

Spectral features of the nonlinear response of high-temperature superconductor films in methods of degenerate four-photon spectroscopy

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Abstract. The electronic nonlinear response $\chi_{ee}^{(3)}$ of high-temperature superconductor films in methods of degenerate four-photon spectroscopy is calculated. It is shown that the model considering interband electronic transitions in a ‘real’ band structure with the only fitting parameter (the electron–phonon relaxation rate) well describes all the experimental spectral features of the response, and the methods of degenerate four-photon spectroscopy themselves can reveal, due to the interference character of $\chi_{ee}^{(3)}$, the presence of the energy gap in the spectrum of states.

Keywords: degenerate four-photon spectroscopy, ultrathin HTSC films, electronic nonlinear susceptibility, spectral features of a nonlinear response.

1. Introduction

The ultrafast relaxation kinetics of electronic excitations in metals and high-temperature superconductors (HTSCs) is commonly studied by a standard probe pulse (PP) method [1–17]. In this method, variations in the reflection [$\Delta R(\tau)$] and (or) transmission [$\Delta T(\tau)$] coefficients of a thin metal or HTSC film caused by its ‘impact’ excitation (due to absorption of an ultrashort pump pulse of duration τ_p) are probed with the help of another, substantially less powerful pulse of the same short duration τ_p , which is delayed by the time τ with respect to the pump pulse. In this case, probing is commonly performed at the excitation wavelength λ_p [1–16]. However, in some papers [17–20] the variations $\Delta R(\tau, \lambda)$ and $\Delta T(\tau, \lambda)$ have been measured in a rather broad spectral range $\lambda \neq \lambda_p$. In this case, for $\lambda_p = \text{const}$, either the wavelength of a narrowband PP can be continuously changed or probing can be performed by using a broadband PP and a monochromator. Then, by employing the well-developed procedures (see, for example, [17–20]), one can obtain the experimental information on the kinetics of PP-induced variations $\Delta \epsilon(\tau, \lambda)$ in the complex dielectric constant.

In any case, the temporal evolution of nonequilibrium

states is most often described with the help of classical thermodynamic parameters – temperatures T_e and T_{ph} of the electron and phonon subsystems, the Fermi-level position E_F , etc., whose instantaneous values (for instants $t = \tau$) are also calculated from $\Delta \epsilon(\tau, \lambda)$ using theoretical models [17–23]. Note, however, that the validity of such an adiabatic approach in some sense, in which the electron subsystem is assumed thermalised at each instant, is disputed [24].

In the method of degenerate four-photon spectroscopy (DFPS), the nonequilibrium spatially inhomogeneous one-dimensional distributions $\Delta \epsilon$, the so-called dynamic gratings [25–28] are probed simultaneously with excitation ($\tau = 0$). Such gratings are written in a sample due to the interference of two comparatively long (usually picosecond) pulses at the same wavelength λ , propagating at an angle to each other. In the experiment, the spectral dependence of the diffraction efficiency $\eta(\lambda)$ is measured for one of the pulses (self-diffraction process) on the $\Delta \epsilon$ grating. Note that the DFPS method was developed to study the resonance mechanisms of nonlinearity, i.e., for measuring the frequencies and spectral widths of well-localised resonance lines in a nonlinear response and also the ratio of their amplitudes, and was not initially intended for the investigation of the kinetics of nonequilibrium states [29]. However, the DFPS probing with preliminary excitation allows one to perform such studies as well.

Already the first PP experiments with metal (Cu) and HTSC (YBaCuO) films revealed distinct spectral features in the dependences $\Delta \epsilon(\lambda)$ [17, 19, 20]. It was shown that there exist points at the λ axis in the vicinity of which $\Delta \epsilon$ is zero and is independent of τ . It was assumed that interband electronic transitions play a key role in the formation of the nonlinear response. The authors of paper [30], which studied the Al and W films by using other modifications of the PP method, made a similar conclusion. A drastic decrease in the amplitude η of the nonlinear response of metal films (Ni, Au, and Pt) in the vicinity of certain points at the λ axis was also observed by the DFPS method [25–28]. It was shown [28] that the model [31], taking into account the saturation of interband electronic transitions in the ‘real’ (see below) band structure of the sample, well describes the experimental dependence $\eta(\lambda)$ using a minimal number of fitting parameters.

In this paper, we show that a similar model [28, 31] can also explain the experimental spectral features of the nonlinear response of HTSC films. The model predicts that due to the interference of the two components of the electronic part $\chi_{ee}^{(3)}$ of the nonlinear susceptibility, there exist points at

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the λ axis in the vicinity of which both the real $\text{Re}\chi_{\text{ee}}^{(3)}$ and imaginary $\text{Im}\chi_{\text{ee}}^{(3)}$ parts of $\chi_{\text{ee}}^{(3)}$ vanish simultaneously. Taking into account the possibility of existence of long-lived (with the lifetime up to 1–3 ns) metastable states of HTSCs with the ‘frozen’ energy gap (‘pseudo-gap’) in the electronic spectrum, predicted in [32], this completely explains the results of PP experiments [17, 19, 20] with impact pulsed excitation.

2. Electronic part of the nonlinear response

Within the framework of our model [28, 31], the electronic part $\chi_{\text{ee}}^{(3)}$ of the nonlinear susceptibility (nonlinear response) of a thin film is written in the form

$$\chi_{\text{ee}}^{(3)} \propto P_0(K_+P_+ + K_-P_-), \quad (1)$$

which is typical for nonlinear spectroscopy. Here, P_0 , P_{\pm} , and K_{\pm} are the resonance factors describing the probabilities of all possible one- and two-photon electronic transitions and depending on the frequency detuning of light wave from resonances. It follows from the expressions for P_0 , P_{\pm} , and K_{\pm} presented in [28, 31] that, when the frequencies ω of all the interacting waves coincide, we have

$$P_+ = P_-^* = P_0^*$$

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

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

Here, subscripts i and i' number the electronic bands involved in the $(i, \mathbf{k}) \rightarrow (i', \mathbf{k}')$ electronic transition with the dipole moment $d_{i,i'}(\mathbf{k}, \mathbf{k}')$; $\Gamma_{i,i'}(\mathbf{k}, \mathbf{k}')$ are the intraband ($i = i'$) and interband ($i \neq i'$) relaxation rates; \mathbf{k} is the electron wave vector; $n_i(\mathbf{k})$ is the occupation number of the (i, \mathbf{k}) electronic state, which is determined in the case of the thermodynamic equilibrium by the Fermi–Dirac distribution; and $\Omega_{i,i'}(\mathbf{k}, \mathbf{k}')$ is the resonance transition frequency. Integration with respect to \mathbf{k} and \mathbf{k}' in (2) and (3) is performed within the first Brillouin zone, and summation is carried out over all the electronic states i and i' . Taking into account the smallness of the photon momentum, we assume that the $(i, \mathbf{k}) \rightarrow (i', \mathbf{k}')$ electronic transitions are ‘direct’ ($\mathbf{k} = \mathbf{k}'$) and will pass in (2) and (3) to single integrals over \mathbf{k} by using the notation $d_{i,i'}(\mathbf{k}, \mathbf{k}') = d_{i,i'}(\mathbf{k})$ and $\Gamma_{i,i'}(\mathbf{k}, \mathbf{k}') = \Gamma_{i,i'}(\mathbf{k})$. We will describe the frequencies of resonance transitions by standard expressions $\Omega_{i,i'}(\mathbf{k}, \mathbf{k}') = \Omega_{i,i'}(\mathbf{k}) = E_{i'}(\mathbf{k}) - E_i(\mathbf{k})$, where $E_i(\mathbf{k})$ is the electron energy in the (i, \mathbf{k}) state normalised to Planck’s constant.

As in [28, 31], we will determine $\Omega_{i,i'}(\mathbf{k})$ by interpolating the known data on the band structure of the sample to the entire Brillouin zone [33]. This allows us to include the ‘real’ (known from the literature) electronic spectrum of the sample to the model (1)–(3), thereby drastically reducing the number of fitting parameters. Below, these parameters will be the temperatures of the electron (T_e) and phonon (T_{ph}) subsystems and the characteristic relaxation time scale $\Gamma_{i,i'}^{-1}(\mathbf{k})$. However, even without any calculations, we can immediately make a very important conclusion, which follows directly from the type of expressions (1)–(3).

Indeed, by introducing the notation

$$P = P_+ = |P| \exp(i\varphi_P), \quad K = K_+ = |K| \exp(i\varphi_K), \quad (4)$$

we can rewrite (1) in the form

$$\chi_{\text{ee}}^{(3)} \propto |P|^2 |K| \cos(\varphi_P + \varphi_K) \exp(i\varphi_P). \quad (5)$$

It follows from (5) that in the degenerate regime, the electronic nonlinear response vanishes not only in the cases when $|P| = 0$ or $|K| = 0$ (in our opinion, this is unlikely because both the real and imaginary parts of the corresponding integrals should simultaneously vanish), but also when $\varphi_P + \varphi_K = (2k+1)\pi/2$, where $k = 0, \pm 1, \pm 2, \dots$. It is very important that this conclusion is of the most general type and is valid for any dependences $d_{i,i'}(\mathbf{k})$, $n_i(\mathbf{k})$, $E_i(\mathbf{k})$, and $\Gamma_{i,i'}(\mathbf{k})$, and requires no specification of these dependences. It is for this reason that we believe that this effect should explain the results of experiments [17, 19, 20], in which the simultaneous vanishing of the real $\text{Re}\chi_{\text{ee}}^{(3)}$ and imaginary $\text{Im}\chi_{\text{ee}}^{(3)}$ parts of the nonlinear response was observed in the vicinities of certain points on the λ axis (more exactly, their reducing below the noise level).

3. Numerical simulation

We performed calculations by using the following simplifications. As in [28, 31], we assumed that $d_{i,i'}(\mathbf{k}) = d = \text{const}$ and is independent of i , i' , and \mathbf{k} ; $\Gamma_{i,i'}(\mathbf{k}) = \Gamma(E_e) = \Gamma_{\text{ee}} + \Gamma_{\text{eph}}$, where $\Gamma_{\text{ee}} = 2\pi(E_e - E_F)^2/(\hbar E_F)$ and $\Gamma_{\text{eph}} = \text{const}$ are the rates of electron–electron and electron–phonon relaxation [34]. The local deviations $\delta n_i(\mathbf{k})$ of the occupation numbers from their equilibrium values $n_i(\mathbf{k}; E_e, T_e)$ determined by the Fermi–Dirac distribution with the electron temperature T_e were assumed small, and the maximum relative saturation level $\delta n_i(\mathbf{k})/n_i(\mathbf{k}; E_e, T_e)$ produced by pump pulses at the frequency ω was limited by the value 0.01. Corrections $\delta n_i(\mathbf{k})$ were calculated by employing the approach developed in [31], which is based on the use of the modified density matrix for an effective two-level system in the dipole approximation [35]. The resonance frequencies $\Omega_{i,i'}(\mathbf{k})$ were calculated by interpolating the known data on the band structure of La_2CuO_4 [36] to the entire Brillouin zone taking into account the corresponding requirements to the symmetry and periodicity. The subsequent integration was performed by the singularity method [37]. Both these procedures (interpolation and integration) were performed for zones covering the electronic energy range of ± 2.5 eV from the Fermi level. The energy gap in the electronic spectrum of the HTSC sample before excitation was assumed constant, depending only on its initial temperature T_0 and the phase transition temperature T_c (the gap with the so-called s -symmetry),

$$\Delta = \Delta(T_0) \equiv 3.12k_B T_c \left(1 - \frac{T_0}{T_c}\right)^{1/2} \quad \text{for } T_0 \leq T_c, \\ \Delta = \Delta(T_0) \equiv 0 \quad \text{for } T_0 > T_c. \quad (6)$$

Here, k_B is the Boltzmann constant. Expression (6) corresponds to the so-called weak-coupling approximation in the BCS theory [38]. The energy gap was introduced into the electronic spectrum (to simulate the phase transition) by

replacing $E_e(\mathbf{k})$ by $E_F \pm \{[E_e(\mathbf{k}) - E_F]^2 + \Delta^2\}^{1/2}$ for the states above [$E_e(\mathbf{k}) > E_F$] and below [$E_e(\mathbf{k}) < E_F$] of the Fermi level. This procedure was used to perform the required redistribution of the density of electronic states in the vicinity of the Fermi level of the interpolated band structure upon a change in the initial (before the action of the pump pulse) temperature T_0 of the sample.

The calculation simulated the following experimental situation. It was assumed that the electron subsystem of a thin HTSC film with the phase transition temperature $T_c = 92$ K, having the initial temperature $T_0 = 80, 100$ or 300 K (i.e., below or above T_c), is instantly ‘heated’ by the pump pulse up to temperature $T_e \sim 600$ K and then cools off. In this case, the temperature of the phonon subsystem T_{ph} does not change significantly due to its large heat capacity, and $T_{ph} \simeq T_0$. It was assumed that after excitation of a sample with the initial temperature $T_0 = 80$ K ($\Delta \neq 0$, ‘superconductor’), two substantially different situations are possible. In the first case, due to the instant heating (the pump pulse duration is τ_p) of the electron subsystem of the sample up to temperatures $T_e \gg T_c$, the phase transition occurs instantly, and the energy gap is absent in excited states ($\Delta = 0$, ‘metal’). In the second case, the energy gap in the spectrum of excited electronic states was assumed ‘frozen’, i.e., it was assumed that its width was determined by the temperature of the phonon subsystem $T_{ph} \simeq T_0$ and did not change (‘superconductor’). This situation corresponds to the existence of long-lived (the lifetime up to 1–3 ns) metastable states of a HTSC with the pseudo-gap in the electronic spectrum after its impact pulsed excitation, which was predicted in [32].

It follows from the above consideration that the only fitting parameter of the described model is the electron–phonon relaxation time Γ_{eph}^{-1} , whose value was selected so that the characteristic scale of spectral features in the dependences of the electronic nonlinear response $\eta \propto |\chi_{ee}^{(3)}|^2$ on λ would be in qualitative agreement with the experimental data [17].

As expected, our calculations confirmed that $|P(\lambda)|$ and $|K(\lambda)|$ monotonically change in the spectral range 625–635 nm of interest to us (see [17]) and never vanish in this range. At the same time, due to variations in φ_P and φ_K , there always exists a point in this spectral region at which $\cos(\varphi_P + \varphi_K) = 0$ and, according to (5), the HTSC sample gives no nonlinear response (Figs 1 and 2). The position of this spectral region on the λ axis depends on the initial temperature T_0 of the sample and changes comparatively weakly with changing the excitation level (upon variations of T_e), in accordance with the available experimental data [6–9]. We found that the dependences $\chi_{ee}^{(3)}(\lambda)$ for samples in the metal ($T_e = 100$ K) and superconducting ($T_e = 80$ K) phases were substantially different (Fig. 1). Moreover, this difference becomes larger when the electronic subsystems of samples are heated up to the same temperature, provided their phase state is remained unchanged (Fig. 2). As a result, the difference $\Delta\chi_{ee}^{(3)}(\lambda) = \chi_{ee}^{(3)}(\lambda; \Delta = 0) - \chi_{ee}^{(3)}(\lambda; \Delta \neq 0)$ of the nonlinear responses of the initially metal ($T_0 = 100$ K) and superconducting ($T_0 = 80$ K) HTSC samples after the action of the pump pulse heating their electronic subsystems up to the temperature $T_e = 300$ K $\neq T_0$, but not changing their phase state, proved to be also comparable with the value $\chi_{ee}^{(3)}(\lambda)$ (Fig. 3). Note that the differences

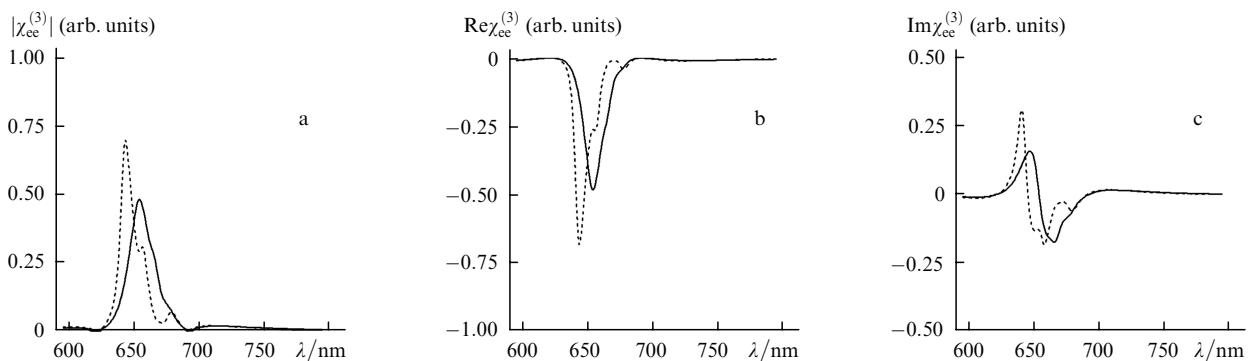


Figure 1. Dependences of the modulus (a), the real (b) and imaginary (c) parts of the nonlinear response $\chi_{ee}^{(3)}$ of the HTSC film on the wavelength λ for a ‘metal’ ($\Delta = 0$, $T_e = 100$ K, solid curve) and a ‘superconductor’ ($\Delta \neq 0$, $T_e = 80$ K, dashed curve).

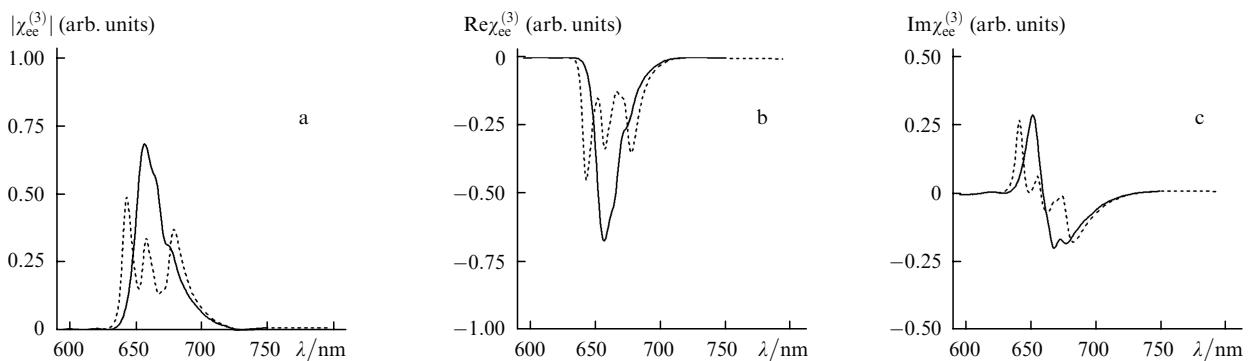


Figure 2. Same as in Fig. 1, after the heating of the electron subsystem of the HTSC film for a ‘metal’ ($\Delta = 0$, $T_e = 300$ K, solid curve) and a ‘superconductor’ ($\Delta \neq 0$, $T_e = 300$ K, dashed curve).

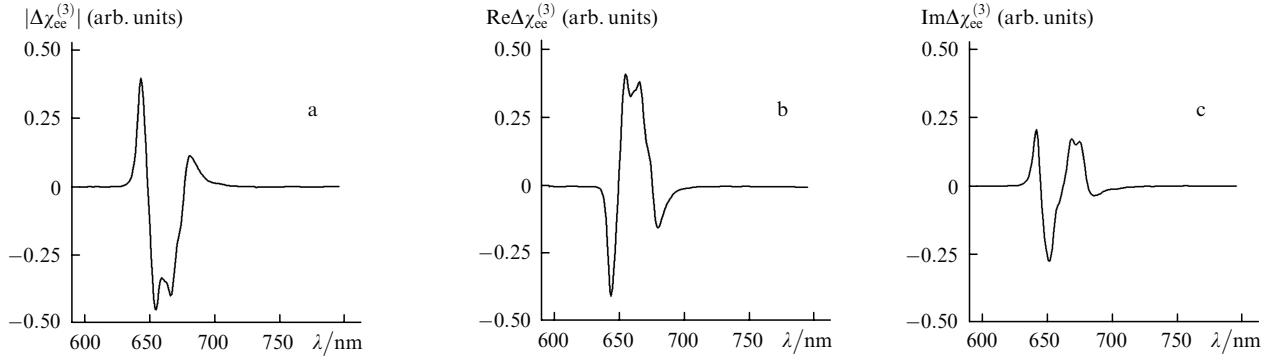


Figure 3. Dependences of the modulus (a), the real (b) and imaginary (c) parts of the difference of the nonlinear response $\Delta\chi_{ee}^{(3)}$ of HTSC films heated by the pump pulse in the ‘metal’ ($\Delta = 0$, $T_e = 300$ K) and ‘superconducting’ ($\Delta \neq 0$, $T_e = 300$ K) phases on the wavelength λ .

$\Delta\chi_{ee}^{(3)}(\lambda)$ of this type are often the main result of such PP experiments, which is typical for the so-called differential measurements.

The dependences $\Delta\chi_{ee}^{(3)}(\lambda) = \chi_{ee}^{(3)}(\lambda; T_e \neq T_0) - \chi_{ee}^{(3)}(\lambda; T_e = T_0)$ were different for all the three situations simulated. In the first case (Fig. 4), the HTSC sample irradiated by the pump pulse experienced the transition from the metal state ($\Delta = 0$) with the initial temperature $T_0 = 100$ K to the similar state ($\Delta = 0$) with the electron temperature $T_e = 600, 450, 300$ K. In the two other cases, the same sample experienced the transition from the superconducting state ($\Delta \neq 0$) with the initial temperature $T_0 = 80$ K either to the metal state ($\Delta = 0$) with $T_e = 600, 450, 300$ K (Fig. 5) or to

the metastable state with the ‘frozen’ pseudo-gap in the electronic spectrum [32] with the same T_e ($\Delta \neq 0$, Fig. 6). Note that, within the framework of our model, a change in the final electron temperature T_e of the sample in all the three above situations (Figs 4–6) can correspond to both a change in its excitation level (change in the pump pulse energy) and an increase in the delay τ of the probe pulse with respect to the pump pulse, thereby characterising the kinetics of the nonlinear response. However, in any case it is most important, in our opinion that despite an extremely narrow width Δ of the energy gap compared to the probe radiation frequency ω , all the three possible situations prove to be readily discernible in real experiments.

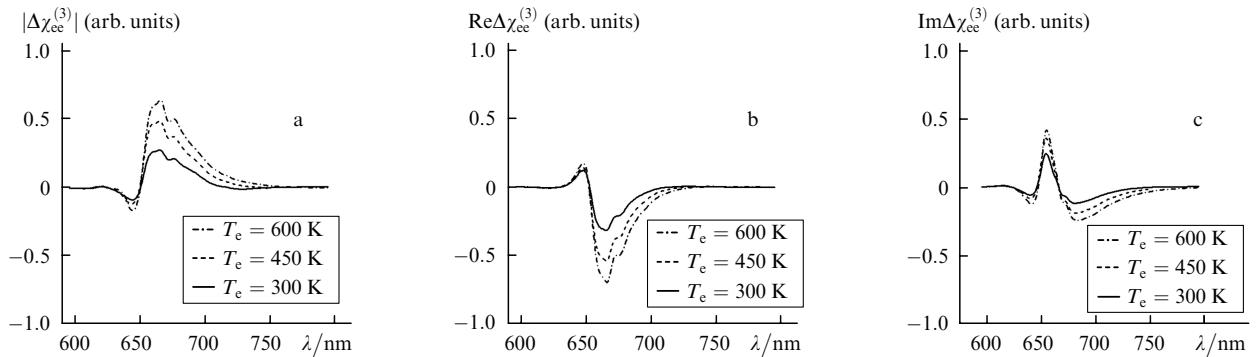


Figure 4. Dependences of the modulus (a), the real (b) and imaginary (c) parts of the change in the nonlinear response $\Delta\chi_{ee}^{(3)}$ of the HTSC film on the wavelength λ caused by the pump pulse for the ‘metal’ ($\Delta = 0$, $T_e = 100$ K) → ‘metal’ ($\Delta = 0$) transition for different T_e .

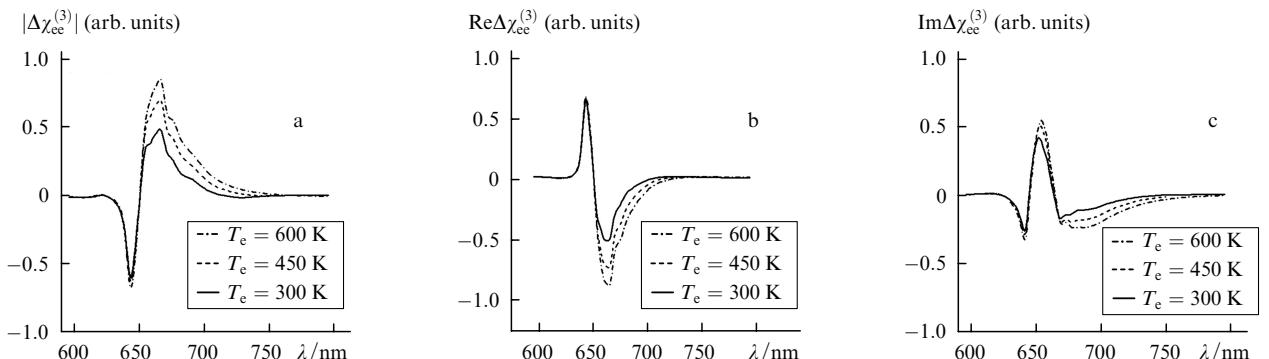


Figure 5. Same as in Fig. 4, for the ‘superconductor’ ($\Delta \neq 0$, $T_e = 80$ K) → ‘metal’ ($\Delta = 0$) transition for different T_e .

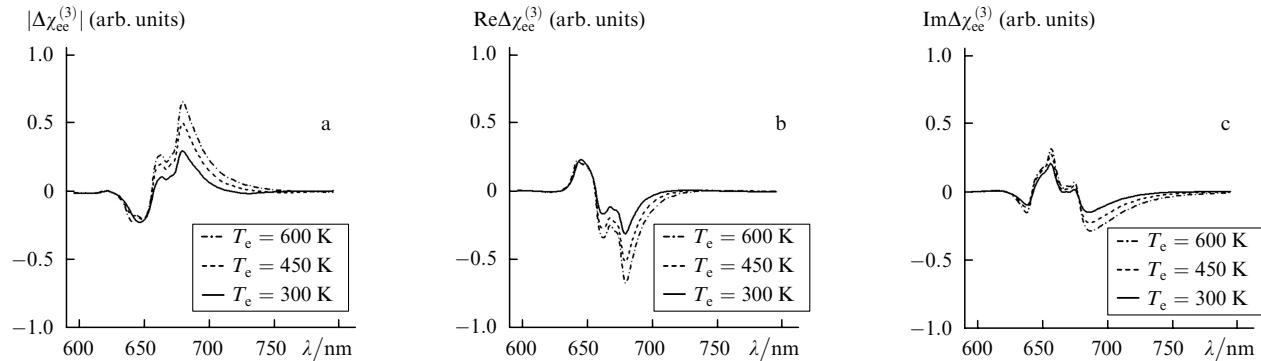


Figure 6. Same as in Fig. 4, for the ‘superconductor’ ($\Delta \neq 0$, $T_c = 80$ K) → ‘superconductor’ ($\Delta \neq 0$) transition for different T_e .

4. Conclusions

We have shown in this paper that the spectral features of the nonlinear response of thin HTSC films are most probably related to variations in phase relations between the two interfering components of the electronic nonlinear susceptibility $\chi_{ee}^{(3)}$. In turn, the presence of these two components in expression (1) for $\chi_{ee}^{(3)}$ is caused only by the existence of the commutation relations in the Liouville equation for the density matrix (see, for example, [29, 35]) and is inevitable in this sense within the framework of any theory. It is for this reason that the effect described in the paper is not related to the specific type of the dependences $d_{i,i'}(\mathbf{k})$, $n_i(\mathbf{k})$, $E_i(\mathbf{k})$, and $\Gamma_{ii'}(\mathbf{k})$, and it should be always observed in the frequency-degeneracy regime. Therefore, this mechanism should be manifested in all the PP experiments [1–17] on the measurement of variations in the reflection [$\Delta R(\tau, \lambda)$] and (or) transmission [$\Delta T(\tau, \lambda)$] coefficients of HTSC samples caused by their pumping. In this case, because of the virtually instant destruction of the coherence of excited electronic states, we study in fact the frequency-degenerate nonlinear response. In our opinion, this removes at least some arguments in favour of the models explaining the features of experimental dependences obtained for HTSCs by the ‘freezing’ of the intraband relaxation on the Fermi surface [12, 17, 20]. Our conclusion is indirectly confirmed by the recent experimental data [16] according to which the decrease in the excitation level (the pump pulse energy) by three-four orders of magnitude leads to the complete disappearance of any sharp features in the temperature dependence of the rate of intraband relaxation at the point $T_0 = T_c$ for a very broad series of cuprate HTSCs studied in [16].

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