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Role of degeneration processes in picosecond nonlinear spectroscopy of HTSCs

Yu.V. Bobyrev, V.M. Petnikova, K.V. Rudenko, V.V. Shuvalov

***Abstract*. It is shown that both spectral-temporal and temperature anomalies of the reflectance and transmittance kinetics of high-temperature superconducting films at high 'impact' excitation levels have a direct analogue in the case when much longer and coincident in time picosecond pulses are used to excite and probe the film state. In this case, due to degeneration, the nontrivial (with a sharp jump in the vicinity of the phase transition point) dependence of the nonlinear response amplitude of a sample on its initial temperature should be observed.**

Keywords: *pump – probe spectroscopy, high-temperature superconductors, picosecond excitation and probing, temperature dependence of the nonlinear response amplitude.*

1. Introduction

Processes of rapid relaxation in low- and high-temperature superconductors (HTSCs) are usually studied by the pump-probe method [1 – 22], which is most often employed to investigate the dependence of the reflectance and (or) transmittance of a preliminary excited sample on the delay time τ of its probing with respect to the instant of its 'impact' pumping [6, 8 – 10]. It is assumed that after excitation, hot carriers first are rapidly thermalised due to electron – electron (e – e) scattering (the thermalisation time is $\tau_{th} < 10$ fs for the electron energy $E_e \sim 1$ eV [23, 24]) and their energy distribution returns to the Fermi – Dirac distribution with the electron temperature T_e, which differs both from the initial temperature T_0 and the lattice temperature T_p [25]. Then, electron – phonon (e – p) scattering equalises the temperatures of the electron and phonon subsystems and $T_e \to T_p$ for the time τ_r. It was found experimentally [2 – 4, 7, 18, 26 – 29] that, in accordance with the theory [1, 30 – 32], the values of τ_{th} and τ_r drastically increase in the vicinity of the superconducting phase transition point ($T_0 \simeq T_c$). However, it was found soon [22] that, surprisingly, this property was observed only at high excitation levels, i.e., only in the cases when a sample virtually instantly should 'forget' any information about its initial state (about its initial temperature T_0).

In [33], the anomalous kinetics of the HTSC nonlinear response at a high excitation level was interpreted within the framework of the model assuming that the energy gap in the state spectrum of a HTSC cannot be rapidly destroyed even when T_e strongly differs from T_p [34] and that the HTSC nonlinear response in the pump-probe spectroscopy is caused by the interband electronic transitions [35]. It was shown that due to 'impact' excitation, the Fermi levels (with energy $E_{e,h}^F$) for free electrons (states over the energy gap) and holes (states under the energy gap) separate (the degeneration process), the energy gap $E_e^F - E_h^F$ drastically increasing with increasing the pump level. Because the state-density distribution in a HTSC provides the suppression of nonradiative three-body relaxation with increasing the energy of recombining carriers, the formation of a gap of width $\Delta(T_0, T_c) \neq 0$ for $T_0 \leqslant T_c$ in the electronic spectrum of the HTSC drastically changes the kinetics $E_e^F(t)$ and $T_{e,p}(t)$, which makes it possible to explain all anomalies observed in experiments.

Below, based on the kinetic model [33], we show that degeneration processes observed in picosecond nonlinear spectroscopy of HTSCs at high excitation levels should play an important role, determining the nontrivial dependence of the nonlinear response amplitude of a HTSC film on its initial temperature.

2. Kinetics of thermodynamic parameters upon picosecond pumping

At the first stage of the solution of the problem by using the kinetic model [33], we calculated the kinetics of the Fermi level shift $\Delta E^F(t) = E_e^F(t) - E_0^F$ from its equilibrium value E_0^F and the kinetics $\Delta T_e(t) = T_e(t) - T_0$ of a change in the electron temperature upon excitation by 20-ps pulses. We assume, as in [33], that the HTSC film (with the phase transition temperature $T_c = 90$ K, the thermal capacity $c_p = 0.9$ J cm^{-3} K^{-1}, the relaxation rate of the heat excess to a substrate $\gamma_{ps}^{(Q)} = 5 \times 10^{-3}$ ps^{-1}, the maximum energy of acoustic phonons $E_{max} \simeq 15$ meV, which corresponds to a $YBa_2Cu_3O_{7-\delta}$ film on a $SrTiO_3$ substrate) absorbs 30 % (the film thickness is ~ 200 nm) of the total energy (4×10^{-7} J) of the 800-nm pump pulse focused into a spot of diameter 150 μm. The rate constants of nonradiative three-body recombination $\tilde{\gamma}_{ehe}^{(R)}$ and $\tilde{\gamma}_{ehp}^{(R)}$ corresponded to the calculation [33].

As in [33], the energy gap $\Delta(T_0, T_c)$ in the state spectrum was assumed a constant independent of t ('frozen' gap)

Yu.V. Bobyrev, V.M. Petnikova, K.V. Rudenko, V.V. Shuvalov
International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia;
e-mail: vsh@vsh.phys.msu.su

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whose value corresponded to the weak coupling limit in the BCS theory. For $T_0 > T_c$, the state-density distribution $g(E_e; \Delta \equiv 0)$ was calculated from the known dependence of the electron energy E_e on its quasi-momentum $\boldsymbol{k}$ for the band structure of La_2CuO_4 [36]. Upon integration with respect to $\boldsymbol{k}$, this dependence was interpolated to the entire first Brillouin zone by the method described in [37]. Then, the dependence $g(E_e; \Delta \equiv 0)$ was approximated by a smooth analytic function in the energy range $|E_e - E_0^F| \leqslant 1$ eV, and for $T_0 \leqslant T_c$ the energy gap was introduced into the distribution obtained in this way, i.e., the electronic states with the energy falling into the gap of width $2\Delta(T_0)$ in the vicinity of E_0^F were redistributed over the rest of the states of the Brillouin zone by the method described in [33].

Figure 1 shows rather drastic transformations of the calculated kinetics of the instant shift $\Delta E^F(t) = E_e^F(t) - E_0^F$ of the Fermi level from its initial position and the jump of the electron temperature $\Delta T_e(t) = T_e(t) - T_0$ of the HTSC film upon variations of its initial temperature. It is easy to verify that upon picosecond excitation, the amplitude of instant deviations of the thermodynamic parameters of a HTSC film from their equilibrium values drastically increases with the opening of the energy gap in the electronic spectrum (in the vicinity of $T_0 \simeq T_c$), which obviously should affect the nonlinear response amplitude.

3. Electronic part of the nonlinear response

As in [33], we assume below that the nonlinear response of the HTSC film is determined by the change $\Delta\varepsilon = \varepsilon(E_e^F; T_e) - \varepsilon(E_0^F; T_0)$ of the electronic part (caused by the consideration of contribution from interband electronic transitions) of its complex permittivity ε due to the deviations of E_e^F and T_e from their equilibrium values E_0^F and T_0. The value of $\Delta\varepsilon$ was calculated from the standard expression

$$\varepsilon \sim \sum_{i \neq i'} \int\int \frac{|d_{ii'}(\boldsymbol{k}, \boldsymbol{k}')|^2 n_i(\boldsymbol{k})[1 - n_{i'}(\boldsymbol{k}')]}{\omega - \Omega_{ii'}(\boldsymbol{k}, \boldsymbol{k}') + i\Gamma_{ii'}(\boldsymbol{k}, \boldsymbol{k}')} \, d\boldsymbol{k} \, d\boldsymbol{k}', \qquad (1)$$

where subscripts i and i' number zones involved in the $(i, \boldsymbol{k}) \to (i', \boldsymbol{k}')$ transition with the dipole moment $d_{ii'}(\boldsymbol{k}, \boldsymbol{k}')$ and the resonance frequency $\Omega_{ii'}(\boldsymbol{k}, \boldsymbol{k}') = E_{i'}(\boldsymbol{k}') - E_i(\boldsymbol{k})$; $E_i(\boldsymbol{k})$ and $n_i(\boldsymbol{k})$ are the normalised electron energy and the occupation number of the $(i, \boldsymbol{k})$ state; and $\Gamma_{ii'}(\boldsymbol{k}, \boldsymbol{k}')$ is the relaxation rate of interband polarisation.

We calculated $\Delta\varepsilon$ by assuming that the $(i, \boldsymbol{k}) \to (i', \boldsymbol{k}')$ transitions are direct ($\boldsymbol{k} = \boldsymbol{k}'$) and values $d_{ii'}(\boldsymbol{k}, \boldsymbol{k}') = d$ and $\Gamma_{ii'}(\boldsymbol{k}, \boldsymbol{k}') = \Gamma = 5 \times 10^{12}$ s^{-1} are independent of i, i' and $\boldsymbol{k}$. The frequencies $\Omega_{ii'}(\boldsymbol{k}, \boldsymbol{k}')$ were calculated by interpolating the same data on $E_i(\boldsymbol{k})$ for La_2CuO_4 at room temperature [36] taking into account the requirements to the symmetry and periodicity [37]. The sample cooling was simulated by the replacement $E_i(\boldsymbol{k}) \to E_0^F \pm \{[E_i(\boldsymbol{k}) - E_0^F]^2 + \Delta^2(T_0)\}^{1/2}$ for $E_i(\boldsymbol{k}) > E_0^F$ and $E_i(\boldsymbol{k}) < E_0^F$, respectively [35], which corresponds to the redistribution of the state density in the vicinity of the Fermi surface simulating the phase transition. The integration was performed by the singularity method [38] over zones lying in the energy range $|E_i \pm E_0^F| \leqslant 2.5$ eV. Unlike [33], the occupation numbers $n_i(\boldsymbol{k})$ in the excited state were assumed specified by the Fermi–Dirac distribution with thermodynamic parameters $\langle E_e^F \rangle_t$ and $\langle T_e \rangle_t$ averaged over the pulse duration $\tau_p = 20$ ps (Fig. 1), which simulated the situation with pump and probe pulses coincident in time.

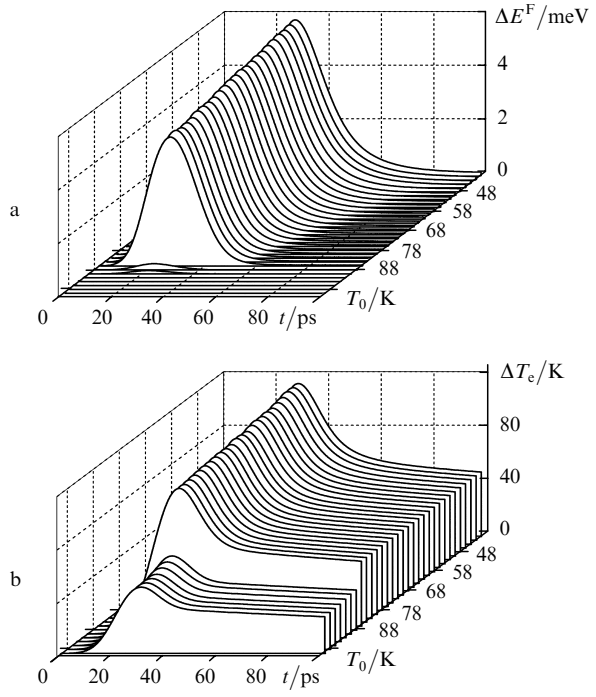

Figure 1. Transformation of the kinetics of the Fermi level instant shift $\Delta E^F(t) = E_e^F(t) - E_0^F$ (a) and the electron temperature $\Delta T_e(t) = T_e(t) - T_0$ (b) induced by the 20-ps pump pulse with changing the initial temperature T_0 of the HTSC sample.

Figure 2 illustrates a drastic jump in the calculated amplitude of variations induced by a pump pulse in the modulus of permittivity $\delta\varepsilon = |\varepsilon(E_e^F; T_e)| - |\varepsilon(E_0^F; T_0)|$ of the HTSC film upon variations of the initial temperature in the vicinity of the phase transition point $T_0 \sim T_c$ and probing of the film at wavelengths of 800 and 600 nm. Note that, as in [33], the signs of $\delta\varepsilon$ in these two situations are different. This means that in the picosecond pump-probe spectroscopy there also exist points on the wavelength axis in the vicinity of which $\Delta\varepsilon \equiv 0$, and it is these points that separate spectral regions with different signs of variations of $\Delta\varepsilon$ induced by the pump pulse. Note also that the anomalous temperature dependence of the nonlinear response amplitude disappears when the pump pulse energy is reduced (i.e., when the Fermi levels for free electrons and holes $E_{e,h}^F$ coincide with the real Fermi level E_0^F).

4. Conclusions

Thus, the spectral-temporal features of the kinetics of nonlinear response of HTSC films observed by the pump-probe method [6–10, 15, 18–22, 31, 32, 39, 40] at high excitation levels and interpreted in [33] have a direct analogue also in the case when the excited state of the HTSC film is excited and probed by substantially longer picosecond pulses coincident in time. In this case, due to degeneracy, the nontrivial (with a sharp jump at the vicinity of the point $T_0 \simeq T_c$) dependence of the nonlinear response amplitude of the HTSC sample on its initial temperature should be observed. In our opinion, this substantially extends the scope of experiments capable of confirming or disproving the correctness of the kinetic model based on the

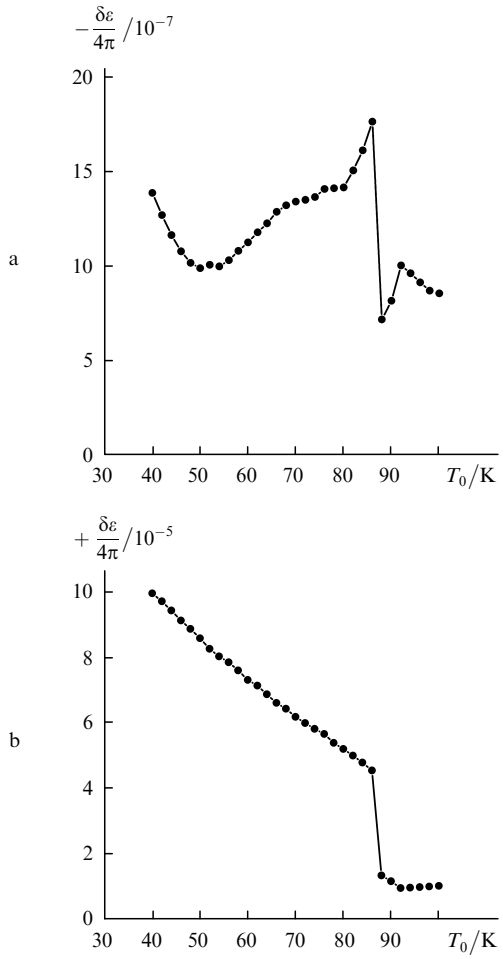

Figure 2. Dependences of the change in the modulus of permittivity $\delta\varepsilon$ of the HTSC sample on its initial temperature T_0 upon excitation by the 20-ps pulse and probing at wavelengths 800 (a) and 600 nm (b).

consideration of the contribution from interband electronic transitions to the nonlinear response of the excited HTSC film with the 'frozen' (metastable [33–35, 41]) energy gap.

Note also that, taking into account the possibility of interference of several contributions to the nonlinear response of the HTSC, the nontrivial temperature dependence of the amplitude of one of them (the resonance part of the response) should considerably affect the data obtained by other methods of picosecond coherent nonlinear spectroscopy of HTSCs, for example, by the biharmonic pump method [42].

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