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Degenerate four-photon probing of the kinetics of a nonlinear response of high-temperature superconductors in pump – probe spectroscopy

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***Abstract*. It is shown that the spectral – temporal and temperature 'anomalies' of the kinetics of reflection and transmission coefficients of high-temperatures superconductors (HTSCs) at high 'impact' excitation levels have direct analogies also in the case when the instant state of samples is probed by using the self-diffraction of probe pulses made coincident in time and propagating at an angle to each other. The possibility of using the transient modification of the method of degenerate four-photon spectroscopy for the HTSC diagnostics substantially expands the scope of experiments that can confirm or refute the correctness of the model based on the consideration of the contribution of interband electronic transitions to the response of an excited HTSC film with the 'frozen' energy gap.**

Keywords: *pump – probe spectroscopy, high-temperature superconductors, degenerate four-photon probing, nonlinear response kinetics.*

1. Introduction

Rapid relaxation processes in low- [1 – 5] and high-temperature [6 – 22] superconductors (LTSCs and HTSCs) are usually studied by the pump – probe method. In this case, the kinetics of the reflection and (or) transmission coefficients of a sample is studied with increasing the time delay τ between the 'impact' pump and probe pulses [6, 8 – 10]. It was assumed until recently that the scheme of processes proceeding in this case is simple. In the first stage, hot carriers are rapidly ($\tau_{th} < 10$ fs for the electron energy $E_e \sim 1$ eV [23, 24]) thermalised due to electron – electron (e – e) scattering and their energy distribution rapidly 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]. And already in the second stage, $T_e \to T_p$ for the time τ_r due to electron – phonon (e – p) scattering. It was established [2 – 4, 7, 18, 26 – 29] that τ_{th} and τ_r in LTSCs and HTSCs drastically increase near the superconducting transition point ($T_0 \simeq T_c$). This was also predicted theoretically [1, 30 – 32] because the formation of the energy gap in the electronic spectrum imposes restrictions on the phase space of scattering events. However, it was shown later [22] that, although these features of the behaviour of τ_r in HTSCs at $T_0 \simeq T_c$ do take place, they are observed only at high excitation levels and become more distinct with increasing the pump pulse energy.

The kinetics of the nonlinear response of a HTSC was interpreted in [33] within the framework of a model based on the four assumptions: (i) the energy gap in the electronic spectrum of cuprates cannot be rapidly destroyed even when T_e substantially differs from T_p [34]; (ii) after 'impact' excitation the positions of the Fermi levels $E_{e,h}^F$ for free electrons (states above the energy gap) and holes (states below the energy gap) in a HTSC do not coincide, the difference $\Delta E^F = E_e^F - E_h^F$ drastically increasing with the pump level; (iii) the distribution of the density of states in a HTSC leads to a strong decrease in the rate of nonradiative three-particle recombination of free electrons and holes in the degeneracy regime; (iv) the nonlinear response of a HTSC in the pump – probe spectroscopy is caused by interband electronic transitions [35]. The authors of paper [33] showed that the appearance of the energy gap in the electronic spectrum (at the point $T_0 \simeq T_c$) for $\Delta E^F \neq 0$ substantially changes the kinetics $E_e^F(t)$, $T_e(t)$, and $T_p(t)$, which allows one to explain all the 'anomalies' of the nonlinear response of HTSCs observed earlier.

Below, we consider for the model kinetics $E_{e,h}^F(t)$ and $T_{e,h}(t)$ in the same real band structure (see below) described in [33] another version of the pump – probe spectroscopy – the transient modification of the method of degenerate four-photon spectroscopy (DFPS) [35]. This version uses the self-diffraction of two short probe pulses made coincident in time but propagating at an angle to each other for probing the instant state of a HTSC sample after its 'impact' excitation by the pump pulse [33, 36, 37].

2. Electronic part of the nonlinear response

Within the framework of our model [35, 37, 38], the structure of the electronic part χ of the cubic nonlinear susceptibility (nonlinear response) of a thin HTSC film is written in the form typical for nonlinear spectroscopy:

$$\chi \propto P_0(K_+P_+ + K_-P_-). \quad (1)$$

Here, P_0, $P_\pm$, and $K_\pm$ are the resonance factors describing the probabilities of one- and two-photon electronic transitions and depending on the frequency detuning of

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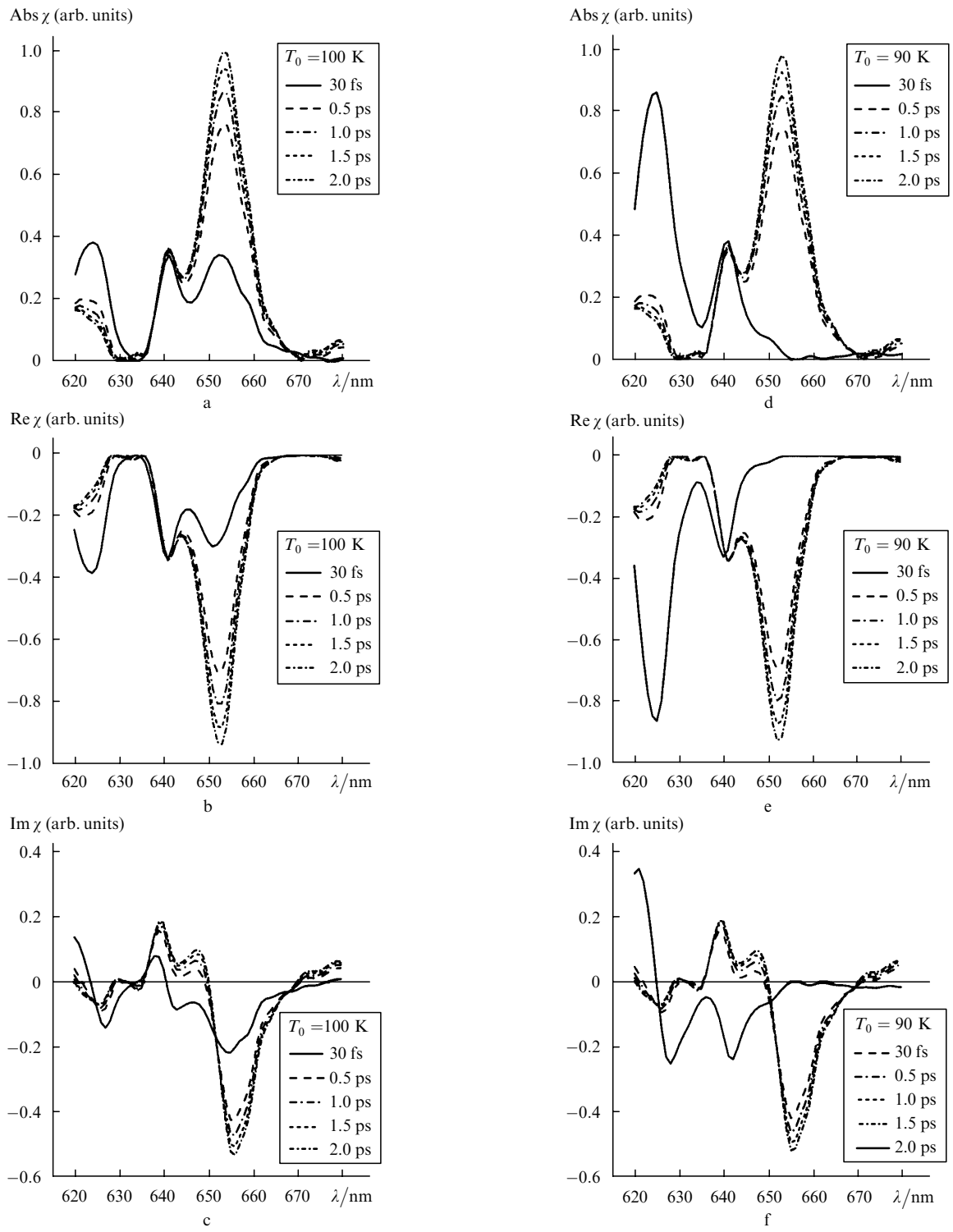


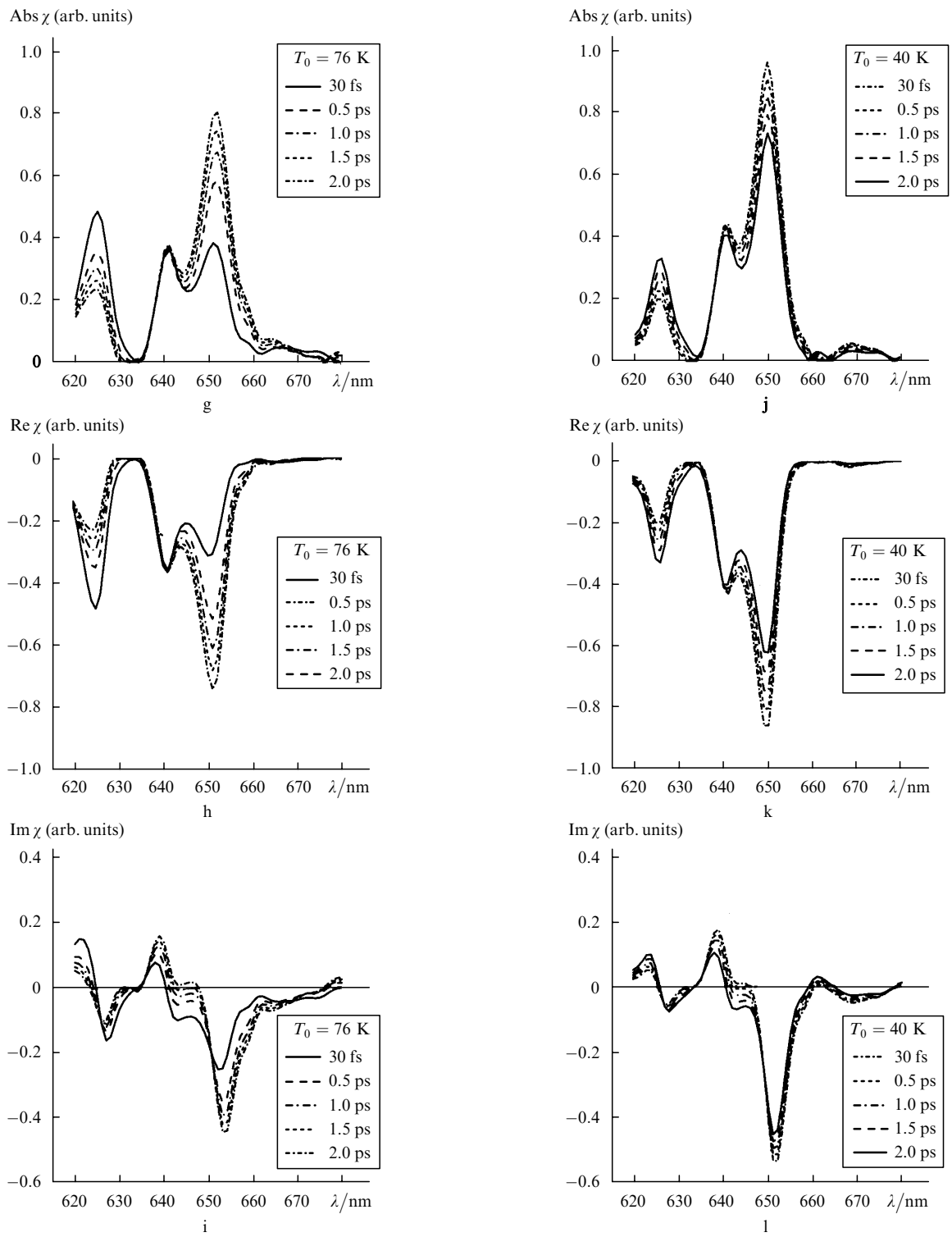
Figure 1. Dependences of the modulus Abs χ (a, d, g, j) of the real Re χ (b, e, h, k) and imaginary Im χ (c, f, i, l) parts of the response of a HTSC film →

light waves from resonances. In the case of coincident frequencies ω of the interacting waves, we obtain

$$P_+ = P_-^* = P_0^* = \sum_{i,i'} \int\int \frac{d_{i,i'}(\boldsymbol{k},\boldsymbol{k}')n_i(\boldsymbol{k})[1-n_{i'}(\boldsymbol{k}')]}{[\omega - \Omega_{i,i'}(\boldsymbol{k},\boldsymbol{k}') + \mathrm{i}\Gamma_{i,i'}(\boldsymbol{k},\boldsymbol{k}')]^2}\,\mathrm{d}\boldsymbol{k}\mathrm{d}\boldsymbol{k}', \quad (2)$$

$$K_+ = K_-^* = \sum_{i,i'} \int\int \frac{d_{i,i'}(\boldsymbol{k},\boldsymbol{k}')n_i(\boldsymbol{k})[1-n_{i'}(\boldsymbol{k}')]}{-\Omega_{i,i'}(\boldsymbol{k},\boldsymbol{k}') + \mathrm{i}\Gamma_{i,i'}(\boldsymbol{k},\boldsymbol{k}')}\,\mathrm{d}\boldsymbol{k}\mathrm{d}\boldsymbol{k}'. \quad (3)$$

Here, the subscripts i and i' numerate the bands involved in the $(i,\boldsymbol{k}) \to (i',\boldsymbol{k}')$ transitions with the dipole moments $d_{i,i'}(\boldsymbol{k},\boldsymbol{k}')$; $\Gamma_{i,i'}(\boldsymbol{k},\boldsymbol{k}')$ are the rates of intraband ($i = i'$) and interband ($i \neq i'$) relaxation; $\boldsymbol{k}$ is the wave vector of an electron; $n_i(\boldsymbol{k})$ is the occupation number of the $(i,\boldsymbol{k})$ state; $\Omega_{i,i'}(\boldsymbol{k},\boldsymbol{k}')$ are the resonance transition frequencies; and integration over $\boldsymbol{k}$ and $\boldsymbol{k}'$ is performed in the first Brillouin zone. Taking into account the smallness of the photon momentum, we will assume below that the $(i,\boldsymbol{k}) \to (i',\boldsymbol{k}')$ transitions are direct ($\boldsymbol{k} = \boldsymbol{k}'$) and will pass in (2) and (3) to



on the DFPS probe wavelength λ for different initial temperatures T_0 at different instants.

single integration by using the notation $d_{i,i'}(\boldsymbol{k},\boldsymbol{k}') = d_{i,i'}(\boldsymbol{k})$ and $\Gamma_{i,i'}(\boldsymbol{k},\boldsymbol{k}') = \Gamma_{i,i'}(\boldsymbol{k})$. We will describe the resonance frequencies by expressions $\Omega_{i,i'}(\boldsymbol{k},\boldsymbol{k}') = \Omega_{i,i'}(\boldsymbol{k}) = E_{i'}(\boldsymbol{k}) - E_i(\boldsymbol{k})$, where $E_i(\boldsymbol{k})$ is the electron energy in the $(i,\boldsymbol{k})$ state normalised to Planck's constant.

3. Numerical simulation

As in [35], the electronic part χ of a nonlinear response was simulated by calculating the frequencies $\Omega_{i,i'}(\boldsymbol{k})$ by the interpolation of the known data on the band structure of La_2CuO_4 [39] to the first Brillouin zone taking into account the symmetry and periodicity requirements. The simulation of the phase transition with changing T_0 was reduced to the forced redistribution of the density of electronic states in the calculated electronic spectrum by making the substitution $E_e(\boldsymbol{k}) \to E_0^F \pm \{[E_e(\boldsymbol{k}) - E_0^F]^2 + \Delta^2\}^{1/2}$ for the states with the energy $E_e(\boldsymbol{k}) > E_0^F$ (free electrons) and $E_e(\boldsymbol{k}) < E_0^F$ (holes), respectively, where E_0^F is the unperturbed position of the Fermi level. The energy gap Δ was assumed a

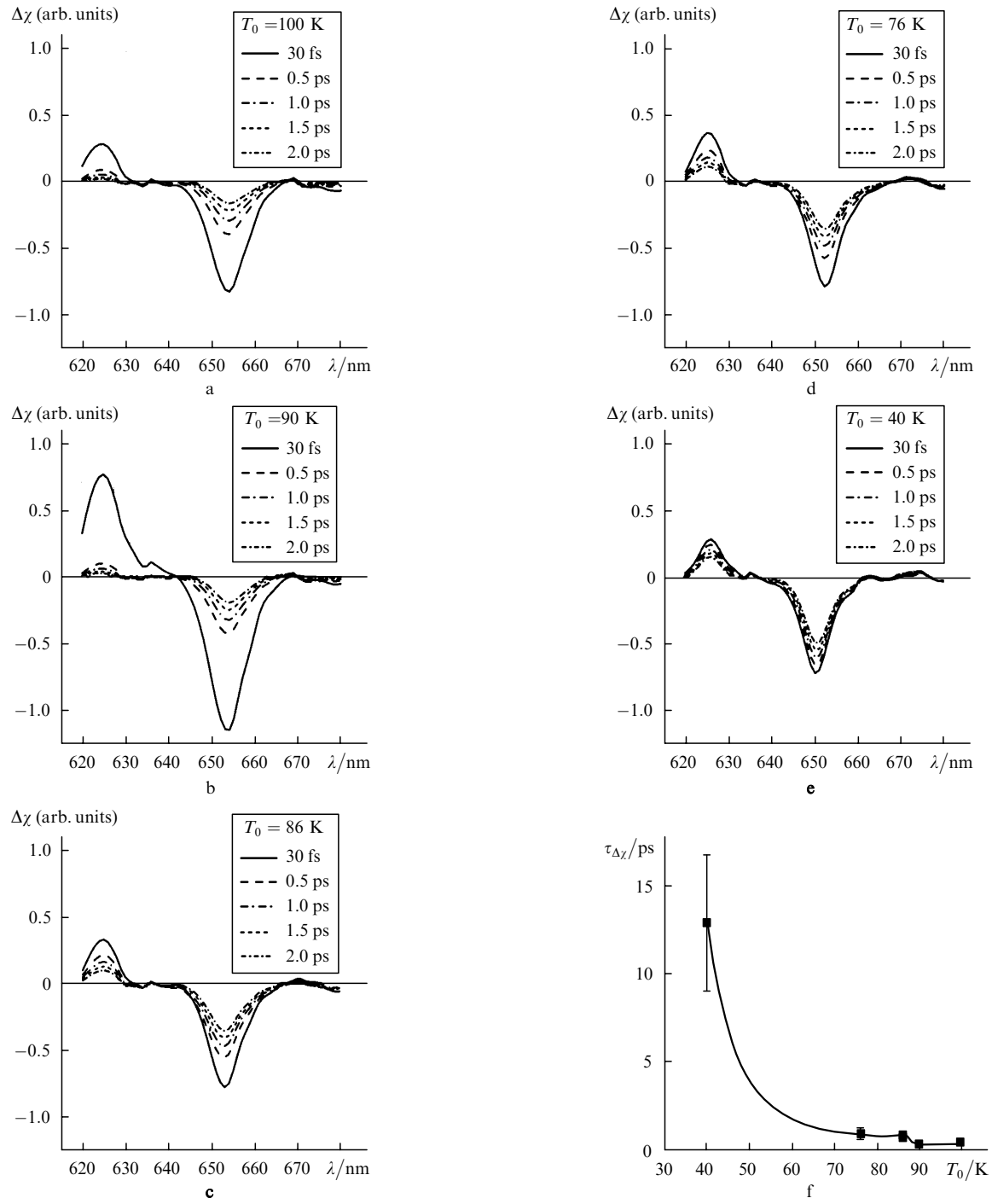


Figure 2. Wavelength dependences of the difference $\Delta\chi(\lambda)$ of responses of a HTSC film in the presence and absence of the pump pulse for different T_0 at different instants (a–e) and the dependence $\tau_{\Delta\chi}(T_0)$ of the relaxation time of variations in χ induced by a pump pulse at $\lambda \approx 653$ nm (f).

constant depending only on T_0 and T_c (the 'frozen' gap of the s symmetry in the weak coupling approximation in the BCS theory [40]):

$$\Delta = \Delta(T_0) \equiv \begin{cases} 3.12 k_B T_c (1 - T_0/T_c)^{1/2} \text{ for } T_0 \leqslant T_c, \\ 0 \text{ for } T_0 > T_c. \end{cases} \quad (4)$$

Here, k_B is the Boltzmann constant.

The calculations were performed by using simplifications similar to those presented in [33]. We assumed that $d_{i,i'}(\boldsymbol{k}) = d = \text{const}$, $\Gamma_{i,i'}(\boldsymbol{k}) = \Gamma = 5 \times 10^{12}$ s^{-1} and are independent of i, i', and $\boldsymbol{k}$. It was also assumed that a $\sim$ 200-nm thick HTSC film absorbed 30 % of the total energy (4×10^{-7} J) of a 30-fs, 800-nm pump pulse focused into a spot of diameter 150 μm. The values of other parameters ($T_c = 90$ K, the specific heat of the film $c_p = 0.9$ J cm^{-3} K^{-1}, the rate of the heat excess relaxation to a substrate, etc.) corresponded to a $YBa_2Cu_3O_{7-\delta}$ film on a strontium titanate substrate. In this connection the calculation of the relaxation kinetics of the nonlinear response $\chi(t)$ was reduced to a simple substitution into (2) and (3) of the instant values of the occupation numbers $n_i(\boldsymbol{k}, t)$ and $[1 - n_i(\boldsymbol{k}, t)]$ of the electron and hole states [for $E_e(\boldsymbol{k}) > E_0^F$ and $E_e(\boldsymbol{k}) < E_0^F$, respectively] specified by the Fermi–Dirac distribution $f_F(E_{e,h}; E_{e,h}^F, T_{e,h})$

with the instant values of the thermodynamic parameters $E_e^F(t) - E_0^F = E_0^F - E_h^F(t)$ and $T_e(t) = T_h(t)$ that we calculated [33] for different initial temperatures T_0 (see Fig. 2 in [33]). Integration in (2) and (3) was performed over all the electronic bands lying in the range $|E_e \pm E_0^F| \leqslant 2.5$ eV by the singularity method [41].

4. Kinetics of the electronic part of a nonlinear response

Figure 1 illustrates the drastic transformation of the dependences of the modulus $\mathrm{Abs}\,\chi$ of the real ($\mathrm{Re}\,\chi$) and imaginary ($\mathrm{Im}\,\chi$) parts of the electronic nonlinear response of a HTSC film on the DFPS wavelength λ in the range 620–680 nm at different initial temperatures T_0 (both above and below the phase transition point $T_0 = T_c$) immediately after the end of the 30-fs pump pulse and at instants $t = 0.5$, 1.0, 1.5, and 2.0 ps. In real experiments, differential measurements are used most often, i.e., the responses of the same sample are compared in the presence (χ) or absence (χ_0 for $E_{e,h}^F \equiv E_0^F$, $T_{e,h} \equiv T_0$) of the pump pulse. Figures 2a–e show the dependences of the difference $\Delta\chi = \mathrm{Abs}\,\chi - \mathrm{Abs}\,\chi_0$ of the results of such two measurements on the wavelength λ for different T_0 and several time delays. It is easy to see that, as in all experiments on diagnostics of variations in the reflection and (or) transmission coefficients [8, 10, 32], upon DFPS probing there also exists singularities ($\Delta\chi \equiv 0$) on the wavelength axis, which separate regions with the opposites signs of $\Delta\chi$ (Figs 2a–e). Note that, because both the real and imaginary parts of $\Delta\chi$ vanish at these points, their existence, in our opinion, can be caused only by a change of the phase relations between the two interfering components of χ, which appear due to the frequency degeneracy of the probe process [35].

The exponential approximation of the decay kinetics of variations in χ induced by the pump pulse in the 653-nm region of the long-wavelength peak of $\Delta\chi$ gives a rather nontrivial dependence of the relaxation time $\tau_{\Delta\chi}$ on T_0 (Fig. 2f) with a distinct sharp step (a jump of $\tau_{\Delta\chi}$ in the vicinity of the point $T_0 = 86$ K, i.e., slightly below the phase transition temperature $T_c \simeq 90$ K) and the subsequent sharp decrease in the relaxation rate at low temperatures. The transformation of $\tau_{\Delta\chi}$ with lowering T_0 is completely similar to that observed experimentally and to the above-discussed 'anomalies' of the nonlinear response kinetics [20, 22, 29, 31, 42, 43]. Note that within the framework of our model, all 'anomalies' of the nonlinear response completely vanish with decreasing the pump pulse energy (i.e., when the positions of Fermi levels $E_{e,h}^F$ for free electrons and holes coincide with its real position E_0^F).

5. Conclusions

We have shown that the spectral–temporal features of the kinetics of the nonlinear response of HTSC films, observed by the pump–probe method in [6–10, 15, 18–22, 29, 31, 32, 43] and interpreted in [33], have at high pump levels their analogues also in the case when the state of a sample is probed by using the self-diffraction of probe pulses made coincident in time and propagating at an angle to each other. The use of the transient modification of the DFPS method substantially expands, in our opinion, the scope of experiments that can confirm or refute the correctness of the model based on the consideration of the contribution of interband electronic transitions to the response of the excited HTSC film with the 'frozen' (metastable [33–35]) energy gap. It is important that this could confirm the fact that all the 'anomalies' of this type appear due to a drastic decrease in the rates of nonradiative recombination of excess free carriers under the conditions of strong degeneracy caused by the unusual distribution of the density of electronic states in a HTSC (compared to the electronic spectrum of narrow-gap semiconductors).

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References

1. Rothwarf A., Taylor B.N. *Phys. Rev. Lett.*, **19**, 27 (1967).
2. Owen C.S., Scalapino D.J. *Phys. Rev. Lett.*, **28**, 1559 (1972).
3. Parker W.H., Williams W.D. *Phys. Rev. Lett.*, **29**, 924 (1972).
4. Schuller I., Gray K.E. *Phys. Rev. Lett.*, **36**, 429 (1976).
5. Bluzer N. *J. Appl. Phys.*, **71**, 1336 (1992).
6. Brorson S.D. et al. *Phys. Rev. Lett.*, **64**, 2172 (1990).
7. Han S.G. et al. *Phys. Rev. Lett.*, **65**, 2708 (1990).
8. Gershenzon M.E. et al. *Pis'ma Zh. Eksp. Teor. Fiz.*, **52**, 1189 (1990).
9. Kazeroonian A.S. et al. *Solid State Commun.*, **78**, 95 (1991).
10. Chekalin S.V. et al. *Phys. Rev. Lett.*, **67**, 3860 (1991).
11. Bluzer N. *Phys. Rev. B*, **44**, 10222 (1991).
12. Hegman F.A. et al. *Appl. Phys. Lett.*, **62**, 1158 (1993).
13. Zherikhin A.N. et al. *Physica C*, **221**, 311 (1994).
14. Zherikhin A.N. et al. *Kvantovaya Elektron.*, **21**, 574 (1994) [*Quantum. Electron.*, **24**, 529 (1994)].
15. Brorson S.D. et al. *Phys. Rev. B*, **49**, 6185 (1994).
16. Buhleier R. et al. *Phys. Rev. B*, **50**, 9672 (1994).
17. White J.O. et al. *Physica C*, **235-240**, 2025 (1994).
18. Stevens C.J. et al. *Phys. Rev. Lett.*, **78**, 2212 (1997).
19. Smith D.C. et al. *J. Low Temp. Phys.*, **117**, 1059 (1999).
20. Demsar J. et al. *Phys. Rev. B*, **63**, 054519 (2001).
21. Segre G.P. et al. *Phys. Rev. Lett.*, **88**, 137001 (2002).
22. Schneider M.L. et al. *Eur. Phys. J. B*, **36**, 327 (2003).
23. Allen P.B. *Phys. Rev. Lett.*, **59**, 1460 (1987).
24. Nessler W. et al. *Phys. Rev. Lett.*, **81**, 4480 (1998).
25. Sun C.K. et al. *Phys. Rev. B*, **50**, 15337 (1994).
26. Lucas G., Stephen M.J. *Phys. Rev.*, **154**, 349 (1967).
27. Woo J.W.F., Abrahams E. *Phys. Rev.*, **169**, 407 (1968).
28. Schmid A., Schoen G. *J. Low Temp. Phys.*, **20**, 207 (1975).
29. Demsar J. et al. *Phys. Rev. Lett.*, **82**, 4918 (1999).
30. Tinkham M., Clarke J. *Phys. Rev. Lett.*, **28**, 1366 (1972).
31. Kabanov V.V. et al. *Phys. Rev. B*, **61**, 1477 (2000).
32. Farztdinov V.M. et al. *Brazilian J. Phys.*, **26**, 482 (1996).
33. Bobyrev Yu.V. et al. *Kvantovaya Elektron.*, **35**, 720 (2005) [*Quantum. Electron.*, **35**, 720 (2005)].
34. Voronov A.V. et al. *Zh. Eksp. Teor. Fiz.*, **120**, 1256 (2001).
35. Bobyrev Yu.V. et al. *Kvantovaya Elektron.*, **35**, 102 (2005) [*Quantum. Electron.*, **35**, 102 (2005)].
36. Kuznetsova L.P. et al. *Kvantovaya Elektron.*, **30**, 175 (2000) [*Quantum. Electron.*, **30**, 175 (2000)].
37. Bobyrev Yu.V. et al. *Kvantovaya Elektron.*, **32**, 789 (2002) [*Quantum. Electron.*, **32**, 789 (2002)].
38. Kornienko A.G. et al. *J. Appl. Phys.*, **80**, 2396 (1996); Petnikova V.M. et al. *Kvantovaya Elektron.*, **28**, 69 (1999) [*Quantum. Electron.*, **29**, 626 (1999)].
39. Perry J.K. et al. *Phys. Rev. B*, **63**, 144501 (2001).
40. Lifshits E.M., Pitaevskii L.P. *Statistical Physics, Part 2. Theory of Condensed State* (Oxford: Butterworth-Heinemann, 1991; Moscow: Nauka, 1978).
41. Chadi D.J., Cohen M.L. *Phys. Rev. B*, **8**, 5747 (1973).
42. Demsar J. *cond-mat/0305597* (2003).
43. Schneider M.L. et al. *Europhys. Lett.*, **60**, 460 (2002).