RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 41, NUMBER 16 1 JUNE 1990

Anomalous Hall effect in superconductors near their critical temperatures

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(Received 5 March 1990)

Measurements on epitaxial $YBa_2Cu_3O_7$ thin films show that the Hall voltage just below the superconducting transition temperature has sign opposite to the Hall voltage in the normal state. This can result either from quasiparticle effects, or from unusual vortex motion. Because we observe a similar effect in Nb thin films, we argue that vortex motion is responsible. A simple interpretation of the Hall effect then requires that the vortex velocity have a component *antiparallel* to the transport current. This motion, while contrary to existing flux flow models, could result from vortex motion damping similar to that expected in superfluid ^{4}He.

Transport measurements[1,2] on the high-temperature superconductors $RBa_2Cu_3O_7$ (R=Y,Er) and $Bi_2Sr_2CaCu_2O_8$ have shown an unexpected reversal of the sign of the Hall voltage V_H at temperatures below the superconducting transition. Although the Hall voltage in the normal state is positive (i.e., holelike) and linear in magnetic field, V_H in the mixed state is negative for small field and positive for larger field, in contradiction of microscopic[3] and flux flow[4,5] theories for the Hall effect in superconductors. Our measurements on $YBa_2Cu_3O_7$ and Nb thin films provide evidence that this sign reversal below T_c results from vortex motion with a velocity component *opposite* to the direction of the superfluid transport current.

We measured the Hall resistivity ρ_{xy} and magnetoresistivity ρ_{xx} of thin films of epitaxial c-axis-oriented $YBa_2Cu_3O_7$ (thickness = 1500 Å) in magnetic fields up to 70 kG oriented perpendicular to the film. The films were dc magnetron sputtered from a stoichiometric target onto rotating substrates of (100) $SrTiO_3$, MgO, $LaAlO_3$, or cubic zirconia. A layer of Au sputtered onto the leads *in situ* provided low-resistance (~ 1 Ω) contacts; the films were then etched into an eight-lead Hall bar pattern (bar area 50 μm × 3.9 mm). The films had $T_c \sim 88$–90 K (midpoint) and $\rho_{xx}(300\ \mathrm{K}) = 450$–$750$ $\mu\Omega$ cm. We measured Hall resistance and magnetoresistance both dc (with current reversal) and ac (phase sensitive at 100 Hz) at current densities $J \sim 10^2$–10^4 A/cm^2, obtaining the Hall resistance from the transverse resistance by subtracting the positive and negative magnetic-field data.

Figure 1 shows typical ρ_{xx} and ρ_{xy} data as a function of field at $T < T_c$. Although ρ_{xy} is nearly temperature independent just above T_c, it decreases to negative values in the superconducting state, eventually becoming zero as temperature or field decreases (Fig. 2). Note that ρ_{xy} changes sign while ρ_{xx} is still close to its normal-state value. Higher fields drive the region of negative ρ_{xy} to lower temperatures (inset of Fig. 1).

Figure 2 shows ρ_{xy} in the immediate neighborhood of T_c(=90 K). At low field, ρ_{xy} becomes negative at tem-

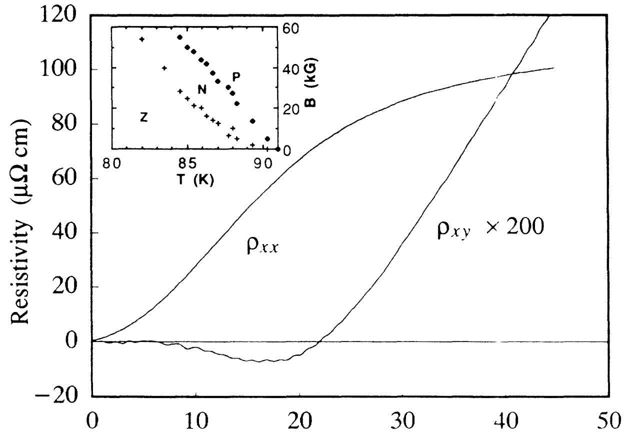

FIG. 1. Hall resistivity ρ_{xy} and longitudinal resistivity ρ_{xx} vs magnetic field in $YBa_2Cu_3O_7$ film at T=86.0 K. Inset: Field and temperature regimes where ρ_{xy} is positive (P), negative (N), and zero (Z) for a film with T_c(midpoint)≅89 K.

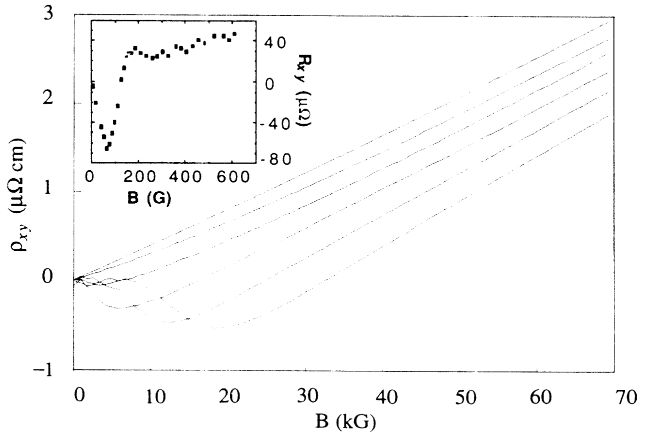

FIG. 2. Hall resistivity ρ_{xy} vs magnetic field in $YBa_2Cu_3O_7$ near T_c (≅90 K) showing linearity at high field. Temperatures from top to bottom are 93.0, 91.5, 90.5, 89.8, 89.1, and 88.4 K. Inset: Hall resistance of Nb film vs field at T=9.16 K.

atures close to T_c; positive ρ_{xy} is then recovered at high field. However, for fields exceeding $H_{c2}(T)$ the full normal state ρ_{xy} is not observed. From the upper critical field slope of -19 kG/K obtained by Welp *et al.*,[6] it is clear that the sample should no longer be in the superconducting state at $B \sim 70$ kG and $(T_c - T) \sim 2$ K. However, $\rho_{xy}(T < T_c)$ remains less than $\rho_{xy}(T > T_c)$ at high fields $H \gg H_{c2}$, even though the slope $d\rho_{xy}/dH$ approaches its normal-state value.

Sign reversal of ρ_{xy} in the mixed state was observed by previous authors[1,2] and attributed to grain effects, possible two-carrier quasiparticle effects, or conventional flux motion. In low-T_c superconductors, many different behaviors were observed in the Hall effect below T_c. Sign reversal, although not always observed, was found in some V foils[7] and In-Pb alloys.[8] Existing flux flow[4,5] and microscopic[3] theories for the Hall effect in the mixed state predict no sign change and therefore do not account for this data.

In order to study the Hall effect in well-characterized samples of the low-T_c superconductor, we prepared Nb thin films. The films (1300 Å thick) were prepared by dc sputtering onto sapphire substrates and etched into the Hall bar pattern. Their resistivity at $T = 273$ K was 13 $\mu\Omega$ cm (cf. bulk value ~ 12.5 $\mu\Omega$ cm), with $\rho_{xx}(300\ \mathrm{K})/\rho_{xx}(9.5\ \mathrm{K}) = 45$ and $T_c = 9.25$ K. The Hall resistivity for $T > T_c$ and $H > H_{c2}$ is positive and linear in field, but for $T < T_c$ and $H < H_{c2}$ we find ρ_{xy} reverses to negative values just as in $\mathrm{YBa_2Cu_3O_7}$ (inset of Fig. 2). Thus the reversal occurs in both low-temperature elemental superconductors and high-temperature oxide superconductors, whose electronic properties are very different, which strongly suggests that the phenomenon is a general property of the vortex state.

Josephson[9] demonstrated that the motion of flux vortices at a velocity $\mathbf{v}_L$ through a superconductor produces an electric field given by Faraday's law,

$$\mathbf{E} = -\frac{n}{c}\mathbf{v}_L \times \bar{\phi}_0 = -\frac{\mathbf{v}_L \times \mathbf{H}}{c}, \tag{1}$$

where n is the areal density of vortices and $\bar{\phi}_0 = \phi_0 \mathbf{H}/H$, so that flux flow in the direction of $\mathbf{J} \times \bar{\phi}_0$ [Fig. 3(a)] generates a field $\mathbf{E}$ parallel to the transport current $\mathbf{J}$ and therefore dissipates energy. Similarly, a component of flux motion along $\mathbf{J}$ produces a transverse field parallel to $-\mathbf{J} \times \bar{\phi}_0$; if the charge carriers are of the same sign in the normal and superconducting states (as expected for a BCS superconductor), this field is observed as a Hall voltage of the *same* sign as in the normal metal [Fig. 3(b)]. Thus a reversal of the sign of the Hall voltage upon entering the mixed state indicates either that the sign of the charge carriers has changed or that the flux-line velocity $\mathbf{v}_L$ has a component *opposite* to the direction of the transport current. Because it is difficult to see why the charge carriers should change sign in the superconducting state not only in copper-oxide superconductors but also in elemental superconductors like Nb and V, we argue that this unexpected flux motion is in fact a more likely explanation of the negative Hall effect near T_c.

In standard models of flux motion,[4,5] the transport current $\mathbf{J}$ produces a force $\mathbf{F}$ that drives the flux motion.

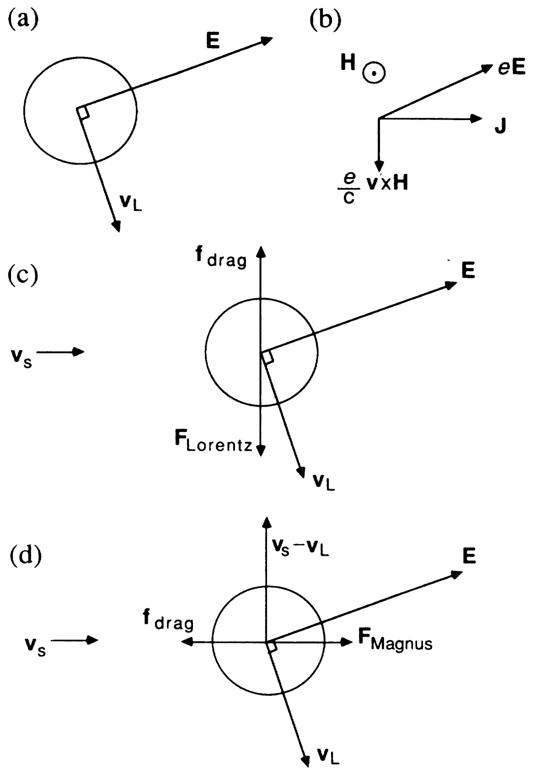

FIG. 3. (a) Electric field generated by vortex motion [Eq. (1)]. (b) Lorentz force $(e/c)\mathbf{v} \times \mathbf{H}$ on free charge carrier $(q = +e)$ balanced by electric field $\mathbf{E}$ to give the normal-state Hall effect. Field $\mathbf{H}$ is directed out of the page. (c) Forces on vortex core in Bardeen-Stephen model for flux flow. The Lorentz force $\mathbf{J} \times \bar{\phi}_0/c$ is balanced by an opposing viscous force. (d) Forces on vortex core in Nozieres-Vinen model. The Magnus force $n_s e(\mathbf{v}_s - \mathbf{v}_L) \times \bar{\phi}_0/c$ is balanced by a drag force antiparallel to the superfluid velocity $\mathbf{v}_s$.

This force can be balanced by a frictional force $\mathbf{f}$ so that the net force $\mathbf{F} + \mathbf{f} = 0$. In the Bardeen-Stephen model,[5] the driving force per unit length is the Lorentz force $\mathbf{F} = \mathbf{J} \times \bar{\phi}_0/c$; the frictional force is assumed to act in the opposite direction [Fig. 3(c)]. Normal electrons passing through the vortex cores experience the usual magnetic force and a Hall voltage results just as in the normal state. Thus the vortex velocity has a small component parallel to $\mathbf{J}$ and the Hall effect in flux flow is of the same sign as in the normal state.

In the Nozieres-Vinen model,[4] the force on a vortex is the Magnus force

$$\mathbf{F} = \frac{n_s e}{c}(\mathbf{v}_s - \mathbf{v}_L) \times \bar{\phi}_0, \tag{2}$$

where n_s is the superfluid electron density and $\mathbf{v}_s = \mathbf{J}/n_s e$ is the superfluid velocity. If $\mathbf{f} = 0$, the vortices move with $\mathbf{v}_L = \mathbf{v}_s$ and produce a transverse field $\mathbf{E} = -\mathbf{v}_L \times \mathbf{H}/c = -\mathbf{J} \times \mathbf{H}/n_s e c$ in the same direction as for the normal-state Hall effect. A frictional drag introduced through $\mathbf{f} \propto -\mathbf{v}_s$ does not affect the sign of the Hall effect [Fig. 3(d)].

Because neither of these models can produce a sign change in the Hall effect near T_c, we suggest an analogy to the case of vortex motion in superfluid ^{4}He. In superfluid ^{4}He, relative motion between the vortices and the superfluid produces a Magnus force

$$\mathbf{F}=\frac{\rho_s h}{m}(\mathbf{v}_s-\mathbf{v}_L)\times\mathbf{z}\,, \qquad (3)$$

where ρ_s is the superfluid density and $\mathbf{z}$ is a unit vector indicating the vortex circulation. Hall and Vinen, and Ambegaokar, Halperin, Nelson, and Siggia (AHNS) discuss a general drag force of the form[10]

$$\mathbf{f}=-\eta\mathbf{v}_L-\eta'\mathbf{z}\times\mathbf{v}_L\,. \qquad (4)$$

The condition $\mathbf{F}+\mathbf{f}=0$ then produces an equation of motion

$$\mathbf{v}_L=\frac{h\rho_s}{m}D\mathbf{v}_s\times\mathbf{z}+C\mathbf{v}_s\,. \qquad (5)$$

Here

$$D=\frac{\eta}{(h\rho_s/m-\eta')^2+\eta^2}\,, \qquad (6a)$$

$$C=\frac{h\rho_s}{m}\,\frac{h\rho_s/m-\eta'}{(h\rho_s/m-\eta')^2+\eta^2}\,. \qquad (6b)$$

AHNS note that in the limit of no drag forces $\eta=\eta'=0$ and the coefficients $D=0$ and $C=1$. The vortices then flow with the superfluid and at the same velocity, as in the Nozieres-Vinen superconductor with $\mathbf{f}=0$. However, we observe that for large damping, if $\eta'>h\rho_s/m$, we have $C<0$ and $\mathbf{v}_L$ has a component *opposite* to the direction of the superfluid flow. Such motion is consistent with energy conservation, as the force in η' does no work. Therefore a similar damping force

$$\mathbf{f}=-\eta\mathbf{v}_L-\eta'\bar{\phi}_0\times\mathbf{v}_L \qquad (7)$$

acting against the Magnus force in a superconductor would cause "upstream" vortex motion for $\eta'>n_se/c$ and reverse the sign of the Hall voltage.

Therefore we propose that under weak pinning the vortices in the mixed state of a superconductor move under the influence of the Magnus force (2) described by Nozieres and Vinen and a frictional force (7). Near the superconducting transition the number of superconducting electrons is small and the drag term in η' may be large enough to produce a flux velocity component opposite to the direction of the transport current: thus the Hall effect has sign opposite to that of the normal state. [We note that a Lorentz force $\mathbf{F}=\mathbf{J}\times\bar{\phi}_0/c$ combined with a frictional force (7) will produce a sign change in ρ_{xy} for any $\eta'>0$.] If the flux lines become pinned to the crystal lattice the vortex motion slows and the Hall voltage vanishes. This picture is consistent with Fig. 1, where ρ_{xx} is close to zero at the field where ρ_{xy} vanishes. We note that the temperatures and fields at which ρ_{xy} disappears in $YBa_2Cu_3O_7$ are comparable[11] to those of the "irreversibility" or "melting" lines[12] observed in other measurements on this material. Thus the appearance of nonzero Hall voltage may mark an increase in flux motion in $YBa_2Cu_3O_7$. By contrast, at high field or temperature, large numbers of quasiparticles will be excited and contribute a positive Hall effect, driving the Hall voltage positive towards its normal-state value. In the case of copper-oxide superconductors, fluctuations[2] persisting to high fields and temperatures may prevent the Hall voltage from fully recovering its normal-state (linear in H) behavior. Thus the fact that $\rho_{xy}(T\leq T_c)$ in $YBa_2Cu_3O_7$ becomes positive and linear at high fields but remains offset from its normal-state value (Fig. 2) can be attributed to the presence of strong fluctuations.

In summary, we suggest that the sign reversal of the Hall effect observed near the superconducting transition in both the high-temperature copper-oxide superconductors and some low-temperature superconducting metals indicates a component of flux-flow velocity *opposite* to the direction of the superfluid transport current.[13] Such a velocity component could result from vortex-motion damping resembling that in superfluid ^{4}He.

We acknowledge useful discussions with D. R. Nelson, who suggested the analogy between vortex motion in thin ^{4}He films and superconductors, and with S. Yip. We also received assistance with high-T_c film growth from J. Talvacchio and J. R. Gavaler, and programming assistance from W. Jiang. Sample fabrication at Westinghouse was supported by U.S. Air Force Office of Scientific Research Contract No. F49620-88-C-0039.

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[12] See, for example, *Strong Correlation and Superconductivity: The Proceedings of the IBM Japan International Symposium,* edited by H. Fukuyama, S. Maekawa, and A. P. Malozemoff (Springer-Verlag, Berlin, Heidelberg, 1989), and references therein.

[13] While this manuscript was being completed, we received a

preprint on the Hall effect in the mixed state of $YBa_2Cu_3O_7$ by T. R. Chien, N. P. Ong, and Z. Z. Wang. These authors emphasize the regime of positive Hall voltage, which they attribute to flux flow, but assert that negative Hall voltages are due to a transition either to flux creep or else to exotic processes such as the nucleation of vortex loop excitations. While we agree that a positive Hall voltage can result within the conventional flux flow model of Nozieres and Vinen, resistivity and magnetization measurements have shown that the fields and temperatures at which the negative Hall effect appears are more likely characterized by flux flow than flux creep. Also, while a transition from flux flow to flux creep must affect the rate of vortex motion, it should not appreciably change the forces acting on a moving vortex or (consequently) the *direction* of the resulting motion. Thus one should not expect a sign change in the Hall voltage to accompany such a transition.