Ellipsometric measurement of the superconducting transition in a $YBa_2Cu_3O_{7-\delta}$ *ab*-oriented film at a wavelength of 119 μm

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The first measurements of the temperature dependence of the complex dielectric function $\epsilon_{ab}(\omega_0,T)$ for an *ab*-oriented (with the *c* axis perpendicular to the film) $YBa_2Cu_3O_{7-\delta}$ film on a single-crystal $SrTiO_3$ substrate are presented. The measurements were obtained using 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 coupling. No peaks below T_c were observed at the frequency which was used.

Many experimental and theoretical studies have recently focused attention on 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 (15–80 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 a plane surface, then the in-plane component of the electric vector **E** and the component perpendicular to the plane of incidence acquire different amplitude attenuations and phase shifts. Because of this effect, the linear incident polarization becomes an elliptical polarization. The optical constants of the substance can be unambiguously computed from two measured ellipsometric parameters.

Ellipsometry must be developed substantially to be useful for the study of high-temperature superconductors in the far-infrared. The far-infrared involves the use of wide beams of long-wavelength radiation and small, highly reflecting samples. To minimize diffraction effects from the sample's edges, we focused the radiation on the sample surface. To take into account the effect of convergence of the beam, we have found an original resolution of the problem, which is based on the lens's ability to perform a Fourier transform and which is valid for an arbitrary wavelength.[6] In our formalism both the direct and 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 $2\alpha \approx 7°$), a polarizer, a warm window, a cold window, a cold filter, the sample (incidence angle 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 a 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 the polarization[7] than does the sample. We have mounted 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 at room temperature the polarization ellipse with and without the windows and filters at the same alignment. The curvature of the mylar was ignored. The sample was placed in a continuous-flow helium or nitrogen cryostat. The temperature of the sample was measured by a thermocouple which was pressed slightly to the back surface of the sample with an In/Ga eutectic.

The sample was a thick film of $YBa_2Cu_3O_{7-\delta}$ with the *c* axis perpendicular to the film's surface. The substrate was cut from a $SrTiO_3$ single crystal which had a (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 thickness of the film was controlled by the frequency shift of a quartz resonator. The film was annealed *in situ*. The film thickness was 0.40 ± 0.01 μm. In the x-ray patterns only $(00n)$ picks with $n = 3,...,9$ were observed. Using an empirical formula, we have obtained $\delta \simeq 0.07$. The SEM microphotographs show a homogeneous structure whose twinning planes run in the diagonal direction.[8]

As is usually the case in ellipsometry, the polarizer angle is fixed at +45° or at −45° with respect to the plane of incidence and the two-zone average is taken. The analyzer is rotated an angle A and the $I(A)$ curve is recorded near 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.

2) The ratio of the minimum signal to the maximum signal, $I_{\min}/I_{\max}$, 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 in such a way 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 real-axis formalism using the $\alpha^2F(\omega)$ function described in Ref. 11. That function has been used successfully to describe the optical spectra of Y–Ba–Cu–O crystals.[12] In the superconducting state the experimental data can be described by means of the temperature-

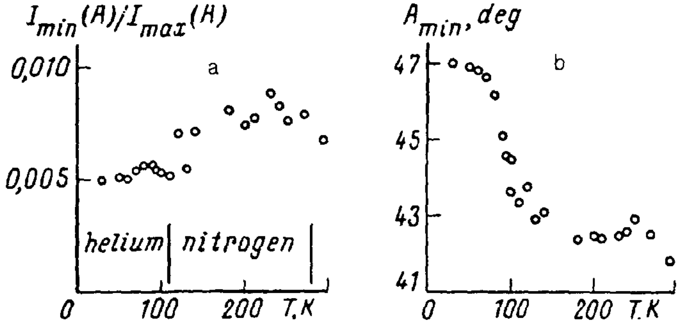

FIG. 1. Measured ellipsometric parameters. (a) Average ellipticity; (b) average azimuth.

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.

In Fig. 1 the measured parameters are shown as a function of temperature. Figure 2 shows ϵ which was 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, which was measured before the preparation of the film, were taken into account in the solution of the ellipsometric inverse problem. The experimental data have not been smoothed out before solving the inverse ellipsometric problem. The circles denote the experimental data. The values measured in the normal state are characterized by a larger error than those measured in the superconducting state. That error was caused by the movement of the cold part of the cryostat during the first half of the cooling process. The error bars in Fig. 2a correspond to ±10%

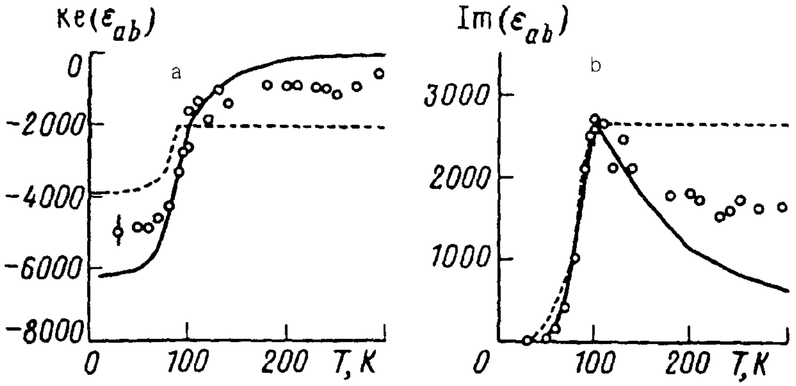

FIG. 2. Complex dielectric function of $YBa_2Cu_3O_{7-\delta}$ film. Circles—experiment; solid lines—strong-coupling model; dashed lines—BCS model.

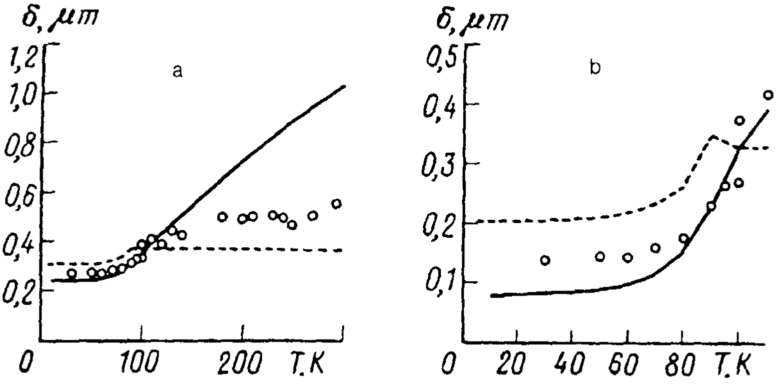

FIG. 3. Penetration depth of the electromagnetic field. The notation is the same as in Fig. 2.

variations in the measured values in Fig. 1a due to the fluctuation of the beam intensity. The error bars in Fig. 2b are due to the measurement accuracy of ±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 $\omega_p = 11\,900$ cm^{-1}, and damping frequency due to impurities $\gamma_{\text{imp}} = 50$ cm^{-1}. Variations of γ_{imp} in the range 1–100 cm^{-1} have virtually no effect on the curves because of the larger electron–phonon scattering. The dashed lines are calculated for the BCS model with a plasma frequency $\omega_p = 6180$ cm^{-1} and $\gamma_{\text{imp}} = 107$ 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, the BCS and the strong-coupling curves agree roughly equally well with the experimental curves, we prefer the strong-coupling model because of the more realistic value of the plasma frequency.

In conclusion, we note that at the frequency 84 cm^{-1} (or 119 μm) our results for the superconducting state are in qualitative agreement with the BCS and the strong-coupling models. The discrepancy between the experimental and theoretical data are attributable to the imperfections of the film and to the simplicity of the model. We believe that ellipsometric measurements at other frequencies and the use of perfect samples can give important information for the solution of the fundamental problem of high-T_c superconductivity.

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