Hall coefficients in the superconductor-insulator transition of $Bi_{2+x}Sr_{2-x}CuO_y$: Measurement of Hall-bar shaped single crystals fabricated by excimer-laser cutting

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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 and the disorder of the system in the superconductor-insulator transition. Excimer-laser cutting technique was applied to fabricate the Hall-bar shaped samples in order to determine precisely the transport properties of the single crystals. The Hall coefficients were found to be independent of the temperature, which suggests Coulomb interaction was negligible in the localized regime of $Bi_{2+x}Sr_{2-x}CuO_y$. We found the Ioffe–Legel condition $k_F l \sim 1$ laid around $n_0 \sim 8\times10^{20}$ cm^{-3} in this system, and the carriers in the localized regime were essentially ''metallic.'' © *1996 American Institute of Physics.* [S0003-6951(96)02946-4]

The normal state of the superconducting cuprate can be explained as a two-dimensional doped Mott insulator. The CuO_2 plane becomes metallic when the sufficient amount of carriers are doped on it, that is, the superconductor-insulator (S-I) transition occurs at a certain threshold carrier concentration. In the cuprates, however, the doped carriers also introduce disorder into the CuO_2 plane. It is well known that the nature of transport phenomena is changed by the disorder of system: in noninteracting two-dimensional electron system, any amount of disorder can prevent the system from being metallic.[1] Consequently, in investigating the transport phenomena in S-I transition of cuprates, it is necessary to distinguish the effects due to varying of carrier concentration and those of disorder.

$Bi_{2+x}Sr_{2-x}CuO_y$ is an ideal system to observe the S-I transition in cuprates, since it has the properties of lower superconducting transition temperature ($T_c \sim 8$ K) and anisotropic electric conduction ($\sigma_{ab}/\sigma_c > 10^4$).[2] This system changes from superconductor to insulator by doping excess Bi and Sr sites into stoichiometric $Bi_2Sr_2CuO_6$.[3,4] The excess Bi^{3+} ions are thought to compensate carriers in the CuO_2 planes. They are also regarded as the source of random potential.[5–7] To date, the relation between the carrier concentration and the disorder remains unknown.

In this letter, we present the experimental results of resistivity and Hall coefficient in the S-I transition of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals, aiming to investigate the relation between the disorder and the carrier concentration. To determine both of sheet resistance and Hall coefficient precisely, it is necessary to know accurate dimensions of the sample. Recently, excimer laser become available to apply for material processing, such as drilling and cutting. We used laser cutting technique to fabricate the Hall bars of $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals. We, thereby, obtained accurate control of the dimensions of the single crystals. We were able to observe the S-I transition in a series of crystals, by varying the Bi/Sr ratio. We found the relation between the disorder and the carrier concentration in the insulator side of S-I transition of $Bi_{2+x}Sr_{2-x}CuO_y$.

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 $SrCO_3$ to 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 reacted in a furnace at 1000 °C for 4 h. The furnace was cooled at the rate of 2 °C/h. All the processes were carried out under standard atmosphere. The resulting crystals were black and slab-shaped.

The samples were cut into the shape of the Hall bar by repeating step-to-step laser drilling. The wavelength of the laser we used to fabricate to the Hall bars was 248 nm. The laser passed through an attenuator and a lens ($f = 1{:}300$). The beam was focused to a 50×50 μm^2 spot. The sample was fixed on a computer-controlled x-y stage. The x-y stage was moved along the programmed position with an accuracy of 2 μm. $Bi_{2+x}Sr_{2-x}CuO_y$ crystals of 10–200 μm thickness were able to be pierced by 50–1000 shots of laser pulse. Figure 1 shows a picture of the fabricated Hall bar of

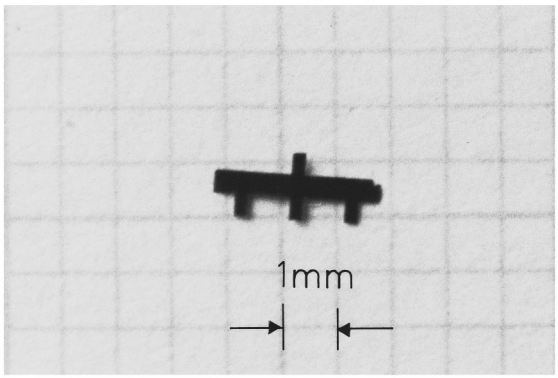

FIG. 1. A Hall-bar shaped $Bi_{2+x}Sr_{2-x}CuO_y$ single crystal. The sample was cut by repeating step-by-step laser drilling. Sample size was 2.7 mm in length and 0.5 mm in width.

$Bi_{2+x}Sr_{2-x}CuO_y$ single crystal. The size of sample shown in Fig. 1 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 0.3×0.4 mm^2. Sample thickness t was calculated from 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 sample was measured by the standard four probe method. The passing current was less than 100 μA. The voltage drop across the sample was piked up by a nanovoltmeter (Keithley 182). In the Hall measurement, magnetic fields up to 4 T were applied perpendicular to the CuO_2 plane. The voltages across the Hall terminals were measured by the nanovoltmeter. We measured the Hall voltages of both direction in order to eliminate the offset voltage at zero field. The passing current through the samples was 5 mA for all Hall measurement. We used a battery-operated current generator to minimize the line noise induced by the power lines.

Before we proceed to the experimental results, we briefly discuss the required accuracy of sample dimensions. Magnetoresistance will appear on Hall terminals, if the terminals are not positioned on the same equipotential line. When the Hall terminals have misalignment of the length Δl, the 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 $V_H=R_HIB/t$, we compared it to the voltage drop across the Hall terminals:

$$\frac{\Delta V}{V_H}=\frac{\rho}{R_HB}\,\frac{\Delta l}{w}. \tag{2}$$

In cuprates, the typical resistivity $\rho\sim10^{-6}$ Ω m, and the Hall coefficient $R_H\sim10^{-9}$ C^{-1} m^3. To make $\Delta V/V_H$ comparable, thus $\Delta l/wB\sim10^{-3}$ T^{-1} is required. Assuming the magnetic field of a several teslas, and sample width $w\sim1$ mm, one should align the Hall terminals with $\Delta l<10^{-3}$ 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 Δl was, however, the order of 10^{-2} mm. Typically, $\Delta V/V_H\sim100$ was obtained.

Figure 2 shows that the temperature dependence of the resistivity in $Bi_{2+x}Sr_{2-x}CuO_y$ single crystals with $x=0.0$ [one is metallic (A) and the other weakly-localized (B)], 0.2, 0.3, 0.4 (hereafter C, D, and E, respectively). It is shown that the resistivities are dependent systematically of the Bi/Sr ratio. Samples A and B showed superconducting transition at 8 K, whereas C, D, and E showed no superconducting transition down to 1.8 K. We found that the S-I transition lays between the samples B and C in the observed $Bi_{2+x}Sr_{2-x}CuO_y$ system. We calculated the sheet resistance per CuO_2 plane, considering the separation between CuO_2 planes is 12 Å. The sheet resistance of 1.0 kΩ corresponds to the resistivity 1.2×10^{-4} Ω cm. It is proposed that the superconductor-insulator transition has a universal critical resistance $h/4e^2$ (6.45 kΩ).[8] The critical sheet resistance obtained here was about two times of this value.

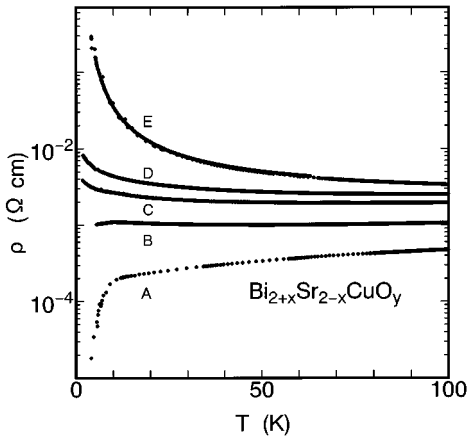

FIG. 2. Resistivities of the samples with different Bi/Sr ratio. The sheet resistance of 1.0 kΩ corresponds to the resistivity 1.2×10^{-4} Ω cm. The residual resistivity becomes higher when much Sr is substituted by Bi. Sample B exhibits the weak localization, whereas C, D, and E show the variable range hopping conduction.

We evaluated the disorder of the crystals from the residual resistivity. Randomness of a system is characterized by a metallic parameter k_Fl, where k_F is a fermi wave vector and l is an elastic mean-free path. The metallic parameter k_Fl is related with residual resistivity:

$$\sigma_0=1/\rho_0=(e^2/h)k_Fl. \tag{3}$$

We obtained $k_Fl=4.0$ for sample B, 2.5 for sample C, and 1.5 for sample D. This result clearly shows that the Bi/Sr substitution enlarges the degree of disorder of the system. If much Bi/Sr substitution is carried out up to the Ioffe–Legel limit $k_Fl\sim1$, the carriers cannot extend over the sample due to disorder, hence the sample changes to insulator.

Sample B 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:[1] $\sigma(T)=\sigma_0+ap(e^2/2\pi^2\hbar)\ln T$, where α is a parameter of the order of unity, and p is determined as $\tau_i\propto T^{-p}$. We obtained $\sigma_0=9.24\times10^{-5}$ Ω^{-1} and $\alpha p(e^2/2\pi^2\hbar)=7.17\times10^{-6}$ Ω^{-1}, hence $\alpha p=0.58$. Consequently, it is shown that the behavior of sample B is described in terms of the weak localization. The residual resistivities of samples C, D, and E are larger than the weakly-localized sample B. The carriers in these samples are, hence, thought to affect the stronger disorder due to much Bi/Sr substitution. Instead of the logarithmic behavior, the electric conduction in these sample exhibited the ''variable range hopping'' behavior: $\sigma\propto\exp(-T_0/T)^\alpha$, where $\alpha=1/(d+1)$, d is the dimension of system.[5]

Figure 3 shows the carrier concentration of samples B, C, D, and E, determined as $n=1/R_He$, where R_H is the Hall coefficient. We found the two characteristics in the Hall coefficients as follows: the carrier concentration became smaller when much Sr sites were substituted by excess Bi atoms; and the temperature dependence of each sample has

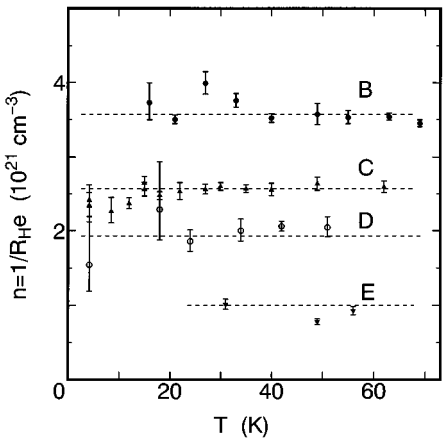

FIG. 3. Carrier concentration of samples B, C, D, and E, determined as $n=1/R_{\mathrm{H}}$, where R_{H} is the Hall coefficient. The carrier concentration becomes smaller when much Sr is substituted by Bi. It is shown that the Hall coefficient of each sample is independent of temperature.

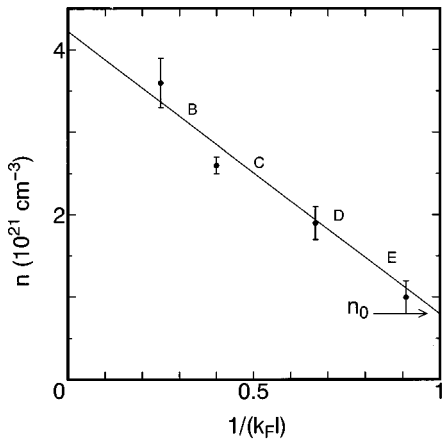

FIG. 5. Relation between disorder and carrier concentration. The disorder is indexed by the metallic parameter $1/k_{\mathrm{F}}l$. The Ioffe–Legel limit $k_{\mathrm{F}}l\sim 1$ lays around $n_0\sim 8\times 10^{20}$ cm^{-3}.

the same behavior. It is surprising that the Hall coefficients are temperature-independent even in the ''insulating'' crystals studied here.[9] We suggest that the Coulomb interaction is negligible in the localized regime of $Bi_{2+x}Sr_{2-x}CuO_y$, as well as the weak localized $Bi_2Sr_2CuO_y$ which we previously reported.[7] It is known that the Anderson localization gives no change in the Hall coefficient.[10] On the contrary, Hall coefficient can be influenced by the resistivity change, if other mechanism is dominant. The Coulomb interaction gives the correction for the Hall resistance R_{H}, which is proportional to the temperature dependence of resistance:[11] $\delta R_{\mathrm{H}}(T)/R_{\mathrm{H}}=2\,\delta R(T)/R$. We observed the temperature independent Hall coefficients in the samples B, C, and D (see Fig. 4). The result of Hall measurement shows that the resistivity rise at the low temperatures should be attributable to the Anderson localization.

We demonstrated that $Bi_{2+x}Sr_{2-x}CuO_y$ changes from superconductor to insulator by Bi/Sr substitution. Both of the disorder of the system and the carrier concentration were subject to the Bi/Sr substitution. Figure 5 shows the carrier concentration as a function of the metallic parameter $1/k_{\mathrm{F}}l$. By extrapolating the n vs $1/k_{\mathrm{F}}l$ plot, we show the Ioffe–Legel limit $k_{\mathrm{F}}l\sim 1$ lays around $n_0\sim 8\times 10^{20}$ cm^{-3}. The average spacing between carriers at n_0 is calculated as 10 Å, corresponding to $\sim$3 times as long as the spacing between Cu sites (3.8 Å). The system may lose metallic behavior at n_0 since the wavefunction cannot extend over the system. In this sense, our result implies that the carrier concentration of samples B, C, D, and E are larger than n_0 and still remain ''metallic,'' even the carriers are localized by the disorder.

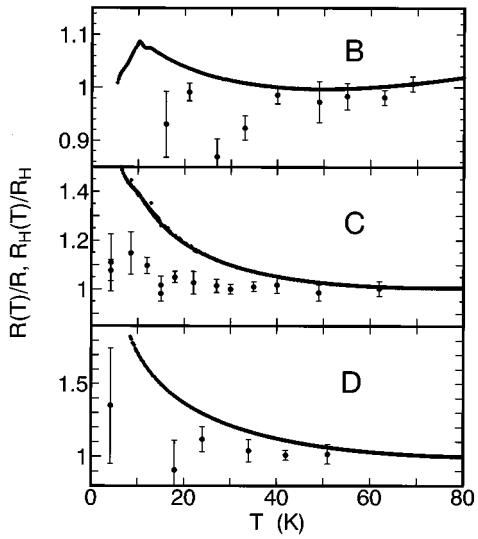

FIG. 4. Normalized resistivity $\delta R(T)/R$ (solid lines) and Hall coefficient $\delta R_{\mathrm{H}}(T)/R_{\mathrm{H}}$ (solid circles with error bars) of samples B, C, and D. If the Coulomb interaction plays a major role in resistivity upturn, the Hall coefficient should depend on the resistivity: $\delta R_{\mathrm{H}}(T)/R_{\mathrm{H}}=2\,\delta R(T)/R$. The data shown here demonstrate no such dependency.

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