PACS numbers: 42.62.Fi; 78.47.+p; 74.25.Gz; 78.20.Bh
DOI: 10.1070/QE2007v037n02ABEH013324

# Asymmetry of the Stokes and anti-Stokes components of the nonlinear response of high-temperature superconductors in the method of picosecond biharmonic pumping

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***Abstract*. It is shown in the model of the nonlinear response of high-temperature superconductors (HTSCs) caused by interband transitions in the electronic spectrum with a metastable energy gap that the Stokes and anti-Stokes components of the HTSC response to picosecond biharmonic pumping are asymmetric, and for the excitation-pulse frequency detuning exceeding 100 cm$^{-1}$, the efficiency of self-diffraction in the directions corresponding to these two components is different.**

***Keywords***: *nonlinear response of high-temperature superconductors, picosecond biharmonic pumping, efficiency of the Stokes and anti-Stokes components of self-diffraction.*

## 1. Introduction

In [1 – 5], the spectral, temporal, and temperature properties of the nonlinear response of high-temperature superconductors (HTSCs) observed at different excitation levels by different methods of femtosecond and picosecond spectroscopy [6 – 17] were interpreted within the framework of a model based on two assumptions. It was assumed that the energy gap in the electronic spectrum of a HTSC is metastable [18] and the electronic part of the nonlinear response is caused by interband transitions [12, 19 – 26]. It was shown, in particular, in [4, 5] that the characteristic dip in the dependence of the self-diffraction efficiency $\eta$ (generation of the field at frequency $2\omega_1 - \omega_2$) on the detuning $\Delta\omega = \omega_1 - \omega_2$ of the frequencies $\omega_{1,2}$ of components of picosecond biharmonic pumping (BP) (Fig. 1a), which was observed near the point $T_0 \simeq T_c$ (transition to the superconducting state) in [7, 9, 10], makes is possible to measure the dependence of the gap width $\varDelta$ on the initial temperature $T_0$ of a sample.

In the absence of the frequency degeneracy ($\Delta\omega \neq 0$, the BP method), the model proposed in [4, 5] admits the appearance of a certain asymmetry of the nonlinear response, i.e. the different dependences $\eta(\Delta\omega, T_0)$ for two different directions ($2\boldsymbol{k}_{1,2} - \boldsymbol{k}_{2,1}$) and frequencies ($2\omega_{1,2} - \omega_{2,1}$) of the self-diffraction signal (Fig. 1a), where $\boldsymbol{k}_{1,2}$ are the wave vectors of the BP components. The asymmetry appears because the electronic part of the dependence $\Delta\varepsilon(\omega)$ has spectral singularities [6, 12] caused by one-photon interband resonances [1 – 5]. Here, $\Delta\varepsilon(\omega) = \varepsilon(\omega; E_e^F, T_e) - \varepsilon(\omega; E_0^F, T_0)$ is the change in the dielectric constant $\varepsilon$ of a sample at frequency $\omega$ caused by the deviation of the Fermi level $E_e^F$ and the electron temperature $T_e$ from their equilibrium values $E_0^F$ and $T_0$, respectively. This is illustrated qualitatively in Fig. 1b, which shows the asymmetry of positions of the frequencies $\omega_{1,2}$ and $2\omega_{1,2} - \omega_{2,1}$ of interacting waves on the axis $\omega$ (for $\Delta\omega \neq 0$) with respect to the point $\omega_0 = \omega_1$ of the one-photon resonance of width $\varGamma_0$. Below, according the adopted terminology, we will call the

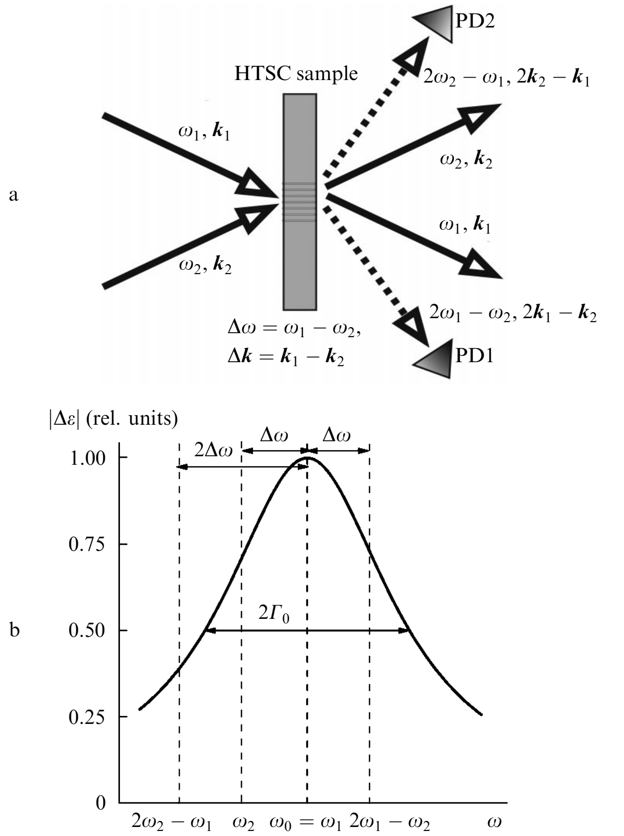


**Figure 1.** Interaction of pump components at frequencies $\omega_{1,2}$ with the wave vectors $\boldsymbol{k}_{1,2}$: self-diffraction signals at frequencies $2\omega_{1,2} - \omega_{2,1}$ detected with photodetectors PD1,2 in directions $2\boldsymbol{k}_{1,2} - \boldsymbol{k}_{2,1}$ (a) and shifts of frequencies $\omega_{1,2}$ and $2\omega_{1,2} - \omega_{2,1}$ with respect to the one-photon resonance singularity at $|\Delta\varepsilon|$ of width $\varGamma_0$ caused by the frequency detuning $\Delta\omega = \omega_1 - \omega_2$ (b).

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Received 5 July 2006
*Kvantovaya Elektronika* **37** (2) 162 – 166 (2007)
Translated by M.N. Sapozhnikov

components of the nonlinear response at frequencies $2\omega_2 - \omega_1$ and $2\omega_1 - \omega_2$ the Stokes and ant-Stokes components (see Fig. 1b). Note that the asymmetry of these components was observed earlier in picosecond BP experiments with thin narrow-gap $PrBa_2Cu_3O_{7-\delta}$ semiconductor films [27].

By using the kinetics $E_e^F(t)$ and $T_e(t)$ calculated in [3, 5] and the model of the nonlinear HTSC response in the BP method described in [4, 5], we will show below that this asymmetry does exist when the pump pulses are made coincident in time $t$, but only in the region of large frequency detunings ($\Delta\omega > 100$ cm$^{-1}$).

## 2. Nonlinear response model

The self-diffraction process was described by using the model of the coherent four-photon response developed in [4, 5]. It is assumed that the total cubic nonlinear susceptibility of a HTSC sample includes several components [28]:

$$\chi = \chi_{nr} + \chi_r + \chi_s + \chi_0. \qquad (1)$$

Here, $\chi_{nr}$ and $\chi_r$ are the nonresonance and resonance (see [4, 5]) components of the electronic nonlinear response caused by direct and indirect interband transitions; $\chi_s$ is the component related to excitation of acoustic phonons; and $\chi_0$ is a constant caused by the errors of the model. The calculation of contributions of these components to $\chi$ for the anti-Stokes component of the response at frequency $2\omega_1 - \omega_2$ completely repeats procedures described in detail in [5], and therefore we will not consider it here.

The value of $\chi_{nr}$ for the Stokes component at frequency $2\omega_2 - \omega_1$ was calculated similarly; however, the replacement $\omega_1 \to \omega_2 = \omega_1 + \Delta\omega$ was made in the resonance denominators of $P_0$ and $P_\pm$ [in expressions (58) and (59) in [5]]. In this case, the Raman resonance frequency $\Delta\omega$ in denominators of $K_\pm$ [expression (60) in [5]] did not change, of course. The one-photon resonance frequencies in $P_0$ and $P_\pm$ were found by interpolating the same data [29] for the band structure [the electronic energy $E_i(\boldsymbol{k}_e)$ in the $(i, \boldsymbol{k}_e)$ state, where $i$ numerates the electronic bands and $\boldsymbol{k}_e$ is the quasi-momentum] of $La_2CuO_4$ at room temperature taking into account the symmetry and periodicity requirements [30]. The cooling of a HTSC sample was simulated by the same substitution $E_i(\boldsymbol{k}_e) \to E_0^F \pm \{[E_i(\boldsymbol{k}_e) - E_0^F]^2 + \Delta^2(T_0)\}^{1/2}$ for $E_i(\boldsymbol{k}_e) > E_0^F$ and $E_i(\boldsymbol{k}_e) < E_0^F$, respectively, describing the redistribution of the density of states near the Fermi surface upon the phase transition. The energy gap width

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

was assumed a constant depending only on $T_0$ and $T_c$ ('frozen' [18] gap of the s symmetry in the weak coupling approximation of the BCS theory [31]). Here, $k_B$ is the Boltzmann constant. The bands lying in the range $|E_i(\boldsymbol{k}_e) \pm E_0^F| \leqslant 2.5$ eV were taken into account. As in [4, 5], $\chi_r$ was calculated by using the model of the effective two-level system, and $\chi_s$ – from the traditional relation for the Mandelstam–Brillouin nonlinearity [28] convoluted with the spectrum of BP components of width $\delta\omega = 1.5$ cm$^{-1}$ [8, 9] taking into account the low sound decay velocity.

It was assumed in numerical calculations that both of the BP components at the point $\Delta\omega = 0$ had wavelengths $\lambda_0 = 625$, 630, and 650 nm. As in [3, 5], we simulated the situation with BP pulses of duration $\tau_p = 20$ ps (the values of $E_e^F$ and $T_e$ averaged over $\tau_p$ were used) incident simultaneously on a $\sim$200-nm-thick $YBa_2Cu_3O_{7-\delta}$ film ($T_c = 91$ K) on a $SrTiO_3$ substrate absorbing 30 % of the total energy $4 \times 10^{-7}$ J of pump pulses focused into a spot of diameter 150 μm [7, 9, 10]. The values of all free parameters of the model corresponded to those used in [5], i.e. the relative amplitudes of the components $\chi_r$, $\chi_s$, and $\chi_0$ were specified so that the dependence $\eta(\Delta\omega, T_0) \propto |\chi(\Delta\omega, T_0)|^2$ for the anti-Stokes component of the nonlinear response for $\lambda_0 = 625$ nm corresponded to experimental data [9], i.e. had dips at points $\Delta\omega = 10$ and 63 cm$^{-1}$ for $T_0 = 90$ and 80 K. The polarisation relaxation rates used in calculations of $\chi_{nr}$ and $\chi_r$ were $\Gamma = 150$ and 50 cm$^{-1}$, as in [4, 5], which provided good agreement between the calculated and experimental widths of $\Delta\varepsilon(\omega)$ spectral features [6, 12].

## 3. Results of simulations

Figures 2 and 3 show variations in the real (Re) and imaginary (Im) parts of $\chi_{nr}$ in the plane $(\Delta\omega, T_0)$ for the anti-Stokes and Stokes components of the nonlinear response and the BP components with coincident frequencies at points $\lambda_0 = 625$, 630, and 650 nm. It is easy to verify that the dependences $\chi_{nr}(\Delta\omega)$ for the response components at frequencies $2\omega_2 - \omega_1$ and $2\omega_1 - \omega_2$ are identical in the frequency detuning range $\Delta\omega < 100$ cm$^{-1}$ for all values of $T_0$ and $\lambda_0$. And only for $\Delta\omega > 100$ cm$^{-1}$, the asymmetry appears, which is especially noticeable for the imaginary part of $\chi_{nr}$ (Fig. 3) and increases with increasing $\Delta\omega$. At first glance, taking into account the value $\Gamma = 150$ cm$^{-1}$ used in calculations, this result seems quite obvious. However, this is not exactly so, because we are dealing here with integral relations describing $\chi_{nr}(\Delta\omega)$, in which even a weak asymmetry of many interfering terms can cause a strong asymmetry of the result of interference.

The dependences $\chi_r(\Delta\omega, T_0)$ and $\chi_s(\Delta\omega, T_0)$ are not presented here because they do not change with $\lambda_0$, are symmetric with respect to the replacement $\omega_1 \leftrightarrow \omega_2$, and simply repeat dependences presented in [5]. In principle, this is not surprising because the models used to describe the contributions of these components of the nonlinear response have a symmetric structure.

Figure 4 shows the calculated dependences of the modulus of the nonlinear response $|\chi(\Delta\omega, T_0)|$ for the anti-Stokes and Stokes components at the same frequency-coincidence points $\lambda_0 = 625$, 630, and 650 nm. It is reasonable that dependences $|\chi(\Delta\omega)|$ for the response components at frequencies $2\omega_2 - \omega_1$ and $2\omega_1 - \omega_2$ are also identical in the frequency detuning range $\Delta\omega < 100$ cm$^{-1}$ for all values of $T_0$ and $\lambda_0$. And only for $\Delta\omega > 100$ cm$^{-1}$, the asymmetry appears, which increases with increasing $\Delta\omega$ and is caused by the asymmetry of the contribution of $\chi_{nr}$ to the total nonlinear response.

Note that this result differs from experimental data [27], according to which the dependences $\eta(\Delta\omega)$ for the anti-Stokes and Stokes components of the nonlinear response of narrow-gap $PrBa_2Cu_3O_{7-\delta}$ semiconductor films noticeably differ from each other already for comparatively small ($\Delta\omega \sim 10$ cm$^{-1}$) frequency detunings of the BP components.

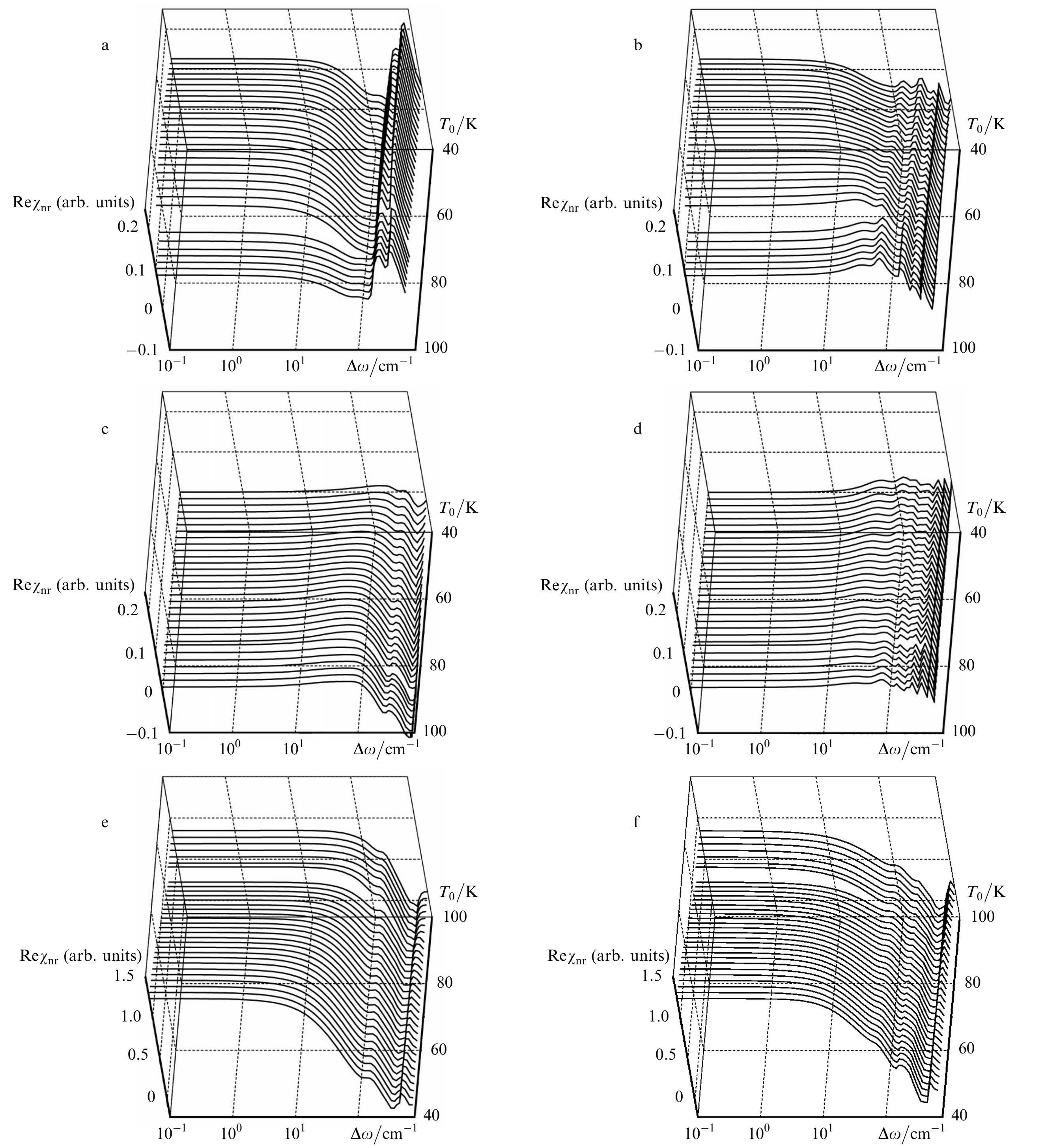


**Figure 2.** Variations in the real part $\mathrm{Re}\chi_{\mathrm{nr}}$ of the nonresonance component of the total nonlinear susceptibility of a sample at frequencies $2\omega_1 - \omega_2$ (a, c, e) and $2\omega_2 - \omega_1$ (b, d, f) in the plane ($\Delta\omega$, $T_0$). The point $\Delta\omega = 0$ corresponds to $\lambda_0 = 625$ (a, b), 630 (c, d), and 650 nm (e, f).

## 4. Conclusions

By using the model of the HTSC nonlinear response to biharmonic pumping [4, 5] and taking into account variations in the Fermi level $E_{\mathrm{e}}^{\mathrm{F}}$ and electron temperature $T_{\mathrm{e}}$ averaged over the duration of coincident pump pulses calculated in [3, 5], we have shown that the Stokes and anti-Stokes components of the HTSC nonlinear response are asymmetric. This means that for frequency detunings of the BP components $\Delta\omega = \omega_1 - \omega_2 > 100\ \mathrm{cm}^{-1}$, the self-diffraction signals are generated in directions $2\boldsymbol{k}_{1,2} - \boldsymbol{k}_{2,1}$ at frequencies $2\omega_{1,2} - \omega_{2,1}$ with different efficiencies $\eta$ due to the presence of one-photon interband resonances. Nevertheless, because of the symmetry of the HTSC nonlinear response in the region of small frequency detunings ($\Delta\omega < 100\ \mathrm{cm}^{-1}$), it is possible to determine the dependence of the energy gap width $\Delta$ in the electronic spectrum of a HTSC sample on its initial temperature $T_0$ from the experimental dependences $\eta(\Delta\omega, T_0)$ for the Stokes and anti-Stoked components by the presence of the characteristic two-photon resonance.

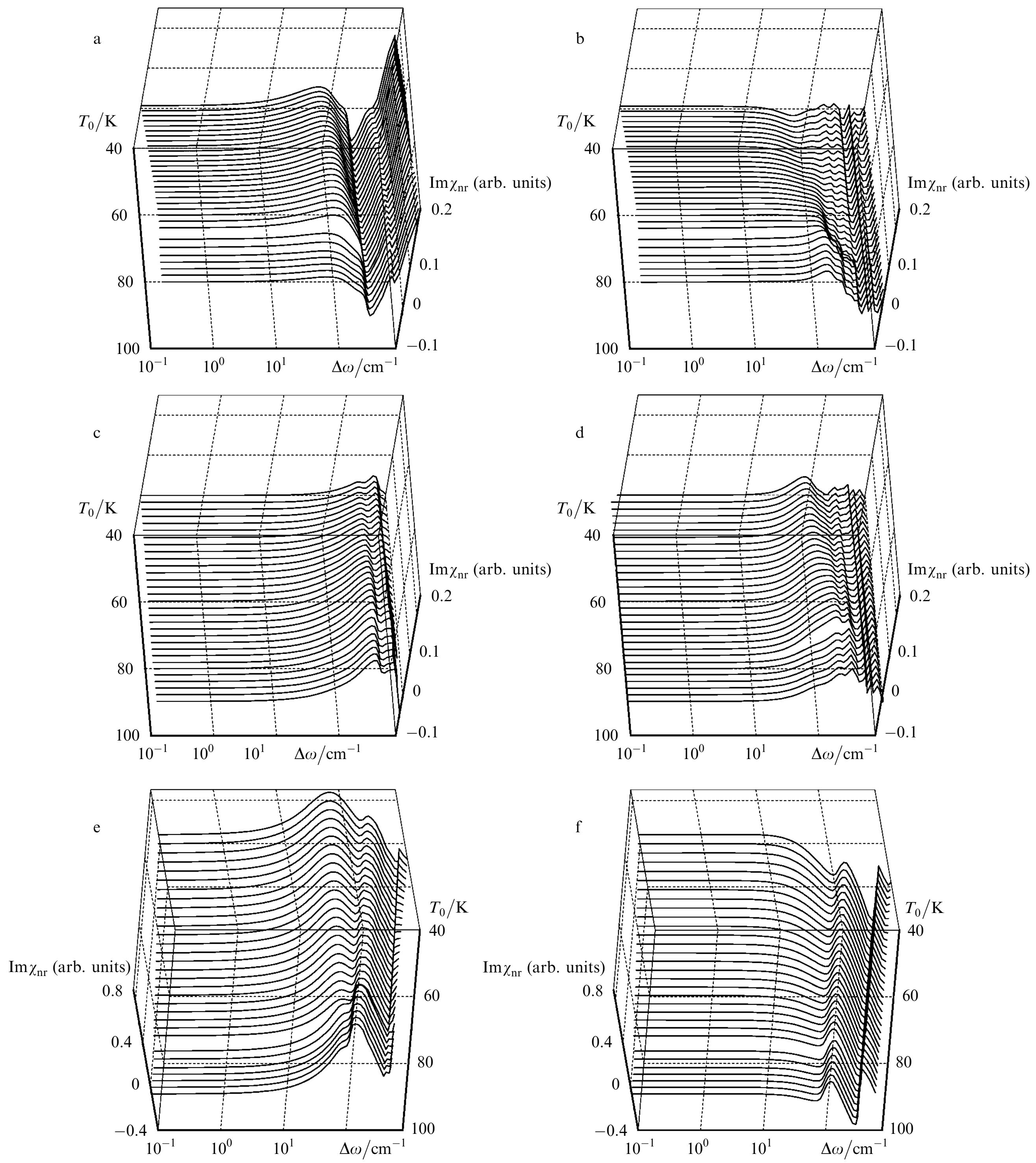


**Figure 3.** Variations in the imaginary part $\mathrm{Im}\chi_{\mathrm{nr}}$ of the nonresonance component of the total nonlinear susceptibility of a sample at frequencies $2\omega_1 - \omega_2$ (a, c, e) and $2\omega_2 - \omega_1$ (b, d, f) in the plane ($\Delta\omega$, $T_0$). The point $\Delta\omega = 0$ corresponds to $\lambda_0 = 625$ (a, b), 630 (c, d), and 650 nm (e, f).

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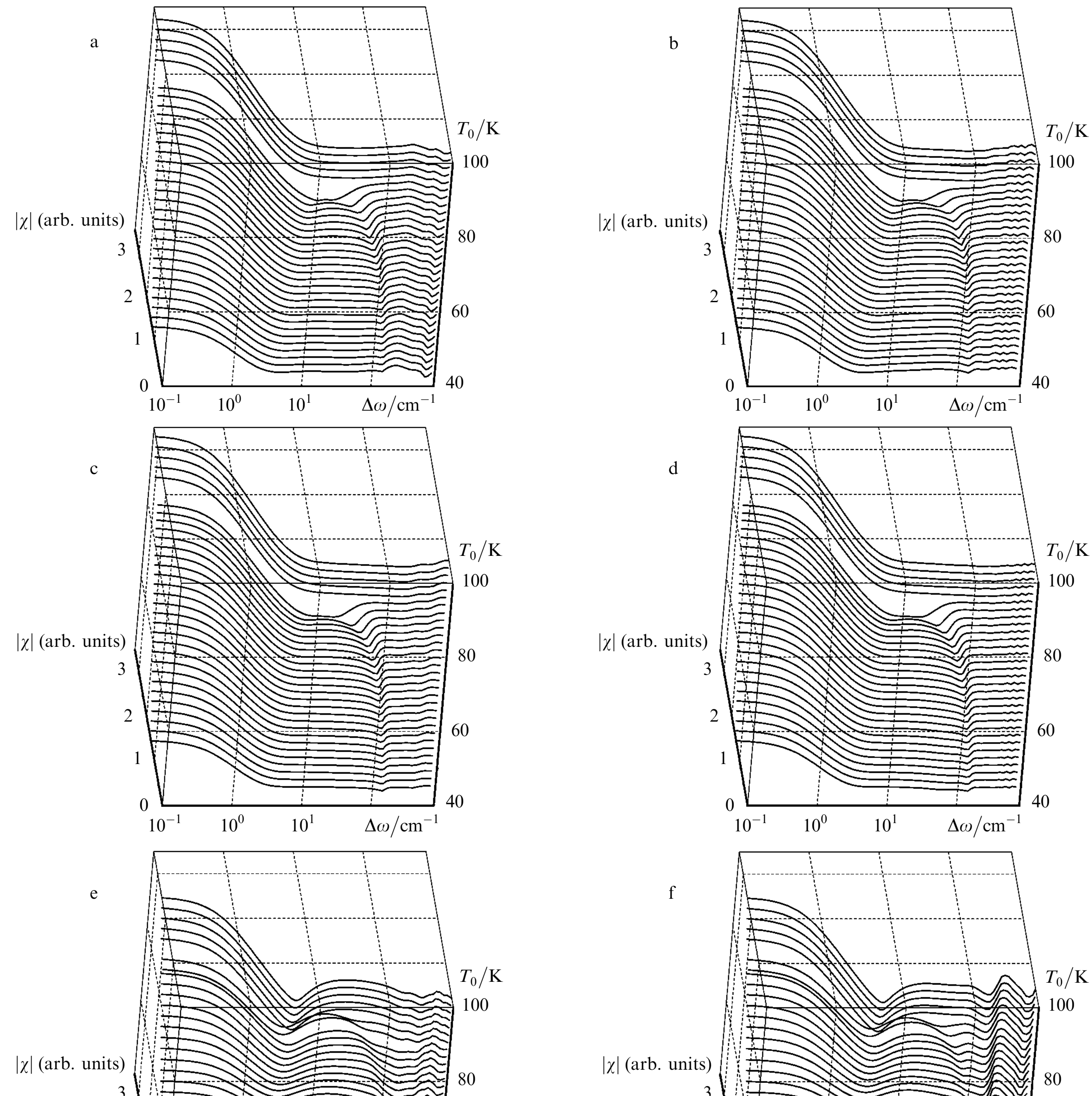


**Figure 4.** Variations in the modulus $|\chi|$ of the total nonlinear susceptibility of a sample at frequencies $2\omega_1 - \omega_2$ (a, c, e) and $2\omega_2 - \omega_1$ (b, d, f) in the plane ($\Delta\omega$, $T_0$). The point $\Delta\omega = 0$ corresponds to $\lambda_0 = 625$ (a, b), 630 (c, d), and 650 nm (e, f).

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