Double sign reversal of the Hall effect in the mixed state of $YBa_2Cu_3O_x$

K. Nakao, K. Hayashi, T. Utagawa, Y. Enomoto, and N. Koshizuka
Superconductivity Research Laboratory, International Superconductivity Technology Center, 10-13 Shinonome 1-chome, Koto-ku, Tokyo 135, Japan
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The Hall effect in a $YBa_2Cu_3O_x$ thin film was measured using high current densities of the order of 10^6 A/cm^2, and the second sign reversal was observed clearly in the irreversible regime. The shape of the boundary between the positive and the negative Hall effect plotted on a B-T plane is similar to those of Bi- and Tl-based materials reported previously. Feigel'man *et al.*'s theory reproduces the experimental results qualitatively. [S0163-1829(98)04814-0]

According to the conventional models of the vortex dynamics,[1,2] the Hall effect in the mixed state of superconductors should have the same sign as that in the normal state. However, many high-T_c materials exhibit a sign reversal of the Hall effect near T_c. Although the origin of the sign reversal has not been clarified yet,[3–8] the mechanism seems not to be related to peculiarities of oxide high-T_c materials, because the sign reversal has also been observed for low-T_c materials.[9–11]

In addition to the sign reversal near T_c, a second sign reversal has been observed in $Bi_2Sr_2CaCu_2O_x$,[12] $Tl_2Ba_2CaCu_2O_8$,[4,13] $(BiPb)_2Sr_2Ca_2Cu_3O_d$,[14] $Tl_2Ba_2Ca_2Cu_3O_d$,[14] and $HgBa_2CaCu_2O_{6+d}$.[15] In these materials, the sign of the Hall effect is positive in the normal state and changes to negative near T_c and becomes positive again at a lower temperature that depends on magnetic field. In the case of $YBa_2Cu_3O_x$ (YBCO), however, such behavior has not been studied systematically so far, although some data in the literature indicate the existence of the second sign reversal.[16–18] One possible reason for this difference is that, in YBCO, the second sign reversal is masked by the pinning effect that is so strong at temperatures where it should take place.[19] In order to investigate this possibility, we must explore the irreversible regime using high current densities larger than the critical current densities.

There have been some pioneering works in the nonlinear region of the Hall effect for YBCO. Kunchur *et al.*[16] measured the Hall effect just below T_c using high current densities up to 0.7 MA/cm^2. Wöltgens and co-workers[20] observed the scaling relation between the longitudinal and the Hall voltages in the region of the negative Hall effect. Both groups, however, did not search for the second sign reversal at lower temperatures. In the present work, we measured the Hall effect in a YBCO thin film in the nonlinear region using pulsed high currents and searched for the possible second sign reversal.

Films were grown on MgO substrates by a laser ablation technique. The c axis was oriented perpendicular to the film surface, and the thickness was 200 nm. The superconducting transition temperature T_c was 87 K. Films were patterned photolithographically into a Hall bar, whose width was 100 μm. Magnetic fields were applied along the c axis. Figure 1 shows the temperature dependence of the Hall resistivities measured by a conventional technique. The Hall resistivity exhibits sign reversals and stays negative until it becomes smaller than our experimental resolution at lower temperatures, which is the typical behavior of YBCO.

For the measurements with large current densities in the irreversible regime, we used triangular current pulses whose width was as short as 33 μs in order to avoid excessive heating. For such a short current pulse, the total dissipation induces negligible temperature increase if the generated heat is absorbed by the substrate that has a large heat capacity. In this situation the temperature increase of the film is mainly controled by the thermal boundary resistance, which exists between the film and the substrate.[21] It is known that the thermal boundary resistance of YBCO films is nearly independent of temperature and the substrate material.[22] Therefore, we can rather easily estimate the temperature increase during the current pulse as was done in Ref. 21. Before each measurement of the Hall voltage, we measured the longitudinal voltage in the same experimental condition and estimated the temperature increase. The maximum temperature increase we allowed in the present work was 0.2 K.

In order to achieve the sufficient signal to noise ratio, measurements were repeated with a low frequency (4–20 Hz) and 1000 pulses were recorded and averaged in a digital oscilloscope (Nicolet Pro92). Measurements were performed in positive and negative magnetic fields and in normal and reversed current directions. The true Hall voltage was calculated from a combination of these four records, which can be symbolically expressed as

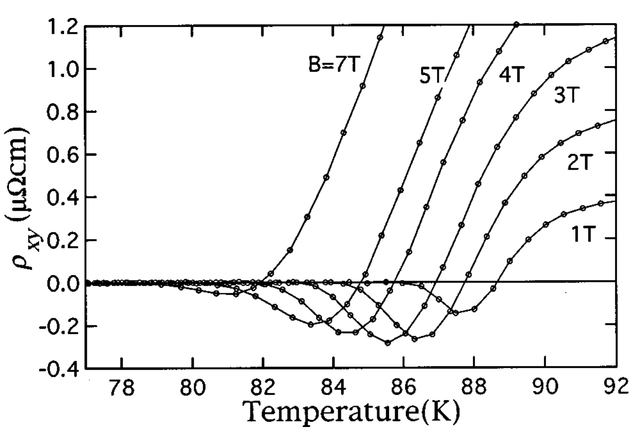

FIG. 1. Hall resistivity as a function of temperature in 1, 2, 3, 4, 5, and 7 T, measured by a conventional technique.

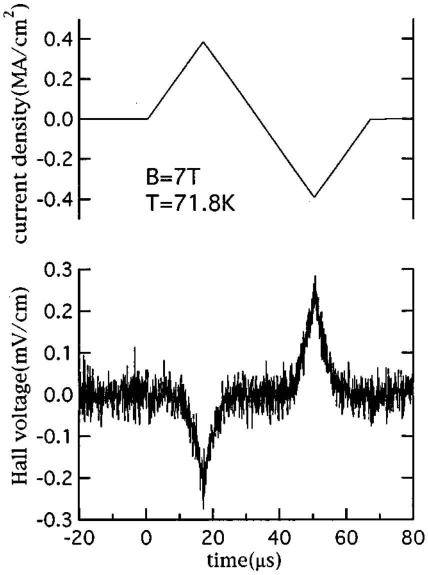

FIG. 2. An example of the measurement using a pulsed high current.

$$V_{xy}=(\tfrac{1}{4})[V_{xy}(+B,+I)-V_{xy}(+B,-I)-V_{xy}(-B,+I)+V_{xy}(-B,-I)]. \tag{1}$$

The reversal of the magnetic field was definitely necessary in order to cancel the residual longitudinal voltage because the typical Hall angle in the irreversible regime was only $\sim 10^{-4}$. The reversal of the current turned out to be very effective to cancel out spurious voltages due to the possible motion of the sample and/or the voltage lead wires induced by a strong Lorentz force exerted on a large current. It was crucial to perform the measurements of four components in Eq. (1) in exactly constant experimental circumstances, especially the temperature, in order to achieve reproducible results. Temperature was stabilized within ± 30 mK. Note that the possible temperature increase up to 0.2 K mentioned above does not harm the data quality provided the four components in Eq. (1) are measured in exactly the same conditions.

Figure 2 shows an example of the measurement. One period of the triangular current wave form, rather than a half period, was used for the purpose of confirming the performance of the measuring system. It was verified that the first and the second half of the current wave form gave a consistent result.

Figure 3(a) shows the Hall voltages in 7 T plotted against the current density at several temperatures in the irreversible regime. The current-voltage relations are strongly nonlinear, but they are monotonic. Therefore, the sign of the Hall effect can be defined without any ambiguity at each temperature. A sign reversal takes place at about 67.6 K. This is the second sign reversal back to the positive Hall effect. The results of the measurements in 5 T are shown in Fig. 3(b). The evolution of the Hall voltage is very similar to that in 7 T, except that the second sign reversal takes place at a lower temperature in 5 T. The same measurements were also performed in 3, 4, and 6 T.

Thus defined sign reversals as well as the first sign reversals observed by a conventional technique in the reversible regime are plotted on a B-T plane in Fig. 4. The data on $Bi_2Sr_2CaCu_2O_x$ and $Tl_2Ba_2CaCu_2O_8$ taken from Refs. 12 and 13, respectively, are also plotted in the figure. The Hall effect is negative only in the region below the boundary. The shape of the boundary is similar for three materials. The difference is that, for YBCO, the boundary extends from the reversible to the irreversible regime. The similarity of the shape strongly indicates the existence of a common mechanism for the sign reversal for these material. The mechanism must not be related to the pinning effect because the location of the irreversibility line does not affect the shape of the boundary.

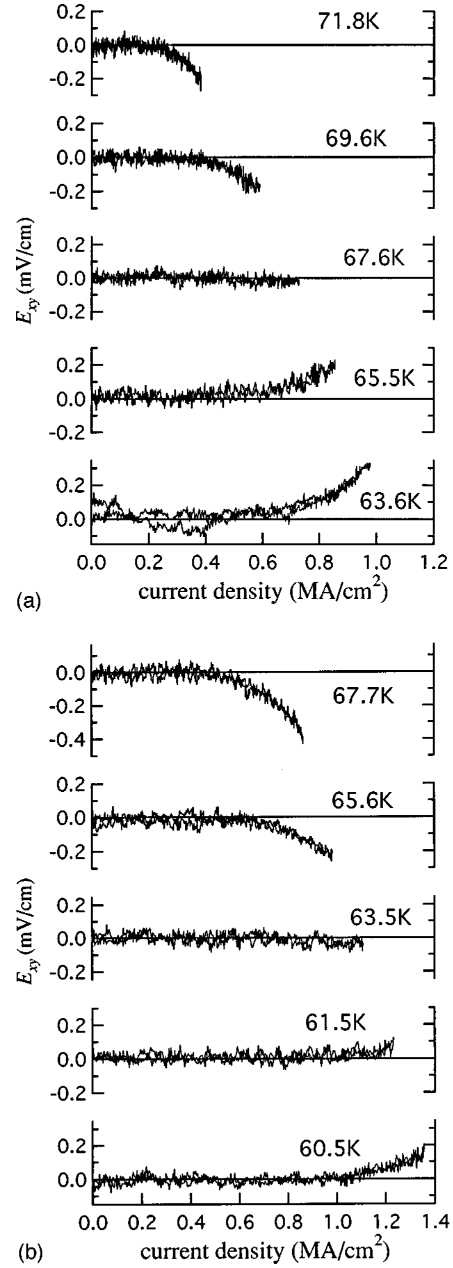

FIG. 3. Hall voltage against current density in (a) 7 T and (b) 5 T at several temperatures in the irreversible regime.

There have been many proposals of mechanisms for the sign reversal of the Hall effect. Among them, only a few can account for the existence of the second sign reversal. Hagen *et al.*[4] modified the model proposed by Nozières and Vinen,[1] so that a frictional force proposed by Bardeen and Stephen[2]

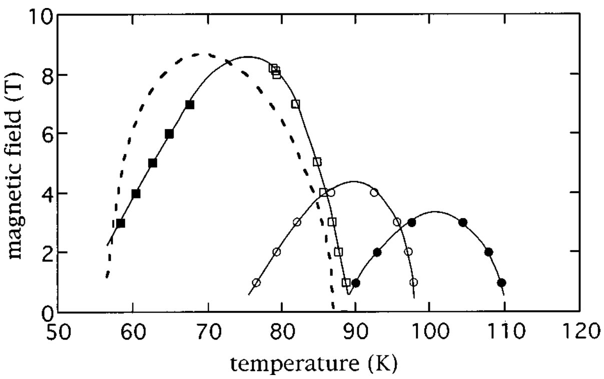

FIG. 4. Boundaries between the positive and the negative Hall effect. Closed circles are for $Tl_2Ba_2CaCu_2O_8$ (Ref. 12), open circles are for $Bi_2Sr_2CaCu_2O_x$ (Ref. 11), and open and closed squares are for $YBa_2Cu_3O_x$ (present work) obtained by a conventional technique and by the pulse current technique, respectively. Solid curves are guides to the eye. The broken curve shows the calculation according to Eq. (5). See text for details.

could be included. According to this model, the boundary of the positive and the negative Hall effect is determined by a delicate balance of two types of frictional forces. If we can assume that the effect of the pinning is equivalent to a renormalization of the Bardeen-Stephen-type frictional force,[23] then the boundary should depend on the strength of the pinning effect. Our experimental results, however, strongly indicate that the boundary is determined by a common and intrinsic mechanism, and independent of the pinning effect. Therefore, in our opinion, it is difficult to explain the present results by this model.

Freimuth and co-workers' explanation[5] for the sign reversal of the Hall effect is based on the assumption that the transverse motion of vortices develops a transverse temperature gradient, and the Seebeck effect due to this temperature gradient modifies the apparent Hall voltage. Using the experimental results on the Seebeck effect, they succeeded in explaining the double sign reversal of $Bi_2Sr_2Ca_2Cu_3O_d$. However, the temperature gradient developed by this mechanism should strongly depend on experimental situations, i.e., single crystals or thin films, dc measurements or pulse measurements, and so on. In this respect, this model seems to be inconsistent with the experimental facts that the double sign reversal is rather common and robust.

In the present article, we tentatively adopt the model proposed by Feigel'man *et al.*[19] This model attributes the sign reversal to the effect of the excess charge of the vortex core. In general, superconducting transition induces a change of the chemical potential, and if the vortex core is in the normal state, the difference in the chemical potential must be compensated by a redistribution of charge carriers. The magnitude of the difference of the carrier density $\delta n/n_0$ is of the order of $(\Delta/\varepsilon_F)^2$, where $\delta n = n_0 - n_\infty$, n_0 and n_∞ being carrier densities in the core and far outside the core, respectively, Δ is the superconducting energy gap, and ε_F is the Fermi energy.[24] According to this model, the sign of the Hall effect is determined by δn and the relaxation rate τ of the normal carrier in the vortex core.[25] In the refined version of this model[19] the Hall conductivity σ_{xy} in the mixed state is expressed as

$$\sigma_{xy} = \frac{n_0 ec}{B} \frac{\Delta^2}{\varepsilon_F^2} [(\Delta\tau/\hbar)^2 g - \sin(\widetilde{\delta n})] + \sigma_{xy}^n (1-g), \quad (2)$$

where g is a parameter which expresses the superconducting portion of the carrier, $\widetilde{\delta n}$ is related to the excess charge of the vortex core and modified by the screening effect, and σ_{xy}^n is the Hall conductivity of the normal state.

This model, as many other theories, assumes the absence of the pinning effect, which is the dominant factor for the vortex motion in the irreversible regime. Vinokur *et al.*[23] pointed out phenomenologically that the Hall conductivity in the mixed state should be independent of the pinning effect. Although this idea works well very often,[20,26] it is not supported by any microscopic theory. In the analysis below, we concentrate on the boundary between the positive and the negative Hall conductivity, which can be more safely expected to be independent of the pinning effect. This assumption is supported by our observation that the evolution of the Hall voltage with the increase in the current density is monotonic and the sign of the Hall effect can be unambiguously defined as a function of temperature and magnetic field.

First, we consider the boundary in the small magnetic field limit, where the second term in Eq. (2), which should be proportional to B, is negligible. Just below T_c, where Δ and g are very small, and in a small magnetic field,

$$\sigma_{xy} \approx -\frac{n_0 ec}{B} \frac{\Delta^2}{\varepsilon_F^2} \sin(\widetilde{\delta n}). \quad (3)$$

As long as sign reversals are observed experimentally we must assume $\sin(\widetilde{\delta n}) = +1$. Then the Hall effect just below T_c is negative. Thus, this model predicts the first sign reversal in the limit of zero magnetic field at T_c, which agrees with the experimental fact.

At lower temperatures and in small magnetic fields, the condition $(\Delta\tau/\hbar)^2 g = 1$ gives the second sign reversal. Experimentally, the second sign reversal in small magnetic fields takes place at 50–60 K, which is well below T_c. Therefore, as a reasonable approximation we can put $g=1$ and $\Delta(T) = \Delta_0$, where Δ_0 is the energy gap at $T=0$. Then the condition for the second sign reversal is reduced to $\tau = \hbar/\Delta_0$. If we adopt 20 meV as the value of Δ_0, we obtain $\tau = 0.33 \times 10^{-13}$ s. Gao *et al.*[27] and Bonn *et al.*[28] estimated τ below T_c from their microwave measurements. They observed a rapid increase in τ with a decrease in temperature. Their estimation of τ at 50–60 K is about one order larger than the above estimation. Taking into account the fact that both the sample and the measuring technique are different in two estimations, the difference of one order is not unreasonable.

Next, we consider the boundary in finite magnetic fields. Because the normal state Hall conductivity should be proportional to magnetic field B, σ_{xy}^n can be written as

$$\sigma_{xy}^n = s_{xy}^n B, \quad (4)$$

where s_{xy}^n is positive and independent of B. Substituting Eq. (4) and $\sin(\widetilde{\delta n}) = +1$ into Eq. (2) and putting $\sigma_{xy} = 0$, we obtain

$$B^2 = \frac{n_0 e c \Delta^2 [1-(\Delta\tau/\hbar)^2 g]}{\varepsilon_F^2 s_{xy}^n (1-g)}. \tag{5}$$

This relation gives the boundary between the positive and the negative Hall effect on a B-T plane. For the calculation of the boundary according to Eq. (5), we need temperature dependences of $\Delta(T)$ and $g(T)$. For $\Delta(T)$, we simply assume that[29]

$$\Delta(T) = 1.74\Delta_0 \left(1-\frac{T}{T_c}\right)^{1/2}, \tag{6}$$

where the magnetic field dependence of $\Delta(T)$ is neglected because the magnetic fields where the second sign reversals occur are much smaller than the upper critical field B_{c2}. For $g(T)$, we adopt the two-fluid model[29] as a simplest approximation,

$$g(T) = 1-(T/T_c)^4. \tag{7}$$

It was observed that the Hall conductivity of the present sample in the normal state has an approximate temperature dependence $\propto T^{-3}$, as was reported for many other samples.[30] For s_{xy}^n below T_c, we assume that the temperature dependence can be extrapolated from the normal state, and obtain,

$$s_{xy}^n = 3.75\times 10^{14}\,\frac{1}{T^3}. \tag{8}$$

At the second sign reversal in small magnetic fields, the relation $(\Delta\tau/\hbar)^2 g = 1$ holds, and $\Delta_0\tau/\hbar$ should be slightly larger than 1. For simplicity, $\Delta_0\tau/\hbar$ is fixed to be 1.1 rather arbitrarily in the calculation. Then the fitting parameter in Eq. (5) is only $n_0(\Delta_0/\varepsilon_F)^2$. The broken curve in Fig. 4 shows the boundary calculated according to Eq. (5), where $n_0(\Delta_0/\varepsilon_F)^2 = 7\times 10^{17}$ is substituted. Widely accepted values are $n_0 = 10^{21-22}$ cm^{-3},[31] and $\Delta_0/\varepsilon_F = 0.1$–$0.2$.[32] Therefore, if we use these value for $n_0(\Delta_0/\varepsilon_F)^2$ in the calculation, we obtain magnetic fields for the boundary larger than the experimental results by more than one order. Furthermore, the shape of the boundary is not perfectly reproduced. However, the disagreement does not seem to rule out the applicability of the theory if we consider large ambiguity in the evaluation of physical parameters of this material and approximations in the theory.

It is reported that τ has stronger temperature dependence below T_c,[28] and it is naturally expected that s_{xy}^n at temperatures below T_c, which is not experimentally accessible directly, has also a stronger temperature dependence than that above T_c.[17] Assuming a steeper temperature dependence than $\propto T^{-3}$ for s_{xy}^n in Eq. (5), we obtain a better agreement between the theory and the measurement. However, we would not pursue this possibility further, because it is not sure whether the present stage of both the theory and the experimental determinations of basic constants of the physical property of YBCO allow more quantitative comparison between the theory and the experiment.

In summary, the Hall effect of $YBa_2Cu_3O_x$ was measured using high current densities at temperatures down to 58 K, and the second sign reversals were observed. The sign of the Hall effect does not depend on the current density and can be defined unambiguously at each point on a B-T plane. The boundary of the sign of the Hall effect is qualitatively similar for Y-, Bi-, and Tl-based high T_c materials. This fact strongly indicates that the existence of the region of the negative Hall effect is the very intrinsic property of high T_c materials and should be explained without including the pinning effect. Feigel'man *et al.*'s theory can explain reasonably well the character of the boundary, but the quantitative agreement with the experiment is not satisfactory.

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