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Optical properties of a semiconductor upon two-photon excitation of biexcitons by a powerful pump pulse and one-photon probing in the M band

A V Corovai, P I Khadzhi

***Abstract*. It is shown that upon two-photon excitation of biexcitons from the ground state by a powerful optical pulse, a weak laser pulse is amplified within the M band.**

Keywords: *exciton, biexciton, susceptibility, pump, probing.*

The pump–probe method has acquired a new significance in connection with experimental studies of the optical Stark effect in the exciton spectral region. The method uses two laser beams: a high-power pump beam and a weak probe beam. The weak beam probes variations in the optical properties of a crystal caused by the high-power pump beam, which are determined by the pump-beam amplitude and frequency and the crystal parameters.

This method was used for studying the kinetics of radiative recombination of excitons, the nonlinear response of a system of excitons and biexcitons of high density [1–3], the red and blue shifts of exciton bands upon picosecond pumping [4–7], and an analogue of the Authler–Townes effect for biexcitons in CuCl [8]. In paper [8], the splitting of the biexciton absorption band into two lines was observed in CuCl between the Z_3-exciton polariton and a biexciton state upon high-power excitation. The biexciton state was probed by two-photon absorption resulting in excitation of biexcitons from the ground state of the crystal upon pumping by a high-power laser beam into the M band, which mixes a pump photon, an exciton, and a biexciton. The authors of paper [8] determined the dipole transition moment from the splitting and showed experimentally that two-photon absorption spectra are determined by the intensity and frequency of the pump pulse.

The theory of the pump–probe method for a system of excitons and biexcitons of high density was developed in papers [9–12]. It was shown [12] that the susceptibility of a semiconductor in the exciton region of the spectrum, taking into account the exciton–photon and elastic exciton–exciton interactions, exhibits a bistable dependence on the frequency and intensity of the pump pulse, as well as on the frequency of the probe pulse.

A V Corovai, P I Khadzhi Department of Physics and Mathematics, T G Shevchenko Pridnestrie State University, ul. 25 Oktyabrya 128, 278000 Tiraspol', Moldova; e-mail: tdsu@tirastel.md

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Analysis of the experimental results [8] suggests the use of an alternative approach within the framework of the same system. Let us assume that a high-power pump pulse with the field amplitude E_0 and frequency $\omega_{las} \approx \Omega_0/2$ excites biexcitons from the ground state of a crystal upon two-photon absorption (Ω_0 is the frequency of the biexciton level). We assume that the probe pulse with the field amplitude E and frequency $\omega \approx \omega_M = \Omega_0 - \omega_0$ acts within the M band (exciton–biexciton conversion band), where ω_0 is the exciton transition frequency (Fig. 1).

Therefore, the roles of the beams are reversed compared to [8]. We use the pump beam in [8] as a probe beam, whereas the probe beam in [8] is used as pump beam, by changing appropriately their intensities. The interaction Hamiltonian in the resonance approximation containing the term of two-photon excitation of biexcitons by the field E_0 of the high-power pulse and the term of optical exciton–biexciton conversion produced by the field E of the weak pulse can be written in the form [13]

$$H = -\hbar\mu\left(b^+E_0^+E_0^+\mathrm{e}^{-2\mathrm{i}\omega_{las}t} + bE_0^-E_0^-\mathrm{e}^{2\mathrm{i}\omega_{las}t}\right)$$

$$-\hbar\sigma\left(a^+bE^-\mathrm{e}^{\mathrm{i}\omega t} + ab^+E^+\mathrm{e}^{-\mathrm{i}\omega t}\right), \qquad (1)$$

where a and b are the amplitudes of exciton and biexciton waves of the polarised medium, respectively; $E_0^+(E^+)$ and E_0^- (E^-) are the positive-frequency and negative-frequency components of the pump field (probe radiation), respectively; σ and μ are the constants of optical exciton–biexciton conversion and two-photon excitation of biexcitons from the ground state of the crystal, respectively [13–15].

Assuming that the exciton, biexciton, and photon states are macroscopically filled, we can readily obtain from (1) the Heisenberg (constitutive) equations for the amplitudes a and b

$$\mathrm{i}\dot{a} = (\omega_0 - \mathrm{i}\gamma_1)a - \sigma bE^-\mathrm{e}^{\mathrm{i}\omega t}, \qquad (2)$$

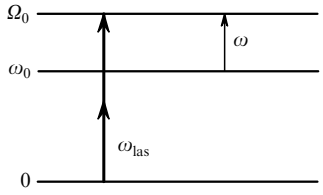

Figure 1. Energy level diagram and quantum transitions in a semiconductor.

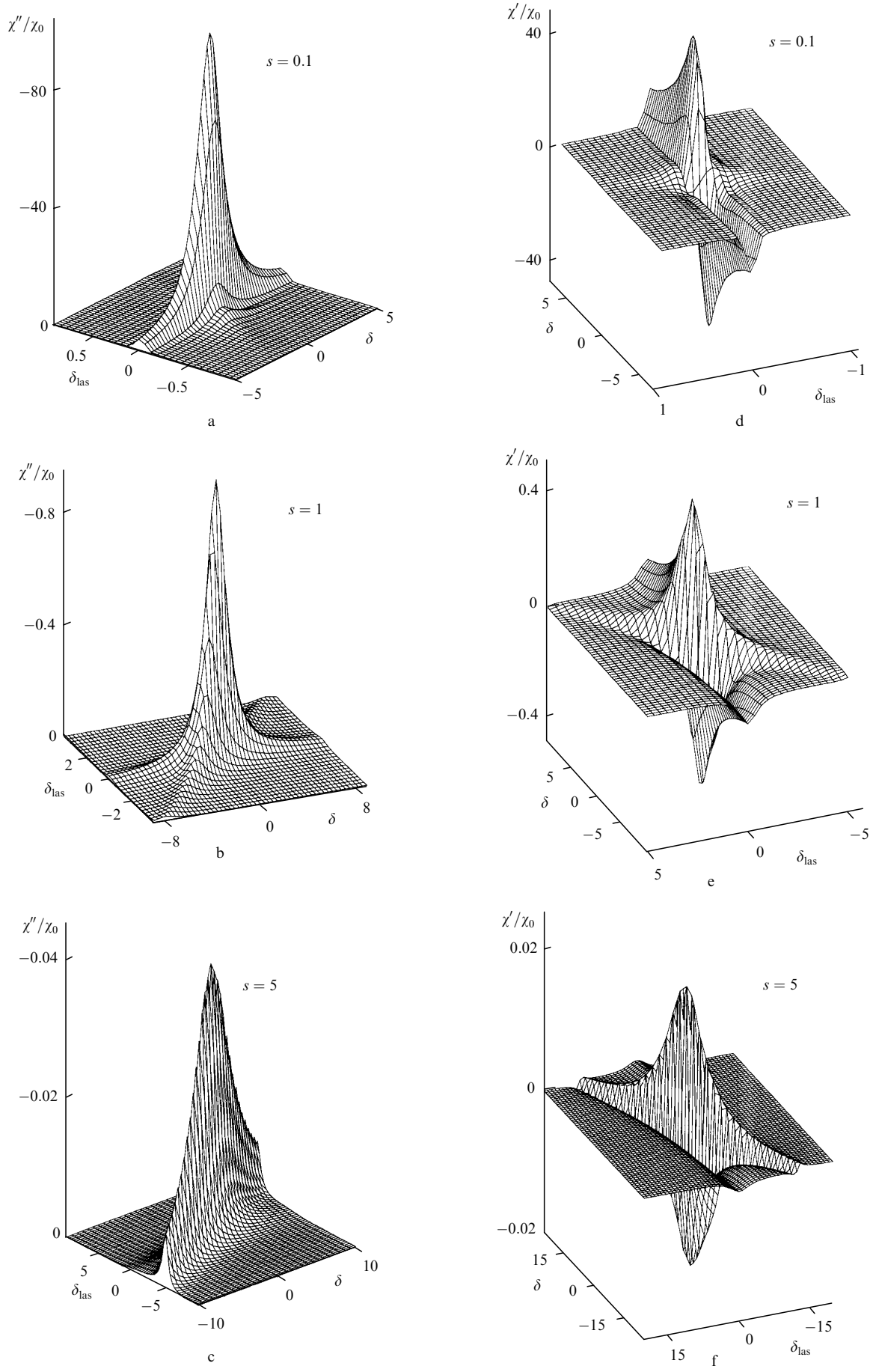

Figure 2. Profiles of the absorption (χ'') and dispersion (χ') components in the plane δ, δ_{las} of resonance detunings for different s.

$$i\dot{b} = (\Omega_0 - i\gamma_2)b - \sigma a E^+ e^{-i\omega t} - \mu E_0^+ E_0^+ e^{-2i\omega_{las} t}, \quad (3)$$

where γ_1 and γ_2 are phenomenological constants that take into account the decay of exciton and biexciton energy levels, respectively, caused by scattering.

Consider the response of the system in the first order of the perturbation theory over the weak-pulse amplitude E in the stationary regime. We will seek the solutions of equations (2) and (3) in the form

$$a = A e^{-i(2\omega_{las} - \omega)t} + A^* e^{i(2\omega_{las} - \omega)t}, \quad (4)$$

$$b = B e^{-2i\omega_{las} t} + B^* e^{2i\omega_{las} t}, \quad (5)$$

where A and B are the time-independent amplitudes. By substituting (4) and (5) in (2) and (3), we obtain for the amplitudes A and B

$$A = -\frac{\sigma B E^-}{2\omega_{las} - \omega - \omega_0 + i\gamma_1}, \; B = -\frac{\mu E_0^+ E_0^+}{2\omega_{las} - \Omega_0 + i\gamma_2}. \quad (6)$$

Because the polarisation $P^+ = \chi E^+$ of the medium with respect to the probe-pulse field is $\hbar\sigma a^+ b e^{i\omega t}$, we obtain for the complex susceptibility of the medium, taking (6) into account,

$$\frac{\chi'}{\chi_0} = -\frac{2\delta_{las} - \delta}{(4\delta_{las}^2 + s^2)[(2\delta_{las} - \delta)^2 + 1]}, \quad (7)$$

$$\frac{\chi''}{\chi_0} = -\frac{1}{(4\delta_{las}^2 + s^2)[(2\delta_{las} - \delta)^2 + 1]}, \quad (8)$$

where $\chi_0 = \hbar\sigma^2\mu^2 E_0^4/\gamma_1^3$; $\delta = \Delta/\gamma_1$; $\delta_{las} = \Delta_{las}/\gamma_1$; $s = \gamma_2/\gamma_1$; $\Delta = \omega - \omega_M$; $\Delta_{las} = \omega_{las} - \Omega_0/2$.

One can see from (7) and (8) that χ' and χ'' are proportional to the square of the pump intensity. In addition, it follows from (8) that $\chi'' < 0$ for any detunings of the resonance of the pump and probe fields. Therefore, the probe field is not absorbed but is amplified instead. The gain of the probe radiation is proportional to the square of the pump-pulse intensity. In the absence of the pump field, the probe radiation propagates through the crystal as if it is absolutely transparent because excitons and biexcitons are absent. Upon pumping, the system becomes inverted with respect to the probe-radiation frequency, resulting in amplification.

The gain function χ'' is a product of two Lorentzians. The Lorentzian $(4\delta_{las}^2 + s^2)^{-1}$ in (8) is caused by two-photon absorption of the pump filed and determines the density of biexcitons being formed, while the Lorentzian $[(2\delta_{las} - \delta)^2 + 1]^{-1}$ is responsible for negative absorption (amplification) of the probe radiation taking into account the resonance detunings both for the pump field (δ_{las}) and probe field (δ). This means that the shape of the gain band is determined by the shapes of both Lorentzians. Because the second Lorentzian does not contain the parameter s, its amplitude is much smaller than that of the first Lorentzian for s and $\delta_{las} \to 0$, and it is in fact masked by the first Lorentzian. Therefore, the gain band for small s has a small half-width over the pump frequency and a large half-width over the probe frequency. As the parameter s increases, the half-width of the first Lorentzian increases and the gain band in the plane δ, δ_{las} becomes distinctly manifested alone the straight line $\delta = 2\delta_{las}$ (Fig. 2).

Fig. 2 shows the spectral dependences of the dispersion and absorption components of the crystal susceptibility in the plane of resonance detunings δ, δ_{las}. The susceptibility χ'' shows a sharp gain peak of the crystal. The component χ' also indicates the amplification of probe radiation. One can see from Fig. 2d–f that the profile of χ' gradually turns in the plane δ_{las}, δ with increasing s, which is caused by different contributions of each of the factors in (7). If the pump radiation is resonant with the biexciton level ($\delta_{las} = 0$), then is follows from (8) that the absorption component of the susceptibility χ'' is described by the Lorentzian

$$\frac{\chi''}{\chi_0} = -s^{-2}\left(\delta^2 + 1\right)^{-1},$$

the half-width of the gain band being equal to the decay constant γ_1 of the exciton level.

It follows from (8) and Fig. 2 that the magnitude of the ratio χ''/χ_0 at the maximum is determined by the parameter s. This ratio equals -90, -0.8, and -0.04 for $s = 0.1$, 1, and 5, respectively. A similar conclusion is valid for the extrema of the function χ'/χ_0.

Therefore, the components of the susceptibility of a medium upon two-photon excitation of biexcitons from the ground state of the crystal by a high-power pump pulse and probing by a weak pulse within the M band predict the amplification of the weak pulse. It was shown earlier [12] that the probe pulse can be absorbed or amplified in different spectral regions due to the elastic exciton–exciton interaction. In our case, the probe pulse is not absorbed but only amplified, which is caused by the inversion of the system at the probe-pulse frequency produced by the pump pulse.

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