

Hall effect in the mixed state of superconducting $L_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ ($L = \text{Nd, Sm}$) single crystals

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The temperature and field dependencies of the Hall voltage in the mixed and in the normal state of superconducting $L_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ ($L = \text{Nd}$ and Sm) single crystals have been measured. Crystals having different critical temperatures ($15 < T_c < 20$ K) and transition widths ($0.5 < \Delta T_c < 3$ K) have been used in order to explore the effects of crystal quality on the controversial mixed-state Hall effect. In spite of the cerium doping, the normal-state Hall coefficient is found to be positive at low temperatures ($T \gtrsim T_c$) for all measured crystals. At higher temperatures ($T \gg T_c$), the Hall coefficient becomes negative. In the mixed state, the Hall resistivity displays an anomalous $\rho_{xy}(H)$ peak. However, in contrast to what has been found in other high- T_c superconductors, the polarity of the anomaly can have, depending on temperature, either the same sign or opposite to the Hall effect in the normal state, just above T_c . The appearance of the $\rho_{xy}(H)$ anomaly is not ubiquitous but is only observed in crystals of narrow transition widths. To account for these results we explore several of the possible explanations that have been already proposed.

INTRODUCTION

The vortex dynamics and particularly the observation of the Hall effect in the mixed state of a type-II superconductivity has been a challenging matter of research. Experiments on low- T_c superconductors soon revealed that the Hall effect displays a variety of behaviors, the sign reversal with respect to its normal state being one of the most intriguing observations.¹⁻³

High-temperature superconductors (HTSC's) also show an anomalous sign of the Hall resistivity (ρ_{xy}): ρ_{xy} is positive in the normal state, of single crystals and c -axis oriented films, owing to the hole-type conductivity; in the mixed state, it becomes negative at small fields and changes to positive at higher fields. Although the origin of this effect is still unclear, experimental evidence is now available for most of hole-doped HTSC cuprates: $\text{YBa}_2\text{Cu}_3\text{O}_7$,^{3,4} BiSrCaCu (2212 and 2232),⁵ and $\text{Tl}_2\text{Ba}_2\text{CaCu}_3\text{O}_{10}$.⁶

Within the scope of the usual flux-flow models, the Hall effect in the mixed state arises because the hydrodynamical forces acting on moving vortices.^{6,7} In the presence of a transport current \mathbf{J} , vortices move in response to a Lorentz force $\mathbf{f}_L = \mathbf{J} \times \mathbf{B}$ and acquire a velocity \mathbf{v}_L . As a consequence an induced electric field $\mathbf{E} = -\mathbf{v}_L \times \mathbf{B}$ occurs, which has a large resistive component E_x and a small Hall component E_y . The longitu-

dinal resistivity and the Hall resistivity are defined by $\rho_{xx} = E_x/J$ and $\rho_{xy} = E_y/J$, respectively. The Hall angle θ is defined as $\tan\theta = E_y/E_x$.

Classical theories of single vortex motion were modeled by Bardeen and Stephen and Nozieres and Vinen^{7,8} but most experimental data did not agree with theoretical predictions. Specially intriguing was the observation that, when the applied field is not very strong and the temperature not very low, the Hall angle in the mixed state was of opposite sign than in the normal state. To account for the common observation of the sign reversal in HTSC's, several models have been proposed. Some of them, based on the conventional models, consider a two-type-of-carriers system (holes and electrons) with different field-dependent effective masses,⁹ others, modify the hydrodynamical vortex motion equations.³ Attempts have also been made to explain the data by including large thermomagnetic effects¹⁰ and it has been suggested that for samples with negative Seebeck coefficient no change of the sign of the Hall voltage should occur. Recently, some models stressing the importance of the pinning forces on the vortex motion have been proposed:¹¹ it has been argued that Hall voltage of opposite sign to its normal-state value can result from the backflow flux motion in presence of a strong flux pinning.^{11,12} However, other authors¹³ have suggested that the observation of a Hall voltage of inverted polarity is a result of intrinsic properties of the superconductor.

In principle, measurements of ρ_{xy} in electron doped $L_{2-x}\text{Ce}_x\text{CuO}_4$ superconducting cuprates, can provide an unique opportunity to test some of the proposed hypothesis because at room temperature the majority charge carriers are electrons and correspondingly the Seebeck coefficient and Hall effect are negative.¹⁴ Similarly, the microscopic defects leading to flux pinning centers in these materials may possibly be different because of the remarkable crystallographic differences between these cuprates and the rest of HTSC cuprates. Indeed, whereas in the hole-type cuprates the coordination of the copper ions is five or six, in the $L_{2-x}\text{Ce}_x\text{CuO}_4$ oxides of T' structure the coordination is square planar. Finally the pinning force strength can also be distinct when compared to the rest of superconducting cuprates because of the in-plane coherence lengths is almost an order of magnitude longer [$\approx 80 \text{ \AA}$ (Ref. 15)].

In this paper we examine the onset of dissipation, i.e., the appearance of a measurable voltage both longitudinal and transverse, when a dc current and a dc magnetic field are applied to several superconducting $L_{2-x}\text{Ce}_x\text{CuO}_4$ ($L = \text{Nd, Sm}$) single crystals. As a preliminary characterization we measured the Hall effect in the normal state. The 300 to 1.5 K temperature range has been explored for magnetic fields high enough to drive the crystal to the normal state at any temperature. We will show that whereas at high temperature the Hall effect is negative, it changes to positive at low temperature. In the mixed state, we will show that, in the same way that the results obtained for hole-doped superconductors, at low magnetic fields, the Hall resistivity presents a peak. It is of major interest that the observed $\rho_{xy}(H)$ peak can have either the same polarity or opposite than in the normal state depending on temperature and crystal. However, the $\rho_{xy}(H)$ anomaly is not ubiquitous but is only observed in crystals having the narrower transition widths. Several currently proposed explanations (contribution of backflow currents due to pinning forces, thermomagnetic effects in the flux-flow region, two-bands models, and inhomogeneous superconductivity) are considered and discussed.

EXPERIMENTAL

Single crystals of $L_{2-x}\text{Ce}_x\text{CuO}_4$ ($L = \text{Nd, Sm}$) ($x \approx 0.15$) were grown by the self-flux method.¹⁶ The crystals have a platelet shape, with the c axis, along the shortest direction. The neodymium crystals (Nd1 and Nd2) used in this study were of about $1.3 \times 1.3 \times 0.1 \text{ mm}^3$ in size whereas the size of the samarium crystal (Sm1) was $1.2 \times 0.7 \times 0.025 \text{ mm}^3$. Four Au dots were painted on the corners of the crystal and cured at 400°C for 10 min. Pt wires ($\Phi = 50 \mu\text{m}$) were attached to the Au dots, by using silver paint. Contact resistance was smaller than 50Ω . Absolute values of the resistivity were obtained by the van der Pauw method. Resistivity versus temperature measurements give a critical temperature of: $T_c(50\%) = 15.9 \text{ K}$ and $\Delta T \approx 0.5 \text{ K}$ for Nd1; $T_c(50\%) = 20 \text{ K}$ and $\Delta T \approx 2.3 \text{ K}$ for Nd2, and $T_c(50\%) = 13.5 \text{ K}$ and $\Delta T \approx 0.5 \text{ K}$ for Sm1 crystals; ΔT is the transition width defined by the 10–90 % drop of the

resistivity. The normal-state resistivities ρ_n just above T_c and the resistivity ratios [$RR = \rho(300 \text{ K})/\rho(30 \text{ K})$] are 500, 740, and $500 \mu\Omega \text{ cm}$ and $RR = 6, 4, \text{ and } 4$ for Nd1, Nd2, and Sm1 crystals, respectively.

Hall effect and magnetoresistance were measured by a standard dc technique using a commercial 9-T superconducting magnet. A dc current of 7 mA was used in the experiments, which corresponds to a current density of about 7 A/cm^2 . The magnetic field, parallel to the c axis, was slowly swept (0.05 T/min) from -9 to 9 T , while the temperature was kept constant, using a Lake Shore 91C temperature controller, with 30-mK stability. In the temperature-dependent experiments, the temperature was swept slowly (0.5 K/min), while the superconducting coil was operated in the persistent mode. All measurements reported here have been performed using an identical protocol: after cooling the sample in zero field. By using a Hall probe, we have verified that for the experimental conditions used, the remnant field in the bore never exceeds 20 G.

The Hall resistivity (ρ_{xy}) was obtained from the antisymmetric part of the transverse voltage V_{xy} (with current reversal) under magnetic field reversal, i.e., to determine ρ_{xy} four $V_{xy}(H \pm, J \pm)$ data are needed. Typically Hall voltages are below $10 \mu\text{V}$ and the background noise of the experimental setup is of about 20 nV . It should be pointed out that we have verified that experimental Hall voltages are independent (within 10%) on changing the particular contact configuration used for current and voltage leads. Similarly repeated runs provide Hall voltages within 10%.

RESULTS

In Fig. 1(a) we display the field dependence of the resistivity (ρ_{xx}) of the Nd1 single crystal ($T_c \approx 15.9 \text{ K}$) at several temperatures. In contrast to what is common in hole-type HTSC cuprates, ρ_{xx} shows a fast increase with field until the normal-state resistivity value is reached at the upper critical field $H_{c2}(T)$, where the resistivity becomes essentially field independent. Data of Fig. 1(a) also reveals that the normal-state resistivity has a small positive slope ($d\rho_{xx}/dT > 0$) for $T < T_c$ and a weak magnetoresistance in the normal state. Figure 2 shows the normal-state resistivity of the Nd1 crystal above and below T_c ; it can be appreciated that when superconductivity is suppressed by the field, $\rho_{xx}(T < T_c)$ is almost saturated. Again this behavior is in contrast with p -type HTSC's where the normal-state resistivity monotonously decreases below T_c .

In the lower panel of Fig. 1 the field dependence of the Hall resistivity (ρ_{xy}) is shown [Fig. 1(b)] at several temperatures above and below T_c . Similarly to what has been already found in hole-type superconductors, the Hall resistivity shows a striking peak at weak fields, eventually becoming zero as the field or temperature decrease because any flux motion and thus dissipation is suppressed. At all temperatures, in the high-field region ρ_{xy} becomes positive and almost linear with field, thus indicating that in the normal state, above $H_{c2}(T)$, the Hall

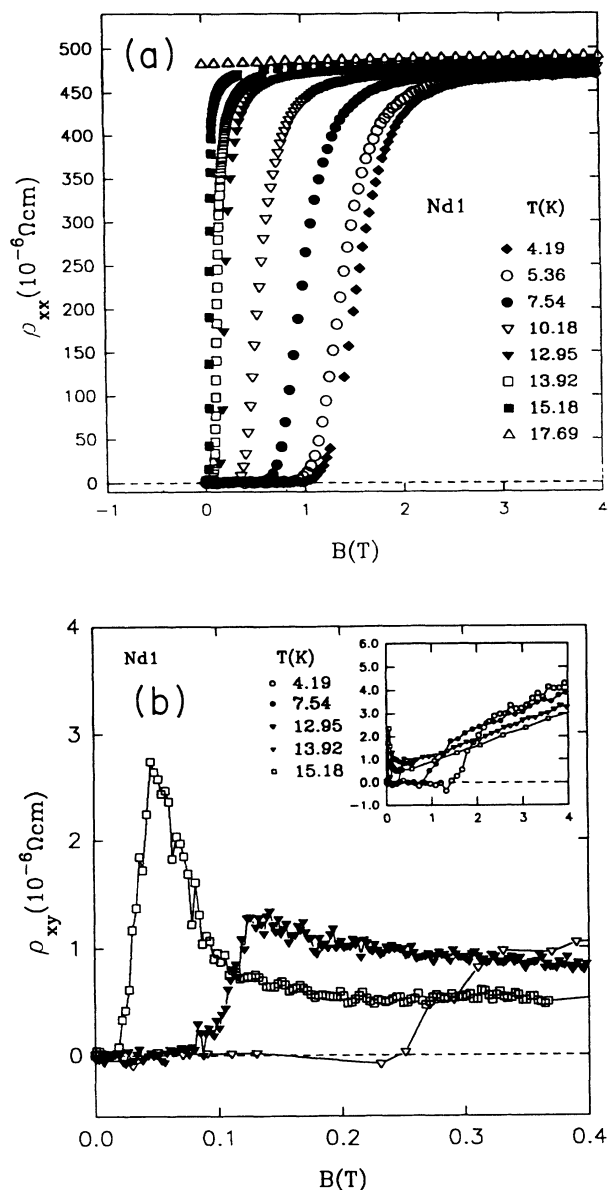


FIG. 1. (a) Field dependence of the resistivity ρ_{xx} of the Nd1 single crystal at various temperatures ($T < T_c$). (b) Field dependence of the Hall resistivity ρ_{xy} ($T < T_c$). The lines connecting experimental points are only eye guides.

coefficient R_H in this crystal is positive and field independent.

The normal-state Hall coefficient $R_H(T)$ measured at several temperatures in the 300–1.5 K temperature range is shown in Fig. 3. All these Hall coefficient values have been obtained at $\mu_0 H = 9$ T because of at this field the material becomes normal [$H > H_{c2}(0)$]. The first prominent feature of the $R_H(T)$ plot is that at high temperature $R_H(T) < 0$ thus signaling electronlike (n -type) conductivity but at lower temperature ($T \lesssim 40$ K) $R_H(T)$ becomes positive and it remains positive down to T_c . Observation of a positive Hall coefficient at low temperature is a common characteristic of all measured $L_{2-x}Ce_xCuO_4$ single crystals.¹⁷

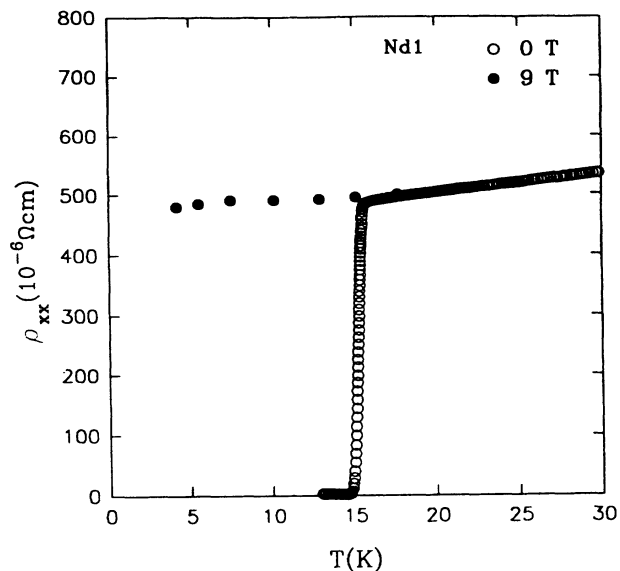


FIG. 2. Resistivity versus temperature at zero field (open symbols) and 9 T (solid symbols) for the Nd1 crystal.

The most remarkable feature in Fig. 1(b) is that the peak of $\rho_{xy}(H)$ is positive, i.e., it has the same polarity as in the normal state. The amplitude of the $\rho_{xy}(H)$ peak depends on temperature and increases with it. We have found that repeated measuring runs lead to slightly different amplitude values (differing less than 20%). The onset of nonzero Hall resistivity occurs at the same field (within the experimental resolution) than the onset of magnetoresistivity (ρ_{xx}) [see Fig. 1(a)]. The $\rho_{xy}(H)$ peak spans over a field range which is similar to that of the width of the transition in $\rho_{xx}(H)$.

In Fig. 4(a), the field dependence of ρ_{xx} for Sm1 crystal at several temperatures below T_c (≈ 13.5 K) is shown. The transition in $\rho_{xx}(H)$ towards the normal state are

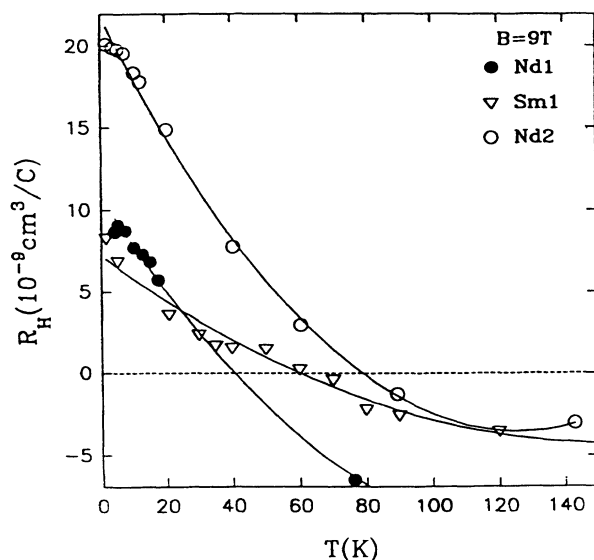


FIG. 3. Normal-state Hall coefficient $R_H(T) = \rho_{xy}/B$ versus temperature measured at 9 T for: Nd1 (solid circles), Sm1 (triangles) and Nd2 (open circles) single crystals. The solid lines through the experimental points are only eye guides.

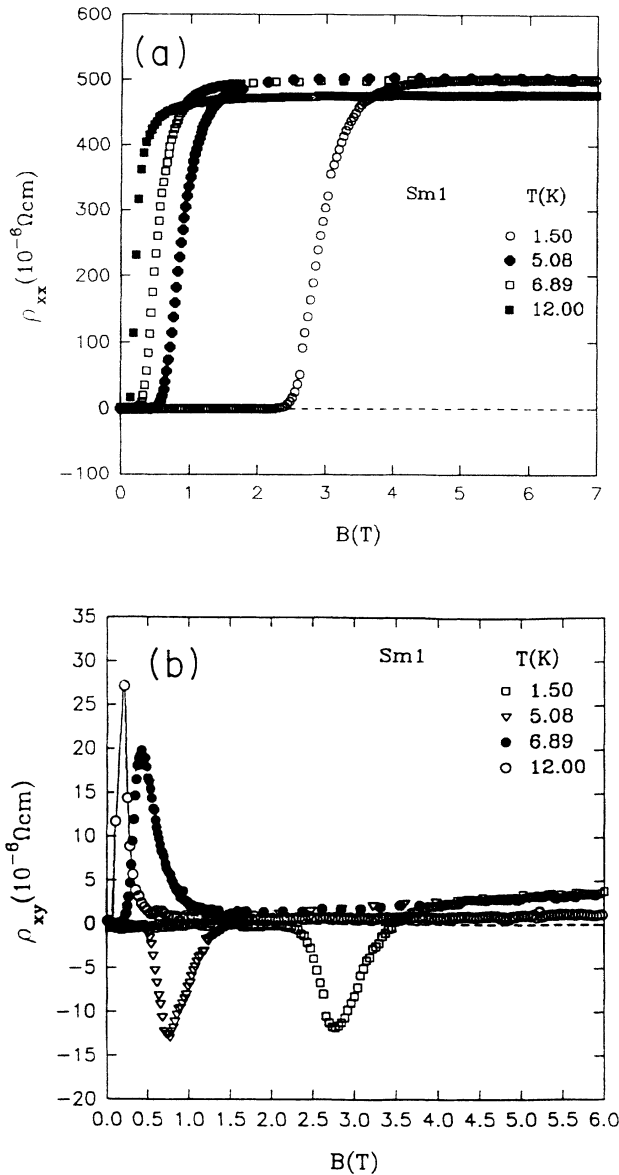


FIG. 4. (a) Field dependence of the resistivity ρ_{xx} of the Sm1 single crystal at various temperatures ($T < T_c$). (b) Field dependence of the Hall resistivity ρ_{xy} ($T < T_c$).

very sharp, especially at high temperature; the normal-state resistivity has a small negative slope, i.e., $d\rho_{xx}/dT < 0$ for $T < T_c$. The high-temperature normal-state Hall coefficient R_H is also negative, and similarly to the Nd1 crystal, it changes to positive at higher temperature (≈ 60 K). See Fig. 3.

Although both $\rho_{xx}(H)$ and $R_H(T)$ of the Nd1 and Sm1 crystals are similar, the transverse voltage $\rho_{xy}(H)$ of Sm1 crystal displays a richer phenomenology. In Fig. 4(b), we show $\rho_{xy}(H)$ at several temperatures below T_c . The appearance of the anomalous $\rho_{xy}(H)$ is clear but now, whereas at temperatures close to but below T_c , the anomalous peak has the same polarity as the Hall effect in the normal state (positive) at lower temperatures the peak has an opposite polarity. The $\rho_{xy}(H)$ peak spans over a field range which is similar to that of the field width of the

transition observed in $\rho_{xx}(H)$.

Data of Figs. 1 and 4 clearly show evidence that the Nd1 and Sm1 crystals have sharp and monotonous in-field transitions; this observation, as already suggested by the zero-field resistive transitions, is an indication of the quality of the crystal. It is only in these crystals, of the highest quality, that the anomalous Hall effect is observed. To allow comparison, we present in the following data obtained from a crystal of lower quality: Nd2. It is well known that crystal homogeneity is a major issue in the electron-doped superconductors.¹⁸

The field dependence of the resistivity (ρ_{xx}) of the Nd2 single crystal at several temperatures below T_c (≈ 19 K) is shown in Fig. 5(a). In contrast to what was observed for Nd1 and Sm1 crystals, the $\rho_{xx}(H)$ curves recorded at the lowest temperatures show a double step transition which reflects the inhomogeneous microstructure of this

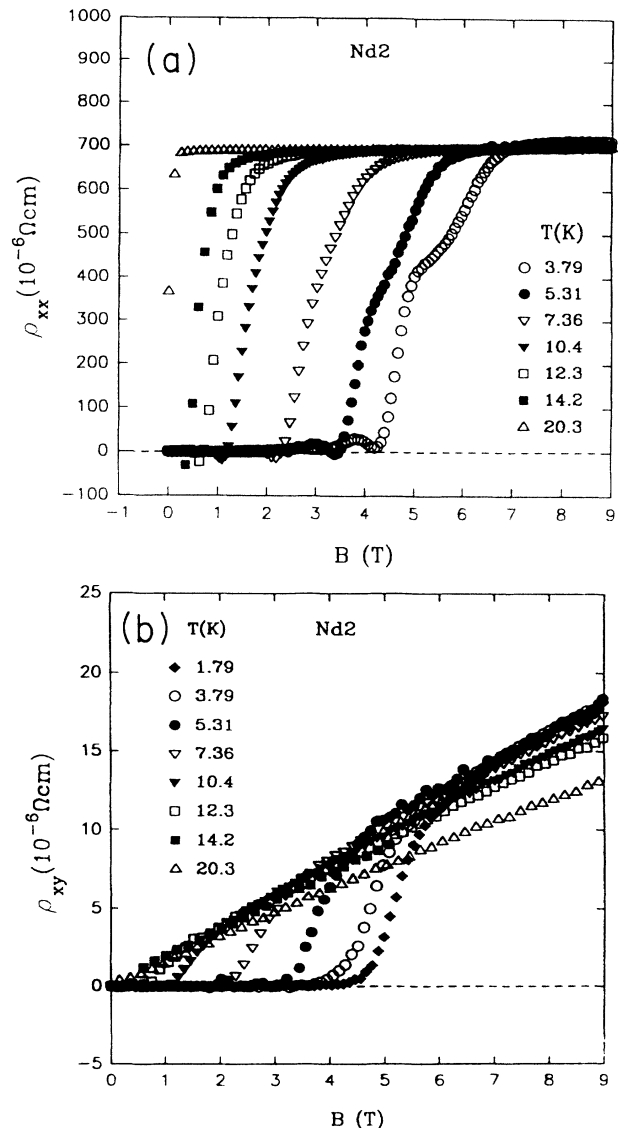


FIG. 5. (a) Field dependence of the resistivity ρ_{xx} of the Nd2 single crystal at various temperatures ($T < T_c$). (b) Field dependence of the Hall resistivity ρ_{xy} ($T < T_c$).

crystal. In fact the large transition width ($\Delta T \approx 2.3$ K) was already a clear indication of the nonhomogeneous character of this crystal. The higher value of the normal-state resistivity (740 vs 500 $\mu\Omega$ cm found in Nd1) also points to the same direction. Therefore this crystal is a good example of an inhomogeneous superconductor. As will be discussed later, we will take advantage of this characteristic to analyze the possible role of inhomogeneities and percolation effects on the appearance of the anomalous Hall peak.

The normal-state Hall coefficient $R_H(T)$ measured at several temperatures and using a field high enough to drive the sample to its normal state at any temperature ($\mu_0 H = 9$ T), is also included in Fig. 3. Similar to what was found for rest of the crystals, at high temperature $R_H(T) < 0$ but at lower temperature ($T \lesssim 80$ K) $R_H(T)$ becomes positive and it remains positive down to the lowest temperature.

In Fig. 5(b) the field dependence of the Hall resistivity (ρ_{xy}) is shown at several temperatures above and below T_c . Similarly to what has been already found for the other crystals, the Hall resistivity is positive just above T_c . However, the remarkable difference with Nd1 and Sm1 crystals is that now, the mixed-state Hall resistivity does not show any evidence of the anomalous peak. Above a threshold field, transverse dissipation (ρ_{xy}) due to the vortex movement suddenly appears but in this case ρ_{xy} is a monotonous increasing function of field. Wang *et al.*¹⁹ in earlier experiments on a Nd_{1.85}Ce_{0.15}CuO₄ single crystal, also observed a similar behavior of the Hall voltage. When the normal state is reached, $\rho_{xy}(H)$ becomes linear. As found for Nd1 and Sm1 crystals, the onset of nonzero Hall resistivity occurs at the same field (within the experimental resolution) that the onset of magnetoresistivity (ρ_{xx}) (see Figs. 5).

All these experimental results can be summarized as follows: (a) Although at high temperature R_H is negative, at the onset of the superconducting transition R_H is positive in all measured crystals. (b) The crystals of higher quality (narrower transition width, higher RR and smaller resistivity at T_c) show an anomalous $\rho_{xy}(H)$ peak in the mixed state. (c) The $\rho_{xy}(H)$ peak has the same polarity (positive) as the Hall coefficient in the normal state just above T_c ; however, in one of the crystals, it changes sign at lower temperatures and thus a Hall voltage appears of opposite polarity that in the normal state.

DISCUSSION

The observation of a sign reversal and a peak in $\rho_{xy}(H)$ in the mixed state of the electron-doped HTSC is a result which, shares some similarities with the reported behavior of $\rho_{xy}(H)$ in hole-type superconductors and some low- T_c materials. However detailed comparison of our data with the behavior of $\rho_{xy}(H)$ in the hole-type cuprates reveals an important difference which is worth mentioning. In the hole-type HTSC's, the $\rho_{xy}(H)$ peak of inverted polarity (with respect to the normal-state one) typically occurs close to the irreversibility line $H_{irr}(T)$ and for temperatures close to T_c . At lower temperature the peak is vanishingly small, and $\rho_{xy}(H)$ has the same

polarity as in the normal state.³⁻⁵ Previously reported data¹³ of Hall effect on a Nd_{2-x}Ce_xCuO₄ crystal, revealed a peak of $\rho_{xy}(H)$ of opposite polarity than in the normal state, at any temperature. In our case, although the peak clearly appears below $H_{c2}(T)$ it has the same polarity than in the normal state at $T \gtrsim T_c$. Only at temperatures well below T_c , $\rho_{xy}(H)$ has an opposite polarity to the normal state ($T \gtrsim T_c$) Hall coefficient. Therefore our experimental data display a sign reversal of the Hall peak, which is a new feature of these electronic superconductors.

Wang and Ting¹¹ developed a general treatment of flux flow, based on the normal-core model of Bardeen and Stephen⁷ and the approach of Nozières and Vinen,⁸ but including the backflow current due to both pinning forces and other vortices. Wang and Ting have demonstrated that, for fields slightly larger than the depinning field $H_p = F_p/J$ (F_p is the pinning force and J is the current density), $\rho_{xy}(H)$ is negative changing to positive for higher fields. For instance, when pinning is included in the Bardeen-Stephen limit, ρ_{xx} and ρ_{xy} are given by^{11,12}

$$\rho_{xx}(H) = \rho_n / H_{c2} [H - H_p], \quad (1)$$

$$\rho_{xy}(H) = \rho_{xx}(H) \mu H_{c2} [H / H_{c2} - 2H_p / H], \quad (2)$$

where ρ_n is the normal-state resistivity and μ the mobility of the charge carriers. Equation (2) predicts that, for a field-independent pinning force, i.e., constant H_p , at low fields the pinning term H_p/H will dominate over the Lorentz term (H/H_{c2}) and thus $\rho_{xy}(H)$ will have a sign opposite to that of $\rho_{xx}(H)$.

Within the Wang and Ting (WT) model, the appearance of Hall voltages of opposite polarity to that of the normal state relies on the existence of pinning forces which were not included in classical theories^{7,8} of flux motion above the depinning field. In the derivation of WT, the field and temperature dependences of F_p were omitted, but according to WT it can be expected that backflow flux motion should be more important at lower temperatures, where pinning forces are enhanced; consequently, the reverse flux motion should be easier observed at lowest temperatures. Dorsey²⁰ has argued that the WT model hardly can account for the anomalous Hall effect commonly observed in hole-type HTSC's, because of the anomalous peak is observed close to $T_c(H)$ where the fluctuations should reduce substantially pinning forces thus leading unobservable the reverse flux motion. This criticism cannot be used in the present case because of the anomalous peak appears at low temperatures where pinning forces are stronger. It can be also argued that the WT model is built under the assumption that the coherence length is smaller or comparable to the mean-free path l_0 of the charge carriers ($\xi \lesssim l_0$) and thus it finds its natural regime of validity at the lowest temperatures. This observation is specially critical when superconducting materials of longer coherence lengths are considered. In fact electronic superconductors of the L_{2-x} Ce_xCuO₄ family are known to have coherence lengths ($\xi \approx 80$ Å) remarkably longer than hole-type cuprates and their electronic properties have typical

features of a metallic system close to localization. For instance, at low temperatures the in-plane resistivity measured under a magnetic field high enough to drive the sample normal [$H > H_{c2}(T)$], increases.²¹ Consequently in this system the $\xi/l_0 < 1$ condition may only be fulfilled at low temperature. To what extent the WT model can be used in the $\xi > l_0$ case is at present not clear enough. Indeed, in the case of hole-type superconductors²² recent experiments have found a clear correlation between the amplitude of the anomalous Hall peak and the particular value of the ξ/l_0 ratio.

The presence of a positive peak is not predicted by Eqs. (1) and (2) if the dependence on field of the pinning force density is omitted. It may be worthwhile to recall that these equations are, in fact, a limiting case of the more general expressions derived by WT, which, however, were still derived in the particular case of field-independent pinning force. Therefore, in its present form, the WT model cannot easily incorporate the existence of a positive Hall resistivity peak in the mixed state.

However, some arguments can be given about the expected role of the inclusion of the a field dependence of $F_p(H)$. It is usually found in superconducting materials, that $F_p(H)$ has a bell-shaped dependence on the field; therefore it could be expected that, at fields close to the maximum F_p , $\rho_{xy}(H)$ to be slightly reduced by the increasing importance of the H_p term in Eq. (2). Thus a shallow dip in $\rho_{xy}(H)$ may appear after its maximum value at the peak. Indeed this is the observed behavior [see Fig. 1(b)].

In the following we will explore to that extent inclusion of the field-dependent pinning-force model can be useful to get some insight into the origin of $\rho_{xy}(H)$ and its sign. Attempts to fit the experimental data to the general expression of WT are not expected to be useful because of the large number of fitting parameters; this objection is even more important if the explicit field dependence of $F_p(H)$ should be considered. It will be proved to be more successful to use the simplest approach given by Eqs. (1) and (2). Notice that these equations are derived in the particular limiting case of the hard-core model of Bardeen and Stephen; we are going to use them because of its extreme simplicity. It is clear that the actual physical situation may be somewhere in between this limit and the Nozieres-Vinen limit. Consequently it cannot be expected that these expressions can provide more than a first-order approximation to the vortex dynamics.

We have attempted to fit the experimental data by using Eqs. (1) and (2). To that purpose, first, $H_p(H)$ is obtained from $\rho_{xx}(H)$ and Eq. (1). In a second step $H_p(H)$ is introduced into Eq. (2) to evaluate $\rho_{xy}(H)$. This procedure allows to qualitatively reproduce the appearance of the negative $\rho_{xy}(H)$ peak. However, positive peaks can only be obtained by using unphysical negative $H_p(H)$. Consequently, it appears that the WT model, at least in the approximation of Jia *et al.*,¹² cannot account for the observation of a Hall peak of the same polarity as the normal state.

Some recent reports on the anomalous Hall effect in $Tl_2Ba_2Ca_2Cu_3O_{10}$ films with radiation-induced columnar defects,²³ have found that the amplitude of the negative

$\rho_{xy}(H)$ peak decreases when increasing the radiation fluency. On the basis of this observation it was concluded that the pinning strength cannot be at the origin of the negative $\rho_{xy}(H)$. Although this is an important piece of experimental work, the effects of irradiation on the mean free path (l_0) and on other superconductor properties could be relevant. Indeed, as it was mentioned in Ref. 23, at least for some fluences, T_c sharply drops.

Next we would like to comment on the possibility that the anomalous $\rho_{xy}(H)$ peak and its sign could be the result of two-band effects. Recently, it has been found that in the mixed state of a $YBa_2Cu_4O_8$ single crystal, the Hall effect changes its sign twice.²⁴ This observation was interpreted in terms of a two band model, which was predicted in this material. In the present case, angular-resolved photoelectron spectroscopy measurements of the Fermi surface have revealed the presence of pockets of holes and electrons²⁵ and the normal-state Hall coefficient has been found to be also consistent with a two-band model.¹⁷ Consequently, the two-band structure lying at the heart of the complex behavior of $\rho_{xy}(H)$, is at present a suggestive possibility, which cannot be easily confirmed by comparison with theoretical models.

We turn now to the model suggested by Freimuth, Hohn, and Galfy,¹⁰ in which the observation of a negative $\rho_{xy}(H)$ peak is interpreted in terms of the existence of large thermomagnetic effects in the mixed state. The model has the following physical basis. When vortices move along a given direction, say y (Hall field direction) carry with them an entropy s_f (per unit length) due to thermal core excitations. This leads to a temperature gradient $(\Delta T)_y$ in the y direction and thus to a Seebeck voltage, which adds to the transverse Hall voltage. It has been argued that, because in HTSC's the Seebeck coefficient is large, its contribution to the measured transverse voltage cannot be neglected. Specifically¹⁰

$$\rho_{xy} = \rho_{xy}^n [\tan\theta_n - (S_S s_f / \kappa_s \Phi_0) T], \quad (3)$$

where ρ_{xy}^n and θ_n are the Hall resistivity and angle in the normal state, S_S , s_f , and κ_s are the Seebeck coefficient, the transport entropy, and the thermal conductivity, respectively. If $S_S > 0$, because of the contributions of the two terms in the right-hand side of Eq. (3) have different signs, than a sign change of ρ_{xy} may be expected. $S_S > 0$ is the actual situation in hole-doped HTSC's.

For electron-doped materials, if in the vicinity of T_c , the Seebeck coefficient were negative then, according to Eq. (3), no sign change of ρ_{xy} is expected to occur. However, we have found that although at room temperature the Hall coefficient is negative it becomes positive below 60 K; we should expect that the same holds for the Seebeck coefficient.²⁶ If so, the situation in electron-doped superconductors with regard to the hole of the Seebeck coefficient on the sign change of ρ_{xy} is similar to that found in p -type materials and thus, on the basis of this model it could be also expected also a sign change of ρ_{xy} .

However, in electron-doped HTSC's, large thermal conductivities have been reported,²⁷ which together with the low temperatures involved ($T < 20$ K) may lead to a small contribution of the second term in Eq. (3). Direct

measurements of the entropy are not available yet but an estimation can be obtained from the predictions of the Ginzburg-Landau theory:

$$s_f = (\Phi_0/\mu_0 T) \{ (B_{c2} - B) / [1.16(2\kappa^2 - 1) + 1] \},$$

where κ is the Ginzburg-Landau parameter. For $\kappa \approx 10$ (Ref. 15) and a reasonable field range, this expression leads to $s_f \lesssim 3.5 \times 10^{-13}$ J/K m. Using this s_f value, $\kappa_S \approx 40$ W/K m (Ref. 27) and $T \approx 10$ K, the second term in Eq. (3) is only of about 10^{-5} , which is much smaller than $\tan\theta_n$ ($\approx 10^{-3}$). Consequently, the thermomagnetic contribution to the Hall angle is much smaller than the normal-state one, its contribution to the Hall resistivity is negligible, and thus it cannot account for the observation of the anomalous peak in ρ_{xy} .

The observed behavior of the Sm1 crystal is especially remarkable. In a recent paper, Shoenes, Kaldis, and Karpinski²⁴ have reported a double sign change of the Hall resistivity in a $\text{YBa}_2\text{Cu}_4\text{O}_8$ single crystal when current was applied along the a or b axis. Similarly, it has been recently shown that in $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films, the sign of the Hall anomaly is related to the direction of the applied field with respect to the crystallographic axis.²² It is clear that the reported behavior is connected to the crystallographic anisotropy and it should be absent in our tetragonal crystals with the field applied along the c axis. Consequently, more experimental data are needed to clarify this important result.

Finally, a comment on the nonobservance of the anomalous Hall effect in the Nd2 crystal is in order. Another suggestion on the origin of this anomaly could be the influence of material inhomogeneities on $\rho_{xy}(H)$. According to this picture, it is clear that the Nd2, because of its obvious lower quality, could display enhanced anomalous behavior. Instead of that, $\rho_{xy}(H)$ is monotonous and it has the same sign that the normal-state Hall coefficient. Consequently, our data indicates that macroscopic inhomogeneities may not be the cause of the anomalous Hall effect.

CONCLUSIONS

We have presented longitudinal and Hall resistivity measurements in the normal and mixed state of several $\text{L}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ ($L = \text{Nd, Sm}$) single crystals. Crystals of different quality from the point of view of transition widths and resistivity ratios are studied and compared. For all of them, in the normal-state the Hall coefficient $R_H(T)$ is found to change from negative at room temperature towards positive at lower temperatures.

The Hall effect in the mixed state reveals some interesting features which, in part, are in striking contrast to those observed in other hole-doped HTSC's: Whereas at temperatures close to T_c the Hall voltage displays a peak of sign identical to that observed in the normal state, at temperatures well below T_c the anomalous $\rho_{xy}(H)$ peak has a polarity opposite to the normal state. This double sign reversal is the most significant result reported here. However, the controversial anomalous Hall peak is not observed in all crystals but only in those of higher quality.

We have shown that thermomagnetic effects are not strong enough to severely modify the Hall angle and thus cannot account for the sign variation of the Hall effect in the mixed state. Explicit calculations based on the hydrodynamical vortex motion model, including pinning, have shown that the essential features of the experimental data, mainly the appearance of positive and negative Hall peaks when changing temperature, cannot be reproduced. However, two-band effects cannot be excluded as a possible explanation for the observed double sign of $\rho_{xy}(H)$.

The Hall effect in the mixed state of electron-doped compounds shows an experimental behavior even richer than the hole-doped cuprates and the models so far proposed cannot give a complete picture of this complicated phenomenon.

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