Optical study of $YBa_2Cu_3O_{7-x}$ thin films by means of synchrotron radiation in the region 4–30 eV

V. G. Stankevich, N. Yu. Svechnikov, K. V. Kaznacheev, R. A. Kink, Kh. É. Niédrais, V. N. Golubev, V. Ya. Kosyev, Yu. N. Simirskiĭ, and M. B. Tsetlin

I. V. Kurchatov Institute of Atomic Energy Moscow; Institute of Physics, Academy of Sciences of the Estonian SSR; Institute of Applied Physics, Academy of Sciences of the USSR

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The reflection, emission, and luminescence excitation spectra of a superconducting $YBa_2Cu_3O_{7-x}$ film have been studied with the help of synchrotron radiation for the first time. The results reveal a broad luminescence band of an exciton type at $E \sim 2.8$ eV. The intensity of this band has a structural feature at the temperature of the superconducting transition. This intensity decreases with a further increase in the temperature.

In most of the many studies of the properties of high-temperature superconductors which have been reported, the optical spectra of these substances have been studied in the IR and visible regions (e.g., Refs. 1 and 2). The luminescence excitation spectrum in the near-vacuum-UV region (up to 10 eV) has been studied only by

Lushchik *et al.*[3] In the present letter we report a study of the reflection, luminescence excitation, and emission spectra and their temperature dependence.

In the experiments we used a film with a thickness of 1500 ± 200 Å, synthesized on a $SrTiO_3$ substrate by pulsed laser deposition in a oxygen medium. The superconducting transition temperature of the film was $\overline{T}_c = 85.7$ K; the width of the transition was 1.6 K. The critical current density of a freshly deposited film at $T = 77$ K was on the order of 10^6 A/cm^2.

The measurements were carried out on the vacuum-UV spectrometer of the SIBIR'-1 synchrotron-radiation source.[4] The luminescence spectra were measured with the help of an MDR-23 analyzing monochromator, operating in the photon-counting mode.

The sample was held in a vacuum $\sim 10^{-8}$ Torr. There was no preliminary heating of the apparatus. Measurements were taken over the temperature range 9–200 K. In the experiments with the film, its resistance was monitored in a four-lead arrangement directly in the beam of synchrotron radiation.

Figure 1 shows the spectra of reflection at an angle of 45° for a film at a temperature of 30 K (1) during the first exposure to the synchrotron radiation and (2) after 2 h of preliminary irradiation. The reflection coefficient of our sample was $R \sim 10^{-3}$, so we can draw only general conclusions regarding the reflection spectrum. The structure of this spectrum includes many maxima, some of which vary during the irradiation. The positions of the peaks at energies of 7.0, 8.5, 19.0, and 24 eV, however, vary only slightly. Guang Lin Zhao *et al.*[5] have recently published calculations of ϵ_2 for an yttrium superconductor in the region up to 8 eV. Their calculations reveal a rounded maximum at 7 eV, which coincides with the peak at 7.0 eV in our own reflection spectrum.

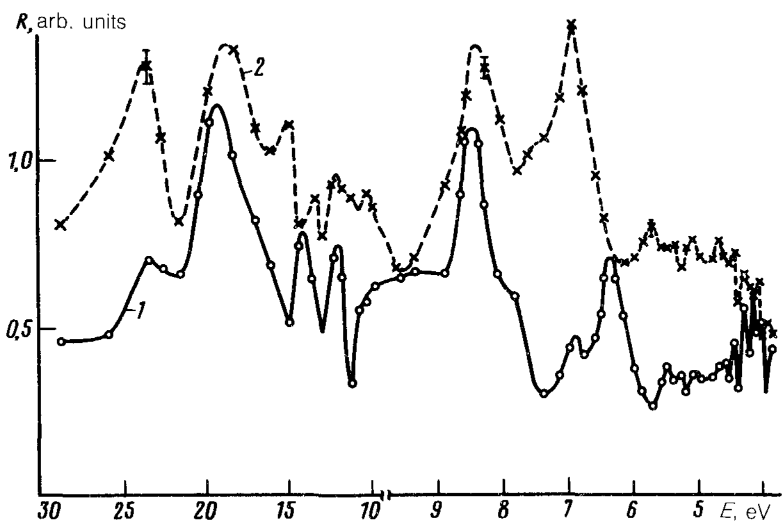

FIG. 1. Reflection spectra of the yttrium film.

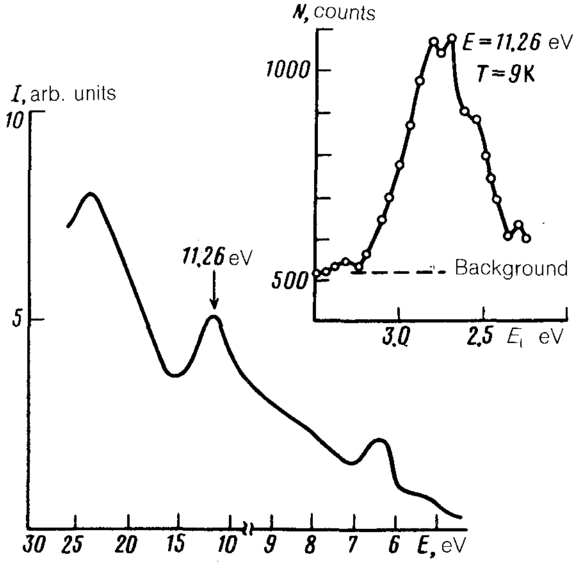

FIG. 2. Luminescence excitation spectrum of the film. The inset shows the emission spectrum of the film during excitation by a synchrotron-radiation beam with an energy of 11.26 eV.

Figure 2 shows a luminescence excitation spectrum of the film measured through an SZS-22 filter (the band 2.2–3.6 eV). We see from this figure that at excitation energies above 10 eV there is a relatively high luminescence yield ($\sim 10^{-2}$.) These spectra, like the reflection spectra, vary during the irradiation, but not as much. The changes consist primarily of a decrease in the height of the peak at $E \sim 12$ eV.

It can be seen from this figure that the luminescence intensity begins to grow at an excitation energy of 6–7 eV. This growth is probably due to a breeding of electronic excitations in the crystal (photon multiplication). The inset in Fig. 2 shows a luminescence spectrum of the film recorded at $T \sim 9$ K and at an excitation energy of 11.26 eV. A similar spectrum was found at an excitation energy ~ 22 eV.

Figure 3 shows the temperature dependence of the luminescence intensity near

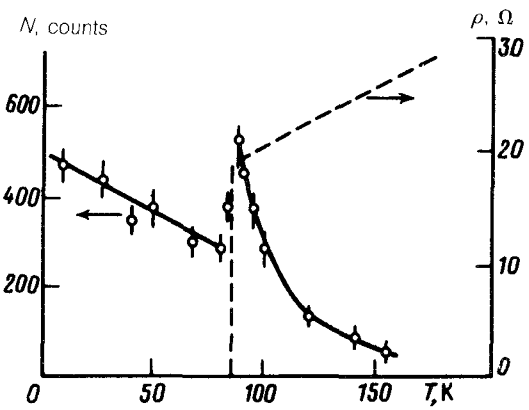

FIG. 3. Temperature dependence of the luminescence intensity and the resistance of the yttrium film.

the maximum of the spectrum ($E_L = 2.7$ eV) at the same excitation energy, 11.26 eV. Also shown here is a $\rho(T)$ dependence measured at the same time as that of the luminescence intensity. We see from this figure that the luminescence intensity initially varies only slightly with increasing temperature, has a structural feature near the superconducting transition, and falls off rapidly in the region of metallic conductivity.

The emission intensity thus correlates with the position of the superconducting transition. We take this experimental fact, combined with the circumstance that the luminescence spectra agree at different excitation energies, as evidence that the observed luminescence belongs to the compound $YBa_2Cu_3O_{7-x}$.

Time-resolved measurements of the luminescence showed that the decay time near the maximum of the spectrum in Fig. 2 is $\gtrsim 10^{-6}$ s.

From the experimental data reported above we can draw several conclusions.

1. The existence of luminescence with $E \sim 2.8$ eV and the beginning of photon multiplication at an energy of 6–7 eV indicate the existence of a band gap of the dielectric type in this yttrium superconductor. If the observed luminescence is interpreted as corresponding to indirect interband transitions, we conclude that the bottom of the conduction band lies ~ 3 eV from the Fermi level (this conclusion follows from the position of the high-frequency edge of the spectrum). This estimate agrees with most of the band-structure calculations (e.g., Ref. 6).

2. The relatively large width ($\gtrsim 0.1$ eV) of the luminescence peaks and the long luminescence formation time ($\gtrsim 10^{-6}$ s), however, are also consistent with the suggestion that the observed luminescence is due to an emission of an exciton type (self-trapped excitons or excitons trapped at defects).

3. The temperature dependence of the luminescence intensity indicates that excitations of this type may be related to the mechanism for the high-temperature superconductivity.

Detailed studies of the luminescence spectra which we plan to carry out may yield information on the temperature dependence of the structure of the electron and hole states near the Fermi level and thus take us closer to the solution of the question of the mechanism for the high-temperature superconductivity.

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