

# Explosive nature of absorption upon the resonant formation of biexcitons

Kh. V. Nerkararyan

*Scientific-Research Institute of the Physics of Condensed Media, Erevan State University*

(Submitted 29 August 1986)

Pis'ma Zh. Eksp. Teor. Fiz. **44**, No. 7, 329–330 (10 October 1986)

The possibility of observing an exponential increase over time in the absorption coefficient during the resonant formation of dissociating excitonic molecules in direct-gap semiconductors is discussed

The various possibilities for experimentally observing an excitonic molecule (a biexciton) in direct-gap semiconductors are based on optical processes, primarily because the huge oscillator strengths typical of transitions to a biexciton state make the various optical processes quite easy to observe.<sup>1-3</sup>

In the present letter we show that an explosive (avalanche) intensification of the absorption may be observed in a system of dissociating excitonic molecules. This time-varying process is characterized by an increase in the absorption coefficient which is exponential over time.<sup>4</sup>

In this case, the explosive absorption of light can be described in the following way. We assume that an electromagnetic wave whose frequency is at resonance with a

transition from an exciton state to a biexciton state is incident on a crystal. The absorption of a photon may then result in the conversion of an exciton into a biexciton. After this event, dissociative processes lead to the decay of the excitonic molecule into two excitons. The light wave converts these excitons into biexcitons; etc. This cycle over time in the densities of both excitons and biexcitons. These events can occur during strong absorption of wave energy.

Within the range of applicability of the equations for the rates of change of the densities of excitons and biexcitons, the following expression can be derived for the absorbed power at a given wave intensity:

$$\Delta P = \Delta P_0 e^{st}, \quad (1)$$

where

$$s = -w - \frac{1}{2} (\Gamma + \gamma_0 + \gamma_1) + \sqrt{\left[ w + \frac{1}{2} (\Gamma + \gamma_0 + \gamma_1) \right]^2 + (\Gamma - \gamma_0)w - (\Gamma + \gamma_1)\gamma_0}. \quad (2)$$

The constant  $\Delta P_0$  depends on the initial density of excitons in the semiconductor;  $w$  is the probability per unit time for an induced transition between an exciton and a biexciton state;  $\Gamma^{-1}$  is the scale time for dissociation of an excitonic molecule;  $\gamma_0^{-1}$  is the lifetime of a free exciton; and  $\gamma_1^{-1}$  is the radiative lifetime of an exciton in a two-exciton complex (the condition  $\gamma_1 \gg \gamma_0$  will ordinarily hold<sup>1</sup>). In (1) we have ignored a term which decays rapidly over time.

It follows from (2) that the explosive absorption occurs when a threshold is reached. In other words, it can be observed only beginning at a certain intensity of the incident radiation, under the condition

$$w > \frac{(\Gamma + \gamma_1)\gamma_0}{\Gamma - \gamma_0}. \quad (3)$$

Confirmation that the absorption is of an avalanche nature can come from an asymmetric change in the shape of a light pulse which is transmitted through the semiconductor if the length ( $\tau$ ) of this pulse is sufficiently large:

$$\tau \gg [2w + \Gamma + \gamma_0 + \gamma_1]^{-1}. \quad (4)$$

One attractive feature of this situation is that there are two easily adjustable parameters. In addition to the parameter  $w$ , whose value is proportional to the intensity of the incident radiation, the parameter  $\Gamma$  is also easily adjustable. Since the binding energy of a biexciton is usually considerably smaller than the binding energy of an exciton, by choosing the thermal conditions appropriately we can significantly increase the value of  $\Gamma$ , while, keeping  $\gamma_0$  and  $\gamma_1$  constant.

An excitonic molecule may undergo dissociation because of the absorption of an optical LO phonon. In particular, in CuCl, where the binding energy of an exciton (28 meV) is close to the energy of an LO phonon (26 meV), this dissociation mechanism may play a governing role. Furthermore, an excitonic molecule can be broken up by free carriers.<sup>5</sup> This process is very sensitive to an external electrostatic field, so that we have one more tool for controlling the change in  $\Gamma$ .

In the derivation of (1) it was assumed that essentially all of the excitons participate, with equal probabilities, in the absorption. This assumption requires justification, since the resonant frequency for the transition from the exciton state to the biexciton state depends strongly on the momentum ( $\mathbf{K}$ ) of the exciton.<sup>1</sup> If we ignore the momentum of the photon, as is permissible in this case, we find that with increasing  $\mathbf{K}$  the transition frequency decreases by  $\mathbf{K}^2/4m$ , where  $m$  is the mass of the exciton. Consequently, the spectral band of biexciton absorption will be quite broad. Since the shift occurs only toward longer waves, there is no circumstance under which the exciton and biexciton bands will overlap.

Explosive absorption can be realized only if the spectral width of the incident wave is such that a significant fraction of the excitons can participate in the process. In CuCl, the spectral width of the wave which would be required should be about 3.5 nm. The most convenient sources of radiation for studying transitions to a biexciton state are tunable dye lasers, in which there would be no fundamental difficulty in arranging the conditions required here.<sup>6</sup>

The exponential growth of the absorbed power cannot, of course, proceed continuously. At sufficiently high densities of the quasiparticles, various effects due to interactions between the quasiparticles will come into play. These effects will cause a substantial shift of the resonant level and also changes in the relaxation parameters. As a result, condition (3) will be violated.

I wish to thank A. L. Gyulamiryan for discussions.

<sup>1</sup>E. I. Rashba, *Fiz. Tekh. Poluprovodn.* **8**, 1241 (1974) [*Sov. Phys. Semicond.* **7**, 835 (1974)].

<sup>2</sup>J. B. Grün, B. Henerlage, and R. Levy, in *Excitons* (Russ. trans. ed. E. I. Rashba and M. D. Sterdkha, Nauka, Moscow, 1985).

<sup>3</sup>V. D. Kulakovskii, V. G. Lysenko, and V. B. Timofeev, *Usp. Fiz. Nauk* **147**, 3 (1985) [*Sov. Phys. Usp.* **28**, 1 (1985)].

<sup>4</sup>A. N. Oraevskii and I. E. Pokrovskii, *Kvant. Elektron.* (Moscow) **12**, 2290 [*Sov. J. Quantum Electron.* **15**, 1509 (1985)].

<sup>5</sup>V. D. Kulakovskii, I. V. Kukushkin, and V. B. Timofeev, *Zh. Eksp. Teor. Fiz.* **78**, 381 (1980) [*Sov. Phys. JETP* **51**, 191 (1980)].

<sup>6</sup>F. P. Schaefer (ed.), *Dye Lasers*, Springer-Verlag, New York, 1974 (Russ. transl. Mir, Moscow, 1976).

Translated by Dave Parsons