Spectral dependence of relaxation in $YBa_2Cu_3O_{7-\delta}$ with femtosecond laser excitation

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A spectral dependence of the energy relaxation time of nonequilibrium charge carriers was determined by means of time measurements of the optical density of a $YBa_2Cu_3O_{7-\delta}$ film with the help of femtosecond laser spectroscopy. The energy relaxation time increases rapidly in the spectral range corresponding to transitions into the vicinity of the Fermi level. This provides a new method for determining the position of the Fermi level. Excitation of coherent phonons with frequencies in the range 10–250 cm^{-1} was observed in a wide spectral probe range $\hbar\omega_{probe}=1.6-3.0$ eV.

Femtosecond laser spectroscopy has been found to be useful for investigating the characteristics of fast processes in solids, for obtaining from these characteristics the fundamental interaction constants of the electron and phonon subsystems, and for making real-time observations of the kinetics of phase transitions (see, for example, Refs. 1 and 2).

In the present work this method is employed to investigate the excited state of the high-T_c superconductor $YBa_2Cu_3O_{7-\delta}$ and to make a detailed study of the relaxation of nonequilibrium charge carriers in the femtosecond range. We observed a rapid growth of the electron energy relaxation time in certain spectral ranges associated with a transition into the vicinity of the Fermi level. The relation which we found opens up a possibility for a new method of determining the position of the Fermi level (and establishing its existence in the system of strongly correlated electrons). The use of 50-fs excitation and probe pulses enabled us to detect the excitation of coherent phonons in $YBa_2Cu_3O_{7-\delta}$ by means of the evolution of the photoinduced response in the femtosecond range.

The experimental sample consisted of a 0.05-μm-thick $YBa_2Cu_3O_{7-\delta}$ ($T_c=89$ K) film on a 0.5-mm-thick $SrTiO_3$ substrate. Excitation was produced with 2.5×10^{11} W/cm^2 50-fs optical pulses with photon energy $\hbar\omega_{pump}=2.34$ eV. The diameter of the excitation

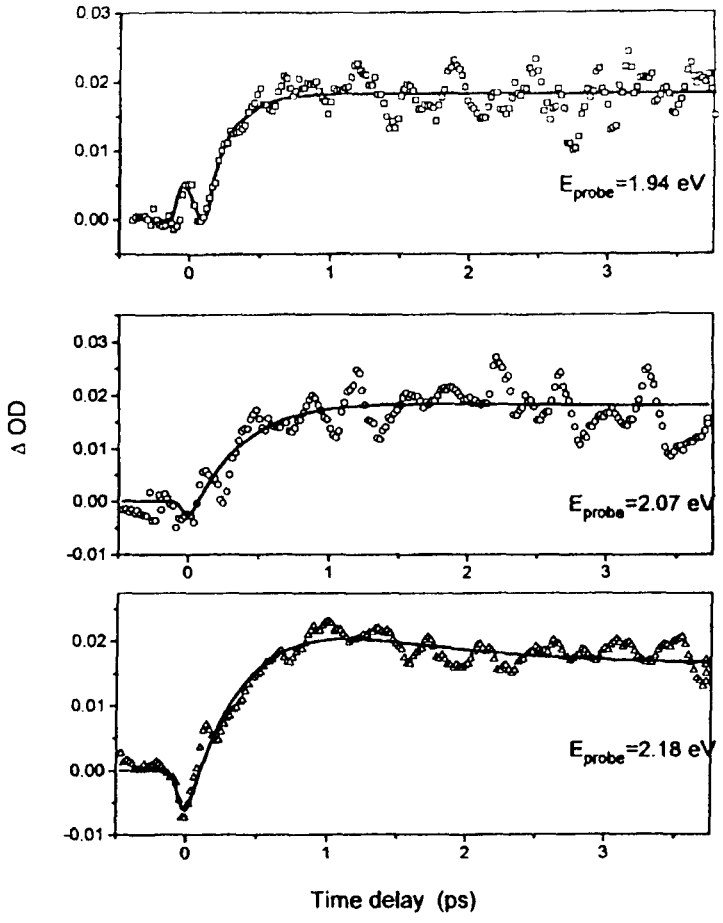

FIG. 1. Kinetics of the photoinduced change in the optical density of the $YBa_2Cu_3O_{7-\delta}$ film.

spot was equal to 100 μm. The transmission coefficient was investigated with the aid of a 50-fs probe pulse in the range 1.6–3.0 eV. The diameter of the probe spot was equal to ≈ 80 μm. The repetition frequency of the excitation and probe pulses was equal to ≈ 1 Hz. The delay time step was equal to 7 fs. The maximum delay in the present experiment reached ≈ 4 ps. The measurements were performed at room temperature.

The temporal dynamics of the measured optical density spectra $\Delta D_{\exp}(\omega,t)$ is shown in Fig. 1. The spectral dependence of the relaxation rate τ_1 of the optical density was obtained by fitting the experimental kinetic curves with adjustable functions $\Delta D_{fit}(t)$. It was found that τ_1 depends strongly on the wavelength of the probe pulse: It assumes the value $\tau_1 = 180 \pm 50$ fs in the range $1.7 < \hbar\omega < 2$ eV, increases up to a maximum value at $\hbar\omega \approx 2.1$ eV, and assumes the value $\approx 300 \pm 200$ fs in the region $2.4 < \hbar\omega < 3$ eV. The time dependences of the optical density spectra – rapid decrease of the optical density over a time determined by the duration of the excitation pulse followed by rapid relax-

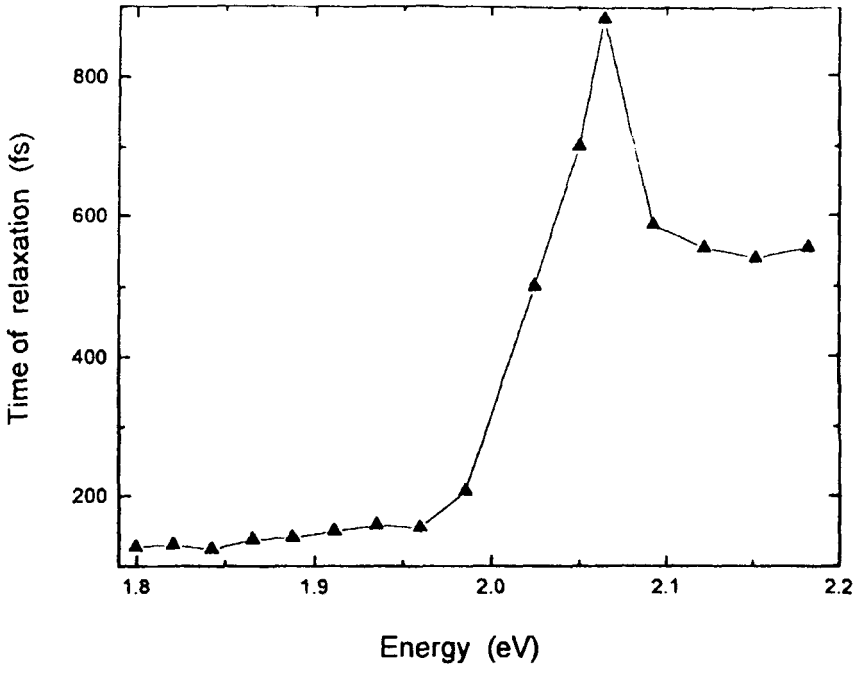

FIG. 2. Energy relaxation time of photoexcited carriers in a $YBa_2Cu_3O_{7-\delta}$ film as a function of the probe photon energy.

ation – indicate that at times much shorter than τ_1 the electron response makes the main contribution to the change in the optical properties. It is therefore possible, in principle, to observe the Fermi level in this region of the spectrum in the indicated time interval.[3] At times of the order of τ_1 and longer the contribution of the electron response is small and the lattice response principally accounts for the change in the transmission.

In this connection, the existence of a narrow peak in the energy dependence of the energy relaxation time τ_1 (see Fig. 2) is, in our opinion, most interesting. This peak can be interpreted as an increase in the inelastic scattering time near the Fermi level as a result of a decrease of the phase volume of the final states. This fact is an important characteristic of a Fermi liquid (see Refs. 3 and 4) and essentially determines the possibility of introducing quasiparticles for a Fermi liquid, whose damping decreases sharply near the Fermi surface. The electron energy relaxation time near the Fermi level at temperatures $T_e \ll \hbar\omega_{\rm ph}$, where $\hbar\omega_{\rm ph}$ is the characteristic phonon energy, behaves as $\tau_1 \sim (\varepsilon - \varepsilon_F)^{-3}$ (for interaction with acoustic phonons see Refs. 3 and 4). In the opposite case of high temperatures, $T_e \gg \hbar\omega_{\rm ph}$, the energy relaxation time does not depend on $\varepsilon - \varepsilon_F$. Since in our case the maximum heating of the electron subsystem amounts to $\Delta T_e \sim 270$ meV, and since it decreases to ~ 30 meV over a time of the order of τ_1, we find that the average electron temperature is $T_e \sim 180$ meV at times $t < \tau_1$. A comparison of this value with the maximum phonon energy in $YBa_2Cu_3O_{7-\delta}$ $\hbar\omega_{\rm ph} \sim 80$ meV shows that we are in the intermediate regime $T_e \gtrsim \hbar\omega_{\rm ph}$. Consequently, the energy dependence

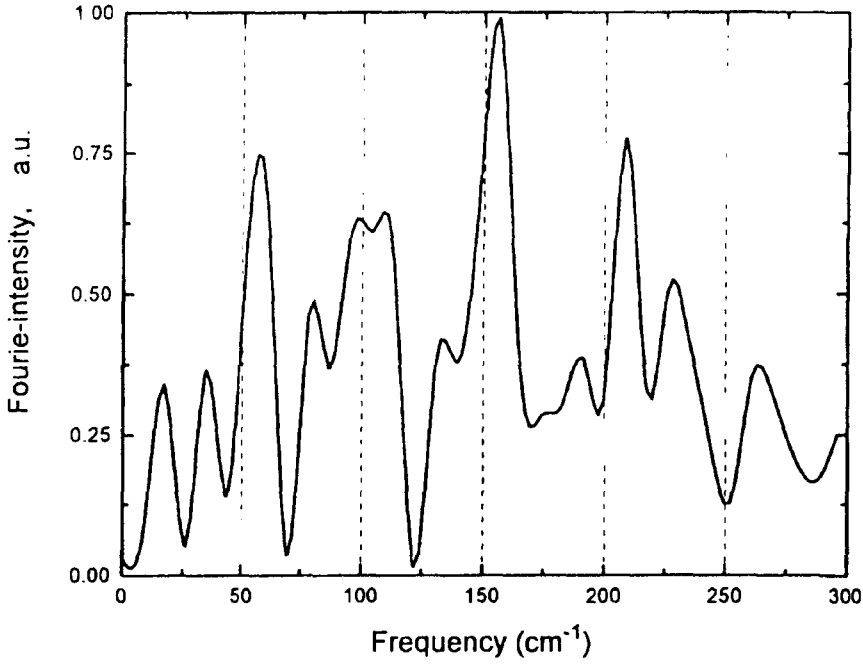

FIG. 3. Fourier spectrum of coherent phonons excited in a $YBa_2Cu_3O_{7-\delta}$ film by a femtosecond laser pulse.

$\tau_1(\varepsilon)$ is found to be not as sharp as one would expect for low temperatures.

The following fact attests to the interpretation given above for the peak in τ_1. It seems that the position of the Fermi level determined from the peak value of τ_1 agrees quite well with a nonmonotonic energy dependence of the photoinduced response for transitions near the Fermi level (see also Refs. 1 and 5). The above-indicated nonmonotonic dependence is attributable to the photoinduced emptying of the states below the Fermi level and to the enrichment of the states above the Fermi level. As a result, $\Delta\varepsilon_2(\omega)$ — the imaginary part of the change produced in the dielectric function by photoexcitation — is a nonmonotonic function of $\hbar\omega$. In this case $\Delta\varepsilon_2(\omega)$ vanishes for transitions exactly into the Fermi level. Correspondingly, the energy at which the peak value of τ_1 is reached agrees well with the energy at which $\Delta\varepsilon_2$ vanishes. We note that the energy dependence of τ_1 is very sharp, which makes it possible to determine the Fermi level more accurately than from the vanishing of the function $\Delta\varepsilon_2(\omega)$ vs the probe photon energy. (It should be noted that additional data are required to determine the curve of $\Delta\varepsilon_2$.)

The electron-phonon interaction parameter $\lambda\langle\omega^2\rangle$ was determined from the time dependence of the optical density. It was found that in the spectral range $1.7<\hbar\omega_{\text{probe}}<2.0$ eV $\lambda\langle\omega^2\rangle=500\pm150$ $(\text{meV})^2$, which agrees well with the results of Refs. 1 and 6.

The use of ultrashort optical pulses made it possible to observe the coherent lattice

vibrations excited by these pulses. These vibrations modulate the change in the permittivity and they change the intensity of the transmitted or reflected light. Fourier analysis of the time dependence of the change in the optical density $[\Delta D_{\exp}(t) - \Delta D_{\text{fit}}(t)]$ was used to determine the characteristic lattice vibration frequencies in the range 10–250 cm^{-1}. It was found that coherent phonons are excited in the entire spectral probe range $\hbar\omega_{\text{probe}} = 1.6-3.0$ eV. To determine the frequencies of the coherent phonons, which are excited efficiently in the entire probed region of the spectrum, all Fourier spectra were multiplied together, i.e., the geometric-mean spectrum of the vibrations was calculated.[7] As a result, it was found that vibrations with the frequency ~ 152 cm^{-1} predominate in this spectral range. The vibrations with frequencies ~ 57, ~ 110, and ~ 210 cm^{-1} and a number of other modes are observed with a lower amplitude. The excitation of modes with frequencies of ~ 116 and ~ 150 cm^{-1} was observed in Ref. 8 with the aid of femtosecond pulses ≈ 100 fs with excitation and probe photon energies $\hbar\omega \approx 2$ eV; the most intense mode (at room temperature) was the mode ~ 150 cm^{-1}, just as in our case. It is interesting that not only Raman-active (see Refs. 9–11 and the literature cited there) phonon modes (~ 116, ~ 150, and ~ 210 cm^{-1}), but also modes which are characteristic of infrared spectra,[9,12] ~ 190 and ~ 210 cm^{-1}, are observed in the Fourier spectra of the coherent phonons. The infrared-active mode with frequency ~ 230 cm^{-1} was observed in the Raman spectra[11] under resonance excitation with photons, $\hbar\omega \approx 2.2 \pm 0.2$ eV. The observation of the modes which are characteristic of infrared spectra is probably attributable to the removal of the alternative exclusion due to the defects in the material which are associated with oxygen vacancies.[11]

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[1] S. V. Chekalin, V. M. Farztdinov, V. V. Golovlev *et al.*, Phys. Rev. Lett. **67**, 3860 (1991); Yu. E. Lozovik, Yu. A. Matveetz, A. G. Stepanov *et al.*, JETP Lett. **52**, 221 (1990); I. E. Kardash, V. S. Letokhov, Yu. E. Lozovik *et al.*, JETP Lett. **58**, 138 (1993).
[2] Ultrafast Phenoeman VIII, Springer Series in Chem. Phys. **55** (1992).
[3] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskiĭ, *Methods of Quantum Field Theory in Statistical Physics*, Prentice-Hall, Englewood Cliffs, N. J., 1963 [Russian original, Fizmatgiz, Moscow, 1962].
[4] V. F. Gantmakher and I. B. Levinson, *Carrier Scattering in Metals and Semiconductors*, North-Holland, N. Y., 1987 [Russian original, Nauka, Moscow, 1984].
[5] A. L. Dobryakov, V. M. Farztdinov, Yu. E. Lozovik, and V. S. Letokhov, Opt. Commun. **105**, 309 (1994); A. L. Dobryakov, V. V. Golovlev, V. S. Letokhov *et al.*, Opt. Spektrosk. **76**, 975 (1994).
[6] S. D. Brorson, A. Kazeroonian, D. W. Face *et al.*, Solid State Commun. **74**, 1305 (1990).
[7] A. L. Dobryakov, S. A. Kovalenko, V. S. Letokhov *et al.*, Pisma Zh. Éksp. Teor. Fiz. **61**, 957 (1995) [JETP Lett. **61**, 895 (1995)].
[8] W. Albrecht, T. Kruse, and H. Kurz, Phys. Rev. Lett. **69**, 1451 (1992).
[9] R. Feile, Physica C **159**, 1 (1989).
[10] K. F. McCarty, J. Z. Liu, R. N. Shelton, and H. B. Radousky, Phys. Rev. B **41**, 8792 (1990).
[11] D. R. Wake, F. Slakey, M. V. Klein *et al.*, Phys. Rev. Lett. **67**, 3728 (1991).
[12] K. Kamaras, S. L. Herr, C. D. Porter *et al.*, Phys. Rev. Lett. **64**, 84 (1990); C. C. Homes, T. Timusk, R. Liang *et al.*, Phys. Rev. Lett. **71**, 1645 (1993).

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