PHYSICAL REVIEW B

## Sign change of the flux-flow Hall resistance in high- $T_c$ superconductors

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We consider a contribution to the flux-flow Hall effect that arises from the existence of large thermomagnetic effects in the mixed state of the high- $T_c$  superconductors. From the measured values of the Seebeck and Nernst-Ettingshausen effects we find that this contribution is as large as the direct flux-flow contribution to the Hall effect, but has the opposite sign for a positive Seebeck coefficient. Therefore it leads to a sign change of the Hall resistance. A calculation of the temperature and field dependence of the Hall resistivity yields good agreement with the data.

In the mixed state of the Bi- and Y-based high- $T_c$  superconductors (HTSC) the Hall resistance  $\rho_H$  shows a change of sign at low-magnetic fields and temperatures near  $T_c$ .<sup>1,2</sup> This behavior is not yet understood. In particular an explanation in terms of vortex dynamics seems difficult, since the change of sign requires a reversion of the direction of motion of the vortices.

The usual flux-flow scenario for the occurrence of a Hall voltage is as follows: With the magnetic field  $B_z$  in the z direction, an electrical current  $j_x$  in the x direction leads to vortex motion at velocity  $v_{\phi}$  at the Hall angle in the y direction (e.g., Refs. 3 and 4). As a result an induced electrical field  $\mathbf{E} = -\mathbf{v}_{\phi} \times \mathbf{B}$  occurs, which has a large longitudinal resistive component  $E_x$  and a small transverse Hall component  $E_y$ . The Hall angle is defined as  $\tan \alpha \equiv E_y/E_x$  and the Hall resistance  $\rho_H$  is given as  $\rho_H = \rho_s \tan \alpha$ , where  $\rho_s$  is the resistivity in the mixed state. We mention that for simplicity we neglect the influence of pinning throughout the paper.

There are various theories for the behavior of the Hall angle in the case of flux motion.<sup>5-9</sup> Whereas the field dependence of  $\tan \alpha$  is different in these theories, all agree that the order of magnitude and the sign of the Hall angle are the same in the mixed state and in the normal state, i.e., one expects

$$\tan \alpha \approx \tan \alpha_n \,. \tag{1}$$

 $\alpha_n$  is the Hall angle in the normal state, which is typically of order  $\tan \alpha_n \approx 10^{-3}$ . Accordingly for the Hall resistance one does not expect a change of sign in the mixed state in general.

There have been attempts to explain the sign change of  $\rho_H$  on the basis of the conventional models<sup>5–7</sup> by considering a two-carrier system with magnetic field-dependent electrical conductivities (effective masses) of electrons and holes<sup>10</sup> and also by modifying the equation of motion of the vortices in analogy to the dynamics of vortices in superfluid HeII.<sup>11</sup> A contribution to the Hall resistance, in addition to the vortex contribution, due to the dynamics of normal quasiparticles has been considered in Ref. 12. In this paper we want to discuss another possibility, which arises from the existence of large thermomagnetic effects in the HTSC.<sup>13–20</sup>

We consider a dc measurement of the Hall effect with adiabatic conditions: Vortices moving in the y direction

carry with them an entropy  $s_{\phi}$  (per unit length) due to the normal excitations in the vortex cores. This leads to a temperature gradient  $(\nabla T)_y$  in the y direction (Ettingshausen effect), which is large in the mixed state of superconductors (e.g., Refs. 3, 4, and 14).  $(\nabla T)_y$  in turn leads to a voltage in y direction via the Seebeck effect. This voltage may be large in the HTSC: In contrast to conventional superconductors, where the Seebeck effect in the mixed state is negligibly small, in the HTSC a large Seebeck effect in the range  $S \sim 1-10 \ \mu V/K$  is found.<sup>15,16,20</sup> As a rough estimate a transverse temperature gradient of order  $10^{-2}$  K will lead to an additional voltage in the y direction of order 0.01-0.1  $\mu V$ , which is comparable to Hall voltages.

We shall now give a more quantitative estimate of this contribution to the Hall effect, which allows comparison with experiment. The heat current  $j_{h,y}^{k}$  associated with the vortex flow in the y direction is given as

$$j_{h,y}^{v} = n_{\phi} s_{\phi} v_{\phi,y} T , \qquad (2)$$

where  $n_{\phi} = B/\Phi_0$  is the vortex density and  $\Phi_0$  is the flux quantum. The velocity  $v_{\phi,y}$  of vortex motion is related to the flux-flow resistivity  $\rho_s$  via the induced electrical field  $E_x$ 

$$v_{\phi,y} = -\frac{E_x}{B} = -\frac{j_x \rho_x}{B}.$$
 (3)

In a stationary state  $j_{h,y}^{\varepsilon}$  is compensated by normal heat conduction  $j_{h,y}^{n} = -\kappa_{s} (\nabla T)_{y}$ , i.e.,

$$j_{h,y}^{v} = -j_{h,y}^{n} = \kappa_{s} (\nabla T)_{y}, \qquad (4)$$

where  $\kappa_s$  is the thermal conductivity.  $(\nabla T)_y$  leads to an electrical field in the y direction

$$E_{v} = S_{s} (\nabla T)_{v} , \qquad (5)$$

where  $S_s$  is the Seebeck coefficient in the mixed state. Combining Eqs. (2)-(5) we find a contribution  $\tan \alpha_{\epsilon}$  to the Hall angle

$$\tan \alpha_{\epsilon} \equiv \frac{E_{y}}{E_{x}} = \frac{E_{y}}{j_{x}\rho_{s}} = -\frac{S_{s}}{\kappa_{s}} \frac{s_{\phi}}{\Phi_{0}} T.$$
 (6)

Assuming that the direct contribution to the Hall angle is given by the normal-state Hall angle  $\tan \alpha_n$  we find

$$\tan \alpha = \tan \alpha_n - \frac{S_s}{\kappa_s} \frac{s_\phi}{\Phi_0} T \tag{7}$$

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for the Hall angle and

$$\rho_H = \rho_s \tan \alpha = \rho_s \left[ \tan \alpha_n - \frac{S_s}{\kappa_s} \frac{s_\phi}{\Phi_0} T \right]$$
(8)

for the Hall resistance in the mixed state.

The two contributions to the Hall angle are of comparable magnitude. For YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> the maximum value of the transport entropy is of order  $s_{\phi} \sim 10^{-13}$  J/Km for epitaxial films and single crystals.<sup>14,16-18</sup> The thermal conductivity is of order  $\kappa_s \sim 10$  W/Km again for a single crystal.<sup>14</sup> The Seebeck coefficient of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> for epitaxial films is of order  $S_s \approx 10^{-6}$  V/K.<sup>16,21</sup> With these numbers one finds  $\tan \alpha_{\epsilon} \approx 10^{-3}$ , i.e.,  $\tan \alpha_{\epsilon}$  is as large as  $\tan \alpha_n$ . For Bi<sub>2</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> there are less data on thermomagnetic effects. However, the order of magnitude of  $\tan \alpha_{\epsilon}$  should be about the same.

For a positive Seebeck coefficient—as found for Bi<sub>2</sub>-Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> (Ref. 15) and also for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (Refs. 16 and 21)—the two contributions to the Hall resistivity have a different sign and, therefore, a sign change of  $\rho_H$  may occur.

For a calculation of the field and temperature dependence of the Hall resistance from available experimental data we note that the transport entropy is related to the Nernst coefficient Q and to the magnetoresistivity via<sup>4</sup>

$$\frac{s_{\phi}}{\Phi_0} = \frac{QB}{\rho_s} \,. \tag{9}$$

With this we rewrite Eq. (8) as

$$\rho_H = \rho_s \tan \alpha_n - QBT \frac{S_s}{\kappa_s} \,. \tag{10}$$

In Fig. 1 we show calculations of  $\rho_H$  as a function of temperature for three values of the magnetic field using the data for Nernst and Seebeck effects, resistivity, and Hall effect of the same polycrystalline Bi-based HTSC as reported in Refs. 2 and 15. The solid lines are the measured data and the symbols are the calculated values according to Eq. (10).  $\tan \alpha_n$  and  $\kappa_s$  have been used as fitting parameters. In Fig. 2 we show a similar analysis for an epitaxial film of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> using the data of Refs. 16 and 17. One observes good agreement between the measurement and the calculation. We note that with increasing magnetic field  $\tan \alpha_n$  becomes larger than  $\tan \alpha_{\epsilon}$ . Equation (10) then no longer yields a change of sign for the Hall resistance, in agreement with the experiment.<sup>1,2</sup>

In Fig. 3 we show  $\tan \alpha_n$  as obtained from the fits in Fig. 1 as a function of the magnetic field. We find absolute values of a few times  $10^{-4}$ , which is reasonable at low-magnetic fields.  $\tan \alpha_n$  increases with the magnetic field, as expected. We find  $\tan \alpha_n \approx B^{2/3}$ .

The values obtained for  $\kappa_s$  from the fits are of order  $10^{-2}$  W/Km for Bi<sub>2</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> and of order 0.2 W/Km for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. This is about 1-2 orders of magnitude smaller than expected from direct measurements.<sup>14,22</sup> One reason for this may be that the electronic thermal conductivity rather than the overall thermal conductivity is relevant in our analysis. We shall discuss this below. Another reason may be the experimental uncertainty in



FIG. 1. The Hall resistance of a polycrystalline sample of  $Bi_2Sr_2Ca_2Cu_3O_x$  as a function of temperature for fixed magnetic fields B = 0.1, 0.5, and 1 T. The curves are shifted by four units along the y axis. The solid lines are the experimental data. The symbols are calculated from Eq. (10). Data are taken from Refs. 2 and 15.

the absolute values of the transport entropy and the Nernst coefficient. The values of the Nernst coefficient reported in the literature so far, scatter by about 2-3 orders of magnitude [e.g.,  $Q \sim 40 \text{ nV/KT}$ ;<sup>19</sup>  $Q \sim 5 \times 10^3 \text{ nV/KT}$  (Ref. 17) for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-s</sub>]. Furthermore, we note that when using Eq. (9) to calculate  $s_{\phi}$  it is assumed that  $\rho_s$  is



FIG. 2. The Hall resistance of an epitaxial film of YBa<sub>2</sub>-Cu<sub>3</sub>O<sub>7- $\delta$ </sub> as a function of temperature for B=3 T. The solid line is the experimental data. The symbols are calculated from Eq. (10) (see text). Data are taken from Refs. 16 and 17. The thermopower S<sub>s</sub> has been calculated via S<sub>s</sub> = S<sub>n</sub>( $\rho_s/\rho_n$ ) (Refs. 16, 20, and 24) from the magnetoresistance  $\rho_s$  and normal-state resistance  $\rho_n$  reported in Ref. 17. The normal-state thermopower has been taken from Ref. 16.

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10 398



FIG. 3. Logarithmic plot of the normal state Hall angle  $\tan \alpha_n$  as a function of the magnetic field as extracted from fits of the Hall resistance. The solid line is a guide to the eye. One finds that  $\tan \alpha_n$  scales rather well according to  $\tan \alpha_n \propto B^{2/3}$ .

entirely due to vortex motion. However, if there are additional channels of dissipation due to, e.g., weak links in a granular sample<sup>16,18</sup> the measured resistivity, which is commonly used in Eq. (9), is larger than the true flux-flow resistivity. Therefore  $s_{\phi}$  is underestimated, typically by 1-2 orders of magnitude as shown in Ref. 18. It is in favor of this interpretation that small values of  $s_{\phi}$  are found in samples that show a rather wide resistive transition, i.e., in which inhomogeneity plays an important role.<sup>16,18</sup> As a result of these complications it is difficult to extract reliable values of  $\kappa_s$  from the fits: Especially any underestimate of  $s_{\phi}$  leads to values of  $\kappa_s$  which are too small. Thus further measurements of the thermomagnetic effects are desirable.

We point out that a test of the interpretation of the Hall resistance presented here would be a systematic investigation of the Nernst effect, Seebeck effect, resistivity, and Hall effect of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> as a function of oxygen content. Since the Seebeck coefficient is negative for oxygen content very close to 7 and positive otherwise<sup>21</sup> tan $\alpha_{\epsilon}$  would be positive for samples with negative Seebeck coefficient and thus no change of sign of the Hall resistance should occur.

So far we have considered the behavior of the Hall resistance only below  $T_c$ . However, the present interpretation is also relevant at  $T_c$  and in a range of about 10 K above  $T_c$ , since in this temperature range the Nernst-Ettingshausen effect (and of course also the Seebeck

effect) are large in the HTSC, the former due to (presumably critical) fluctuations.<sup>23</sup>

We comment finally on a possible problem of our interpretation: Measurements of the Hall effect (e.g., the ones presented in Fig. 1) are quite frequently carried through as ac measurements at low frequencies of order  $10^2$  Hz. These measurements yield the same results as dc measurements. However, with increasing frequencies of the ac current one expects  $\tan \alpha_{\epsilon}$  to vanish, since the transverse temperature gradient should then approach zero. On the other hand the typical velocity of vortex motion in these experiments near  $T_c$  is of order  $v_{\phi,y} \approx 1-10$  cm/s. With a typical measuring frequency of about 10<sup>2</sup> Hz this corresponds to a distance of order of mm per period traveled by the vortices. This is comparable to the width of the samples. Therefore the measurements may be considered as quasi-dc-measurements. Furthermore, we point out that a true temperature gradient over the entire sample might not be necessary. It could be sufficient that local variations in temperature (or chemical potential) exist to give rise to a Seebeck effect. We note here that the Seebeck effect results from the dynamics of normal quasiparticles not bound to the vortex cores.<sup>16,24</sup> The oscillating motion of the vortex cores leads to oscillating heat transport, to which these quasiparticles may respond fast, i.e., on a time scale given by the Fermi velocity instead of the sound velocity. It should not be necessary that the oscillating heat is delivered to the lattice first and shows up as a measurable temperature gradient-in other words: adiabatic conditions may not be necessary. In this case, however, it seems more appropriate to use the electronic thermal conductivity  $\kappa_{el}$  of the normal quasiparticles instead of the overall thermal conductivity  $\kappa_s$  for the calculation of  $\tan \alpha_{\epsilon}$ . Since  $\kappa_{el}$  is at least 1 order of magnitude smaller than  $\kappa_s$  this may be a reason for the small values of  $\kappa$  coming out of our calculations.

In summary we have presented some evidence that the sign change observed in the Hall resistance in the Y and Bi-based HTSC may be due to an additional contribution to the Hall voltage arising from the large Nernst-Ettingshausen and Seebeck effects found for the HTSC. We point out that this contribution arises from the dynamics of normal quasiparticles, which determines the Seebeck coefficient in the HTSC. Further experiments should be carried through to check the validity of this interpretation.

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