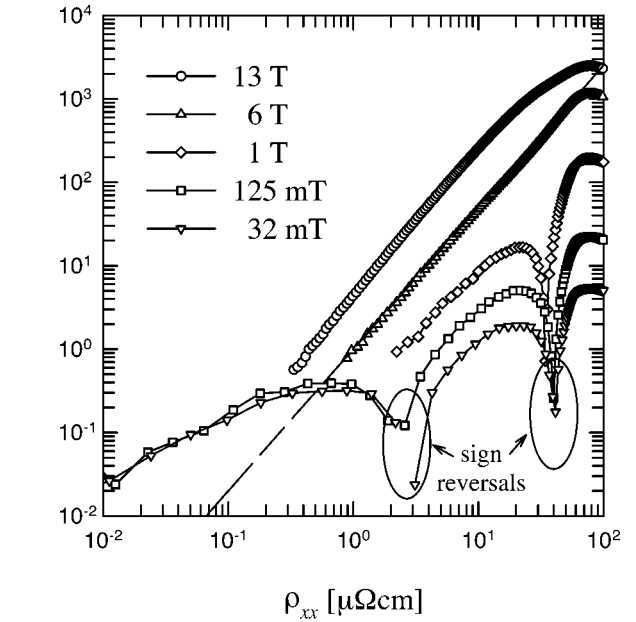

FIG. 4. Scaling of the transverse (Hall) and longitudinal resistivities in various magnetic fields B parallel to the film c axis. The data for 1, 6, and 13 T have been truncated at low dissipation. The broken line represents a fit to the 6-T data according to $|\rho_{yx}| \propto \rho_{xx}^{1.7}$.

fields within the resolution of our measurement. In this respect the impact of the B tilt on σ_{xy} is remarkable. The results presented in Fig. 3 were obtained on films deposited on different substrates, what confirms that the second sign reversal is not associated with the film fabrication and/or sample inhomogeneities, or due to a particular percolation path for the Hall current. It should be finally noted that our results are compatible with a study on clean YBCO single crystals that reveal a significant difference in σ_{xy} measured at B tilted at $\alpha=0°$ and $\alpha=4°$, respectively, although the second sign reversal was there not observed.[26]

The refutation of the pinning independence of σ_{xy} concept by our experiments immediately suggests a test of the scaling law—the another prediction that has been derived in the same theoretical context. Previous work was limited to moderate magnetic fields and Fig. 4 shows (broken line) the commonly observed scaling $|\rho_{yx}| \propto \rho_{xx}^{1.7}$ at, e.g., $B=6$ T over more than four orders of magnitude of ρ_{yx}. In fields $B \leqslant 1$ T, and at temperatures where $\rho_{yx}<0$, scaling can be found only on a limited range. Finally, in the strong pinning regime, where the second sign reversal is observed and $\rho_{yx}>0$, the resistivities are related roughly as $\rho_{yx} \propto \rho_{xx}$, in sharp contrast to $\beta=2$,[22,23] but not incompatible with the scaling law near a vortex-glass transition.[21]

Our experimental results impose several additional interesting constraints on models that attempt to explain the anomalous Hall effect in HTS's. Wang and co-workers[7,8] associate the negative Hall anomaly with the backflow current due to pinning. The increase of the negative anomaly in low magnetic fields seems to support their model, but the second sign change and the breakdown of the scaling in the limit of strong pinning are hardly compatible. Several models based on the time-dependent Ginzburg-Landau theory do not incorporate pinning effects and thus cannot be applied to

the low dissipation limit. Van Otterloo *et al.*[2] and Kopnin[4] have argued that the double sign reversal is an intrinsic electronic phenomenon. While this seems to be applicable for the highly anisotropic HTS's, it is not the case for our YBCO films. Ao[5] has considered vortex lattice defects as the origin of the sign reversal and predicted that the negative anomaly can disappear in strong pinning and near a glass state. This latter notion is in agreement with our experimental results, but contrary to theoretical predictions, we did observe deviations from $\sigma_{xy} \propto 1/B$ dependence even in the range where $\sigma_{xy} < 0$. It has been recently pointed out by Kopnin and Vinokur[27] that the initially proposed pinning independence of σ_{xy}, and $|\rho_{yx}| \propto \rho_{xx}^2$ is not valid in strong pinning case at twin boundaries, in accordance with our observations. The dependence of σ_{xy}^P on the dimensionality of the vortex-pinning disorder, proposed by Ikeda,[28] is supported by our results. Finally, D'Anna *et al.*[34] have interpreted their scaling results on YBCO single crystals with a percolation model that does predict $|\rho_{yx}| \propto \rho_{xx}^\beta$ with $\beta = 2$ for B oriented parallel to the twin boundaries and $\beta = 1.4$ for slightly oblique fields.

The latter exponent is similar to our result when $\rho_{yx} < 0$ in Fig. 4 but is incompatible with our findings in the strong pinning limit.

In summary, we have measured the Hall effect of YBCO thin films in very low magnetic fields and found a double sign reversal that is associated with strong pinning along twin boundaries. The second sign reversal does vanish at high current densities when the pinning is reduced, and also in slightly oblique B fields when the vortices transform from Bose glass to a vortex glass. The scaling law proposed for the Hall and longitudinal resistivities breaks down in the regime of the second sign reversal. Our data endorse that the Hall conductivity is significantly influenced by vortex pinning.

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