Anisotropy of the optical characteristics of the superconducting $YBa_2Cu_3O_{7-\delta}$ crystals and of the Bi–Sr–Ca–Cu–O system

M. P. Petrov, A. I. Grachev, M. V. Krasin'kova, A. A. Nechitaĭlov,
V. V. Poborchiĭ, S. I. Shagin, and S. V. Miridonov
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR

(Submitted 26 May 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **50**, No. 1, 25–29 (10 July 1989)

The data on the anisotropy of the dispersion of optical characteristics of superconducting $YBa_2Cu_3O_{7-\delta}$ and Bi–Sr–Ca–Cu–O crystals in the **ab** plane have been obtained for the first time on the basis of measurements of the polarized reflection spectra in the range 0.07–3.1 (3.6) eV.

The present stage of the ongoing research of high-T_c superconductors puts considerable emphasis on the study of the properties of single-crystal samples, primarily because of the strong anisotropy of the structure of the known high-T_c superconducting materials. Unfortunately, the small size of the synthesized crystals and phenomena such as microtwinning of the structure of $YBa_2Cu_3O_{7-\delta}$ crystals greatly complicate the experimental study of the anisotropy of the physical properties of high-T_c superconductors. In our previous studies[1-3] and also in Refs. 4 and 5 optical methods have been shown to be useful in solving the indicated problem. By expanding considerably the spectral range of measurements of the polarized reflection spectra [and subsequently analyzing them on the basis of the Kramers-Kronig relations and by assigning a model-based function of the dielectric constant $\epsilon(\omega)$] we were able to determine for the first time the anisotropy of dispersion of the optical characteristics of $YBa_2Cu_3O_{7-\delta}$ and Bi–Sr–Ca–Cu–O superconducting crystals in the **ab** plane.

The growth technology and the structural and morphological properties of the test samples were described in Refs. 1–3. We will therefore describe them here only briefly. The $YBa_2Cu_3O_{7-\delta}$ crystals ($\delta \gtrsim 0.1$; $T_c \approx 90$ K) are well-formed wafers with length scales of $200\times200\times5$ μm along the **a**, **b**, and **c** axes, respectively. It is important to point out that there are 100×100-μm regions, essentially free of twinning, in the **ab** plane. The crystals of the Bi–Sr–Ca–Cu–O system ($T_c \sim 82$ K) are thin (~ 5 μm) lamella which are oriented at right angles to the **c** axis. The polarized reflection spectra were recorded in the **ab** plane and they were linked to the crystallographic axes through systematic optical measurements and electron-microscope analysis of the same region of the sample. The measurements in the visible region and the near-IR region were carried out with a custom-built microspectrophotometer. The surface area of the probe was ~ 50 μm^2 in the measurements. The standard model-113V Bruker Fourier spectrometer was used for the measurements in the middle IR region (0.5–0.7 eV).

The reflection spectra of the $YBa_2Cu_3O_{7-\delta}$ crystals are shown in Fig. 1. We

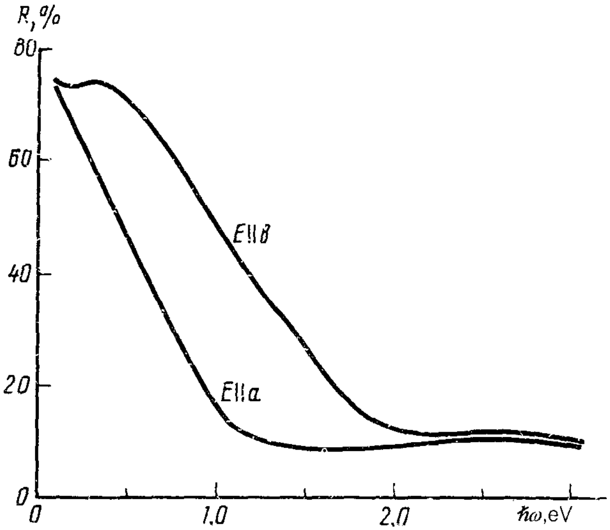

FIG. 1. Reflection spectra of a superconducting $YBa_2Cu_3O_{7-\delta}$ crystal in a plane-polarized light of a single-domain region (**E** is the electric field vector of the light wave).

clearly see the reflection anisotropy, which manifests itself most conspicuously in the IR region, with $R_b > R_a$. A similar result was obtained in Ref. 5 for the $EuBa_2Cu_3O_{7-\delta}$ crystals. In our case, however, the $R(\omega)$ curve reaches a local maximum at $\hbar\omega \sim 0.3$ eV only in the case **E**‖**B**, while at **E**‖**a** the $R_a(\omega)$ curve rises monotonically.

Figure 2 shows the dispersion curves of the real part of the complex dielectric constant $\epsilon_1(\omega)$ and the optical conductivity $\sigma(\omega)$, which were obtained by analyzing the reflection spectra using the Kramers-Kronig relations. The low-frequency and high-frequency extrapolations of the functional dependence $R(\omega)$, which must be carried out for the analysis based on the Kramers-Kronig relations, were chosen by a standard method: by means of the Hagen-Rubens relation in the limit $\omega \to 0$ and by prescribing the relation $R(\omega) = R_0 + \Delta R(\omega_0/\omega)^4$ in the limit $\omega \to \infty$, where ω_0 is the high-frequency limit of the spectral measurements, and $R_0 + \Delta R = R(\omega = \omega_0)$. Analysis of the effect of the parameters R_0 and ΔR on the behavior of the curves of $\epsilon_1(\omega)$, $\sigma(\omega)$, etc. showed that in the region $\hbar\omega < 2.0$ the basic features of the dispersion curves are essentially constant. This behavior also holds for the strong absorption peak which is centered at $\hbar\omega = 0.28$ eV and which is seen only on the $\sigma_b(\omega)$ curve, i.e., only in the case of **E**‖**b** polarization. The presence of an absorption peak in this region was reported in the first experimental studies of the reflection, using ceramic samples of $YBa_2Cu_3O_{7-\delta}$ (Ref. 6). But the exact position of this peak or its polarization dependence (in the **ab** plane) could not be determined in either the ceramic samples[6] or the twinning samples.[7] The polarization dependence of the 0.28-eV peak obtained by us shows unambiguously that it is related to the optical transitons in copper-oxygen

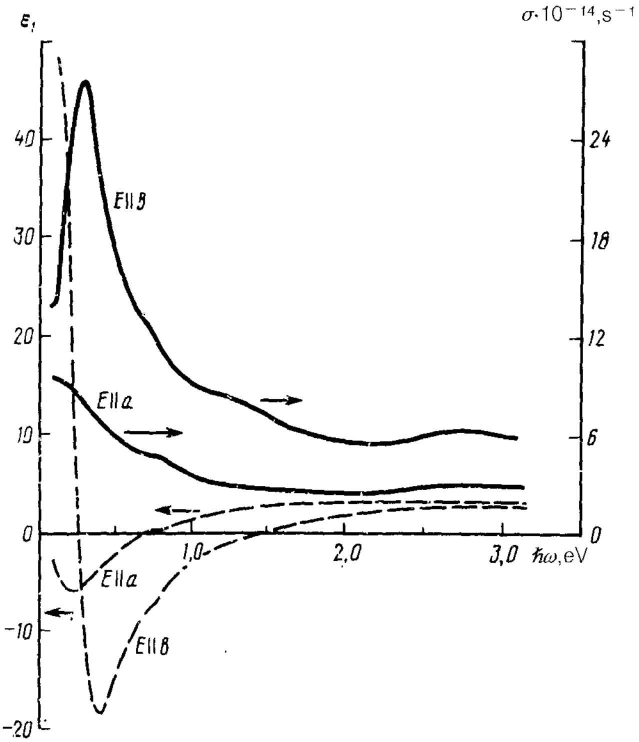

FIG. 2. Spectral dependence of the optical characteristics of $\epsilon_1(\omega)$ and $\sigma(\omega)$ of a superconducting $YBa_2Cu_3O_{7-\delta}$ crystal, obtained from a Kramers-Kronig analysis of the measured reflection spectra.

complexes, which are a part of the composition of the chains. Worth noting is the considerable oscillator strength of these transitions, amounting to ~0.5–0.6 per Cu(1) atom, suggesting that they are of a dipole nature.

It would be highly tempting to link the absorption peak at 0.28 eV with the energy band structure calculated in Refs. 8 and 9; specifically, with the optical transitions near point S between the two bands which lie near the Fermi level. As was shown in Ref. 9, the principal component of the state density of these bands is attributable to the copper-oxygen chains, and below the Fermi level there is a strong state-density peak which is linked with oxygen at the 0(1) and 0(4) sites.

As experimental proof of the conclusion that the 0.28-eV peak belongs to the chain transitions we can also add the data obtained for the Bi–Sr–Ca–Cu–O crystals (Fig. 3), in which there are only the copper-oxygen planes. As can be seen in Fig. 3, the reflection of each polarization resembles the behavior of the $R_a(\omega)$ curve in Fig. 1. Accordingly, the behavior of the $\epsilon_1(\omega)$ curve is similar to that of the $\sigma(\omega)$ curve (Fig. 3b). Note also the presence of anisotropy, though rather slight, of the optical characteristics of the crystals under study, because of the presence in them of a superlattice along the **b** axis.[3]

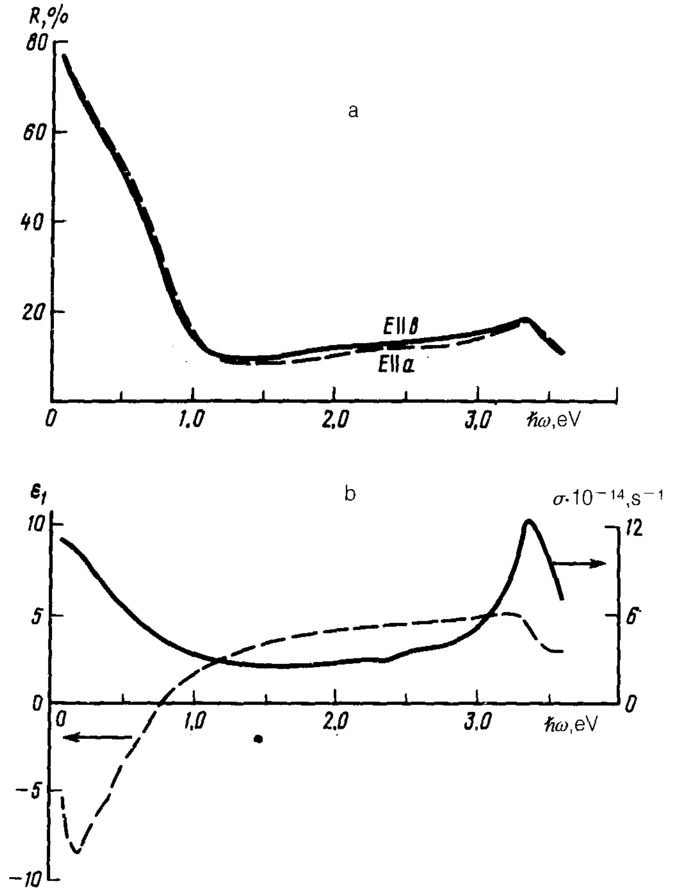

FIG. 3. Spectral dependence of (a) the reflection and (b) the optical characteristics of the superconducting crystals of a Bi–Sr–Ca–Cu–O system (because the $\epsilon_1(\omega)$ and $\sigma(\omega)$ curves behave similarly when E||**a** and E||**b**, we have plotted the curves for only one of the polarizations).

The use of the dependences which we obtained to estimate such parameters of the material as the plasma frequency is complicated by the presence in the region of the intraband transitions of free carriers in the auxiliary absorption bands (for $YBa_2Cu_3O_{7-\delta}$ with $\hbar\omega \sim 0.75$, 1.4, and 2.8 eV), whose nature is not yet clearly understood. In addition to the Kramers-Kronig analysis, we have accordingly made use of a method of prescribing the model-based function $\epsilon(\omega)$, in which its parameters are fitted by the method of least squares. Joint use of the two indicated independent methods of analyzing the results has enabled us to more accurately estimate the parameters of the material, on the one hand, and to put the extrapolations in a more suitable form for the Kramers-Kronig analysis, on the other. The values of the plasma frequency ω_p, of the relaxation frequency Γ of the carriers, and of the high-frequency dielectric constant ϵ_∞ which we found by a fitting procedure for the $YBa_2Cu_3O_{7-\delta}$

and Bi–Sr–Ca–Cu–O crystals are $(\omega_p)_a = 2.3$ eV, $\Gamma_a = 0.7$ eV, $\epsilon^a_\infty = 4.4$; $(\omega_p)_b = 3.1$ eV, $\Gamma_B = 0.95$ eV, $\epsilon^b_\infty = 4.8$; $(\omega_p)_a \approx (\omega_p)_b = 2.35$ eV, $\Gamma_a = \Gamma_b = 0.68$ eV, and $\epsilon_\infty = 4.5$, respectively.

Using the values of ω_p which we obtained, we can estimate the effective mass of the carriers, m^*. Taking into account the paucity of data on the concentration of the free carriers, however, we can legitimately estimate only the degree of the anisotropy of the effective mass in the **ab** plane, which for $YBa_2Cu_3O_{7-\delta}$ is $m^*_a/m_b = (\omega_p)^2_b/(\omega_p)^2_a \approx 2.0$.

We note in conclusion that the relationship established between the absorption band at 0.28 eV in $YBa_2Cu_3O_{7-\delta}$ and the optical transitions in copper-oxygen chains strongly suggests that certain corrections should probably be introduced into those theoretical models in which the dominating role in the carrier-pairing mechanism is attributed to the optical excitations with an energy on the order of a few tenths of an electron volt.

[1] M. P. Petrov *et al.*, Pis'ma Zh. Tekh. Fiz. **14**, 748 (1988) [Sov. Tech. Fiz. Lett. **14**, 333 (1988)].
[2] M. P. Petrov *et al.*, Sol. State Commun. **67**, 1197 (1988).
[3] M. P. Petrov *et al.*, Pis'ma Zh. Tekh. Fiz. **14**, 2097 (1988) [Sov. Tech. Fiz. **14**, No. 6 (1988)].
[4] I. Bozovic *et al.*, Phys. Rev. **B38**, 5077 (1988).
[5] J. Tanaka *et al.*, Physica **C153–155**, 1752 (1988).
[6] K. Kamaroš *et al.*, Phys. Rev. Lett. **59**, 919 (1987).
[7] Z. Schlesinger *et al.*, Physica **C153–155**, 1734 (1988).
[8] J. Yu *et al.*, *Novel Superconductivity*, Plenum Press, New York, 1987, p. 367.
[9] H. Krakauer *et al.*, *Journal of Superconductivity* **1**, 111 (1988).

Translated by S. J. Amoretty