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IR SPECTROSCOPY OF METAL–OXIDE SUPERCONDUCTORS IN NORMAL AND SUPERCONDUCTING STATES

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Normal state reflectivity of $YBa_2Cu_3O_{6.9}$ and $Bi_2Sr_2CaCu_2O_{8+y}$ single crystals has been studied by means of polarized light spectroscopy in mid- and far-infrared. The obtained data show that inplane conductivity of both compounds is metal-like and differs drastically from the out-of-plane one thus giving the direct evidence of electronic structure of these systems to be quasi two dimensional (2D). The analysis of low-temperature reflectivity of $YBa_2Cu_3O_{6.9}$ crystalline films and ceramic samples indicates the anisotropy of the energy gap with 2Δ value in (*ab*) plane and perpendicular to (*ab*) plane being equal $7.5kT_c$ and $3.5kT_c$ respectively. The obtained anisotropy of the energy gap and of normal state reflectivity proves the hypothesis of 2D superconductivity in $YBa_2Cu_3O_{6.9}$.

1. INTRODUCTION

THE DATA on the anisotropy of the electronic properties in normal and superconducting states of $Bi_2Sr_2CaCu_2O_8$ and $YBa_2Cu_3O_{6.9}$ compounds studied by means of IR spectroscopy are reported. On the basis of qualitative analysis of normal state reflectivity anisotropy and the anisotropy of the energy gap the proof of superconductivity in the systems under study to be quasi 2D one is obtained. The data on the dimensionality of superconductivity, as it was recently proposed, seems to be one of the key points to understand the physical origin of high T_c, observed in metal oxide materials [1].

2. MATERIALS AND METHODS

Anisotropy of normal state reflectivity was studied by means of polarized light IR spectroscopy on single crystalline samples. $YBa_2Cu_3O_{6.9}$ single crystals ($T_c \cong 90\,K$, $\Delta T_c \cong 1\,K$) with typical size $1 \times 2 \times (0.03\text{–}0.12)\,mm^3$ were grown from a non-stoichiometric melt with further annealing in oxygen [10]. $Bi_2Sr_2CaCu_2O_8$ single crystals with typical size $6 \times 2 \times 0.5\,mm^3$ ($T_c = 68\text{–}90\,K$) were grown using zone melting technique.

The mid IR reflectivity spectra of the single crystals were taken from the lateral surface of the sample using IR microscope or mirror condensors. The "edge technique" makes it possible to measure reflectivity both in $\mathbf{E} \parallel \mathbf{c}$ and $\mathbf{E} \perp \mathbf{c}$ light polarizations.

Both the available sizes of single crystals and the achieved level of experimental technique make so far unreliable the data obtained in far IR at low temperatures aimed to determine 2Δ values. So the other types of high T_c materials were used to study the anisotropy of the gap: single crystalline thin films with different crystallographic orientations and ceramic samples. Thin films with thickness of $d = 4000\,Å$, produced by *in situ* laser epitaxy on $SrTiO_3$ substrates, demonstrate sharp transition at $T_c = 90\,K$ with $\Delta T_c = 0.5\,K$.

The in-plane gap $2\Delta_{ab}$ was measured for *c*-axis crystalline films on (1 0 0) substrates. Non-polarized light reflectivity of these films with almost normal light incidence corresponds to $\mathbf{E} \perp \mathbf{c}$ polarization and can be well compared with that of the single crystal, measured in the same polarization (Fig. 1 curves A, B).

To determine the out-of-plane gap value $2\Delta_c$ we used the crystalline "1 2 3" films with the angle between normal to the substrate and *c*-axis being equal 10° and perfect ceramic samples (Fig. 1 curves C and D respectively). The non-polarized reflectivity spectra of both systems incorporate contribution of $\mathbf{E} \parallel \mathbf{c}$ polarization because of a certain angle between **c** and normal to substrate in case of thin film and because of random orientation of microcrystals in the ceramic samples.

3. NORMAL STATE REFLECTIVITY

Figure 2 shows the reflectivity spectra of $Bi_2Sr_2CaCu_2O_8$ and $YBa_2Cu_3O_{6.9}$ single crystals,

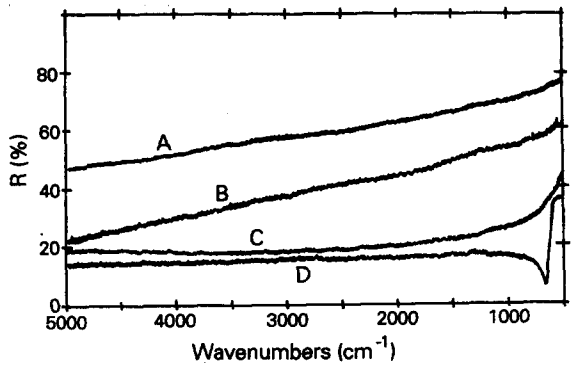

Fig. 1. Room-temperature polarized light reflectivity of $YBa_2Cu_3O_{6.9}$ single crystals at $T = 300$ K – curves A and C for $\mathbf{E} \perp \mathbf{c}$ and $\mathbf{E} \parallel \mathbf{c}$ polarizations respectively and for $Bi_2Sr_2CaCu_2O_8$ single crystal – curves B and D for $\mathbf{E} \perp \mathbf{c}$ and $\mathbf{E} \parallel \mathbf{c}$ polarizations respectively.

measured both in $\mathbf{E} \parallel \mathbf{c}$ and $\mathbf{E} \perp \mathbf{c}$ light polarizations. For E vector being parallel to copper–oxygen planes the obtained dependences demonstrates Drude-like conductivity (curves A and B for $YBa_2Cu_3O_{6.9}$ and $Bi_2Sr_2CaCu_2O_8$ crystals respectively). When E vector is perpendicular to these planes the reflectivity spectrum is typical for dielectrics with phonon peaks in Far-IR in case of $Bi_2Sr_2CaCu_2O_8$ compounds (curve D) or metal-like but with low reflectivity coefficient in case of $YBa_2Cu_3O_{6.9}$ compounds (curve C). These data show that carriers are much more mobile in Cu–O planes and can be almost localized within the planes as it takes place in $Bi_2Sr_2CaCu_2O_8$ crystals in which free carriers give no contribution to $\mathbf{E} \parallel \mathbf{c}$ spectrum. So reflectivity anisotropy together with data on the anisotropy of dc conductivity and critical currents [2, 3] prove quasi 2D electronic structure of the systems under study and give indications on possible low-dimensionality of the superconducting properties in the above mentioned compounds.

4. SUPERCONDUCTING ENERGY GAP

The analysis of the reproducibility of the IR data shows that the most reliable 2Δ value arise from the normalized R_s/R_n spectra where R_s and R_n are reflectivities of the sample in the superconducting and normal states respectively [4–6]. The reason is that normalized spectra are less affected by the quality of the surface and by other extrinsic properties. The improved version of the method based on the energy gap temperature dependence, determines 2Δ value from the normalized spectrum R_1/R_2 where R_1 and R_2 are reflectivities measured at two temperatures $T_1 \leqslant T_2 \ll T_c$ [7]. Figure 3 demonstrates the R_1/R_2 spectra, measured with the accuracy of 0.03% on the crystalline film with 10° angle between c-axis and normal to the substrate (curve A), on the ceramic sample (curves B and C) and on the c-axis film (curve D). The in-plane gap $2\Delta_{ab}$ was determined from the peak position of the structure featuring c-axis film and of the peak position of high-frequency pecularity observed in the spectra for other samples. The obtained value of $2\Delta_{ab} = 470\,\mathrm{cm}^{-1} = 7.5kT_c$ is in good agreement with the single crystals data [4].

The out-of-plane gap $2\Delta_c$ was determined from the peak position of low-frequency pecularities, observed in the samples with $\mathbf{E} \parallel \mathbf{c}$ contribution to the

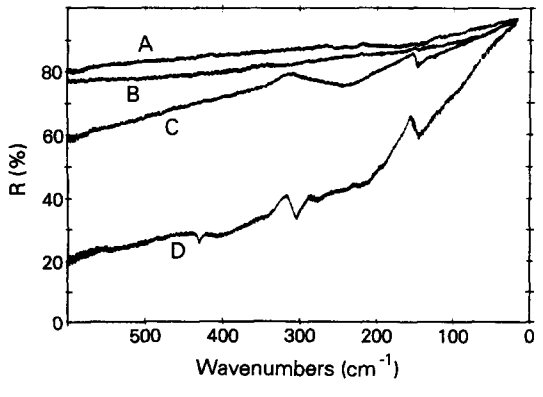

Fig. 2. Reflectivity spectra of $YBa_2Cu_3O_{6.9}$ at $T = 300$ K: the c-axis film – curve A; single crystal ($\mathbf{E} \perp \mathbf{c}$) – curve B; thin film with 10° angle between c-axis and the normal to the substrate surface – curve C; ceramic sample – curve D.

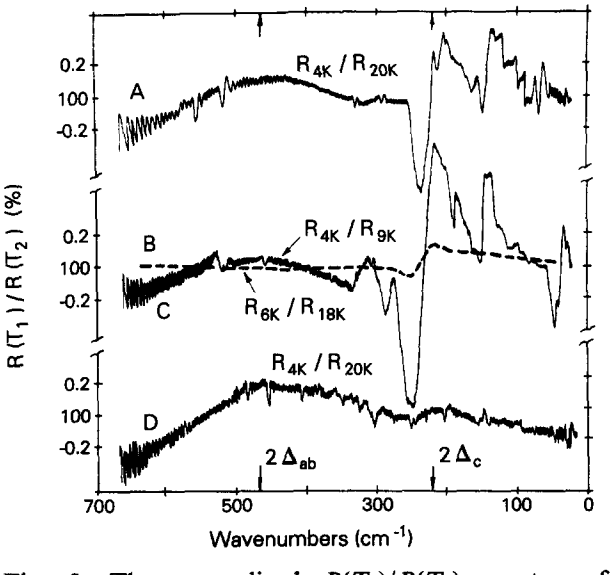

Fig. 3. The normalized $R(T_1)/R(T_2)$ spectra of $YBa_2Cu_3O_{6.9}$: thin film with 10° angle between c-axis and the normal to the substrate surface – curve A; ceramic sample – curves B and C; the c-axis film – curve D.

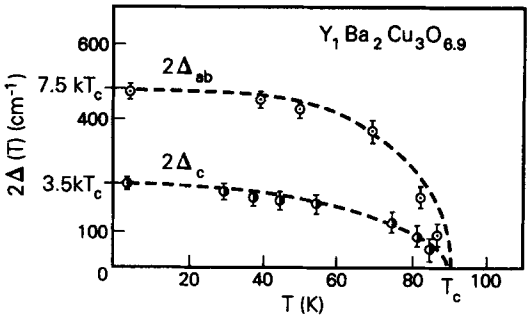

Fig. 4. Temperature dependence of the in-plane and out-of-plane gap pecularities $2\Delta_{ab}$ and $2\Delta_c$ respectively for $YBa_2Cu_3O_{6.9}$ compound, obtained from R_1/R_2 spectra.

non-polarized light spectra (Fig. 2 curve B). The obtained value of $2\Delta_c = 220\,cm^{-1} = 3.5kT_c$.

Both features demonstrates temperature dependence when R_1 and R_2 are measured at temperatures lying between 4 K and T_c (Fig. 4). The threshold behaviour of both pecularities at temperature close to T_c and the behaviour of peak position dependence on temperature prove that they can be attributed to 2Δ values for corresponding polarizations. So the energy gap in $YBa_2Cu_3O_{6.9}$ compounds is anisotropic with factor of anisotropy $f = \Delta_{ab}/\Delta_c = 2$ that is consistent with that of obtained in single crystals [4].

5. DISCUSSION

Determination of 2Δ value from normalized spectra R_s/R_n following Tinkham [8] or from R_1/R_2 spectra, as it was proposed recently [7], has a serious disadvantage besides of mentioned above benefits. The obtained value actually is the energy interval between maxima in $N(E)$, the density of states of the superconductor, that determines pair binding energy in the condensed state [9]. The absolute value of $N(E)$ at the Fermi level cannot be derived from normalized spectra. In other words, R_s/R_n data cannot separate the gap-less case from the case of the energy gap superconductivity. Still, we discuss the results using the term "energy gap" though we are not quite sure about absolute $N(E)$ at Fermi level.

There are several possible microscopic explanations of the energy gap anisotropy with one common disadvantage they hardly explain why the k-space anisotropic gap leads to 2Δ anisotropy observed in IR experiments. The assumption of 2D superconductivity in metal oxide systems resulted from their normal state 2D electronic structure give the direct explanation of 2Δ IR anisotropy..

Within the framework of 2D superconductivity there is only one energy gap, associated with Cu–O planes and its value $2\Delta_{ab}$ is measured with $\mathbf{E} \parallel \mathbf{c}$ light polarization. The second gap-like feature is the result of optical observation of proximity effects and Josephson-type current, excited by radiation with $\mathbf{E} \parallel \mathbf{c}$ polarization between Cu–O superconducting layers, separated by normal metal.

6. CONCLUSIONS

Normal state reflectivity anisotropy of high T_c superconductors under study and the anisotropy of superconducting energy gap in $YBa_2Cu_3O_{6.9}$ compounds prove the hypothesis of 2D superconductivity in metal oxides.

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