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Out-of-plane transport properties of Bi₂Sr₂CaCu₂O₈ single crystals in normal and mixed states

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We have investigated c-axis transport in the normal and mixed states of Bi₂Sr₂CaCu₂O₈ single crystals. The temperature dependence of the normal-state resistivity along the c axis is significantly affected by thermal annealing, but all data are well described by the relation $\rho = \rho_0 T \exp(E/k_B T)$, suggesting a thermally activated hopping motion. In the mixed state, the behavior of $\rho_c(T,H)$ and $J_c(H)$ is observed to be closely related to the behavior of $\rho_{cN}(T)$. Some implications of these results are discussed.

One of the striking features of high-temperature superconductors is the large anisotropy, which is especially pronounced in the **Bi-based** materials. In $Bi_2Sr_2CaCu_2O_8$, for example, the normal-state resistivity¹ and the magnetoresistivity for $H \parallel c$ (Refs. 2 and 3) show dramatic anisotropies between the c axis and the abplane. In order to explain large anisotropy, several models assuming interlayer tunneling or weak coupling^{2,4-6} have been proposed for c-axis conduction, and the presence of Josephson coupling between layers has been experimentally seen in Bi₂Sr₂CaCu₂O₈ single crystals.⁷ However, it should be pointed out that most reported data^{2,3,8} consistent with the interlayer weak-coupling model are obtained with samples whose c-axis resistivity ρ_{cN} in the normal state displays semiconductorlike behavior, but ρ_{cN} of some samples⁹ are reported to show metallic behavior.

In this paper, we describe an experiment to study the c-axis transport properties in normal and mixed states of Bi₂Sr₂CaCu₂O₈ single crystals possessing different temperature dependences of ρ_{cN} . It is found that $\rho_{cN}(T)$ in $Bi_2Sr_2CaCu_2O_8$ single crystals changes with thermal annealing from semiconducting behavior to more metallic behavior and that this $\rho_{cN}(T)$ may be accounted for by a thermally activated hopping motion. In the mixed state, different behaviors of $\rho_c(T,H)$ are observed for samples having different temperature dependence of ρ_{cN} . For the sample where ρ_{cN} increases near T_c with decreasing temperature, $\rho_c(T,H)$ shows the depression of onset transition temperature T_c and the enhancement of ρ_c , which have been observed previously in Bi₂Sr₂CaCu₂O₈ single crystals^{2,3} and qualitatively explained by interlayer weak-coupling models.^{2,4} On the other hand, for the sample exhibiting linear temperature dependence of ρ_{cN} , $\rho_c(T,H)$ is found to be explained by a flux-motion model¹⁰ rather than by the interplane weak-coupling model, in spite of the absence of a macroscopic Lorentz force. From these results it is inferred that the layers in Bi₂Sr₂CaCu₂O₈ may be intrinsically well coupled. However, the magnitude of interplane coupling may be easily reduced by structural defects or stresses so that the effects of interlayer weak coupling are observed in many

samples. In fact, the measured $\rho_c(T,H)$ of samples having different $\rho_{cN}(T)$ may be understood if the excess resistivity in the mixed states has the origin in the motion of nonrigid flux lines, but as the strength of interlayer coupling becomes weak by defects or stresses, additional dissipation caused by interlayer weak coupling is added to dissipation due to flux motion. We have also measured the critical current density J_c as a function of magnetic field applied perpendicular to the c axis for two samples which show different behaviors of $\rho_c(T,H)$. Different magnetic field dependences of J_c are found in two samples. In the sample where T_c depression and enhanced magnetoresistance are observed, a kind of oscillation is observed that indicates the presence of interlayer weak coupling; in the other sample, where T_c is not changed and only the transition width is broadened by the application of magnetic field, J_c exhibits the bulk property.

Single crystals of Bi₂Sr₂CaCu₂O₈ used in our measurements were grown by a traveling-solvent-floating-zone method described elsewhere.¹¹ The crystals were cut into bars with typical dimension of $4 \times 2 \times 0.2$ mm³. Low-resistance contact (<1 Ω) was made to the sample using silver paint. The resistivity and critical current-density measurements were carried out using a standard four-probe technique. The independence of exact contact configuration was checked by exchanging the role of current and voltage probes and confirming that the data were unaffected. The current density used in the resistivity measurements was 0.0125 A/cm². The magnetic field was applied parallel to the *c* axis in the critical current measurement.

Figure 1(a) is a plot of ρ_c vs T in zero field for a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystal (sample I) annealed at several temperatures for 30 min in air and cooled to room temperature in a furnace. The zero-resistivity temperature T_{c0} is not changed by thermal annealing. However, the magnitude of ρ_{cN} is reduced and the temperature dependence of ρ_{cN} is affected by thermal annealing. ρ_{cN} of the as-grown sample increases monotonically with decreasing temperature for T < 300 K, while ρ_{cN} of the annealed sample exhibits a minimum followed by a resistivity upturn, and its

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minimum temperature T_0 decreases as the annealing temperature T_a increases. Similar resistivity upturn has been also observed in other high-temperature superconductors.¹² We have tried to fit these data to several models assuming the tunneling between planes.^{5,6} However, neither of these models fits the curves in Fig. 1(a).

Figure 1(b) shows the same data replotted in the form $\ln(\rho_c/T)$ vs 1/T. For the most part, $\rho_{cN}(T)$ is well represented by the functional form

$$\rho = \rho_0 T \exp(E/k_B T) . \tag{1}$$

Parameters ρ_0 and *E* obtained by fitting the data are presented in Table I. Interestingly, the value of ρ_0 converges to 0.0026-0.0030 Ω cm/K independently of T_a , while the value of *E*, which is consistent with $k_B T_0$, decreases with increasing T_a as shown in the inset of Fig. 1(b).

Figure 2 shows the resistive transition curves in zero field for three $Bi_2Sr_2CaCu_2O_8$ samples annealed at 700 °C for 30 min in air and cooled to room temperature in a furnace. The temperature dependence of ρ_{cN} seems to be very dependent on the sample. However, $\rho_{cN}(T)$ of three different samples can be well fitted by Eq. (1) as in Fig. 1(b). Even ρ_{cN} of sample III, which is linearly dependent on T, can be described by Eq. (1), since for $T > T_c$, Eq. (1) is approximated to be



FIG. 1. (a) Temperature dependence of the c-axis resistivity for a Bi₂CaCu₂O₈ single crystal (sample I): (a) as grown, (b) annealed at 200 °C, (c) at 400 °C, (d) at 500 °C, (e) at 600 °C, and (f) at 700 °C for 30 min in air. (b) Data of Fig. 1(a) replotted as ρ_c/T vs 1/T. The inset shows annealing temperature dependence of E obtained by fitting the data to Eq. (1).

TABLE I. Parameters obtained by fitting the data for $Bi_2Sr_2CaCu_2O_8$ single crystals to an equation of the form $\rho = \rho_0 T \exp(E/k_B T)$.

	Annealing temperature (°C)	$ \rho_0 $ $(\Omega \ cm/K)$	E (meV)
	As-grown	0.0030	27.4
	200	0.0027	23.0
Sample I	400	0.0029	18.8
	500	0.0026	17.2
	600	0.0028	16.0
	700	0.0027	14.6
Sample II	As-grown	0.0025	25.1
	700	0.0024	8.53
Sample III	As-grown	0.0024	22.6
	700	0.0022	6.32

$$\rho \simeq \rho_0 T + \rho_0 E / k_B \tag{2}$$

if E/k_B is smaller than T_c as presented in Table I. Before annealing, $\rho_{cN}(T)$ of all samples showed the semiconducting behavior.

Equation (1) reminds one of the transport via a thermally activated hopping motion of spatially localized charge carriers observed in several metallic oxides.¹³ If the carriers are localized due to the absence of any long-range order, or lattice distortion resulted from a strong electron-phonon interaction, the carriers can move to a neighboring site via hopping when the thermal fluctuation reduces the depth of the potential well and creates an empty well at a neighboring site. In this case, if the number of carriers is constant or slowly varying with respect to T, the conductivity σ is given by

$$\sigma \propto T^{-1} \exp(-\varepsilon/k_B T) \tag{3}$$

where ε is the enthalpy for hopping. In Bi₂Sr₂CaCu₂O₈, it is considered that the carriers may be localized at the Cu-O planes. Then, the *c*-axis conduction may be interpreted in terms of an activated hopping motion. The reason that the value of *E* is dependent on T_a or the sample is not clear. However, since Bi₂Sr₂CaCu₂O₈ single crystals are easily cleaved along the *ab* plane and strong



FIG. 2. Temperature dependence of the *c*-axis resistivity for three $Bi_2Sr_2CaCu_2O_8$ single crystals (samples I, II, and III) annealed at 700 °C for 30 min in air.

stresses may be applied to the sample during cutting, it is guessed that a kind of distortion between layers may be released by thermal annealing, and this leads to a reduction of the value of $E(=k_BT_0)$. Similarly, sample dependence of E may be explained by differences in the defect density in the crystals.

Figure 3(a) is a plot of ρ_c vs T in magnetic fields of 0,1,3,5,7 T oriented in the *c* direction for sample I whose ρ_{cN} displays the peak near T_c . T_c appears to decrease and the peak of ρ_c near T_c increases as the magnetic field increases. These behaviors are very similar to what was observed previously in Bi₂Sr₂CaCu₂O₈ crystals,^{2,3} which



FIG. 3. Temperature dependence of the c-axis resistivity in magnetic fields of 0, 1, 3, 5, and 7 T oriented parallel to the c axis for three Bi₂Sr₂CaCu₂O₈ single crystals: (a) sample I, (b) sample II, and (c) sample III. The dashed curves are theoretical fits described in the text. The insets of Figs. 3(a) and 3(b) show Arrhenius plots of $\Delta \rho$ obtained by subtracting the dashed curves from the experimental data.

have been explained by interlayer weak-coupling models.^{2,4} The dashed curves in Fig. 3(a) represent $\rho_c(T,H)$ calculated from the model proposed in Ref. 2, where $\rho_c(T,H)$ is interpreted by thermally activated phase slip across a weak link described by

$$\rho_{c}(T,H) = \rho_{cN} [I_{0}(h/4\pi ek_{B}T)J_{c}(0) \\ \times (1-T/T_{c})^{3/2} \Phi_{0}/H]^{-2}$$
(4)

where $\rho_{cN}[=\rho_0 T \exp(E/k_B T)]$ is the normal-state resistivity, I_0 the modified Bessel function, $J_c = J_c(0)(1 - T/T_c)^{3/2}$ the intrinsic Ginzburg-Landau depairing critical current density, and Φ_0 the flux quantum. In calculations, $T_c = 85.3$ K, $\rho_0 = 0.0027 \ \Omega \text{ cm/K}$, E = 14.6 meV, and $J_c(0) = 5.4 \times 10^6$ A/cm² were assumed, where the values of T_c , ρ_0 , and E were obtained from the experimental data and the value of $J_c(0) = 5.4 \times 10^6$ A/cm², which is consistent to that reported in Ref. 2, was obtained by fitting the data to Eq. (4). The experimental data is explained qualitatively by Eq. (4), but the data show more dissipation than that predicted by the theory.

Figure 3(b) is a plot of ρ_c vs T in various magnetic fields for sample II where $\rho_{cN}(T)$ exhibits a resistivity minimum at T = 99 K. Similarly to sample I, T_c is shifted to the lower temperature and ρ_c near T_c is slightly enhanced by the magnetic field. The $\rho_c(T,H)$ calculated from Eq. (4) with $T_c = 84$ K, $\rho_0 = 0.0024 \ \Omega \text{ cm/K}$, E = 8.53 meV, and $J_c(0) = 5.4 \times 10^6 \text{ A/cm}^2$ is represented by the dashed curves. The T_c depression is well accounted for by Eq. (4), but, similarly to sample I, the measured magnetoresistance is higher than the calculated one from Eq. (4) at low temperatures. Surprisingly, however, the data in Figs. 3(a) and 3(b) and the data reported in Ref. 2 are all best fit with the same value of $J_c(0) = 5.4 \times 10^6 \text{ A/cm}^2$ despite the different behaviors of $\rho_{cN}(T)$. It may be a kind of indication that phase slippage is preferrable to interlayer Josephson tunneling⁴ as the origin of the c-axis excess dissipation in the mixed state.

Figure 3(c) is a plot of ρ_c vs T in several magnetic fields for sample III where $\rho_{cN}(T)$ is linear. Contrary to samples I and II, T_c is not changed by the magnetic field and the magnetoresistance is not enhanced near T_c . The ρ_c calculated from Eq. (4) with $T_c = 96$ K, $J_c(0) = 5.4 \times 10^6$ A/cm², $\rho_0 = 0.0022$ Ω cm/K, and E = 6.32 meV is represented by the dashed curves. A large discrepancy is observed between the experimental data and the prediction of Eq. (4). Equation (4) yields $\rho_c(T)$ with a negative slope. However, the measured ρ_c exhibits a positive slope with respect to T for $T < T_c$ and H > 0. It implies that the broadening of the resistive transition shown in Fig. 3(c) may be caused by vortex motion rather than interlayer weak coupling. In our experiment, H is parallel to J so that a macroscopic Lorentz force is zero and the Lorentz-force-driven flux motion is not expected. However, if the flux lines are neither rigid nor straight¹⁴ and there is some sort of misalignment between H and J, dissipation due to flux motion may be produced even in the absence of a macroscopic Lorentz force.

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FIG. 4. Magnetic field dependence of the critical current density normalized by the critical current density in zero field at T = 78 K for Bi₂Sr₂CaCu₂O₈ single crystals (samples I and III). In this measurement, the magnetic field is applied perpendicular to the c axis and the current parallel to the c axis. The solid curves are guides for the eye.

The result of sample III shown in Fig. 3(c) suggests that dissipation caused by vortex motion also may be present in samples I and II in addition to the dissipation, due to interlayer weak coupling, and its magnitude may not be negligible. Indeed, more dissipation than that predicted from Eq. (4) is observed in Figs. 3(a) and 3(b). The insets of Figs. 3(a) and 3(b) show the difference $\Delta \rho$ between the experimental data and the calculated curves from Eq. (4) as a function of 1/T for samples I and II, respectively. These Arrhenius plots support the result that $\Delta \rho$ at low temperatures is from thermally activated flux creep. The values of U_0 (63.4–22.8 meV) estimated from $\Delta \rho$ of samples I and II in the insets of Figs. 3(a) and 3(b) are comparable to U_0 (58.8–18.1 meV) obtained from ρ_c of sample III in Fig. 3(c), but slightly higher than U_0 (27.8-8.62 meV) obtained from the data of ρ_{ab} which were measured simultaneously with ρ_c .

Figure 4 shows the normalized $J_c(H)$ by $J_c(0)$ at T = 78 K as a function of magnetic field applied perpendicular to the c axis for samples I and III. In sample I, J_c is reduced by about half in H = 0.02 T and a kind of oscillation is observed, suggesting the presence of weak coupling. However, $J_c(H)$ of sample III remains almost constant in low magnetic fields. It verifies the above arguments that the layers of sample I are connected via weak coupling, while the layers of sample III are well coupled so that the effects of weak coupling are not observed.

In summary, we have measured the c-axis resistivity ρ_c of Bi₂Sr₂CaCu₂O₈ single crystals in normal and mixed states. In the normal state, although $\rho_{cN}(T)$ shows semiconductorlike or metallic behavior depending on the sample preparation, $\rho_{cN}(T)$ is well described by Eq. (1) for all samples. From the analogy of Eq. (1) to the transport via a thermally activated hopping process, it is inferred that the c-axis transport in $Bi_2Sr_2CaCu_2O_8$ may occur by hopping motion of charge carriers localized at the Cu-O planes. In the mixed state, depending on the value of Eobtained by fitting $\rho_{cN}(T)$ to Eq. (1), different behaviors of $\rho_c(T,H)$ are observed. For $E < k_B T_c$ (sample III), $\rho_c(T,H)$ exhibits similar resistive transition broadening to that of the well-known in-plane resistivity ρ_{ab} usually interpreted in terms of dissipative flux motion. For $E > k_B T_c$ (samples I and II), however, $\rho_c(T, H)$ displays the T_c depression and the enhanced magnetoresistance near T_c , followed by a broadening of the resistive transition. The data of samples I and II are found to be well explained by the sum of dissipations caused by nonrigid vortex motion and thermally activated phase slip across the layers consisting of weak links. These results imply that the layers in Bi₂Sr₂CaCu₂O₈ may be well coupled intrinsically as seen in sample III. However, since the strength of interlayer coupling is easily weakened by structural defects or stresses, the effects due to interplane weak coupling are observed in many samples. In addition, we have also measured J_c at 78 K as a function of magnetic field oriented perpendicular to the c axis for samples I and III. As expected from the magnetoresistivity measurements, J_c of sample I shows a kind of oscillation, while J_c of sample III shows no indication of interlayer weak coupling.

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