Hall coefficients and resistivities of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals: The presence of a mobility edge in correlated two-dimensional electron system

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Hall coefficients and resistivities of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals: The presence of a mobility edge in correlated two-dimensional electron system

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Hall coefficients and resistivities of a series of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals were measured to investigate the relation between the carrier concentration n and the resistivity-minimum temperature $T_{\min}$. The $T_{\min}$ was found to obey a simple linear relation: $T_{\min} \propto n_c - n$, where $n_c \simeq 4.2 \times 10^{21}\,\mathrm{cm}^{-3}$ is a critical carrier concentration. A model of two-dimensional electron systems with a mobility edge was introduced to explain this linear relation, and the presence of a mobility edge in two-dimensional copper-oxide-based metals was suggested. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1509842]

I. INTRODUCTION

Recent studies have revealed that a new kind of two-dimensional (2D) metallic phase can exist, according to the results of the dynamical scaling theory.[1,2] Such 2D metallic phases have been experimentally observed: Longitudinal resistivity in the quantum Hall system varies from infinity (quantum Hall insulator) to zero (quantum Hall liquid) by the tuning of the applied field, and a metal phase emerges between the two;[3] and superconductivity of a molecular-beam-epitaxy-grown $Nd_{2-x}Ce_xCuO_{4-y}$ crystal is suppressed by the applied field, and a metal phase is observed just at the superconductor-insulator (SI) transition.[4] Similar results are also obtained in the SI transition of ultrathin films of bismuth.[5] In addition, a different type of 2D metal-insulator transition at zero field has been proposed to explain the experimental results in metal-oxide-semiconductor field-effect transistors.[6] These theoretical and experimental developments exhibit a striking contrast to the well-known nature of conventional noninteracting 2D systems, in which all electron states are localized and there is no 2D metallic state.[7] These discussions seem analogous to those of minimum conductivity, first proposed by Mott,[8] and later clearly denied by the scaling theory of noninteracting 2D electron gas.[7]

High-T_c superconducting cuprates provide a stage for the investigation of a 2D electron system with strong correlation. The normal state of cuprates can be explained as a 2D doped Mott insulator. The CuO_2 plane of the cuprates show metallic behavior when carriers are doped. Since the word ''metallic'' here implies the electron system at finite temperatures exhibits a positive temperature coefficient of resistivity ($dR/dT>0$), it is important to deduce the behavior of the electron system at zero temperature from the data obtained at finite temperatures. $Bi_{2+x}Sr_{2-x}CuO_y$ is an ideal material to study a 2D electron system in the cuprates because of low-superconducting transition temperature ($T_c \sim 8\,K$) even at the optimum carrier concentration and its highly anisotropic electric conduction ($\sigma_{ab}/\sigma_c > 10^4$).[9] In this article, we provide experimental results for resistivity and Hall coefficient of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals of different carrier concentrations. Accurate control of geometry by use of the excimer-laser cutting technique allows us to evaluate the resistivity and the carrier concentration of each crystal and to investigate the evolution of the resistivity as a function of the carrier concentration. We found that the temperature of the resistivity-minimum $T_{\min}$ obeys a simple relation to the carrier concentration n: $T_{\min} \propto n_c - n$, where $n_c \simeq 4.2 \times 10^{21}\,\mathrm{cm}^{-3}$ is a critical carrier concentration. This relation suggests the presence of a mobility edge in the two-dimensional CuO_2 planes of the superconducting cuprates.

II. EXPERIMENT

The $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals used in this study were synthesized by the self-flux method. We changed the ratio of starting powders Bi_2O_3 and $SrCO_3$, so that we could control Bi/Sr substitution. A composite powder consisting of Bi_2O_3, $SrCO_3$, and CuO was placed in an MgO crucible, and the mixture was heated in a furnace at 1000 °C for 4 h. The furnace was cooled at the rate of 2 °C/h. All processes were carried out under standard atmosphere. The resulting crystals were black and slabshaped.

The obtained crystals were cut into the shape of the Hall-bar by repeating step-by-step laser drilling. The laser system consists of a Kr-F excimer laser source (wavelength 248 nm), an attenuator, a lens (f=1:300), and a computer-controlled x-y stage. The beam was focused to a $50 \times 50\,\mu\mathrm{m}^2$ spot. The sample was fixed on the x-y stage, which can be positioned with an accuracy of 2 μm. Shortly we will discuss why such accuracy is required for this study. $Bi_{2+x}Sr_{2-x}CuO_y$ crystals of 10 to 200 μm thickness were able to be pierced by 50 to 1000 laser pulses. The typical size of the sample is 2.7 mm in length, and 0.5 mm in width. The sample has two potential terminals and two Hall terminals, the size of which is $0.3 \times 0.4\,\mathrm{mm}^2$. Sample thickness t was calculated from the sample area and density. Weight of the sample was measured by a microbalance (Mettler M3) up to the order of 0.1 μg. The typical thickness was ~10 μm.

The resistivity of the samples was measured by the standard four probe method with a current of less than 100 μA.

The Hall voltage and the voltage drop across the sample was monitored with a nanovoltmeter (Keithley 182). In the Hall measurement, magnetic fields up to 4 T were applied perpendicular to the CuO_2 plane. We measured the Hall voltages of both directions in order to eliminate the offset voltage at zero field. We used a battery-operated current generator to minimize the line noise induced by the power lines.

It is important to evaluate if the geometry of the Hallbars have the accuracy required for this study. Magnetoresistance will appear on Hall terminals, if the terminals are not positioned on the same equipotential line. When the Hall terminals are misaligned by Δl, a voltage drop across the Hall terminals occurs

$$V = I\rho\frac{\Delta l}{tw}, \tag{1}$$

where ρ is resistivity, t is thickness, and w is width. Magnetoresistance effect may change ρ in Eq. (1) as much as a few percent. Since the Hall voltage is represented as $V_H = R_H IB/t$, we compared it to the voltage drop across the Hall terminals

$$\frac{\Delta V}{V_H} = \frac{\rho}{R_H B}\frac{\Delta l}{w}. \tag{2}$$

In cuprates, the typical resistivity $\rho \sim 10^{-6}\,\Omega$m, and the Hall coefficient $R_H \sim 10^{-9}\,C^{-1}\,m^3$. To make $\Delta V/V_H$ comparable, thus $\Delta l/wB \sim 10^{-3}\,(\text{tesla})^{-1}$ is required. Assuming a magnetic field of several teslas, and a sample width $w \sim 1$ mm, one should align the Hall terminals within $\Delta l < 10^{-3}$ mm to satisfy the above conditions. We attained this accuracy by means of the laser cutting technique described above. We also estimated Δl by the voltage drop across the Hall probes at zero magnetic field. The electrically estimated misalignment of Δl was, however, the order of 10^{-2} mm. Typically, $\Delta V/V_H \sim 100$ was obtained.

III. RESULTS

Figure 1 shows that the temperature dependence of the resistivity in $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals with $x=0.0$ (hereafter A), 0.2 (B), 0.3 (C), and 0.4 (D and E). Data for a sample showing a superconducting transition is also shown as a reference (S). It is shown that the resistivity depends systematically on the Bi/Sr ratio of the samples. Sample A showed a superconducting transition at 8 K, whereas B, C, D, and E showed no superconducting transition down to 1.8 K. We found that the SI transition lies between samples A and B in the observed $Bi_{2+x}Sr_{2-x}CuO_y$ system. We calculated the sheet resistance per CuO_2 plane, taking the separation between CuO_2 planes as 12 Å.[10] The sheet resistance of 1 kΩ corresponds to the resistivity $1.2\times10^{-4}\,\Omega$ cm. It was proposed that associated with the superconductor-insulator transition is a universal critical resistance $h/4e^2(6.45\,\text{k}\Omega)$.[1] The critical sheet resistance obtained here was about two times this value.

Sample A exhibits ln T upturn in the resistivity in the temperature regime from superconducting transition temperature to 50 K. In the weak localized regime, the conductivity obeys the following logarithmic form:[7]

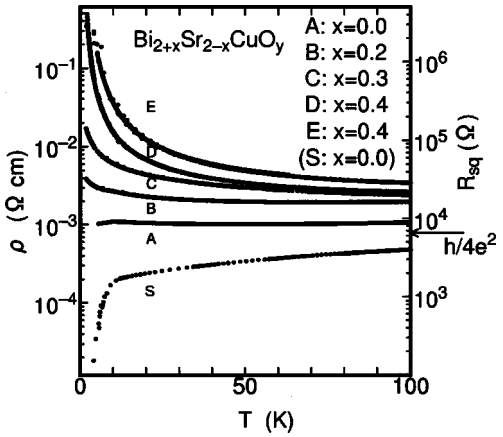

FIG. 1. Resistivities of the samples with different Bi/Sr ratio. The sheet resistance of 1 kΩ corresponds to the resistivity $1.2\times10^{-4}\,\Omega$ cm. The residual resistivity becomes higher when a significant amount of Sr is substituted by Bi.

$$\sigma(T) = \sigma_0 + \alpha p(e^2/2\pi^2\hbar)\ln T, \tag{3}$$

where α is a parameter of the order of unity, and p is determined as $\tau_i \propto T^{-p}$. We obtained $\sigma_0 = 9.24\times10^{-5}\,\Omega^{-1}$ and $\alpha p(e^2/2\pi^2\hbar) = 7.17\times10^{-6}\,\Omega^{-1}$, hence, $\alpha p = 0.57$. Consequently, it is shown that the behavior of sample A is described in terms of the weak localization. The possibility of the resistivity upturn being due to magnetic impurities (the Kondo effect) should also be evaluated. Several authors including us have independently reported anisotropic magnetoresistance in $Bi_2Sr_2CuO_y$.[11,12] The anisotropy of the magnetoresistance is a reliable indicator for Anderson localization. In contrast, for the Kondo effect the magnetoresistance is purely isotropic. We shall therefore, interpret the resistivity upturn on the basis of the Anderson localization.

We evaluated the disorder of the crystals from the residual resistivity. The randomness of a system is characterized by the metallic parameter $k_F\ell$, where k_F is the Fermi wave vector and ℓ is an elastic mean free path. The metallic parameter $k_F\ell$ is related to the residual sheet resistance R_0

$$1/R_0 = (e^2/h)k_F\ell. \tag{4}$$

We obtained $k_F\ell = 4.0$ for sample A, 2.0 for B, 1.6 for C, 1.4 for D, and 1.1 for E. This result suggests that the Bi/Sr substitution effectively enhances system disorder. If much Bi/Sr substitution were carried out up to the Ioffe-Regel limit $k_F\ell \sim 1$, the carriers could not extend over the sample due to disorder. However, since $k_F\ell > 1$ for the all substitutions, our system remains metallic.

Figure 2 shows the carrier concentration of samples A, B, C, D, and E, defined as $n = 1/R_H e$, where R_H is the Hall coefficient. This treatment is justified by applying a single-carrier model as the first approximation. We found the following two characteristics in the Hall coefficients: the carrier concentration became smaller when Sr sites were substituted by excess Bi atoms; and the Hall coefficients of each sample were temperature independent. The observed temperature independent Hall coefficient is consistent with the Anderson localization picture.[13] The carrier concentrations of the samples are 3.6 ± 0.4, 2.6 ± 0.3, 2.0 ± 0.2, 1.1 ± 0.1, and 0.9

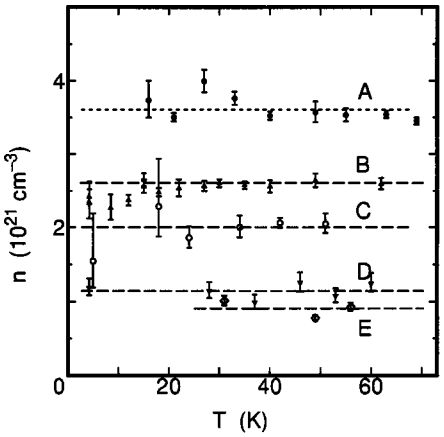

FIG. 2. Carrier concentration of samples A, B, C, D, and E, determined as $n=1/R_{\mathrm{H}}e$, where R_{H} is the Hall coefficient. The carrier concentration becomes smaller when a significant amount of Sr is substituted by Bi. It is shown that the Hall coefficient of each sample is independent of temperature.

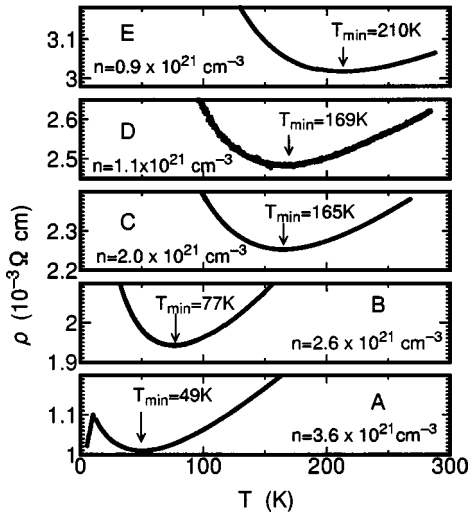

FIG. 3. Resistivity minima of samples A, B, C, D, and E. The temperatures of the resistivity minima depend on the carrier concentration.

$\pm 0.1\times 10^{21}\,\mathrm{cm}^{-3}$ for A, B, C, D, and E, respectively. These are equivalent to the carrier number per each Cu site as 0.25 ± 0.03, 0.18 ± 0.02, 0.14 ± 0.01, 0.08 ± 0.01, and 0.06 ± 0.01.[10]

IV. DISCUSSIONS

A striking result is the discrepancy of temperature dependencies between the resistivity, increasing by several orders, and the Hall coefficient, being almost constant. This discrepancy between the resistivity and the Hall coefficient has been observed in various superconducting cuprates, such as ion-beam-damaged YBCO films,[14] BiSr(Ca, Y)CuO single crystals,[15] polycrystalline BiSr(Ca, Pr)CuO,[16] and BiSrCuO single crystals.[11,17] We focus on the temperatures at resistivity minima T_{min} to understand the discrepancy. Figure 3 is a close up of the resistivity variation with temperature in the region of T_{min} for different carrier concentrations n determined from the Hall coefficients. The temperature T_{min} systematically increases as carrier concentration n decreases.

A few authors have discussed the physical interpretation of the T_{min} in terms of localization theories: the temperature when the inelastic scattering length becomes comparable to the localization length,[18] and the energy difference between the Fermi level and the mobility edge.[16]

We analyze the experimental results, making use of the idea that T_{min} represents the energy difference between the Fermi level and the mobility edge.[16] The resistivity has a metal-like temperature dependence $(d\rho/dT>0)$ above T_{min}, whereas it shows insulating properties below T_{min}. The insulating sample $[d\rho/dT(T\rightarrow 0)<0]$ can also exhibit metallic behavior at finite temperatures for which the thermal energy $k_{\mathrm{B}}T$ exceeds the difference between the mobility edge E_c and the Fermi level E_{F}. Therefore, the temperature of resistivity minimum is given by the following formula:[16]

$$k_B T_{\mathrm{min}} \sim |E_c - E_{\mathrm{F}}|. \tag{5}$$

We now consider two-dimensional electron systems with a mobility edge. The Fermi energy E_{F} is proportional to the carrier concentration, that is,

$$E_{\mathrm{F}} \propto k_{\mathrm{F}}^2 \propto n. \tag{6}$$

Hence, Eqs. (5) and (6) lead to a relation between the T_{min} and n,

$$T_{\mathrm{min}} \propto n_c - n, \tag{7}$$

where n_c is a critical carrier concentration, if the mobility edge E_c is not sample dependent.

To see if the relation (7) is satisfied, we plot T_{min} as a function of n in Fig. 4. It is shown that the relation (7) fits our data well, and the critical carrier concentration is obtained as $n_c \simeq 4.2\times 10^{21}\,\mathrm{cm}^{-3}$. This value corresponds to $5.0\times 10^{14}\,\mathrm{cm}^{-2}$ (or 0.35 per Cu site) in terms of two-dimensional electron systems.

The model of two-dimensional electron systems with a mobility edge describes our experimental results of the temperature dependences of the resistivities and Hall coefficients of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals. By extrapolating our re-

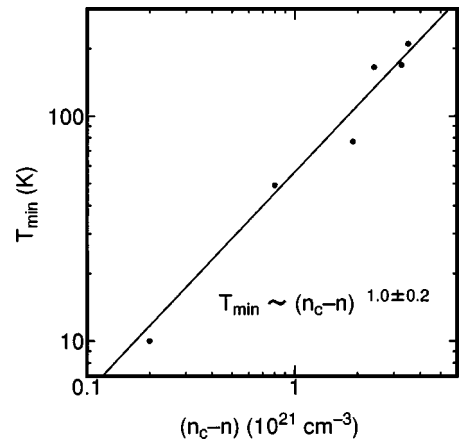

FIG. 4. Relation between the resistivity-minimum temperature T_{min} and the carrier concentration n.

sults to $T_{\min} \rightarrow 0\,\mathrm{K}$, we predict that a metal-insulator transition should take place in $Bi_{2+x}Sr_{2-x}CuO_y$ when E_F is equal to E_c at $n = n_c$, and a two-dimensional metallic system will appear at the phase transition point. We believe further investigation to determine the critical exponent will reveal the details of this noble type of metal-insulator transition, by lower temperature studies of the cuprates whose carrier concentrations lay in the neighborhood of $n = n_c$.

V. CONCLUSIONS

In conclusion, we present the experimental results for resistivity and Hall coefficient of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals of different carrier concentrations. We found that the temperature of the resistivity-minimum $T_{\min}$ obeys a simple relation to the carrier concentration n: $T_{\min} \propto n_c - n$, where $n_c \simeq 4.2 \times 10^{21}\,\mathrm{cm}^{-3}$ is a critical carrier concentration. This relation suggests the presence of a mobility edge in two-dimensional CuO_2 planes of the superconducting cuprates.

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