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On the diagnostics of semiconductors upon two-photon excitation of biexcitons

P.I. Khadzhi, L.Yu. Nad'kin

***Abstract*. A version of the one-photon probing of optical properties of semiconductors is presented under conditions when a high-power pump pulse excites biexcitons from the ground state of a crystal due to two-photon absorption. The existence of a Lorentzian absorption peak is predicted and the quasi-polariton dispersion law is found in the region where a real energy level is absent. The temporal evolution of the absorption band under the action of ultrashort pulses is studied.**

Keywords: *two-photon absorption, biexcitons, semiconductors.*

1. Introduction

The optical properties of semiconductors in the exciton spectral region at high excitation levels can be efficiently studied by the pump-probe method. This method uses two laser beams – a high-power pump beam and a weak probe beam. The weak beam monitors variations in the optical properties of a crystal caused by the action of the high-power pump beam. This method was used to study the recombination kinetics and nonlinear response of a system of excitons and biexcitons [1–3], the red and blue shifts of exciton emission bands upon picosecond pumping [4–6], and the Autler–Townes effect on biexcitons [7]. Different aspects of the application of the pump-probe method for studying a high-density system of excitons and biexcitons were considered in theoretical papers [8–14]. In [8, 9], the dielectric susceptibilities of a CuCl crystal were studied upon pumping by a high-power pulse into the M band and probing two-photon absorption of light with the formation of excitons. It was shown in [13] that the behaviour of the susceptibility of a semiconductor in the exciton spectral region, taking the elastic exciton–exciton interaction into account, can be bistable depending on the pump frequency and intensity.

In this paper, we propose a new version of the pump-probe method with the use of two-photon absorption of light as the main nonlinearity mechanism as applied to a CuCl crystal. It is known [14, 15] that the biexciton binding energy in a CuCl crystal is 30–40 meV. Biexcitons can be easily identified in their spectra of direct two-photon excitation from the ground state of the crystal.

P.I. Khadzhi Institute of Applied Physics, Academy of Sciences of Moldova, ul. Akademiei 5, 2800 Kishinev, Moldova; T.G. Shevchenko Dnester State University, MD 3300 Tiraspol', Moldova;
e-mail: tdsu4@idknet.com;
L.Yu. Nad'kin T.G. Shevchenko Dnester State University, MD 3300 Tiraspol', Moldova

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2. Formulation of the problem, the basic equations and discussion of results

Consider two-photon excitation of biexcitons by a high-power pump pulse with the amplitude E_0 and frequency ω_p (Fig. 1). A weak probe pulse with the amplitude E and frequency ω acts in the same spectral region; however, two-photon absorption of this pulse is vanishingly small. For this reason, we will consider two-photon absorption in the case when biexcitons are generated by one photon of the high-power pulse together with a photon of the weak pulse and, hence, one-photon probing by photons of the weak pulse occurs (Fig. 1). Purely one-photon absorption of the weak pulse without the participation of the high-power pulse at the frequency approximately equal to half the biexciton frequency is impossible [14]. However, one-photon absorption of this pulse proves to be possible in the presence of the high-power pump pulse at the frequency approximately equal to that of the weak pulse. The high-power pump pulse produces a quasi-energy state with the frequency equal to the difference $\Omega_0 - \omega_p$ between the biexciton frequency and the frequency of the high-power pulse, at which photons of the weak pulse are absorbed. Thus, we can assert that one-photon probing of the quasi-energy state by photons of the weak pulse with the frequency ω takes place.

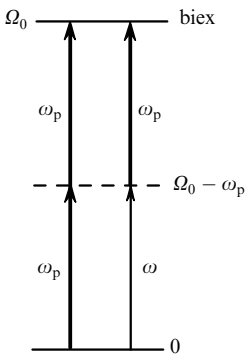

Figure 1. Scheme of quantum transitions upon two-photon excitation of the biexciton state by a high-power pulse ω_p and its one-photon probing by a weak pulse ω.

The Hamiltonian of interaction of biexcitons with light in the resonance approximation has the form

$$H_{\rm int} = -\hbar\mu\{b^+E_0^+E_0^+\exp(-2{\rm i}\omega_{\rm p}t) + bE_0^-E_0^-\exp(2{\rm i}\omega_{\rm p}t) + b^+E_0^+E^+\exp[-{\rm i}(\omega_{\rm p}+\omega)t] + bE_0^-E^-\exp[{\rm i}(\omega_{\rm p}+\omega)t]\}, \quad (1)$$

where b is the amplitude of a biexciton polarisation wave of the medium; μ is a two-photon excitation constant of biexcitons; and $E_0^+(E_0^-)$ and $E^+(E^-)$ are the positive (negative) frequency components of the high-power and weak pulses, respectively. As shown in [16], the constant μ can be determined from the relation $\mu E_{\rm c} = g$, where g is the exciton–photon interaction constant. The characteristic field $E_{\rm c}$ is determined from the energy conservation law $E_{\rm c}^2/8\pi = \hbar\Omega_0 N_{\rm c}/2$, where Ω_0 is the biexciton state frequency. For CuCl and CdS crystals, the value of $N_{\rm c}$ is $\sim 10^{15}$ cm^{-3} [16].

By using (1), we can easily obtain the Heisenberg equation for the amplitude $b(t)$ of the biexciton wave:

$$\dot{\rm i}b = (\Omega_0 - {\rm i}\gamma)b - \mu E_0^+E_0^+\exp(-2{\rm i}\omega_{\rm p}t) - \mu E_0^+E^+\exp[-{\rm i}(\omega_{\rm p}+\omega)t], \quad (2)$$

where γ is a phenomenological constant taking into account the decay of the biexciton state. From (2), we find the stationary amplitude b. Knowing the polarisation $P = \hbar\mu b E_0^-\exp[{\rm i}(\omega_{\rm p}+\omega)t]$ of the system, we can readily determine the complex susceptibility χ with respect to the weak-pulse field

$$\chi = \chi' + {\rm i}\chi'' = -\frac{\hbar g^2(E_0/E_{\rm c})^2(\Delta + \Delta_{\rm p} - {\rm i}\gamma)}{(\Delta+\Delta_{\rm p})^2 + \gamma^2}, \quad (3)$$

where χ' and χ'' are the real (dispersion) and imaginary (absorption) components of the susceptibility; and $\Delta_{\rm p} = \Omega_0/2 - \omega_{\rm p}$ and $\Delta = \Omega_0/2 - \omega$ are the resonance detunings for photons of the strong and weak pulses, respectively. One can see from (3) that the imaginary (absorption) component of the susceptibility $\chi''(\Delta)/\chi_0$ (the absorption band of probe radiation) has a Lorentzian shape (Fig. 2), where $\chi_0 = \hbar g^2/\gamma$. For $\gamma \to 0$, the absorption band is a delta-shaped peak at the frequency $\omega = \Omega_0 - \omega_{\rm p}$. Knowing χ, we can easily obtain the expression for the dielectric function of a crystal at the weak-pulse frequency:

$$\varepsilon = \varepsilon_\infty\left(1 - \frac{\Omega_{\rm LT}}{\Delta + \Delta_{\rm p} + {\rm i}\gamma}\right), \quad (4)$$

where

$$\Omega_{\rm LT} = \frac{\omega_{\rm LT}J_0}{J_{\rm c}} \quad (5)$$

is the effective longitudinal–transverse splitting; $\omega_{\rm LT}$ is the longitudinal–transverse splitting of the exciton state; $J_{\rm c} = cE_{\rm c}^2/8\pi$; and J_0 is the pump intensity. By assuming that $\gamma = 0$ and using the wave equation for the field E, we can obtain the dispersion law at the weak-pulse frequency, which has the form

$$\frac{c^2k^2}{\omega^2} = \varepsilon_\infty\left(1 - \frac{\Omega_{\rm LT}}{\Delta + \Delta_{\rm p}}\right), \quad (6)$$

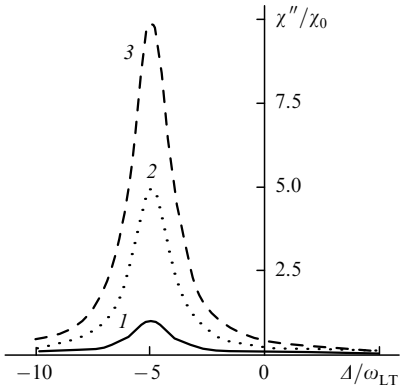

Figure 2. Shape of the absorption band of the probe pulse as a function of the resonance detuning $\Delta/\omega_{\rm LT}$ for the probe pulse at the resonance detuning of the pump field $\Delta_{\rm p}/\omega_{\rm LT} = 5$ and normalised intensities $J_0/J_{\rm c}$ of the pump pulse equal to 1 (*1*), 5 (*2*), and 10 (*3*).

where k is the wave vector. It follows from (6) that the dispersion law predicts the existence of polariton states for photons of the weak pulse in the frequency region where the real levels are absent (Fig. 3). The most intense interaction appears at the frequency $\omega = \Omega_0 - \omega_{\rm p}$, i.e., the frequency equal to the difference of the biexciton frequency Ω_0 and high-power pulse frequency $\omega_{\rm p}$. This means that the interaction of photons of the high-power pulse with biexcitons at this frequency can produce a virtual quasi-energy state interacting with photons of the weak pulse. In this case, optical transitions from the ground state of the crystal on the quasi-energy level are one-photon. They are optically allowed. It follows from (3)–(5) that the oscillator strength of such a transition and the effective longitudinal–transverse splitting in the region of one-photon interaction of the weak-pulse field with the medium are proportional to the high-power pulse intensity J_0. Therefore, the effective longitudinal–transverse splitting increases with increasing the excitation level, which means that the one-photon interaction of the weak-pulse field with the medium is enhanced in the presence of high-power

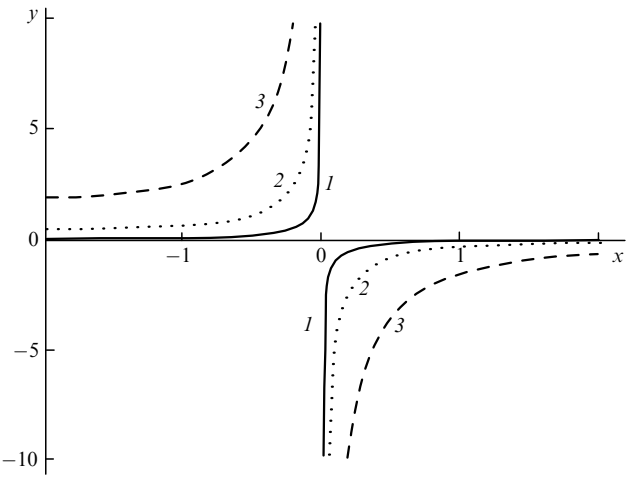

Figure 3. Polariton-like dispersion law for probe-pulse photons under the condition of the exact resonance of the pump field for $J_0/J_{\rm c}$ equal to 0.1 (*1*), 0.5 (*2*), and 5 (*3*); $x = ck/\sqrt{\varepsilon_\infty}(\Omega_0 - \omega_{\rm p})$, and $y = (\Delta + \Delta_{\rm p})/\omega_{\rm LT}$.

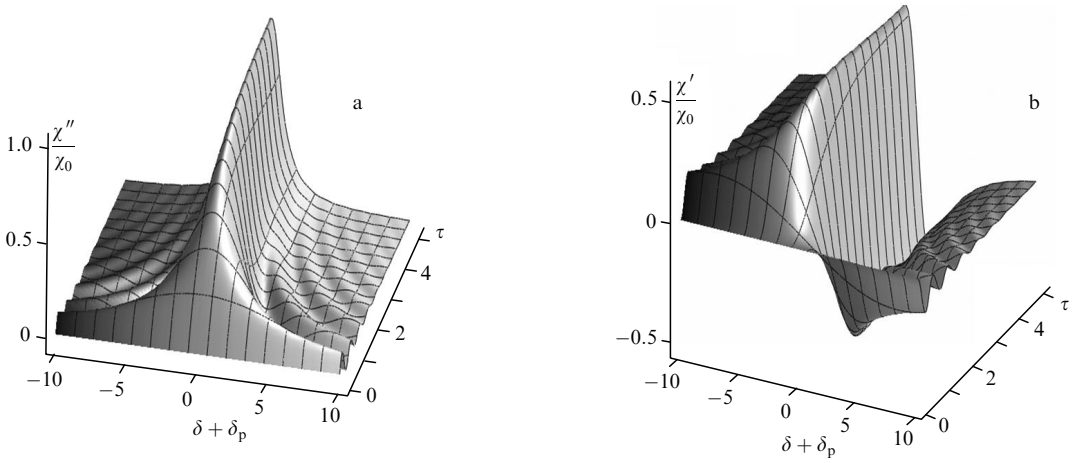

Figure 4. Spectrochronograph representations of the absorption (a) and dispersion (b) components of the susceptibility for probe-pulse photons.

pumping. Note that this interaction takes place in the frequency region where the real energy level is absent.

One can see from (6) and Fig. 3 that the position of the polariton-like branches of the dispersion law is determined by the intensity J_0 and frequency ω_p of the high-power pulse.

Assuming in (2) that the field $E_0(t) = E_0\Theta(t)$ and the field $E(t) = E\Theta(t)$, i.e., the incident pulses are step-like and are switched at the instant $t = 0$, we can easily obtain the expressions for the time dependence of the susceptibilities

$$\frac{\chi'}{\chi_0} = -\frac{j_0}{(\delta+\delta_p)^2+1}\{(\delta+\delta_p)[1-\exp(-\tau) \times \cos[\tau(\delta+\delta_p)]] - \exp(-\tau)\sin[\tau(\delta+\delta_p)]\}, \quad (7)$$

$$\frac{\chi''}{\chi_0} = \frac{j_0}{(\delta+\delta_p)^2+1}\{1-\exp(-\tau) \times \cos[\tau(\delta+\delta_p)] - (\delta+\delta_p)\exp(-\tau)\sin[\tau(\delta+\delta_p)]\}, \quad (8)$$

where $\chi_0 = \hbar g^2/\gamma$; $j_0 = J_0/J_c$; $\delta = \Delta/\gamma$; $\delta_p = \Delta_p/\gamma$; $\tau = \gamma t$.

Figure 4 shows the spectrochronograph behaviour of the absorption and dispersion components $\chi''(\delta+\delta_p)$ and $\chi'(\delta+\delta_p)$, respectively, of the susceptibility. One can see that the stationary value of $\chi''(\tau)$ is established in the oscillating regime (Fig. 4a). For a fixed instant of time, χ'' oscillates depending on δ, while for fixed δ, χ'' oscillates depending on τ at the initial stage of the establishment of a stationary value. During certain time intervals determined by the inequality

$$\exp(-\tau)\cos[\tau(\delta+\delta_p)+\varphi] \geqslant \cos\varphi \equiv [(\delta+\delta_p)^2+1]^{-1/2}, \quad (9)$$

the susceptibility χ'' proves to be negative, i.e., the weak pulse is amplified rather than absorbed. The smaller the decay constant of the biexciton state, the greater the number of such time intervals.

At long times, the stationary regime and stationary value of $\chi''(\delta+\delta_p)$ are established. In this case, the absorption band of probe radiation has a Lorentzian shape. The dispersion component of the susceptibility $\chi'(\delta+\delta_p)$ (Fig. 4b) is also characterised by the oscillating regime of the establishment of its stationary value (Fig. 4b).

3. Conclusions

The existence of the Lorentzian one-photon absorption peak of a weak probe radiation has been predicted upon two-photon excitation of biexcitons from the ground state of a crystal by a high-power pump pulse. The dispersion law for photons of weak radiation is polariton-like, the longitudinal–transverse splitting being proportional to the high-power pulse intensity. The absorption band appears at the frequency equal to the difference of the frequencies of the biexciton transition and high-power pulse, where a real level is absent. The amplification of the probe pulse upon nonstationary excitation of the medium has been discussed.

References

1. Leonelli R., Manar A., Grun J.B., Honerlage B. *Phys. Rev. B*, **45**, 4141 (1992).
2. Smith G.O., Mayer E.J., Kuhl J., Ploog K. *Sol. State Commun.*, **92**, 325 (1994).
3. Finkelstein G., Bar-Ad S., Carmel O., Bar-Joseph I., Levinson Y. *Phys. Rev. B*, **47**, 12964 (1993).
4. Hulin D., Joffre M. *Phys. Rev. Lett.*, **65**, 3425 (1990).
5. Peyghambarian N., Gibbs H.M., Jewell J. L., Antonetti A., Migus A., Hulin D., Mysyrowicz A. *Phys. Rev. Lett.*, **53**, 2433 (1984).
6. Von Lehmen A., Chemla D.S., Zucker J.E., Heritage J.P. *Opt. Lett.*, **11**, 609 (1985).
7. Shimano R., Kuwata-Gonokami M. *Phys. Rev. Lett.*, **72**, 530 (1994).
8. Khadzhi P.I., Korovai A.V., Tkachenko D.V. *Fiz. Tverd. Tela*, **44**, 774 (2002).
9. Khadzhi P.I., Corovai A.V., Korovai O.V., Tkachenko D.V. *Moldavian J. Phys. Sci.*, **1**, 152 (2002).
10. Hanamura E. *Phys. Rev. B*, **44**, 8514 (1991).
11. Moskalenko S.A., Snoke D.W. *Bose-Einstein Condensation of Excitons and Biexcitons and Coherent Nonlinear Optics with Excitons* (Cambridge: Cambridge University Press, 2000).
12. Agarwal G.S. *Phys. Rev. A*, **51**, R2711 (1995).
13. Khadzhi P.I., Tkachenko D.V. *Fiz. Tverd. Tela*, **40**, 934 (1998).
14. Khadzhi P.I. *Kinetika rekombinatsionnogo izlucheniya eksitonov i bieksitonov v poluprovodnikakh* (Kinetics of Recombination Radiation of Excitons and Biexcitons in Semconductors) (Kishinev: Shtiintsa, 1977).
15. Bobrysheva A.I. *Bieksitony v poluprovodnikakh* (Biexcitons in Semiconductors) (Kishinev: Shtiintsa, 1979).
16. Hanamura E. *Sol. State Commun.*, **12**, 951 (1973); *J. Phys. Soc. Jpn*, **39**, 1516 (1975).