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Optical evidence for strong anisotropy in the normal and superconducting states in $Bi_2Sr_2CaCu_2O_{8+z}$

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The optical reflectivity spectra for the *bc* surface of a large $Bi_2Sr_2CaCu_2O_{8+z}$ crystal were investigated over a wide energy range (30–30 000 cm^{-1}) at room temperature and 6 K. In the spectrum for $E\|c$, only the phonon peaks were observed without any electronic contribution down to 30 cm^{-1}, whereas for $E\|b$ a Drude-like spectrum was observed. Even below T_c the spectrum for $E\|c$ does not show any remarkable change except for a small change in the phonon spectrum. This indicates an extremely small plasma frequency for $E\|c$, less than 30 cm^{-1}, and thus quite a large mass anisotropy $m_c^*/m_b^* > 10^4$. Such a small plasma frequency supports the conduction model along the *c* axis, that the supercurrent flows by tunneling through insulating (semiconductor) layers such as the Bi-O layers.

Strong anisotropy in the crystal structure as well as in the electronic states is one of the characteristic properties of the high-T_c superconducting cuprates (HTSC), which may be essential for the mechanism of high-T_c superconductivity. Although the physical properties in the *ab* plane have been intensively investigated from many points of view, there are less reports on the properties in the *c* direction.[1] This is not only due to the fact that the main interest is concentrated on the electronic states within the CuO_2 plane but also due to the limitation of available sample size along the *c* axis. Especially for the measurement of far-infrared spectra, we need sample sizes thicker than 2 mm. To understand the electronic states in the HTSC, the conduction mechanism along the *c* axis should be an important subject of study, being related to the problem of the conduction dimensionality.

Recently large single crystals of $La_{2-x}Sr_xCuO_4$ (LSCO) have been successfully grown by a traveling-solvent-floating-zone (TSFZ) method, which have made it possible to measure the optical spectra for $E\|c$ over wide energy and temperature ranges.[2] The most interesting feature is the abrupt appearance of a sharp edge below T_c in the reflectivity spectrum. Tamasaku, Nakamura, and Uchida attributed this feature not to the superconducting gap but to the plasma edge for $E\|c$.

In $YBa_2Cu_3O_7$ (YBCO), the spectrum for $E\|c$ shows a clear electronic contribution below 0.5 eV even at room temperature,[3] which is consistent with the metallic dc conductivity along the *c* axis.[4] When the temperature decreases below T_c, only the electronic part of the spectrum seems to change dramatically, while the superposed phonon structures change in strength and damping.[5,6] The almost unity reflectivity ($R \sim 1.0$) is observed below 100 cm^{-1}. Therefore, in contrast to the in-plane spectrum, the *c* axis spectrum seems to strongly depend on material, presumably due to the structural difference among the materials.

This fact motivated us to investigate another typical HTSC, $Bi_2Sr_2CaCu_2O_{8+z}$ (BSCCO), which is quite different in the crystal structure from YBCO but exhibits the same T_c value as YBCO. There are a few reports on the room-temperature spectra of BSCCO for $E\|c$.[7] In spite of the use of several pieces of BSCCO crystals buried in epoxy, the range of wave number was limited down to 300 cm^{-1}. In the present work, we have measured the optical reflectivity spectrum for the *bc* surface of BSCCO over a wide wave-number range (30–30 000 cm^{-1}) at 300 and 6 K. Within this covering range, no electronic contribution to the spectrum for $E\|c$ was observed both in the normal and superconducting states, while the in-plane spectrum was dominated by a Drude-like spectrum. It indicates the strong electronic anisotropy in this material and suggests a Josephson current along the *c* axis, flowing through insulating layers such as Bi-O layers in the superconducting state.

A large single crystal with a size of $5\times5\times1.4$ mm^3 was cut out of the BSCCO crystal rod grown by the TSFZ method. The details of the crystal growth are described in Ref. 8. The composition ratio of Bi:Sr:Ca:Cu, analyzed by an electron-probe microanalyzer, was 2.05:1.79:0.93:2.0, which is a little nonstoichiometry. The as-grown crystal exhibits a sharp superconducting transition at 90 K with $\Delta T_c = 2$ K, as shown in the susceptibility data in Fig. 1. The small difference between the field cooling and the zero-field cooling data guarantees that there are only a few pinning centers such as dislocations and other disorders. The temperature dependence of the in-plane resistivity is shown in the inset of Fig. 1. This indicates a mixing of a small amount of 2:2:2:3 phase ($Bi_2Sr_2Ca_2Cu_3O_z$), which presumably exists in the crystal as a stacking fault. However, such a small amount of the secondary phase has little effect on the optical spectrum, unless the secondary phase covers the sample surface.

The reflectivity spectra were measured for the mirror-polished surface with polarized light, using a Fourier transformation-type spectrometer in the far-infrared and infrared region (30–9000 cm^{-1}) and a grating-type spec-

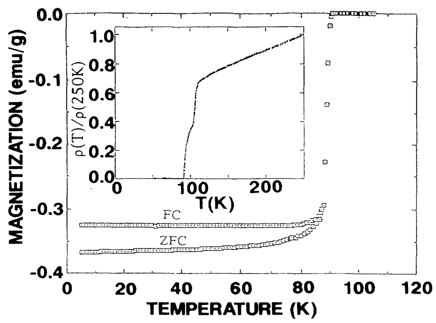

FIG. 1. Temperature dependence of the magnetization for the $Bi_2Sr_2CaCu_2O_{8+z}$ single crystal under the magnetic field of 10 Oe. The inset is the temperature dependence of the in-plane resistivity normalized to the data at 250 K.

trometer in the infrared and visible region (4000–30 000 cm^{-1}). The absolute reflectivity value was calibrated to the data measured by a microscope system in the range from 700 to 4000 cm^{-1}. A gold- (or Al-) evaporated mirror was used as a reference in the far-infrared (or the visible) region.

Figure 2 shows the room-temperature reflectivity spectra for $E\|b$ and $E\|c$. For $E\|b$ a Drude-like spectrum is observed with the reflectivity minimum around 10 000 cm^{-1}, as reported by many other groups.[9,10] A small but clear deviation from a simple Drude profile is also established now, although its origin still remains an unresolved problem. On the other hand, in the spectrum for $E\|c$, only phonon peaks are observed, without any component associated with free carriers down to 30 cm^{-1}. Although seven phonon modes are expected for the tetragonal $I4/mmm$ symmetry, much more phonon peaks are observed in Fig. 2. This is due to the lower symmetry ($A2aa$) in the actual material, in which there is a modulated structure along the b axis.

The effective mass ratio m_c^*/m_b^*, estimated from the plasma frequency $\omega_p \sim (n/m^*)^{1/2}$, is larger than 10^4, using an in-plane plasma frequency $(\omega_p)_b \sim 10\,000$ cm^{-1} (Ref. 10) and a far-infrared dielectric constant for $E\|c$ $\varepsilon_{FIR}=12$. This ratio is extraordinarily larger than in the case of LSCO ($m_c^*/m_{ab}^* \sim 300$) (Ref. 11) and YBCO ($m_c^*/m_a^* \sim 6$).[12] (In all cases, we adopt the in-plane ω_p values estimated in the two component model, which gives a smaller ω_p than the one component model, for the observed in-plane spectra.[13]) Such a large mass anisotropy may imply that the effective mass approximation breaks down in the c direction and the plasmon cannot be well defined for $E\|c$ in the usual sense.

When the temperature decreases below T_c, no dramatic change was observed for $E\|c$ even at 6 K, as shown in Fig. 3. Figures 4(a) and 4(b) show the imaginary part of the dielectric function $\varepsilon(\omega)$ and the loss function $\mathrm{Im}[-1/\varepsilon(\omega)]$, which were calculated from the reflectivity spectra in Fig. 3 by a Kramers-Kronig analysis. Also in these figures we cannot see any clear electronic component but only the change in the phonon spectrum, such as the narrowing of the peak widths as well as the splitting of some peaks. It is quite distinct from the results for LSCO (Ref. 2) and/or YBCO,[5,6] which show a dramatic change at the superconducting transition, attributed to shift of the spectral weight of the conductivity into a δ function at zero frequency below T_c.

There might be two possible interpretations for this lack of spectral change. One is that the superconducting gap energy is smaller than 30 cm^{-1} (4 meV), and the other is that the screened plasma energy is smaller than 4 meV. The former seems to be unlikely, because a superconducting gap is expected to be larger than 300 cm^{-1} from the spectrum for $E\perp c$.[9,10] The latter is rather plausible. In LSCO, the plasma edge for $E\|c$ was observed just within the measurable range, i.e., around 40 cm^{-1}. Since the plasma frequency $\omega_p=(4\pi ne^2/m^*)^{1/2}$ is a function of the effective mass m^*, it is speculated that in BSCCO the plasma edge is located at a lower energy than 30 cm^{-1}, if the difference in resistivity ρ_c between these two materials (two orders higher in BSCCO than in LSCO) originates from the difference in m^*.

As pointed out for LSCO by Tamasaku, Nakamura, and Uchida,[2] such a small plasma frequency in the c direction leads us to a picture of the microscopic Josephson array consisting of alternate stacking of insulating layers (the Bi-O planes) and superconducting layers (the CuO_2 planes). In this case, the Josephson plasma frequency is described as $\omega_J^2=4\pi(2e)^2\phi^2/\varepsilon m^*$, ϕ^2 being the number density.[14] Such a conjecture was also made from the direct measurement of the ac and dc Josephson

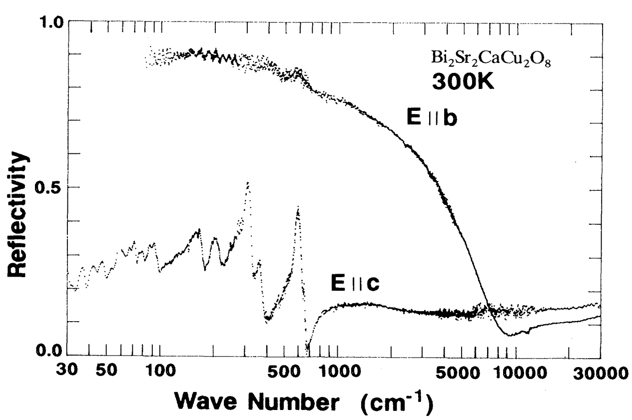

FIG. 2. The room-temperature reflectivity spectra for the $Bi_2Sr_2CaCu_2O_{8+z}$ single crystal with $E\|b$ and $E\|c$.

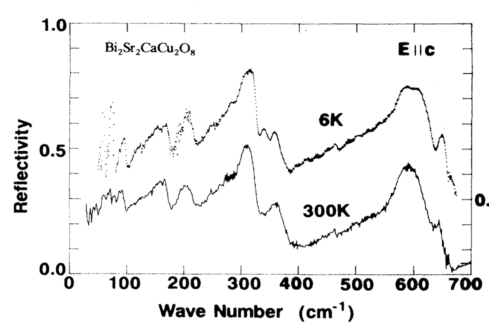

FIG. 3. The reflectivity spectra of $Bi_2Sr_2CaCu_2O_{8+z}$ in the far-infrared region with $E\|c$ at room temperature and 6 K.

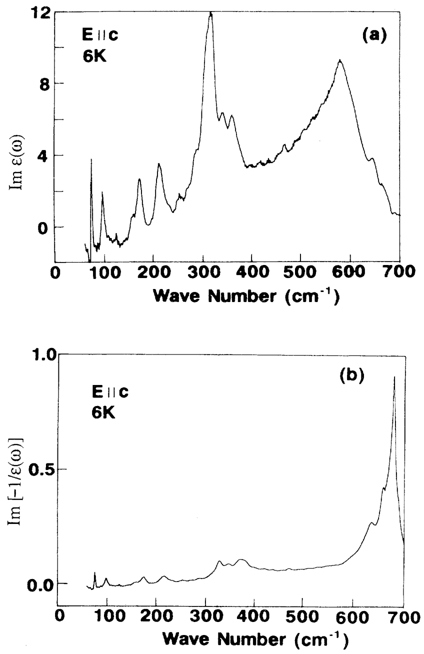

FIG. 4. (a) The imaginary part of the dielectric function $\mathrm{Im}\varepsilon(\omega)$ and (b) the loss function $\mathrm{Im}[-1/\varepsilon(\omega)]$ calculated from the reflectivity spectra in Fig. 3 by the Kramers-Kronig analysis.

effects.[15] If we adopt this superlattice (alternate stacking) picture, it turns out that the normal-state current along the c axis above T_c also flows by tunneling or the temperature-dependent resistivity in the semiconducting layer dominates the total resistivity along the c axis. The results of a direct measurement of dc resistivity by a Montgomery method indicated a semiconducting temperature dependence of $\rho_c(T)$ for BSCCO,[16] which is similar to the case of $Bi_2Sr_2CuO_z$.[4] It would indicate that ρ_c is dominated either by the tunneling matrix which is not constant but which decreases dramatically with reducing temperature, or more simply by the temperature-dependent resistivity in the semiconducting layers.

Another important suggestion given by the superlattice picture is that the infrared phonons associated with the Bi-O layers may not be perfectly screened by the plasmon which is confined to the vicinity of the CuO_2 planes. It means that the Bi-O layers do contribute to the optical spectra almost independently, which has been neglected by most groups in interpreting their observed in-plane spectra. Some of the structure observed in the far-infrared spectra for $E\perp c$ may possibly originate from the excitation within the so-called charge reservoir layers, such as the Bi-O layers, the Tl-O layers, the Nd-O layers, etc.[1]

The remaining and most essential problem is why the electronic state in HTSC is so strongly anisotropic, that is, why the carriers are confined to such a narrow region, the CuO_2 planes. This must be an important factor for understanding HTSC, but it is beyond the scope of this paper.

In summary, we have measured the optical reflectivity spectra of the BSCCO single crystal both for $E\|c$ and $E\|b$ over a wide temperature range. In the spectrum for $E\|c$, only the phonon peaks are observed both at room temperature and at 6 K. This means that the plasma frequency for $E\|c$ is so small that we cannot observe the reflectivity edge. For $E\|b$ the spectrum is dominated by a Drude-like electronic contribution. The estimated mass ratio m_c^*/m_b^* is larger than 10^4. It may be too large to define a plasmon for $E\|c$ in the usual sense. It strongly suggests that the supercurrent along the c axis is a kind of Josephson current which flows by tunneling through the insulating (semiconducting) layers such as the Bi-O layers. This metal-insulator (semiconductor) alternate stacking model also suggests the possibility that some of the structures observed in the far-infrared spectra for $E\perp c$ originate from the excitations within layers other than the CuO_2 planes.

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[12]In calculation, we used the parameters $(\omega_p)_c = 4150$ cm^{-1} [S. L. Cooper *et al.*, Phys. Rev. B **47**, 8233 (1993)] and $(\omega_p)_a = 10\,000$ cm^{-1} [J. Orenstein *et al.*, Phys. Rev. B **42**, 6342 (1990)].

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