

Normal-state Hall effect and the insulating resistivity of high- T_c cuprates at low temperatures

Yoichi Ando

*Bell Laboratories, Lucent Technologies, 700 Mountain Avenue, Murray Hill, New Jersey 07974
and Central Research Institute of Electric Power Industry, Komae, Tokyo 201, Japan*

G. S. Boebinger and A. Passner

Bell Laboratories, Lucent Technologies, 700 Mountain Avenue, Murray Hill, New Jersey 07974

N. L. Wang, C. Geibel, and F. Steglich

Institut für Festkörperphysik, Technische Hochschule Darmstadt, D-64289 Darmstadt, Germany

I. E. Trofimov* and F. F. Balakirev

Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey 08855

(Received 25 June 1997)

The normal-state Hall coefficient R_H and the in-plane resistivity ρ_{ab} are measured in La-doped $\text{Bi}_2\text{Sr}_2\text{CuO}_y$ ($T_c \approx 13$ K) single crystals and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films by suppressing superconductivity with 61-T pulsed magnetic fields. In contrast to data above T_c , the R_H below ~ 10 K shows little temperature dependence in all the samples measured, irrespective of whether ρ_{ab} exhibits insulating or metallic behavior. Thus, whatever physical mechanism gives rise to insulating behavior in the low-temperature normal state, it leaves the Hall conductivity relatively unchanged. [S0163-1829(97)51438-X]

Application of a pulsed high magnetic field to suppress superconductivity in the high- T_c cuprates has opened up the possibility for measurements of normal-state transport at low temperatures. This regime has been rather unexplored due to the extremely high H_{c2} of the cuprates. Thus far the anisotropic normal-state resistivity has been measured in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO)^{1,2} and La-doped $\text{Bi}_2\text{Sr}_2\text{CuO}_y$ (Bi-2201)³ down to subkelvin temperatures using 61-T pulsed magnetic fields.

One of the surprising findings in the low-temperature normal-state resistivity of LSCO is an unusual $\log(1/T)$ divergence of both in-plane (ρ_{ab}) and c -axis resistivity (ρ_c) of underdoped samples. Similar divergence of resistivity that is consistent with $\log(1/T)$ is also found in disordered Bi-2201,³ where the dynamic range of the divergence is generally smaller than in underdoped LSCO, where the increase of ρ_{ab} can be as large as a factor of 3. Although the origin of the unusual $\log(1/T)$ behavior is not yet clear, there are several proposed explanations, some of which involve the localization of the charge-carrying quasiparticles,^{4,5} others of which involve the suppression of the two-dimensional density of states near the Fermi energy. In this latter scenario, the low-temperature insulating behavior in the cuprates might result from conventional disorder-enhanced electron interactions,⁶ from the temperature-dependent impurity scattering time in the marginal Fermi liquid,⁷ or from the existence of a pseudogap in the underdoped high- T_c cuprates.⁸

In conventional physics of disordered metals in two dimensions, both weak localization and electron-electron interactions give rise to identical $\log(1/T)$ corrections to the resistivity. Measurement of the Hall coefficient R_H is useful in separating these two physical mechanisms:⁶ weak localization does not affect R_H , while interactions lead to corrections in R_H which are two times larger than the corrections to

the resistivity. The physics of the cuprates is expected to be very different from that of conventional disordered metals. Indeed, three independent reports of logarithmic behavior in underdoped cuprates provide three separate arguments against interpretations involving conventional weak localization.^{1,9,10} Nevertheless, measurement of the Hall effect down to low temperatures could help resolve which of the proposed physical mechanisms governs low-temperature transport in the cuprates.

In this paper, we present low-temperature measurements of ρ_{ab} and the Hall coefficient, $R_H \equiv \rho_{\text{Hall}}/B$, in Bi-2201 single crystals and LSCO thin films using 61-T pulsed magnetic fields to suppress superconductivity. For Bi-2201, we measured several crystals with nominally the same carrier concentration, for which the low-temperature behavior of ρ_{ab} is either metallic ($d\rho_{ab}/dT > 0$) or insulating ($d\rho_{ab}/dT < 0$), depending upon the apparent amount of naturally occurring disorder in the different samples. The LSCO thin films have very different carrier concentrations, such that the low-temperature ρ_{ab} behavior also varies from metallic to insulating between the samples.

The Bi-2201 single crystals are grown by the flux method in Al_2O_3 crucibles.¹¹ To obtain nearly optimally doped crystals, La is doped onto the Sr site, giving the composition of $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_y$ with nominal $x=0.05$ and T_c (midpoint) around 13 K. The actual La concentration is estimated to be much higher, $x \sim 0.3$, corresponding to slightly overdoped samples.¹² The samples are annealed in flowing oxygen for 1 hour at 400 °C after six silver contacts are painted on the edges of the platelet-shaped crystals. Typical size of the crystals is $1.7 \times 0.7 \times 0.02$ mm³, small enough that the data are not adversely affected by eddy-current heating during the magnet pulses.³

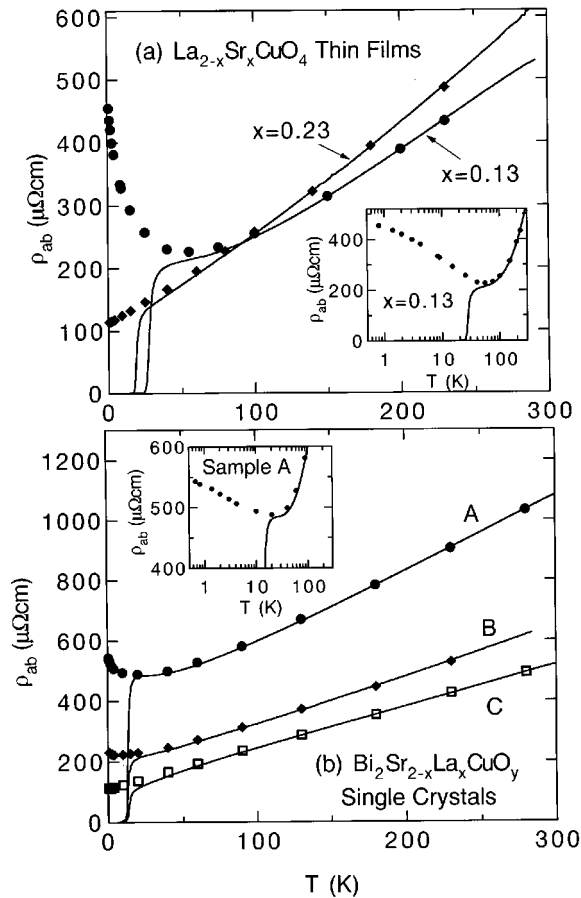


FIG. 1. ρ_{ab} in 0 T (solid lines) and 60 T (symbols) of (a) underdoped ($x=0.13$) and overdoped ($x=0.23$) LSCO thin films and (b) three La-doped Bi-2201 single crystals with varying amounts of disorder. The insets illustrate that the insulating ρ_{ab} is consistent with a $\log(1/T)$ divergence.

The LSCO films are c -axis-oriented, epitaxial films deposited on LaSrAlO_4 substrates by pulsed laser deposition, as described in Ref. 13. One film is overdoped, with a nominal $x=0.23$ and T_c (midpoint) of 19 K. The other is underdoped, with a nominal $x=0.13$. This film was annealed in high-pressure oxygen¹³ yielding a T_c (midpoint) of 28 K. The films were patterned by photolithography into a conventional Hall bar with a center strip 0.2-mm wide. Silver contact pads were evaporated onto the arms and gold wires attached to them using silver epoxy. Since the samples have submicron thickness and are thermally anchored to the insulating substrate, there is no evidence of eddy-current heating in the films during the magnet pulses.

Longitudinal and transverse voltages are measured simultaneously using two lock-in amplifiers (at ~ 100 kHz, using an output time constant = $30 \mu\text{sec}$), whose outputs are recorded on a transient digitizer. The ohmicity of both longitudinal and transverse voltages were confirmed down to the lowest temperatures. Because the contact alignment is generally good, the transverse voltages are typically an order of magnitude smaller than the longitudinal voltages. Nevertheless, at each temperature, it is necessary to record data from a pair of magnetic-field pulses of opposite polarity, where the reported Hall voltage is the asymmetrical component of

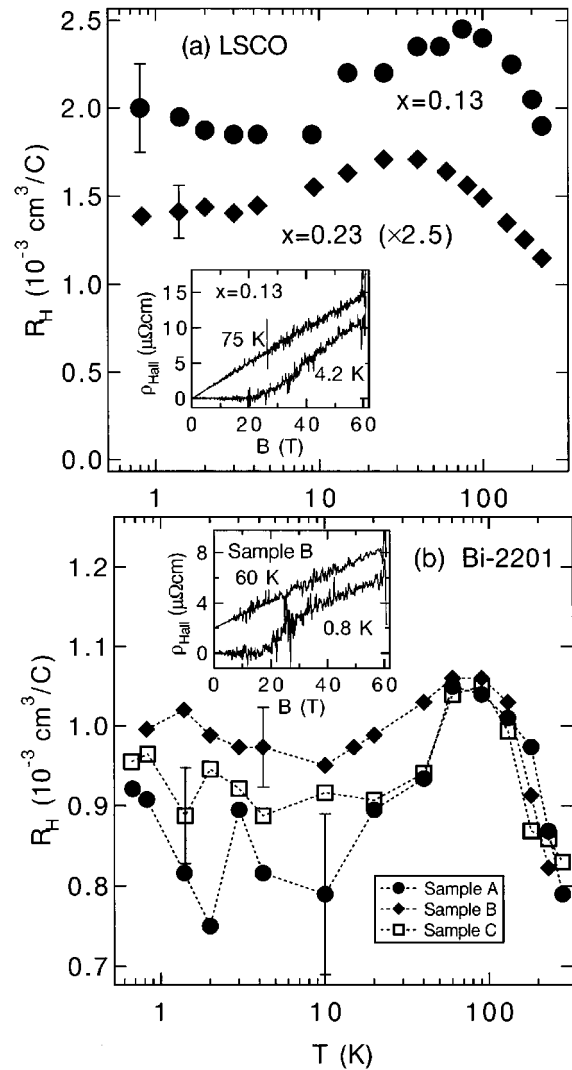


FIG. 2. T dependence of the normal state R_H for (a) the underdoped and overdoped (scaled as indicated) LSCO thin films and (b) the three La-doped Bi-2201 single crystals. A typical error bar is shown for each data set. The insets show examples of the Hall resistivity traces above and below T_c [The 60-K data in the inset (b) is shifted up for clarity].

the transverse voltage. (The asymmetrical component of the longitudinal voltages is always negligible.)

Figure 1(a) shows ρ_{ab} of the underdoped ($x=0.13$) and overdoped ($x=0.23$) LSCO films. Several features of the data reproduce previous measurements on LSCO single crystals:^{1,2,14} The normal state magnetoresistance is small, evidenced in Fig. 1(a) by the superposition of the 60-T data (symbols) on the 0-T data above T_c . Also, the normal-state ρ_{ab} of the underdoped film crosses from metallic behavior above T_c to strong insulating behavior at low temperatures. In contrast, ρ_{ab} of the overdoped film stays metallic down to the lowest experimental temperature.¹⁵ Finally, the insulating ρ_{ab} of the underdoped film appears to diverge as $\log(1/T)$ (inset), the same temperature dependence reported in LSCO single crystals.^{1,14}

Similar data from three different Bi-2201 crystals is given in Fig. 1(b). All three crystals have the same nominal com-

position, the same $T_c \sim 13$ K, and were grown from the same batch in the same crucible. The fact that ρ_{ab} differs by more than a factor of 2 among these crystals presumably reflects significantly varying disorder, which is believed to be due to oxygen vacancies in the CuO_2 planes.¹⁷ Only sample C, with $\rho_{ab} \leq 100 \mu\Omega \text{ cm}$, remains metallic down to the lowest temperatures once superconductivity is suppressed with the magnetic field, a result which is consistent with a previous report on a different series of Bi-2201 crystals.³ The inset to Fig. 1(b) shows that the weakly insulating behavior in samples with higher resistivities is consistent with $\log(1/T)$. This result suggests that the logarithmic divergence of ρ_{ab} arises, in part, from the presence of disorder in the samples.

Figure 2 shows Hall data on the same sets of samples. The inset of Fig. 2(a) shows two traces of the Hall resistivity, ρ_{Hall} , versus magnetic field, B , for the underdoped LSCO film at temperatures above and below T_c . Note that ρ_{Hall} is linear in magnetic field in the absence of superconductivity, which yields an unambiguous value for the normal state R_H for these samples. The main panel of Fig. 2(a) shows the temperature dependence of the normal state R_H for the two LSCO thin films. Above T_c , the underdoped and overdoped films show broad peaks in R_H at ~ 80 and ~ 40 K, respectively, whose magnitude and positions are consistent with data in the literature.¹⁸ The intense magnetic field, however, reveals that both samples exhibit a nearly temperature-independent R_H at low temperatures (the data are consistent with a constant R_H , although there is a $\sim 15\%$ experimental uncertainty). This common low-temperature saturation of R_H occurs despite the drastically contrasting behavior of ρ_{ab} shown in Fig. 1(a). In particular, note that ρ_{ab} in the underdoped film changes by a factor of 2 between 0.8 and 50 K, while the change in R_H is less than 30%.

Figure 2(b) shows the normal-state R_H of the three Bi-2201 crystals, along with representative ρ_{Hall} traces in the inset. Note that, once again, the normal state ρ_{Hall} depends linearly on B , although the noisier traces from the Bi-2201 single crystals yield larger error bars in determining R_H . The large temperature dependence of R_H at high temperatures and the broad peak in R_H near 100 K are, as in the LSCO data, consistent with previous reports.¹⁷ As with the LSCO thin films, however, R_H of the Bi-2201 crystals becomes essentially temperature independent at low temperatures, at least within the experimental resolution.

Although it is not clear how to properly interpret the low-temperature normal-state R_H of the high- T_c cuprates, it is clear that the hole-doped LSCO and Bi-2201 behave in stark contrast to the electron-doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$, in which ρ_{ab} is constant yet R_H continues to be strongly temperature dependent down to low temperatures.¹⁹ Also, there is no obvious accounting for the data of Figs. 1 and 2 in terms of conventional disordered two-dimensional (2D) metals. As discussed earlier, a logarithmically divergent ρ_{ab} can result from disorder-enhanced electron interactions; however, this mechanism is effectively ruled out because the logarithmically divergent ρ_{ab} is not accompanied by a similarly divergent R_H . On the other hand, while conventional weak localization gives a temperature-independent R_H , it has previously been ruled out as a mechanism underlying the observed $\log(1/T)$ dependence of ρ_{ab} .^{1,9,10} Further evidence

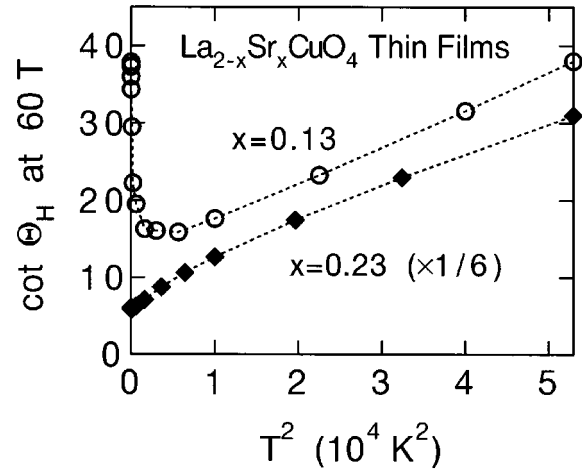


FIG. 3. $\cot \Theta_H$ ($\equiv \rho_{xx}/\rho_{\text{Hall}}$) at 60 T versus T^2 for the underdoped and overdoped LSCO films (scaled as indicated).

against conventional theories comes in an attempt to interpret the magnitude of the low-temperature R_H in terms of a carrier density. Such an approach gives a carrier number per Cu atom of 0.3 (LSCO $x=0.13$), 0.9 (LSCO $x=0.23$), and 0.6 (Bi-2201). While each of these values might suggest a “large” Fermi surface in the cuprates, they are significantly larger than the carrier concentration expected from the nominal doping.

Among the models specific to high- T_c cuprates which have been proposed to account for the logarithmic divergence of ρ_{ab} and ρ_c , some include a constant R_H at low temperatures. In considering disorder effects in 2D Luttinger liquids, Anderson *et al.*⁴ discuss a low-temperature regime in which scattering due to disorder inhibits spin-charge separation. In this regime, ρ_{ab} and ρ_c both diverge with the same weak power law and, more importantly for this data, the Hall resistivity is expected to be temperature independent. In a bipolaron model of high- T_c superconductivity, a random disorder potential leads to carrier localization marked by a logarithmically divergent ρ_{ab} and a constant R_H at low temperatures.⁵ In the marginal Fermi liquid theory, the logarithmically divergent ρ_{ab} results from a suppression in the density of states, yet the R_H will be constant.²⁰

Finally, we discuss our data in terms of the now-well-established T^2 dependence of the cotangent of the Hall angle, $\cot \Theta_H \equiv \rho_{xx}/\rho_{\text{Hall}}$. This behavior has been observed in many high- T_c cuprates above T_c and is believed to be a consequence of two different scattering times for carrier motion along and perpendicular to the Fermi surface.^{18,21} Figure 3 contains plots of $\cot \Theta_H$ at 60 T versus T^2 for our underdoped and overdoped LSCO films.²² The Hall angle of the overdoped sample does not obey the T^2 law, consistent with Hwang *et al.*¹⁸ Rather, there is a slightly sub- T^2 behavior which continues down to the lowest temperatures without any apparent crossover to a low-temperature regime. In contrast, the underdoped film shows a good T^2 behavior above ~ 80 K ($\sim 6 \times 10^3 \text{ K}^2$) and a rapid increase at lower temperatures. Note that ~ 80 K is a characteristic temperature for this sample: in Fig. 1(a), it marks the first deviation of ρ_{ab} from the high-temperature T -linear behavior; while in Fig. 2(a), it marks the maximum in R_H . The break down of the T^2 be-

havior of $\cot\Theta_H$ may suggest that impurity scattering dominates at low temperatures and a *single* impurity-scattering time dominates the two separate scattering times which govern the higher temperature behavior. This is reminiscent of the behavior reported in the heavily overdoped $\text{Ti}_2\text{Ba}_2\text{CuO}_y$ system,²³ where lifetime separation disappears at low temperatures and a single scattering time with an anomalous temperature dependence governs the carrier transport.

To summarize, a 61-T pulsed magnetic field suppresses superconductivity and reveals an essentially temperature-independent normal-state Hall coefficient, R_H , at low tem-

peratures. This behavior is observed in both underdoped and overdoped LSCO thin films, despite the fact that ρ_{ab} in these samples varies from insulating to metallic. The normal-state R_H is also found to be nearly temperature independent in a series of Bi-2201 single crystals in which increasing disorder causes the low-temperature ρ_{ab} to cross from metallic to insulating behavior.

We thank M. Berkowski for growing the LaSrAlO_4 substrates. We gratefully acknowledge helpful discussions with E. Abrahams, P.W. Anderson, A.P. Mackenzie, and C.M. Varma.

*Present address: Department of Electrical Engineering, Princeton University, Princeton, NJ 08544.

- ¹Y. Ando, G. S. Boebinger, A. Passner, T. Kimura, and K. Kishio, *Phys. Rev. Lett.* **75**, 4662 (1995).
- ²G. S. Boebinger, Y. Ando, A. Passner, T. Kimura, M. Okuya, J. Shimoyama, K. Kishio, K. Tamasaku, N. Ichikawa, and S. Uchida, *Phys. Rev. Lett.* **77**, 5417 (1996).
- ³Y. Ando, G. S. Boebinger, A. Passner, N. L. Wang, C. Geibel, and F. Steglich, *Phys. Rev. Lett.* **77**, 2065 (1996).
- ⁴P. W. Anderson, T. V. Ramakrishnan, S. Strong, and D. G. Clarke, *Phys. Rev. Lett.* **77**, 4241 (1996).
- ⁵A. S. Alexandrov, V. V. Kabanov, and N. F. Mott, *Phys. Rev. Lett.* **77**, 4796 (1996); A. S. Alexandrov (unpublished).
- ⁶B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interactions in Disordered Systems*, edited by M. Pollak and A. L. Efros (North-Holland, Amsterdam, 1985), pp. 1-53.
- ⁷G. Kotliar and C. M. Varma, *Physica A* **167**, 288 (1990); C. M. Varma, *Phys. Rev. B* **55**, 14 554 (1997); and (unpublished).
- ⁸Y. Zha, S. L. Cooper, and D. Pines, *Phys. Rev. B* **53**, 8253 (1996); and (private communication).
- ⁹N. W. Preyer, M. A. Kastner, C. Y. Chen, R. J. Birgeneau, and Y. Hikada, *Phys. Rev. B* **44**, 407 (1991).
- ¹⁰T. W. Jing, N. P. Ong, T. V. Ramakrishnan, J. M. Tarascon, and K. Reimann, *Phys. Rev. Lett.* **67**, 761 (1991).
- ¹¹N. L. Wang, B. Buschinger, C. Geibel, and F. Steglich, *Phys. Rev. B* **54**, 7449 (1996).
- ¹²N. L. Wang, Y. Chong, C. Y. Wang, D. J. Huang, Z. Q. Mao, L. Z. Cao, and Z. J. Chen, *Phys. Rev. B* **47**, 3347 (1993).
- ¹³I. E. Trovimon, L. A. Johnson, K. V. Ramanujachary, S. Guha, M. G. Harrison, M. Greenblatt, M. Z. Cieplak, and P. Lindemfeld, *Appl. Phys. Lett.* **65**, 2481 (1994).

- ¹⁴Y. Ando, G. S. Boebinger, A. Passner, K. Tamasaku, N. Ichikawa, S. Uchida, M. Okuya, T. Kimura, J. Shimoyama, and K. Kishio, *J. Low Temp. Phys.* **105**, 867 (1996).
- ¹⁵We note that the $x=0.23$ film is apparently disordered since ρ_{ab} is roughly a factor of 2 larger than single crystals with the same Sr concentration (Refs. 2 and 16). This yields a ρ_{ab} in this overdoped film which is comparable to that of the underdoped film, yet ρ_{ab} remains metallic down to the lowest temperatures. This provides further evidence that the metallic behavior in the overdoped regime is robust against disorder and that carrier concentration plays an important role in determining whether the low-temperature behavior is insulating or metallic (Ref. 2).
- ¹⁶T. Kimura, S. Miyasaka, H. Takagi, K. Tamasaku, H. Eisaki, S. Uchida, K. Kitazawa, M. Hiroi, M. Sera, and N. Kobayashi, *Phys. Rev. B* **53**, 8733 (1996).
- ¹⁷A. P. Mackenzie, S. D. Hughes, J. R. Cooper, A. Carrington, C. Chen, and B. M. Wanklyn, *Phys. Rev. B* **45**, 527 (1992).
- ¹⁸H. Y. Hwang, B. Batlogg, H. Takagi, H. L. Kao, J. Kwo, R. J. Cava, J. J. Krajewski, and W. F. Peck, *Phys. Rev. Lett.* **72**, 2636 (1994).
- ¹⁹Z. Z. Wang, T. R. Chien, N. P. Ong, J. M. Tarascon, and E. Wang, *Phys. Rev. B* **43**, 3020 (1991).
- ²⁰C. M. Varma (private communication).
- ²¹T. R. Chien, Z. Z. Wang, and N. P. Ong, *Phys. Rev. Lett.* **67**, 2088 (1991).
- ²²The $\cot\Theta_H$ of Bi-2201 has been known not to obey the T^2 law (Ref. 17). The complicated $\cot\Theta_H$ temperature dependence exhibited by our Bi-2201 crystals is consistent with this earlier report.
- ²³A. P. Mackenzie, S. R. Julian, D. C. Sinclair, and C. T. Lin, *Phys. Rev. B* **53**, 5848 (1996).