Negative-light-flux spectroscopy of oriented films

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The absorption spectra of oriented $YBa_2Cu_3O_7$ films in the regime of negative light fluxes are reported. The spectra are interpreted on the basis of a model calculation in which it is assumed that there is no gap in the IR conductivity spectrum $\sigma_1^S(\omega)$ of the superconductor.

Several papers on the long-wave IR spectra of ceramics, oriented films, and single crystals of $YBa_2Cu_3O_7$ have reported an energy gap $2\Delta(T)$, which arises in the course of the transition to a superconducting state. The quantity $2\Delta(0)/kT_c$ has varied over the range 0.5–3.5 in studies carried out by different investigators,[1–5] while the corresponding figures for oriented films and single crystals are about 4 (Ref. 6) and 8 (Ref. 7), respectively. In most of the studies, $2\Delta(0)/kT_c$ has been determined approximately, on the basis of an analogy with low-temperature superconductors. It seemed worthwhile to carry out a detailed study of an oriented entity and to determine $2\Delta(0)/kT_c$ more precisely through a calculation based on a model.

The essential feature of negative-light-flux spectroscopy[8] is that a cooled sample is put in the place of an ordinary radiation source. The signal at a thermal radiation detector, at room temperature, is proportional to the difference between the heat flux from the detector to the sample and that in the opposite direction. The signal is thus proportional to the absorptivity of the sample.

This is one of the important advantages of the negative-light-flux method over ordinary reflection spectroscopy of high-temperature superconductors: When the material goes superconducting, the change in the absorption is substantially greater than that in the reflection. A second advantage is that the intensity of the radiation incident on the sample is very low (lower by a factor of about 100 than that in conventional methods). For measurements of the absorptivity A of a film-plus-substrate system it is necessary to carry out three measurements: 1) for the sample itself; 2) for a model blackbody, which serves as an emissivity standard; 3) for a metal mirror. The latter measurement is carried out in order to eliminate parasitic radiation by the sample holder.

In the measurements the sample is mounted on the cold stage of a helium cryostat with temperature regulation with the help of a precalibrated temperature-sensitive transistor, which is cemented directly to the reflecting surface. The temperature is regulated within ± 0.1 K.

The $YBa_2Cu_3O_7$ film samples are produced on substrates of strontium titanate single crystals, oriented in the (001) plane, by pulsed laser evaporation in an oxygen atmosphere. We studied single-phase, single-crystal films 1 μm thick with $T_c \gtrsim 90$ K

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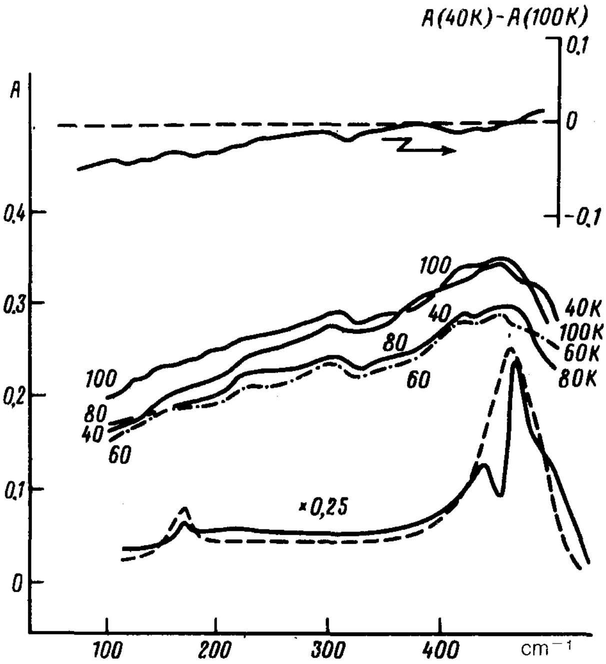

FIG. 1. Absorption spectra of a $YBa_2Cu_3O_7$ film on a strontium titanate substrate. Shown at the bottom are an experimental spectrum (solid line) and a model spectrum (dashed line) of the absorption of the substrate. The substrate parameters are as follows: $\omega_{T_1} = 50\ \mathrm{cm}^{-1}$, $\omega_{L_1} = 800\ \mathrm{cm}^{-1}$, $\gamma_1 = 75\ \mathrm{cm}^{-1}$; $\omega_{T_2} = 500\ \mathrm{cm}^{-1}$, $\omega_{L_2} = 470\ \mathrm{cm}^{-1}$, $\gamma_2 = 15\ \mathrm{cm}^{-1}$; $\omega_{T_1} = 170\ \mathrm{cm}^{-1}$, $\omega_{L_1} = 150\ \mathrm{cm}^{-1}$, $\gamma_3 = 60\ \mathrm{cm}^{-1}$; $\epsilon_\infty = 25$.

and with a transition width <0.5 K (T_c was found by the four-probe resistance method and from the temperature dependence of the magnetic susceptibility).[9]

Figure 1 shows some typical absorption spectra of the film-plus-substrate system. Shown in the same figure is the absorption spectrum of the substrate at $T = 80$ K, which depends weakly on the temperature over the interval 40–100 K. All of the observable changes in the absorption spectrum are thus caused primarily by changes in the optical properties of the film. (We recall that the thermal drift of the parts of the cryostat is taken into account for each value of the temperature by subtracting the spectrum measured when the metal mirror was put in the place of the sample.) It can be seen from Fig. 1 that as T is reduced from 100 to 80 K, there is a decrease in A over the entire spectral range, and at $T < 60$ K there is an increase in A at frequencies $\omega > 150\ \mathrm{cm}^{-1}$. In the present letter we will discuss only the difference between the spectra in the normal state (100 K) and the superconducting state (40 K), with the intention of returning to the analysis of the temperature dependence in a future paper.

To interpret the results, we have carried out a model calculation of the absorption spectra through a solution of the electrodynamic problem of the reflection of light from a two-layer system.[10] The dielectric constant of the substrate was simulated by three oscillators with the parameter values listed in the Fig. 1 caption. The IR conduc-

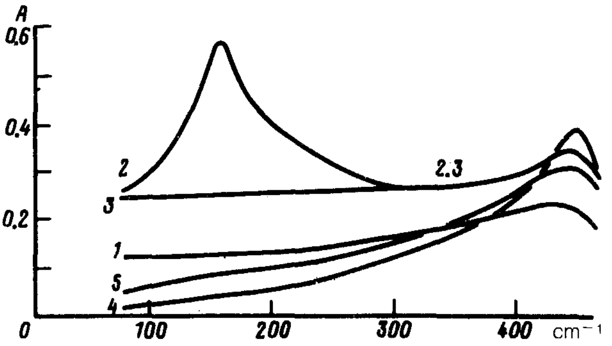

FIG. 2. Model absorption spectra of a metal film ($d = 1$ μm, $\omega_p = 22\,000$ cm^{-1}, $\Gamma_p = 18\,000$ cm^{-1}, $\epsilon_\infty = 20$). 1—Normal state; 2–5) superconducting state; 2—BCS, energy gap of $2\Delta = 150$ cm^{-1}; 3—BCS, "smeared" gap of $2\Delta = 150$ cm^{-1}, $D = 2\Delta$; 4—BCS, gap of $2\Delta = 800$ cm^{-1}; 5—gapless superconductor (the third version of the model; see the text proper).

tivity spectrum of the film was written in the Drude approximation. Three versions of the IR conductivity spectrum were used to simulate the superconducting state: 1) The spectrum found from the BCS model through calculations by Mattis and Bardeen[11]; 2) a first version of a spectrum with a 2Δ gap "smeared over" a Gaussian distribution with a dispersion $D \sim 2\Delta$; 3) a spectrum proportional to the spectrum in the normal state (with a δ-function added at $\omega = 0$, of course). Figure 2 shows model spectra corresponding to all three versions of $\sigma_1^S(\omega)$.

A comparison of the experimental spectra with the model spectra shows that there are no sharp structural features associated with an energy gap at any of the temperatures in Fig. 1, at least in the frequency range $500 > \omega > 50$ cm^{-1}. It can be seen from Fig. 2 that the spectrum calculated from version 3 is the best approximation of the experimental spectrum in the superconducting state. It may be that an energy gap arises at higher energies, at $T = 80$ K. However, a modeling of this situation (line 4 in Fig. 2) shows that in this case the absorption should have fallen off significantly at frequencies $\omega \lesssim 400$ cm^{-1}.

An alternative explanation of the experimental results would have to be based either on the assumption that there is no energy gap at all—this assumption contradicts the results measured by other methods (tunneling conductivity and the spin-lattice relaxation time from NMR)—or the assumption that there are two types of carriers and that the energy spectrum of one type has no gap. The size of the gap in the spectrum of the carriers of the other type should again be large ($\gtrsim 8kT_c$).

We should also mention the possibility that the gap is actually nonuniform (the second version of the modeling). In this case, as we see from line 3 in Fig. 2, the model spectrum again has no sharp structural features in the superconducting state, agreeing with the experimental spectrum in this regard.

In summary, a definite conclusion which follows from our study is that there are no structural features associated with a sharp (unsmeared) energy gap in the interval $(0.8–8)kT_c$. We are naturally led to ask about the nature of the low-frequency anomalies ($\omega \approx 150$–$200\ \mathrm{cm}^{-1}$) in the spectra of $YBa_2Cu_3O_7$ ceramics, which many investigators have linked with a manifestation of a sharp energy gap. We do not rule out the possibility that these anomalies can be explained by a vanishing of the effective dielectric constant $\mathrm{Re}(\epsilon_{\mathrm{eff}})$ at a corresponding frequency. Such an event would be possible if the relation $\mathrm{Re}(\epsilon_{\mathrm{eff}}) > 0$ prevailed in the normal state.

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