Письма в ЖЭТФ, том 58, вып.9, стр.722 - 725 ©1993 г. 10 ноября

ELLIPSOMETRIC MEASUREMENT OF THE SUPERCONDUCTIVE TRANSITION IN $YBa_2Cu_3O_{7-\delta}$ *ab*-ORIENTED FILM AT WAVELENGTH 119 μm

A.B.Sushkov, S.V.Shulga, E.A.Tishchenko[†]

Institute of Spectroscopy, Russian Academy of Science,
142092 Troitzk, Moscow region, Russia
[†]*P.L.Kapitza Institute for Physical Problems, Russian Academy of Science,*
117973 Moscow, Russia

Submitted 3 September 1993

We present the first measurements of the temperature dependence of the complex dielectric function $\epsilon_{ab}(\omega_0, T)$ for *ab*-oriented (with c-axis perpendicular to the film) $YBa_2Cu_3O_{7-\delta}$ film on a single crystal $SrTiO_3$ substrate obtained by far-infrared ellipsometry. The temperature behavior of $\epsilon_{ab}(\omega_0, T)$ qualitatively conforms to that predicted by the model of the strong electron-phonon interaction. At our frequency we did not observe any peaks below T_C.

Recently a lot of investigations both experimental and theoretical have been devoted to the temperature dependence of the low-frequency conductivity of high-T_C superconductors [1-4]. Most measurements have been carried out at microwave frequencies; only those of Ref. 2 were at far-infrared frequencies ($15 \div 80$ cm^{-1}). Both the real and the imaginary part of the complex dielectric function (or conductivity) are of great interest for the understanding of high-T_C superconductivity. The real part of ϵ determines the penetration depth of the electromagnetic field, while the imaginary part is responsible for the energy absorption. Ellipsometry [5] is one of the appropriate techniques for measurement of the complex dielectric function of high-T_C superconductors in the far-infrared.

In ellipsometry the change of the polarization ellipse is recorded. If polarized light is reflected from some plane surface, then the in-plane component of the electric vector $\vec{E}$ and the component perpendicular to the plane of incidence acquire different amplitude attenuations and phase shifts. Due to that effect the linear incident polarization becomes elliptical polarization. The optical constants of the substance can be unambiguously computed from a couple of measured ellipsometric parameters.

Ellipsometry needed an essential development to be useful for the study of high-temperature superconductors in the far-infrared. In the far-infrared one deals with wide beams of long-wavelength radiation and small highly reflecting samples. To minimize diffraction effects from the sample edges we focus the radiation on the sample surface. To take into account the effect of the beam's convergence we have found an original resolution of the problem, based on the lens's ability to perform a Fourier transform, valid for an arbitrary wavelength [6]. In our formalism both the direct and the inverse problems of the convergent beam ellipsometry for an arbitrary reflecting system can be solved.

Far-infrared radiation from a water-vapor electrical discharge laser passes through the following optical system: a chopper, a focusing lens L_1 (angle of convergence $2\alpha \approx 7^\circ$), a polarizer, a warm window, a cold window, a cold filter, the sample (angle of incidence of the beam 80°), a cold filter, a cold window, a warm window, an analyzer, a lens L_2, an aperture, a lens L_3, and the detector. The windows are made of 20 μm mylar. Black polyethylene filters cut off the short-wavelength radiation. When put between polarizers together with a sample, windows and filters can produce larger changes in polarization [7] than the sample can. We mount them in a compensating orientation. To take into account the rest of the polarization effect due to the oblique incidence of the beam on the windows, we measured the polarization ellipse with and without windows and filters at the same alignment at room temperature. We neglect the curvature of the mylar. The sample was placed in a continuous-flow cryostat and an atmosphere of cryoagent (helium or nitrogen). The temperature of the sample is measured by a thermocouple which is lightly pressed to the back surface of the sample with In/Ga eutectic.

The sample is a thick film of $YBa_2Cu_3O_{7-\delta}$ with c-axis perpendicular to the film's surface. The substrate was cut from a $SrTiO_3$ single crystal. It has (100) orientation and dimensions 5×10 mm. The film was prepared by laser ablation to the hot substrate in an atmosphere of argon-oxygen mixture. The film thickness was controlled by the frequency shift of a quartz resonator and was annealed *in situ*. The film thickness is $0.40 \pm 0.01 \mu m$. In X-ray patterns only (00n) picks with $n = 3, \ldots, 9$ were observed. Using an empirical formula we have obtained $\delta \simeq 0.07$. The SEM micro-photographs show a homogeneous structure with twinning planes in the diagonal direction [8].

We fix, as usual in ellipsometry, the polarizer angle at +45° or at −45° with respect to the plane of incidence and take the two-zone average. The analyzer is rotated an angle A and the $I(A)$ curve is recorded in the vicinity of the minimum. We measured two quantities:

1) The angle A_{min} at which the detected signal $I(A)$ has the minimum value (Fig. 1b); this parameter is, in fact, the azimuth of the polarization ellipse averaged over the convergent beam;

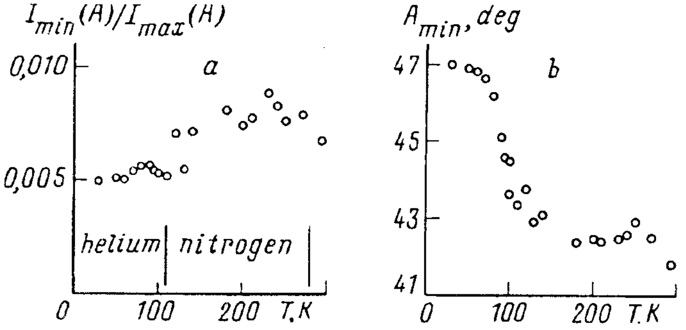

Fig. 1. Measured ellipsometric parameters: *a*) average ellipticity, *b*) average azimuth

2) The ratio of the minimum signal to the maximum one I_{min}/I_{max} which is, in turn, the ellipticity of the polarization ellipse averaged over the convergent beam (Fig. 1a).

These two parameters can also be calculated. Solving the inverse ellipsometric problem, we find the values of ϵ such that the error function has the minimum value [7].

To calculate ϵ for the superconducting and normal states we have used the nonlinear Eliashberg equations [9] and Nam's formalism for the phonon contribution to the optical conductivity [10]. All the calculations were carried out in the realaxis formalism using the $\alpha^2F(\omega)$ function described in Ref. 11. That function has been successfully to describe the optical spectra of Y-Ba-Cu-O crystals Ref. 12. In the superconducting state the experimental data can be described by means of the temperature-dependent pair-breaking effect, which is a natural consequence of the strong electron-phonon interaction [13]. The inelastic pair-breaking scattering leads to a smearing of singularities in the electron density of states, to the appearance of states inside the superconducting "gap," and to strong dumping in different dynamic processes.

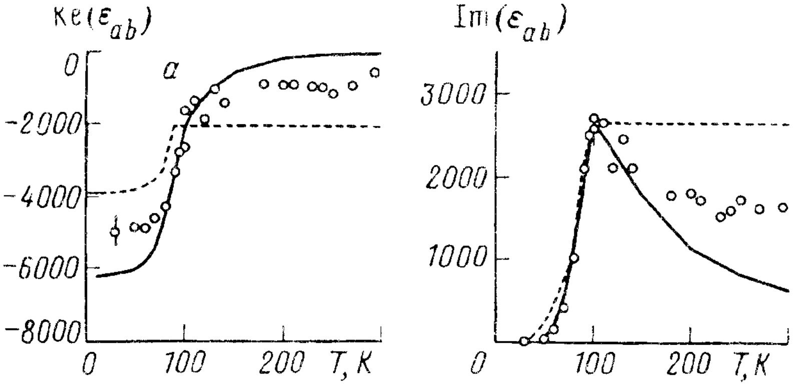

Fig. 2. Complex dielectric function of $YBa_2Cu_3O_{7-\delta}$ film: circles – experiment, solid lines – strong coupling model, dashed lines – BCS model

In Fig. 1 the measured parameters are shown as a function of temperature. In Fig. 2 we show ϵ calculated from the experimental data. The convergence of the beam, the effect of the windows, the thickness of the 1-2-3 film, and $\epsilon_s(\omega_0,T)$ for the $SrTiO_3$ substrate measured before the preparation of the film were taken into account in solving the ellipsometric inverse problem. The experimental data have not been smoothed before solving the inverse ellipsometric problem. The circles denote the experimental data. The values measured in the normal state are characterized by larger error than those measured in the superconducting state. That error is caused by the movement of the cold part of the cryostat during the first half of cooling process. The error bars in Fig. 2a correspond to ±10% variations in the measured value in Fig. 1a due to the fluctuation of the beam intensity. The error bars in Fig. 2b are due to the measurement accuracy ±0.1° in the azimuthal change caused by the windows. The solid lines are calculated for the strong-coupling model with electron-phonon interaction constant $\lambda = 2.5$, plasma frequency $\omega_p = 11900$ cm^{-1}, and damping frequency due to impurities $\gamma_{imp} = 50$ cm^{-1}. Variations of γ_{imp} in the range $1 \div 100$ cm^{-1} practically do not change the curves because of the larger electron-phonon scattering. The dashed lines are calculated for the BCS-model with plasma frequency $\omega_p = 6180$ cm^{-1} and $\gamma_{imp} = 107$ cm^{-1}. In Fig. 3 we show the temperature dependence of the electromagnetic field penetration depth δ. We calculate it as $\delta = \lambda/2\pi\kappa$, where λ is the wavelength and κ is the imaginary part of the complex index of refraction. Although, at first glance, both the BCS and the strong-coupling curves agree roughly equally well with the experimental ones, we prefer the strong-coupling model because of the more realistic value of the plasma frequency.

In conclusion, we should say that our results at the frequency 84 cm^{-1} (or 119 μm) concerning the superconducting state are in qualitative agreement with both the BCS and the strong-coupling models. The discrepancy between the

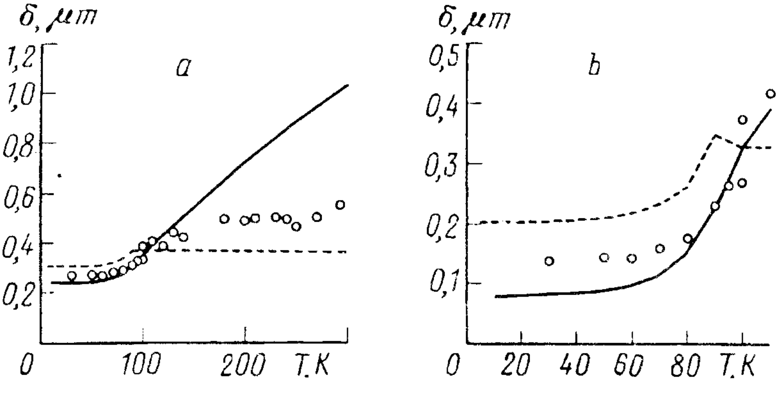

Fig. 3 Penetration depth of the electromagnetic field. All notation as in Fig. 2

experimental and theoretical data can be related both to the imperfections of the film and to the simplicity of the model. We believe that ellipsometric measurements at other frequencies and perfect samples can give important information for the solution of the fundamental problem of the high-T_C superconductivity.

The authors thank G.N.Zhizhin and V.A.Yakovlev for supporting this work and for helpful discussions.

---

1. P.H.Kobrin, J.T.Cheung, W.W.Ho et al., Physica C **176**, 121 (1991).
2. M.C.Nuss, P.M.Mankievich, M.L.O'Malley et al., Phys. Rev. Lett. **66**, 3305 (1991).
3. O.Klein, K.Holczer, G.Gruner and G.A.Emelchenko, Journal de Physique I France **2**, 517 (1992).
4. D.A.Bonn, P.Dosanjh, R.Liang and W.N.Hardy, Phys. Rev. Lett. **68**, 2390 (1992).
5. R.M.Azzam and N.M.Bashara, "Ellipsometry and Polarized Light", North-Holland, Amsterdam, 1977.
6. А.Б.Сушков, Э.А.Тищенко, Оптика и спектроскопия **72**, 491 (1992). [A.B.Sushkov and E.A.Tishchenko, Opt.Spectrosc.(USSR) **72**, 265 (1992)].
7. A.B.Sushkov and E.A.Tishchenko, Submitted to Int. J. Infrared and Millimeter Waves.
8. А.Н.Ивлев, Э.А.Тищенко, Сверхпроводимость: физика, химия, техника **5**, 836 (1992).
9. P.B.Allen and B.Mitrovic, in: Solid State Physics, vol. 37, eds. H.Ehrenreih, F.Zeitzand and D.Turnbull, Academic Press, New York, 1982.
10. S.B.Nam, Phys. Rev. **156**, 470 (1967).
11. S.V.Shulga, O.V.Dolgov, and E.G.Maksimov, Physica C **178**, 266 (1991).
12. O.V.Dolgov, A.A.Golubov, and S.V.Shulga, Phys. Lett. A **147**, 317 (1990).
13. A.A.Mikhailovsky, S.V.Shulga, A.E.Karakozov, et al., Solid State Commun. **80**, 511 (1991).