Hall Anomaly in the Superconducting State of High-T_c Cuprates: Universality in Doping Dependence

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We have measured the flux flow Hall effect in the superconducting state of various high-T_c superconductors (HTSC) from the underdoped to the overdoped regime. We show that the Hall sign is universal and is determined by the doping level; the sign is electronlike in the underdoped regime and holelike in the overdoped regime. This tendency contradicts the prediction of the time dependent Ginzburg-Landau equation based on the *s*-wave weak coupling theory, suggesting that such a theory fails to evaluate the Hall force acting on the vortices in HTSC. This discrepancy may be attributed to the novel electronic structure of the vortex in HTSC. [S0031-9007(98)05831-1]

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The vortex motion in the superfluid electrons has presented a persistent problem in the superconducting state of type II superconductors. Knowledge of the Hall effect enables us to obtain clear and important information on this problem. One of the most puzzling and controversial phenomena is the sign change that has been observed in the Hall effect in the superconducting state in most high-T_c superconductors (HTSC) and some conventional superconductors [1]. The Hall sign is determined by the topology of the Fermi surface in the normal state, while it is determined by the vortex motion in the superconducting state. The classical theories of vortex motion, the Bardeen-Stephen [2] and Nozieres-Vinen [3] models, predict that the superconducting and normal states will have the same Hall sign, and thus cannot explain this anomaly. Recent experiments have ruled out the possibility that some form of pinning induces the sign reversal [4,5]. Moreover, the occurrence of the sign reversal in one-unit-cell-thick ultrathin $YBa_2Cu_3O_{7-\delta}$ film demonstrates that the Hall anomaly occurs in a two-dimensional CuO_2 plane [6]. Several attempts to understand the Hall anomaly have been undertaken, but the microscopic origin of this phenomenon remains a controversial and vexing problem that demonstrates explicitly our incomplete knowledge of vortex dynamics.

A recent phenomenological theory based on the time dependent Ginzburg-Landau (TDGL) equation has been shown to be quite successful in describing the Hall effect in the superconducting state [7,8]. According to the TDGL theory, the vortex Hall conductivity σ_{xy}^V arising from the hydrodynamic contribution plays an important role in determining the Hall sign at low fields. The Hall sign reversal occurs when σ_{xy}^V has a sign opposite that of the normal state Hall effect. In the framework within the BCS theory, several authors have calculated σ_{xy}^V and emphasized the importance of the electronic structure of the materials for understanding the Hall effect. Fukuyama, Ebisawa, and Tsuzuki (FET) [9] have derived the TDGL equation from the microscopic BCS theory and found that σ_{xy}^V appears as a result of the electron-hole asymmetry, which is quantified by $\partial N(\mu)/\partial\mu|_{\mu=\varepsilon_F}$, where $N(\mu)$ is the density of states, μ is the chemical potential, and ε_F is the Fermi energy. Recently, Aronov, Hikami, and Larkin (AHL) [10] have shown that the sign of σ_{xy}^V is universal and is determined by $\partial \ln T_c/\partial\mu$ from a general gauge invariance requirement of the TDGL equation (see also Ref. [11]). More recently, van Otterlo *et al.* [12] have microscopically derived σ_{xy}^V from the effective action for the vortex motion based on the BCS Hamiltonian and pointed out that σ_{xy}^V can be interpreted as the vortex charging effect arising from the difference in electron density between the core and the far outside region of the vortex (see also Ref. [13]). All of these calculations remain valid for *s*-wave weak coupling superconductors regardless of the nature of the interaction. Thus the Hall anomaly does not itself contradict the BCS theory.

As will be shown later, application of these theories to HTSC leads to the conclusion that the Hall sign is holelike in the underdoped regime and electronlike in the overdoped regime. Since the Hall sign in the normal state of HTSC is always holelike, sign reversal is expected to occur in the overdoped regime. (For the sake of simplicity, we do not consider here the electron-doped material $Nd_{2-x}Ce_xCuO_{4-\delta}$.) Therefore measurement of the doping dependence of the Hall effect would contribute important information for understanding the microscopic mechanism of the Hall anomaly in HTSC. In this paper, we have performed systematic measurements on the Hall effect in the superconducting state of various HTSC, including La-, Y-, and Bi-based materials, from the underdoped to the overdoped regime. We show that *the Hall sign in the superconducting state of HTSC is universal and is determined by the doping level;* the sign is electronlike in the underdoped and slightly overdoped regimes and is holelike in the overdoped regime. This behavior is strikingly in contrast with the conclusion inferred from the weak coupling *s*-wave BCS theory, suggesting that such a theory fails to evaluate the hydrodynamic force acting on the vortex of HTSC.

We have measured three different HTSC systems, $La_{2-x}Sr_xCuO_4$ (La:214), $Bi_2Sr_2CuO_{6+\delta}$ (Bi:2201), and $YBa_2Cu_3O_{7-\delta}$ (Y:123), by changing the electronic state from an underdoped to an overdoped regime. The La:214 single crystal thin films ($x = 0.10$, 0.15, 0.20, 0.24, and 0.28) with a thickness of 200–300 nm were grown on $SrTiO_3$ (100) substrates using the rf magnetron sputtering method. They were annealed at 800 °C for 8 h in air after deposition in order to minimize the remaining oxygen vacancy. The [Sr]/[La] concentrations were determined by x-ray fluorescent analysis and Rutherford backscattering. The transition temperatures T_c for $x = 0.10$, 0.15, 0.20, 0.24, and 0.28 were 26.0, 33.3, 29.6, 18.6, and 9.5 K, respectively. The transition temperature for optimally doped crystal $T_c^{\rm opt}$ is 34.0 K. Epitaxial *c*-axis oriented Y:123 thin films ($T_c^{\rm opt} = 92.0$ K) with a thickness of 100 nm were deposited on $SrTiO_3$ (100) substrates by laser ablation. To obtain the overdoped crystals, Y is substituted by Ca with a composition $(Y_{1-x}Ca_x)Ba_2Cu_3O_y$ ($x = 0$, 0.1, 0.2, 0.4). The transition temperatures for $x = 0$, 0.1, 0.2, and 0.4 were 90.3, 80.1, 65.0, and 62.1 K, respectively. Films were annealed at 400 °C for 1 h in oxygen atmosphere after deposition. The calcium contents of the films were the same as that of the target, which was confirmed by inductive coupled plasma spectroscopy. The single crystals of Bi:2201 ($T_c^{\rm opt} = 35.0$ K) were grown by the traveling-solvent-floating-zone technique. To realize an overdoped state in the crystal, we have made a Pb-for-Bi substitution to yield a composition of $Bi_{1.80}Pb_{0.38}Sr_{2.01}CuO_{6+\delta}$. To obtain an underdoped state, the crystals were annealed in a vacuum of $\sim 10^{-4}$ Pa for the reduction of oxygen content. The transition temperatures for the samples measured were 21.1 K for the underdoped and 20.0 and 6.0 K for the overdoped crystals.

Diagonal resistivity ρ_{xx} and Hall resistivity ρ_{xy} were measured simultaneously at ac (17 Hz) current densities $J = 10$–100 A/cm^2 using lock-in amplifiers (PAR124A). We obtained ρ_{xy} from the transverse resistance by subtracting the positive and negative magnetic-field data. Before presenting the data, we will discuss the method of analyzing the Hall sign at low fields. It has been pointed out that $\sigma_{xy}[= \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2)]$ is insensitive to disorder by a general argument of the vortex dynamics [5,14]. We therefore discuss the Hall data in terms of σ_{xy}. To obtain σ_{xy} precisely at low fields, we measured the change of ρ_{xy} for more than 3 orders of magnitude from the normal state value. This was because σ_{xy} stays in the same order of magnitude, even though ρ_{xx} and ρ_{xy} change more than 3 orders of magnitude at some temperatures. In particular, the magnitude of ρ_{xy} of overdoped crystals is small compared with that of underdoped crystals, and changes rapidly with *H*. We therefore used a carefully calibrated Hall sensor and stabilized the temperatures within ± 2 mK in magnetic fields.

Figures 1(a)–1(f) show the field dependence of σ_{xy} for La:214, Y:123, and Bi:2201. The upper (lower) panels represent a typical case for the underdoped (overdoped) crystals. At high fields, σ_{xy} increases linearly with *H* in all crystals. With decreasing *H*, σ_{xy} diverges to $-\infty$ for underdoped crystals but diverges to $+\infty$ for overdoped crystals. These *H* dependences are analyzed in accordance with the TDGL theory, which predicts that the flux flow Hall conductivity σ_{xy} consists of two contributions:

$$\sigma_{xy} = \sigma_{xy}^{Q} + \sigma_{xy}^{V}. \quad (1)$$

The first term describes the contribution arising from the quasiparticles inside and around the vortex core. This term has the same sign as the normal state and is proportional to *H*, which is consistent with the σ_{xy} at high fields in Figs. 1(a)–1(f). The second term is the vortex Hall term discussed earlier. This term is inversely proportional to *H*. Accordingly, at low fields, the Hall sign is determined by σ_{xy}^{V}. For Bi:2201 and underdoped Y:123, σ_{xy} varies as $1/H$ at low fields, but for La:214 and overdoped Y:123, σ_{xy} varies faster than $1/H$ [15,16]. This seems to suggest that σ_{xy}^{V} does not vary as $1/H$ in the regime, where the vortices are very strongly pinned. The detailed analysis will be published elsewhere; here, we focus on the sign of σ_{xy}^{V}, which we determine by the diverging direction of σ_{xy} with decreasing *H* at low fields.

Figure 2 displays the doping dependence of the Hall sign in the superconducting state of HTSC including La-, Y-, Bi-, and Tl-based compounds. In Fig. 2, we display the present results together with the data obtained by other groups. In this figure, T_c is normalized by $T_c^{\rm opt}$ for the corresponding system. The filled circles depict the electronlike Hall sign (Hall anomaly), and the open circles depict the holelike Hall sign (no Hall anomaly). Figure 2 demonstrates that the Hall anomaly occurs always in the underdoped and slightly overdoped regimes. The Hall anomaly is sample dependent near $T_c/T_c^{\rm opt} \sim 0.9$ in the

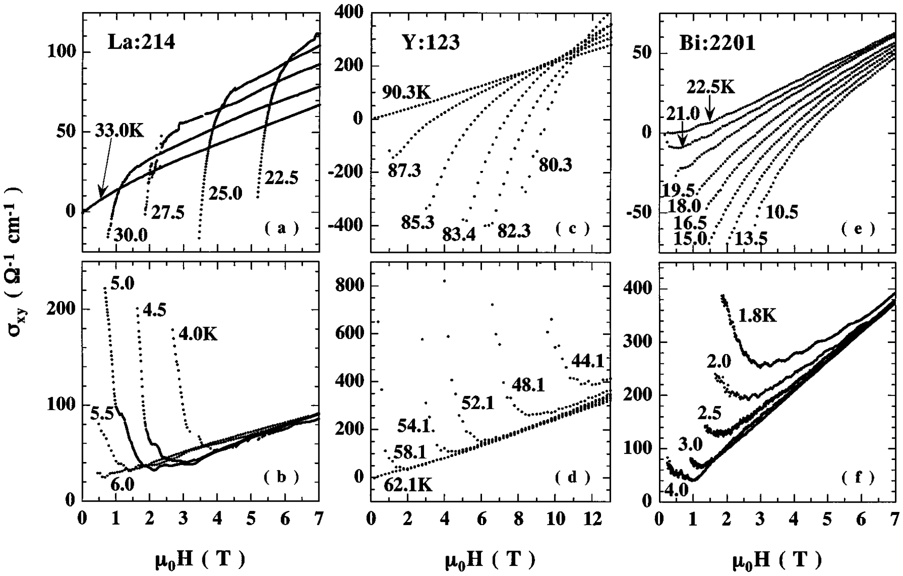

FIG. 1. Field dependence of the Hall conductivity below T_c; (a) $La_{2-x}Sr_xCuO_4$, $x = 0.15$ ($T_c = 33.3$ K); (b) $La_{2-x}Sr_xCuO_4$, $x = 0.28$ ($T_c = 9.5$ K); (c) $YBa_2Cu_3O_{7-\delta}$, ($T_c = 90.3$ K); (d) $(Y_{1-x}Ca_x)Ba_2Cu_3O_{7-\delta}$, $x = 0.4$ ($T_c = 62.1$ K); (e) $Bi_{1.80}Pb_{0.38}Sr_{2.01}CuO_{6+\delta}$ annealed in a vacuum, ($T_c = 21.1$ K); (f) $Bi_{1.80}Pb_{0.38}Sr_{2.01}CuO_{6+\delta}$, as-grown ($T_c = 6.0$ K). The upper panels [(a), (c), and (e)] display σ_{xy} for the underdoped crystals, and the lower panels [(b), (d), and (f)] display σ_{xy} for the overdoped crystals.

slightly overdoped regime, but no Hall anomaly is observed beyond this regime. This behavior is observed in the crystals with monolayer, double layer, or triple layer structures. Moreover, the Hall sign depends neither on the magnitude of anisotropy nor on the pinning strength of the materials. We therefore conclude that the doping dependence shown in Fig. 2 is one of the universal transport properties of HTSC, showing that *the Hall sign in the superconducting state is related closely with the characteristic electronic structure determined by the doping.* This is the main result of this paper.

We now compare the present results with the TDGL description based on the BCS theory. It has been shown that the imaginary part of the complex relaxation time ($\gamma = \gamma_1 + i\gamma_2$) in the TDGL equation gives rise to σ_{xy}^V ($\sigma_{xy}^V \propto -\gamma_2$) [7,8]. According to FET, the sign of σ_{xy}^V is given by the sign of $\mathrm{sgn}(e)\, \partial N(\mu)/\partial\mu|_{\mu=\varepsilon_F}$, where sgn($e$) is the sign of the carrier [23]. We lack the detailed knowledge of the Fermi surface needed to validate this theory. However, according to AHL theory, which is based on a more general background, the sign of σ_{xy}^V is given by the sign of $\mathrm{sgn}(e) d\ln T_c/d\mu$ [10]. The photoemission experiment demonstrates that μ decreases monotonically with doping holes [25]. Thus the BCS theory leads to the conclusion that the Hall sign is positive in the underdoped regime where $\mathrm{sgn}(e) d\ln T_c/d\mu > 0$ and is negative in the overdoped regime where $\mathrm{sgn}(e) d\ln T_c/d\mu < 0$; the sign reversal should occur in the overdoped regime and be absent in the underdoped regime. This is strikingly in contrast to the result displayed in Fig. 2, indicating that the BCS theory yields the wrong Hall signs in both the underdoped and the overdoped regimes.

The present results suggest that a simple *s*-wave weak coupling theory fails to evaluate the hydrodynamic force acting on the vortices of HTSC. Recently, Geshkenbein, Ioffe, and Larkin [26] discussed the Hall effect of HTSC in the presence of the preformed pair that forms a local **k** space pairing above T_c. The authors suggested that, in such a situation, γ_2 term may not be written simply as $\mathrm{sgn}(e) d\ln T_c/d\mu$. Their theory points out the possibility of an electronlike Hall sign in the underdoped materials, but it seems to fail in explaining the holelike Hall effect in the overdoped materials. Another possible origin may be the novel structure of the vortex in HTSC, in which the symmetry of the superconducting gap of HTSC is most likely a $d_{x^2-y^2}$ wave. A recently growing body of theoretical and experimental results indicates that the structure of a vortex of *d*-wave superconductors is quite different from that of *s*-wave superconductors [27]. For instance, the phenomenological Ginzburg-Landau theory has shown that the vortex in the *d*-wave superconductor should contain the *s*-wave component counter-rotating relative to the *d*-wave component near the vortex core [28]. A similar structured *d*-wave vortex has also been predicted quite recently by Himeda *et al.* [29] from microscopic calculations based on the *t*-*J* model. Therefore it seems probable that the contribution of the vortex core to the Hall force in *d*-wave superconductors is different from that in *s*-wave superconductors [30]. A detailed theoretical calculation is greatly needed.

It is well known that the normal state of HTSC in the underdoped regime exhibits unconventional behavior, which is characterized by the unusual excitation spectra observed in NMR-relaxation-rate and photoemission

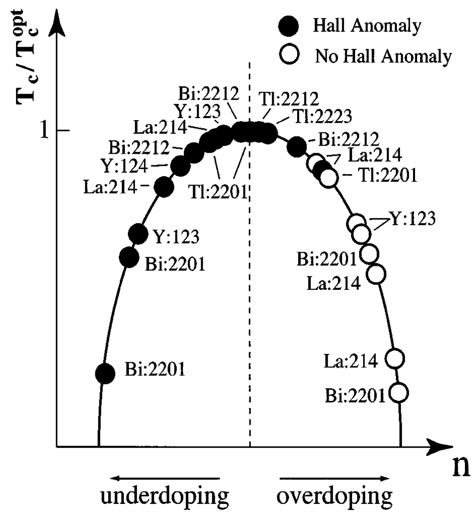

FIG. 2. Doping dependence of the Hall anomaly in the superconducting state for various high-T_c cuprates including La-, Y-, Bi-, and Tl-based compounds. We display the Hall sign determined in the present study (see text), together with the data obtained by other groups; Bi:2201 ($T_c =$ 7.5 K: underdoped) [17], Y:123 ($T_c = 60$ K: underdoped) [18], Y:124 ($T_c = 81$ K: underdoped, $T_c^{opt} = 91$ K) [19], Bi:2212 ($T_c = 86$ K: underdoped, $T_c = 92$ K: optimally doped, $T_c =$ 87 K: overdoped) [20], Tl:2201 ($T_c = 79$ K: underdoped, $T_c =$ 82 K: optimally doped, and $T_c = 72$ K: overdoped) [21], Tl:2212 ($T_c = 104$ K: optimally doped) [22], and Tl:2223 ($T_c = 107$ K: optimally doped) [4]. Filled circles denote presence and open circles denote absence of Hall anomaly. n represents the carrier number, and T_c are normalized by T_c^{opt} (see text).

experiments. On the other hand, the system approaches the normal Fermi liquid with large Fermi surface in the overdoped regime. However, despite the frequent emphasis given to the difference between the normal states of under- and overdoped HTSC, the difference between the superconducting states of the two regimes has been rarely discussed. The fact that the Hall sign changes from electronlike to holelike when going from an underdoped to an overdoped regime implies that the electronic structure of the vortex changes with doping. It should be noted that the quite recent calculation of the vortex structure based on SO(5) symmetry [31] and the *t*-*J* model [29] has revealed a qualitative difference between the vortex structure of under- and overdoped regimes.

In summary, we have measured the doping dependence of the Hall effect in the superconducting state from the underdoped to overdoped regime of HTSC. The Hall sign is universal and is electronlike in the underdoped regime and holelike in the overdoped regime. This tendency is opposite that predicted by the weak coupling *s*-wave theory, suggesting that such a theory fails to evaluate the Hall force acting on the vortices of HTSC. These results on the Hall effect imply also that the electronic structure of the vortex changes with doping.

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[1] A brief review of the literature is given in S.J. Hagen *et al.*, Phys. Rev. B **47**, 1064 (1993).
[2] J. Bardeen and M.J. Stephen, Phys. Rev. **140**, A1197 (1965).
[3] P. Nozieres and W.F. Vinen, Philos. Mag. **14**, 667 (1966).
[4] R.C. Budhani, S.H. Liou, and Z.X. Cai, Phys. Rev. Lett. **71**, 621 (1993).
[5] A.V. Samoilov *et al.*, Phys. Rev. Lett. **74**, 2351 (1995).
[6] Y. Matsuda *et al.*, Phys. Rev. Lett. **69**, 3228 (1992).
[7] N.B. Kopnin, B.I. Ivlev, and V.A. Kalatsky, J. Low Temp. Phys. **90**, 1 (1993).
[8] Alan T. Dorsey, Phys. Rev. B **46**, 8376 (1992); S. Ullah and A.T. Dorsey, Phys. Rev. B **44**, 262 (1991).
[9] H. Fukuyama, H. Ebisawa, and T. Tsuzuki, Prog. Theor. Phys. **46**, 1028 (1971).
[10] A.G. Aronov, S. Hikami, and A.I. Larkin, Phys. Rev. B **51**, 3880 (1995).
[11] A.G. Aronov and A.B. Rapoport, Mod. Phys. Lett. B **6**, 1083 (1992).
[12] A. van Otterlo *et al.*, Phys. Rev. Lett. **75**, 3736 (1995).
[13] G. Blatter *et al.*, Phys. Rev. Lett. **77**, 566 (1996); D.I. Khomskii and A. Freimuth, Phys. Rev. Lett. **75**, 1384 (1995); M.V. Feigel'man *et al.*, JETP Lett. **62**, 834 (1995).
[14] V.M. Vinokur *et al.*, Phys. Rev. Lett. **71**, 1242 (1993).
[15] Y. Matsuda *et al.*, Phys. Rev. B **52**, R15 749 (1995).
[16] J.M. Harris *et al.*, Phys. Rev. B **51**, 12 053 (1995).
[17] R. Jin and H.R. Ott, Phys. Rev. B **53**, 9406 (1996).
[18] J.M. Harris *et al.*, Phys. Rev. Lett. **73**, 1711 (1994).
[19] J. Schoenes, E. Kaldis, and J. Karpinski, Phys. Rev. B **48**, 16 869 (1993).
[20] T. Nagaoka *et al.* (unpublished).
[21] Judy Z. Wu *et al.* (to be published).
[22] S.J. Hagen *et al.*, Phys. Rev. B **43**, 6246 (1991).
[23] We note that Ref. [9] derived a misleading result; the sign of σ_{xy} is opposite that reported. The revised result is given in Ref. [24].
[24] T. Nishio and H. Ebisawa, Physica (Amsterdam) **190C**, 43 (1997).
[25] A. Ino *et al.*, Phys. Rev. Lett. **79**, 2101 (1997). We take μ as the chemical potential of the electron.
[26] V.B. Geshkenbein, L.B. Ioffe, and A.I. Larkin, Phys. Rev. B **55**, 3173 (1997).
[27] N. Schopohl and K. Maki, Phys. Rev. B **52**, 490 (1995); M. Ichioka *et al.*, *ibid.* **53**, 15 316 (1996).
[28] G.E. Volovik, JETP Lett. **58**, 470 (1993); P.I. Soininen, C. Kallin, and A.J. Berlinsky, Phys. Rev. B **50**, 13 883 (1994); Yong Ren, Ji-Hai Xu, and C.S. Ting, Phys. Rev. Lett. **74**, 3680 (1995); A.J. Berlinsky *et al.*, Phys. Rev. Lett. **75**, 2200 (1995).
[29] A. Himeda *et al.*, J. Phys. Soc. Jpn. **66**, 3367 (1997).
[30] Yusuke Kato (private communication).
[31] Daniel P. Arovas *et al.*, Phys. Rev. Lett. **79**, 2871 (1997).