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Anomalous flux-flow Hall effect: $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ and evidence for vortex dynamics

S. J. Hagen, A. W. Smith, M. Rajeswari, J. L. Peng, Z. Y. Li, R. L. Greene,
S. N. Mao, X. X. Xi, S. Bhattacharya, Qi Li, and C. J. Lobb
Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, Maryland 20742-4111

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We report an observation of a sign change in the Hall resistivity ρ_{xy} in the superconducting state of the *n*-type superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-y}$. This anomaly in other superconductors has widely been attributed to extrinsic effects, such as pinning or thermoelectric effects, or else to complicated band structures. However, the behavior of the Hall effect in the *n*-type cuprate $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ and the systematics of the anomaly in other superconducting materials together provide strong evidence against such models. The data instead indicate that ρ_{xy} reveals an intrinsic property of vortex motion.

In the mixed state of a type-II superconductor, Josephson's relation[1] $\mathbf{E}=-\mathbf{v}_L\times\mathbf{B}$ (where $\mathbf{v}_L$ is the velocity of vortex motion and $\mathbf{B}$ is the magnetic induction) shows that dissipative fields ($\mathbf{E}\|\mathbf{j}$) result from flux motion perpendicular to the current density **j**. While this motion and the resulting flux-flow resistivity can be calculated approximately within any of several models,[2-4] the nondissipative Hall electric field (i.e., $\mathbf{E}\perp\mathbf{j},\mathbf{B}$) from vortex motion *along* the direction of **j** has been far more difficult to predict correctly. As a small ($\sim 10^{-3}$ part) transverse correction to the flux velocity, this Hall resistivity $\rho_{xy}(\equiv E_y/j_x$ for $B=B_z)$ thus offers a very sensitive test of vortex motion models, but the observed behavior of ρ_{xy} has often included a sign reversal below T_c that contradicts[5] the existing models. A number of mechanisms have been proposed[5-10] (both in recent and older literature) to explain these puzzling data, but both the origin of this Hall anomaly and the reasons for its presence or absence in particular superconductors remain controversial.

We present here experimental results on the Hall effect in the "electron-doped" superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ that offer a useful test of these interpretations of the Hall anomaly. We also discuss an empirical correlation between microscopic material parameters and the behavior of ρ_{xy} that should provide useful direction for future theoretical work on the flux-flow Hall effect.

While treatments based on time-dependent Ginzburg-Landau theory were quite successful[11,12] in describing transport properties of very dirty superconductors (transport mean free path $l \ll$ BCS coherence length ξ_0), the Josephson relation[1] $E=-\mathbf{v}_L\times\mathbf{B}$ together with a phenomenological model for vortex motion has formed the basis of transport theories[2-4] for cleaner superconductors $l\geq\xi_0$. The Noziéres-Vinen model[3] is typical of such vortex dynamics pictures; a transport current $\mathbf{j}=n_s e\mathbf{v}_T$ results in a Magnus force

$$\mathbf{F}=n_s e(\mathbf{v}_T-\mathbf{v}_L)\times\phi_0 \tag{1}$$

that drives the motion of the vortex (of flux $\phi_0=h/2e$). In the presence of a drag force $\mathbf{f}=\mathbf{f}(\mathbf{v}_T,\mathbf{v}_L)$ the vortex velocity $\mathbf{v}_L$ is determined by the steady-state condition $\mathbf{F}+\mathbf{f}=0$. From the effective inertial mass[13] μ of the vortex and the flux-flow viscosity[2] $\eta(\approx\phi_0 B_{c2}/\rho_n)$ one finds a very short damping time $\tau\approx\mu/\eta$ and distance $d\approx v_L\tau$ (using $\mu\sim 10^8 m_e$/m for $YBa_2Cu_3O_7$ gives $\tau\approx 10^{-14}$ sec and typically $d\sim 10^{-14}$ m $\ll\xi_0$); thus a single vortex in motion always obeys the steady-state condition. (We ignore collective vortex effects.)

Vortex motion treatments by Bardeen and Stephen[2] (BS) and Noziéres and Vinen[3] (NV) obtained the flux-flow resistivity and Hall effect using slightly different models: Both found the same flux-flow resistivity

$$\rho_{xx}=\rho_n B/B_{c2}\ . \tag{2}$$

However, BS found a Hall resistivity

$$\rho_{xy}=(e\tau/m)\rho_n B^2/B_{c2}\ , \tag{3}$$

giving a field-dependent Hall angle

$$\tan(\Theta_H)\equiv\rho_{xy}/\rho_{xx}=[\tan\Theta_H]_{c2}B/B_{c2}\ , \tag{4}$$

(where $[\tan\Theta_H]_{c2}\equiv e\tau B_{c2}/m$), while NV obtained

$$\rho_{xy}=(e\tau/m)\rho_n B\ , \tag{5}$$

giving a field-independent Hall angle

$$\tan\Theta_H=[\tan\Theta_H]_{c2}=\text{const}\ . \tag{6}$$

In both cases the predicted Hall effect was of the same sign as in the normal state (and with comparable magnitude). However, experimental data, initially[14-17] for Nb and V and, subsequently[5,18-20] for high-T_c cuprates, showed striking deviations from this prediction: The Hall resistivity ρ_{xy} often changes sign as B decreases below B_{c2}. Figure 1 shows ρ_{xx} and ρ_{xy} of an epitaxial $YBa_2Cu_3O_7$ film: $\rho_{xy}>0$ above T_c, while $\rho_{xy}<0$ below T_c for low and moderate fields. Higher fields ($B>8$ T typically) suppress this sign reversal and restore $\rho_{xy}>0$ near T_c, while at low temperature $\rho_{xy}\rightarrow 0$.

The origin of this anomalous Hall effect remains controversial. But, while it contradicts both the standard models for vortex motion, it has rarely been seen as a challenge to the validity of those models; rather it has been attributed to phenomena specific to particular materials and unrelated to the vortex dynamics picture generally. For example, two-band models involving both electron and hole carriers[6] were invoked to explain the sign reversal in Nb and V. Once Hall anomalies were observed in many high-T_c cuprates, flux-pinning forces[8] and thermoelectric effects[9] as well as new two-band models[7] were proposed to explain the sign reversal of ρ_{xy}.

However, we have argued[5,19,21] that because the effect occurs in so many materials it should be viewed as a general consequence of vortex dynamics, i.e., that the driving and drag forces acting on a moving vortex are such that the steady-state condition $\mathbf{F}_{\text{driving}} + \mathbf{f}_{\text{drag}} = \mathbf{0}$ requires a small component of vortex velocity *antiparallel* to the superfluid flow. From the relation $\mathbf{E} = -\mathbf{v}_L \times \mathbf{B}$, such a vortex velocity $\mathbf{v}_L$ generates a Hall voltage of sign *opposite* to that of the normal state. The observed field-independence[19,20] of the sign-reversing Hall angle (in the isolated vortex limit $B \rightarrow 0$) in $Tl_2Ba_2CaCu_2O_8$ and $Bi_2Sr_2CaCu_2O_8$, which is expected in this vortex dynamics picture but not in other models, was strong evidence for such an intrinsic origin for the Hall anomaly. Nernst effect measurements in high-T_c systems confirmed[21] that the mobile vortices were of the same sign as the applied field; this rules out models in which *anti*vortices move with $\mathbf{v}_L \cdot \mathbf{v}_T > 0$ to give $\rho_{xy} < 0$.

However, almost all Hall measurements in the superconducting state have been made on materials for which $\rho_{xy}(T > T_c) > 0$; $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ is one of very few type-II systems for which $\rho_{xy}(T > T_c) < 0$. Our ρ_{xy} studies in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ provide additional support for the flux dynamics interpretation, as well as evidence against thermoelectric models for the Hall anomaly.

We measured the in-plane ρ_{xy} and ρ_{xx} on crystal and epitaxial film samples of $Nd_{1-x}Ce_xCuO_{4-y}$ with $x \approx 0.15$ ($T_c \approx 24$ K in the crystal and ≈ 21 K in films). The crystal was grown from CuO flux; details of the growth and characterization have been given elsewhere.[22] The crystal studied was $\sim 1 \times 1 \times .02$ mm^3 in size, with Hall resistivity $\rho_{xy} < 0$ in the normal state $T_c < T \leq 300$ K indicating *n*-type conduction. The films (≈ 5000 Å thick, *c* axis normal to substrate) are grown by pulsed laser deposition and patterned into an eight-lead bar configuration (width ≈ 100 μm). We measure ρ_{xy} of the *ab* planes using a superconducting solenoid with $B \perp ab$; the Hall resistivity is obtained as the part of the transverse resistivity E_y/j_x that is antisymmetric in the applied field B_z.

Figure 2 shows the resistivity and Hall resistivity

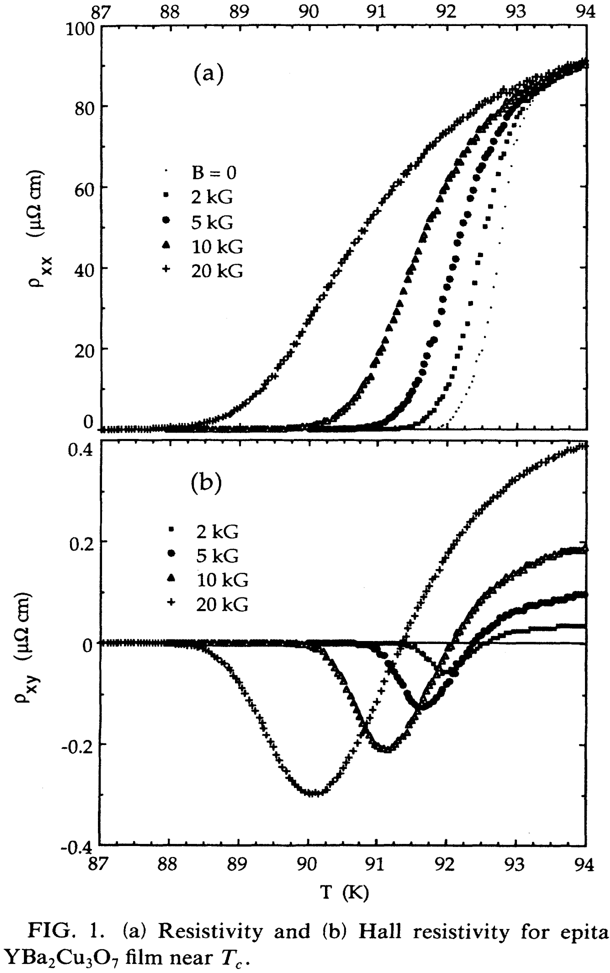

FIG. 1. (a) Resistivity and (b) Hall resistivity for epitaxial $YBa_2Cu_3O_7$ film near T_c.

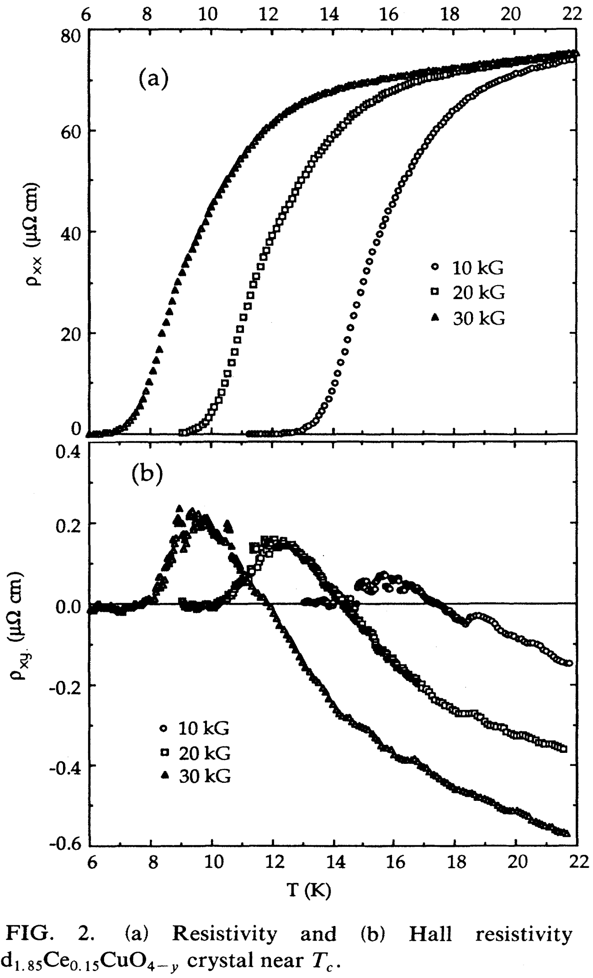

FIG. 2. (a) Resistivity and (b) Hall resistivity for $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ crystal near T_c.

versus temperature near T_c for a crystal of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ at different fields B. Above T_c the Hall resistivity is negative and linear in applied field, as expected for an n-type conductor; however, below T_c as ρ_{xx} falls to $\sim 50-70\,\%$ of its normal state value ρ_{xy} abruptly reverses to positive values and then decreases rapidly to zero at lower T. Similar behavior is seen in film samples. The Hall resistivity in this narrow temperature regime is clearly not linear in field B; increasing field suppresses the region of positive ρ_{xy} to lower temperatures and eventually quenches it entirely.

Aside from the overall sign, the Hall data for $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ is similar to that of the p-type cuprate $YBa_2Cu_3O_7$ (Figure 1). The temperature and field scales are narrower in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$, but the essential behavior of the anomalous Hall effect is clearly *independent* of the sign of the majority charge carriers. The existence of the same Hall anomaly in both n- and p-type materials provides a useful test of models for ρ_{xy}. For example, it supports a vortex dynamics interpretation. A sign reversal of ρ_{xy} for both cases indicates that vortices move opposite to the superfluid flow $\mathbf{v}_T$ rather than to the transport current **j**. In "hydrodynamical" vortex dynamics pictures the vortices should move in the same direction relative to $\mathbf{v}_T$ for the same conditions of B, T, etc., regardless of whether the current consists of electrons or holes. Thus, from the presence of a sign change of ρ_{xy} at T_c in, e.g., $YBa_2Cu_3O_7$ one would expect an anomaly also in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$.

By contrast, the thermoelectric model of Freimuth, Hohn, and Galffy[9] (based on Maki's description[11] of the Hall effect below T_c) predicts no anomaly in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$. In that picture, the sign-reversing ρ_{xy} below T_c results from a temperature gradient $\partial T/\partial y \propto -j_x$ developing transverse to j and B in the sample (through the Ettingshausen effect[23]). Because of the nonzero thermopower S of the flux-flow state, this thermal gradient in turn creates a parallel electric field $E_y = S(\partial T/\partial y) \propto -S j_x$ that appears as a Hall effect. Although the expected magnitude for E_y is small, the positive thermopower S in $YBa_2Cu_3O_7$ indicates that $\rho_{xy} = E_y/j_x \propto -S < 0$ so the Hall effect may become negative below T_c. Since the thermopower S of the $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ samples is also positive,[22] the model also predicts $\rho_{xy} < 0$ below T_c in this system; this is in contrast to the $\rho_{xy} > 0$ we observe. Thus the thermoelectric model does not explain the anomalous ρ_{xy} in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$. (A further problem with the thermoelectric model is that the required temperature gradient $\partial T/\partial y$ would be difficult to sustain in a film sample, where the high thermal conductance of the crystalline substrate will tend to lessen temperature gradients generated within the film. A much smaller anomaly is thus expected in the films, while experimentally a similar anomaly is detected in both film and crystal samples.)

If the Hall anomaly instead simply reflects the force balance on a moving flux line, one should perhaps expect sign reversals of ρ_{xy} to occur in all type-II superconductors. The fact that such behavior is absent in many systems has led some researchers to oppose the vortex dynamics interpretation of the anomaly. We find that a survey of the literature provides a possible explanation of the variation between superconductors. Table I summarizes existing Hall data for both high-T_c and low-T_c superconductors, with samples listed in rough order of increasing "cleanness" l/ξ_0. Samples of all types are included (e.g., films, bulk crystals, polycrystals, etc.). The Hall anomaly is consistently seen in the high-T_c cuprates,[5,18,24] with the curious exception[25] of the otherwise unremarkable $YBa_2Cu_4O_8$. Among low-T_c systems the sign-reversal is well established in some specimens of the elemental type-II superconductors V and Nb but is generally not seen in alloys[12,26–29] ($Ti_{1-x}Mo_x$, $Nb_{1-x}Ta_x$, etc). One can argue that sign reversal of the Hall effect is largely a high-T_c phenomenon, but the trend in the data indicates a different conclusion: The anomalous Hall behavior is pervasive among moderately clean $l/\xi_0 \sim 0.5-5$ superconductors, but does not occur in either the very clean ($l \gg \xi_0$) or dirty ($l \ll \xi_0$) limits. Thus, the sign reversal is not generally seen in the alloys; instead $\tan(\Theta_H)$ in these materials increases steeply for $B < B_{c2}$. The Hall anomaly is widely observed in the high-T_c cuprates, for which $\xi_0 \le l$ near T_c in most cases [e.g., $\xi_0 \approx 20$ Å, $l(T \approx T_c) \approx 90$ Å in[30] $YBa_2Cu_3O_7$]. In V and Nb, sample preparation can make l much larger or smaller than ξ_0 (~ 400 Å in Nb); the Hall behavior then becomes strongly sample dependent.[14–16,31]. In clean-limit materials, where $l \gg \xi_0$ [e.g., Nb with residual resistivity ratio (RRR) = 6500 and $l/\xi_0 \sim 330$], the anomaly disappears and a NV behavior $\tan(\Theta_H) \approx \tan(\Theta_H)_{c2}$ is found.[31,32] From this perspective the absence of a Hall anomaly in the cuprate $YBa_2Cu_4O_8$ is more understandable: The specimen studied had $l \sim 26$ Å ($T \approx T_c$), which is short compared to the estimated[33] $\xi_0 \sim 60$ Å.

The two exceptions[17,34] to this trend are $Pb_{1-x}In_x$, which shows a sign reversal in ρ_{xy} for low In concentrations, and α-MoGe. In $Pb_{1-x}In_x$, the constituent elements have Hall effects of opposite sign above T_c, so the complicated Hall behavior may involve two-band effects. The α-MoGe case clearly deserves closer study.

A connection between sign reversal of the Hall effect and microscopic disorder in Nb and V was noted by Usui, Ogasawara, and Yasukochi[15] and by Noto, Shinzawa, and Muto.[16] Noto, Shinzawa, and Muto, motivated by disagreements in the reported Hall behavior of Nb and V, studied the effects of sample purity on $\tan(\Theta_H)$ and found no ρ_{xy} anomaly in dirty samples ($l/\xi_0 < 0.4$). However they observed that in samples annealed to increase the electronic mean free path l (giving $0.4 \le l/\xi_0 \le 5$) a sign change anomaly did appear in the Hall effect. In the cleanest samples ($l/\xi_0 \sim 100$) the sign change was again suppressed and BS or NV behavior was observed. These authors noted that this interesting behavior is strong evidence against two-band models for the sign changes in ρ_{xy}; significant changes in the electronic structure that could reverse ρ_{xy} can hardly occur as the defect density varies over this range.

Table I shows that the findings of Noto, Shinzawa, and Muto and Usui, Ogasawara, and Yasukochi in fact apply to type-II superconductors generally: Despite widely diverse material and superconducting properties in the systems that have been studied, the single parameter l/ξ_0 largely determines whether or not the Hall anomaly is observed. Material characteristics such as sample type (crystal or film, etc.), transition temperature, layered crystal structure, or flux-pinning strength are apparently far less important to the behavior of ρ_{xy}.

The evident importance of l/ξ_0 should help to clarify the origin of the Hall anomaly. For example, the fact that the sign reversal is most familiar in high-T_c cuprates indicates not that it is essentially a high-T_c phenomenon (as some authors have implied[9,35]), but simply that few studies have been made of noncuprate type-II superconductors with $l/\xi_0 \sim 1$. Also, the data in Table I argue strongly against the theory of Wang and Ting[8] for the Hall effect, in which strong flux pinning generates the sign charge in ρ_{xy}. Clearly pinning is not the key parameter in determining the behavior of ρ_{xy}: $YBa_2Cu_3O_{7-x}$ and $Bi_2Sr_2CaCu_2O_8$ show similar Hall anomalies despite very different pinning strengths in the two materials.[36]. The pinning model gives a sign reversal of ρ_{xy} for $B > H_p$, where H_p is a critical field for the onset of flux motion in the presence of pinning: No sign reversal is expected for $H_p \approx 0$. While this prediction is qualitatively consistent with Figs. 1 and 2, sign reversal of ρ_{xy} is observed[19,20] in $Tl_2Ba_2CaCu_2O_8$ and $Bi_2Sr_2CaCu_2O_8$ at temperatures where no H_p is detected (i.e., $H_p \approx 0$). In addition, the pinning model does not reproduce the field-independent, sign-reversing $\tan(\Theta_H)$ observed[19,20] below T_c in these weak-pinning materials.

However, the importance of l/ξ_0 is entirely consistent with a vortex dynamics interpretation for the Hall effect: The size of l relative to ξ_0 is critical in vortex motion models but quite difficult to treat correctly. ξ_0 sets the scale for the size of the vortex core, while l is a measure of the carrier scattering rate within the core. Both the NV and BS models consider $\xi_0 \ll l$ but are not entirely consistent in this assumption. In the NV model, for example, the vortex core is treated as a cylinder of normal material (radius $a \sim \xi_0$) with a sharp boundary; current entering the core region loses momentum over a characteristic distance that NV assume to be $\sim v_L \tau \ll \xi_0$. However, a distance l may be more appropriate here if $l \gg \xi_0$ and would change the drag force balancing the Magnus force at the core. Similarly, the BS calculation finds a dipolelike surface charge to accumulate at the core boundary, i.e., on the scale $\ll \xi_0$; NV point out that the assumed long mean free path $l \gg \xi_0$ makes this unrealistic. By eliminating this charge layer NV obtain a significantly different ρ_{xy} from that of BS. Thus the size of l relative to ξ_0 affects key steps in the calculation of forces acting on a moving vortex, and its evident influence on the measured Hall effect should probably be understood in this context. A different treatment of l/ξ_0 in these models could result in significantly different predictions for ρ_{xy}.

Therefore we suggest that the sign reversal of ρ_{xy} results from a failure of the NV and BS models in the regime $l/\xi_0 \sim 0.5-5$ due to inappropriate approximations

TABLE I. Summary of published literature on Hall anomaly (sign reversal of ρ_{xy} upon entering superconducting state) for different type-II materials. l = estimated transport mean free path at $T \approx T_c$; ξ_0 = BCS coherence length. Where l/ξ_0 was not given it was determined from H_{c2} in the literature and a free-electron model. This introduces some uncertainty in l/ξ_0.

Material	no ρ_{xy} anomaly	ρ_{xy} anomaly	l/ξ_0	Ref.
α-MoGe		×	$\ll 1$	34
In-InO_x	×		$\ll 1$	28
$Pb_{1-x}In_x$ ($x \le 35$)		×	~ 0.01	17
$Pb_{1-x}In_x$ ($x \ge 40$)	×		~ 0.01	17
$Nb_{1-x}Ta_x$ ($0.1 \le x \le 0.9$)	×		~ 0.01	12
$Ti_{.84}Mo_{.16}$	×		~ 0.01	27
$Nb_{0.8}Mo_{0.2}$	×		$\ll 1$	26
$Nb_{0.99}Zr_{0.01}$	×		$\ll 1$	29
V	×		0.23	15
$Nd_{1.85}Ce_{0.15}CuO_4$ (film)		×	0.26	(This work)
$YBa_2Cu_4O_8$	×		0.43	25
$Nd_{1.85}Ce_{0.15}CuO_4$ (film)		×	0.54	(This work)
V		×	0.56	15
$Nd_{1.85}Ce_{0.15}CuO_4$ (film)		×	0.75	(This work)
$Nd_{1.85}Ce_{0.15}CuO_4$ (crystal)		×	1.0	(This work)
$Tl_2Ba_2CaCu_2O_8$		×	1.0–1.5	19,24
$Bi_2Sr_2CaCu_2O_8$		×	1.8	18,20
Nb		×	2.5	14
Nb		×	4.3	16
$YBa_2\ Cu_3O_7$ (R = Y, Eu, . . .)		×	4.5	18,5
V		×	3.1,4.8,5.5	16
Nb	×		5.6	16
$NbSe_2$	×		6.2	32
Nb	×		330	31

in those models. We argue that a proper treatment of vortex dynamics in this parameter range would find that steady-state motion gives $\mathbf{v}_L \cdot \mathbf{v}_T < 0$, so that a sign reversal occurs in the Hall effect. In the clean limit, however (e.g., Nb samples[31] with RRR $\sim 10^3$ or $Bi_2Sr_2CaCu_2O_8$ well below T_c), the NV model gives the correct motion $\mathbf{v}_L \cdot \mathbf{v}_T > 0$ and $\tan(\Theta_H) \approx \tan(\Theta_H)_{c2}$.

Together with the Hall angle and Nernst data[19–21] on high-T_c superconductors, our $Nd_{2-x}Ce_xCuO_{4-y}$ results and the data of Table I argue very strongly that the Hall anomaly in the mixed state of superconductors can be understood as a general consequence of vortex dynamics: Within a certain range of microscopic parameters, vortex motion antiparallel to the superfluid flow (as indicated by the sign reversal of ρ_{xy}) occurs consistently in a wide variety of superconducting materials. Models in which the anomaly results from flux pinning, thermoelectric effects, band structure, or other properties of particular materials are clearly inconsistent with much of the data, and generally do not address the parameter l/ξ_0 that plays a key role experimentally.

Although the vortex dynamics model of NV does not explain the sign reversal of ρ_{xy}, we believe that an appropriate modification of the model for the regime $l/\xi_0 \sim 1-5$ would reproduce the observed anomaly. Such modification may manifest itself as a new drag force slightly different from the NV form $\mathbf{f}_{\text{drag}} = -(nm\pi a^2/\tau)\mathbf{v}_T$: Because the Hall effect represents such a small correction to the vortex motion, the drag force strongly affects the magnitude and sign of ρ_{xy}.[5,19] In fact, there is still no consensus on the proper calculation of these drag forces. We discussed in Refs. 5 and 19 some forms for additional forces that would result in sign reversal of ρ_{xy}. Recently, Ferrell[10] has calculated from a two-fluid model an additional drag term that would generate sign reversal in the NV model. We expect that such modifications to the NV model, especially those that explicitly treat the parameter range $l/\xi_0 \sim 1$, will show that the Hall anomaly in superconductors can be seen as a consequence of vortex dynamics.

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