Direct measurement of electron–phonon coupling characteristics in $YBa_2Cu_3O_7$ by femtosecond laser spectroscopy

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Femtosecond laser spectroscopy has been used for a real-time study of the relaxation of an induced optical response in a $YBa_2Cu_3O_7$ film in the spectral interval 620–680 nm. The relaxation time τ varies with the wavelength, over the interval 0.5–1.5 ps. The electron–phonon coupling parameter is found from the measurement of τ: $\Lambda\langle\omega^2\rangle = 150\,\text{meV}^2$.

Despite major progress in research on the oxide superconductors, several of their fundamental properties in both the superconducting and normal states remain unclear. This is true, in particular, of the electron-phonon coupling parameter $\Lambda\langle\omega^2\rangle$, knowledge of which might shed light on the superconductivity mechanism of the oxide superconductors (Λ is the electron–phonon coupling constant, and $\langle\omega^2\rangle$ is the second moment of the normalized weight function in Eliashberg's theory).[1,2]

The characteristics of the electron-phonon interaction can be studied directly by femtosecond laser spectroscopy (a thermomodulation method[2–4]). This method was used in the present study to learn about the carrier relaxation in a $YBa_2Cu_3O_7$ film and to find the parameter $\Lambda\langle\omega^2\rangle$.

The test sample, a $YBa_2Cu_3O_7$ film 150 nm thick, was deposited on an $SrTiO_3$ substrate and then covered with a protective layer of MgO about 5 nm thick. The film, synthesized by rf magnetron sputtering, was polycrystalline with good grain orientation. The crystalline size was about 300 nm; the c axis ran normal to the surface. The superconducting transition temperature of this sample was $T_c = 80$ K.

The idea underlying this method is that a femtosecond laser pulse creates nonequilibrium carriers in a sample. The time evolution of these carriers is studied over a wide spectral range by measuring the reflection from the sample of a probing pulse applied after a time delay. The laser oscillator supported the generation of a continuous train of pulses 90 fs long at a wavelength of 610 nm with a repetition frequency of 100 MHz. The output from the laser oscillator was sent to the input of a four-stage dye laser amplifier, which was pumped by the beam from an excimer laser. The laser amplifier was pumped at a frequency of 3 Hz; this frequency determined the pulse repetition frequency at the output of the laser systems. From the amplifier output the laser pulses went to the measurement part of the experimental apparatus. After passage through the laser amplifier, the length of the laser pulses was 150 fs; the energy of

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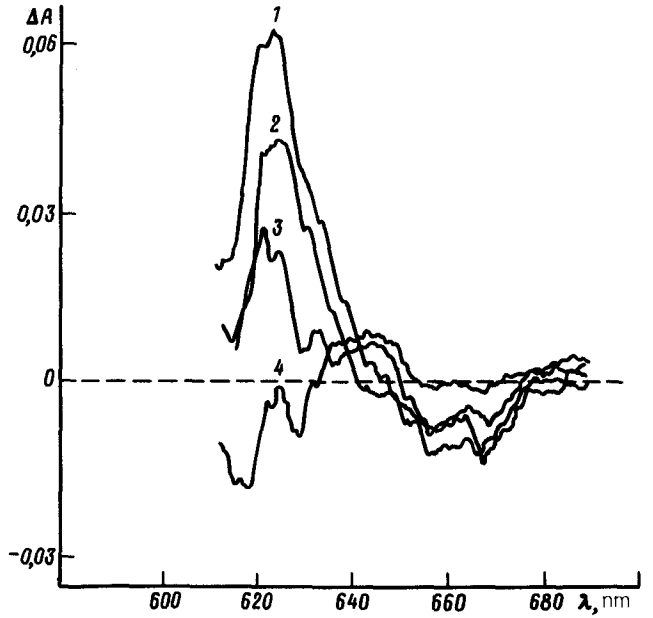

FIG. 1. Difference spectra found at several delay times. *1*—200 fs; *2*—340 fs; *3*—5600 fs; *4*—0 fs. [$\Delta A = \log(R + \Delta R/R)$].

an individual pulse ranged up to 400 μJ; and the pulse repetition frequency was 3 Hz.

In the measurement part of the apparatus, the laser beam was split in two: an excitation channel and a probing channel. In the excitation channel, the laser pulses were focused directly onto the test sample; the diameter of the excitation spot was 100 μm. In the probing channel the original laser beam was focused in a cell filled with heavy water in order to generate light with a spectral continuum; this light was then used to probe the sample over a wide spectral range. In the experiments, the change in the transmission (ΔT) or reflection (ΔR) of the sample was detected. The probing pulse was focused inside the excitation spot; the diameter of the spot of the probing pulse on the sample was 50 μm. The probing was carried out over the spectral interval 620–680 nm. An optical multichannel analyzer based on two linear arrays of charge-coupled devices was used to detect the probing pulse in order to achieve a spectral resolution. The error of the measurements was 0.01 of an optical density unit; the corresponding change in the reflection coefficient is $\Delta R/R = 2\%$.

The difference reflection spectrum of the film, measured at an initial temperature of 55 K (Fig. 1), was of variable sign: $\Delta R > 0$ in the interval 620–640 nm and $\Delta R < 0$ in the interval 640–680 nm. The value (λ^*) of the wavelength, at which the change in the reflection coefficient vanished [$\Delta R(\lambda^*) = 0$], was essentially independent of the time delay. The maximum change in R ($\Delta R/R \simeq 15\%$) was observed at $\lambda = 630$ nm.

Figure 2 shows the behavior of the reflection coefficient according to a recon-

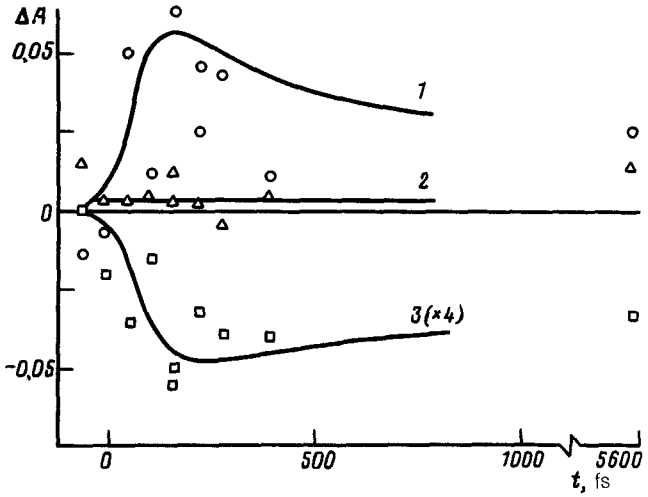

FIG. 2. Behavior of the change in the reflection coefficient, $\Delta R/R$, at three wavelengths. O—625 nm; Δ—640 nm; □—660 nm [*sic*]. Lines 1 and 3 correspond to fast relaxation processes with time scales of 0.5 and 1.5 ps.

struction from the experimental difference spectra. The relaxation time increases significantly with increasing λ: $\tau(\lambda = 628\text{ nm}) \simeq 0.5$ ps and $\tau(\lambda = 650\text{ nm}) \simeq 1.5$ ps. This effect may be due to a change in the filling of states near the Fermi level as a result of the pump pulse.

The change in reflection, $\Delta R/R$, can be written in the form

$$\Delta R/R = \alpha \Delta \epsilon_1 + \beta \Delta \epsilon_2, \tag{1}$$

where $\Delta\epsilon_1$ and $\Delta\epsilon_2$ are small changes in the real and imaginary parts of the dielectric constant (in the spectral interval of interest, we have $\alpha = 0.15$ and $\beta = 0.25$). The spectrum of the optical response ΔR can be explained in terms of the contribution to the dielectric constant from transitions between bands with an identical dispersion law, one of which (the band of the heavier quasiparticles) contains the Fermi level. Since α and β are of the same order of magnitude, it is necessary to consider the changes in both the real and imaginary parts of the dielectric constant (in contrast with the case of, for example, Cu, in which the relation $|\alpha| \ll |\beta|$ holds). Instead of considering the entire complex band diagram of $YBa_2Cu_3O_7$ (see the review by Pickett[5]), it is sufficient to adopt the simple but realistic model which incorporates the contribution to ϵ_1 and ϵ_2 of transitions near the photon energy $E_0 \simeq 2$ eV. Specifically, the contribution of transitions between the heavy-hole band, near whose top the Fermi level lies, to the two higher-lying bands was taken into account. The latter two bands are slightly split bands of lighter holes. Values of the parameters (the effective mass, the distance between bands, and the Fermi energy) close to those found from band-theory calculations were adopted. In addition, we used experimental values[6] for ϵ_1 and ϵ_2 for a photon energy $E_0 \simeq 2$ eV. For an estimate we used the value $\gamma^{-1} \simeq 50$ fs for the

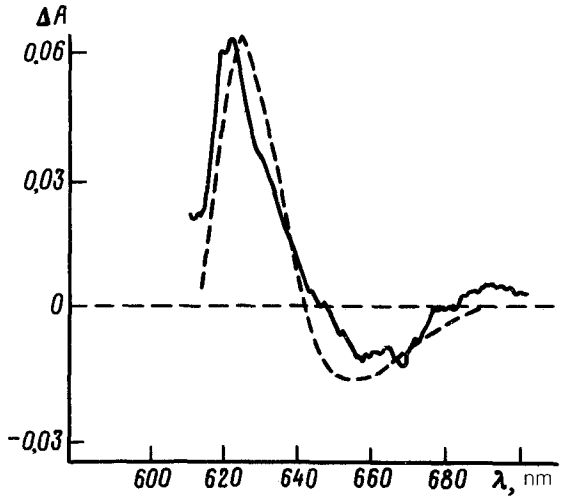

FIG. 3. Difference reflection spectra. Dashed line—Theoretical; solid line—experimental, at a delay time of 200 fs.

intraband relaxation time. The initial temperature of the sample was $T_0 = 55$ K. An estimate of the final temperature of the electron subsystem, $T_f \simeq 10^3$ K, was found under the assumption that all the absorbed energy is expended on heating this subsystem. The calculated difference spectrum agrees with the experimental spectrum (Fig. 3). The wavelength of 640 nm, at which the sign of the optical response changes, corresponds to transitions into the region of the Fermi level (or out of this region). The response relaxation time ($\tau \simeq 1$ ps) corresponding to this spectral region can be used to estimate the rate of electron–phonon relaxation near the Fermi level. Using the expression $1/\tau = 3\hbar\Lambda\langle\omega^2\rangle/(\pi k_B T)$, we find the electron-phonon coupling parameter for $YBa_2Cu_3O_7$ to be $\Lambda\langle\omega^2\rangle \simeq 150$ meV^2. The value of this parameter may prove important for reaching an understanding of the superconductivity in the oxide superconductors.

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