Effect of Nonlinear Exciton-Phonon Interaction on the Intrinsic Optical Bistability in Layered Semiconductors

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Abstract

Theoretical investigation of the role of bending waves in the conditions for observing optical bistability in layered semiconductors is performed using the Green function method. By the example of 2H-polytype of PbI_2, we show that efficient exciton scattering by the oscillations of this type leads to a short-wave shift of the frequency region of optical bistability, decrease in the size of this region, widening of temperature interval of its observation, a shift of the hysteresis loop towards higher intensities and a decrease in its height and width.

1. Introduction

Optical bistability (OB) phenomenon has been recorded for the first time by McCall [1]. It has become very important from the viewpoint of various practical applications, including creation of optoelectronic devices (memory elements, optical transistors, discriminators, switches, etc.) [2]. The phenomenon of OB, which is essential for a wide class of materials and devices, is based on a nonlinear response of system to intense external electromagnetic radiation. Under certain conditions, interaction of nonlinear resonance system with the radiation can bring the system into one of two or several steady, dynamically stable states that differ by the energy absorption levels for the incident wave. In the case of OB one deals with two different states (characterized with low and high absorption, respectively), while in the case of multistability there are more than two states. It is possible to switch over any of them at the same parameters (frequency, polarization and intensity) of the incident wave. OB represents a common property of nonlinear optical systems with the feedback. Different processes [2–5] can be responsible for its realization in semiconductors, including a dynamic nonlinearity of optical absorption in the exciton frequency region (the so-called intrinsic OB) [7–9].

A series of theoretical works [10–14] has been devoted to investigation of different aspects of the nature of intrinsic OB. It has been found out that the nonlinearities, which promote OB under considerable light intensities, are explained by competition of processes responsible for the excitation of exciton states under illumination of crystal and their relaxation caused by exciton-phonon interactions [10]. Essential influence of external factors on the processes mentioned above enables one to control the OB region by changing the temperature of crystal [11], polarization of the excitation wave [12] or the external magnetic field [13]. Interactions with specific phonon modes of the type of bending wave (BW)

peculiar to layered crystals (LC) could promote indirect vertical phototransition responsible for arising of multistable states [14].

In this paper we study the role of nonlinear exciton-phonon interaction in the processes of OB realization in LC, the point that has not been sufficiently clarified in the works mentioned above. We reveal that the exciton scattering on the BW may have different efficiency and is essentially anisotropic. It influences greatly the position and size of the frequency and temperature regions where the OB and the hysteresis loop appear.

2. The model

Layered semiconductors typically manifest a considerable anisotropy of physical character-istics, e.g., the effective mass of charge carriers [12,13] and the optical spectra characteristics [13,14]. The difference in the forces of inter-action among the atoms inside the layer packet and the interlayer interaction is the cause of anomalies present in the phonon spectra of those crystals. It is responsible for appearance of specific types of oscillations, the low-energy optical phonons and the BW [13]. It is these oscillations that form the optical spectrum of LC in the exciton frequency region at low temperatures. This is evidenced by a large variety of warm-up changes in the exciton absorption band of LC, when compare with isotropic semiconductors.

First and foremost, many spectral characteristics of layered semiconductors, in contrast to isotropic ones, are dependent on temperature in the low-temperature region (see, e.g., the position and height of the absorption peak). Second, since the sign of the nonlinear exciton-phonon interaction constant depends on the light propagation direction with respect to crystallographic axis [16], the sign of the low-temperature shift of the exciton absorption maximum may change when the LC is heated. At the same time, the maximum shift occurring in isotropic crystals is always directed towards

the long-wave region [15]. The anomalous short-wave exciton band shift has been observed at low temperatures for different directions of light propagation in 2H-polytype PbI_2 [17]. It is conditioned by the fact that the sign of nonlinear exciton-phonon coupling function for the BW has an extra dependence on the relation between the effective electron and hole masses [16]. The lead iodide differs from the other LC by a strong anisotropy of these parameters $(m_e^{\parallel} > m_h^{\parallel}, m_e^{\perp} < m_h^{\perp})$, so that the sign of the coupling function is the same for both light propagation directions [16–18].

The distinctive features of the exciton spectrum dynamics in LC caused by the peculiarities of their exciton-phonon interactions could manifest themselves in the OB taking place in those crystals. In order to study this problem, we investigate the model for excitation of three-dimensional exciton with the quadratic dispersion law

$$\hbar\omega_{\vec{k}} = \hbar\omega_0 + \frac{\hbar^2 k^2}{2m_{ex}}$$

in the LC under its illumination, where $\vec{k}$, $\hbar\omega_{\vec{k}}$ and m_{ex} denote respectively the wave vector, the energy and the effective mass of the exciton. We account for the two possible relaxation mechanisms for the exciton excitation energy, related to the interactions with the BW $(\Omega_{\vec{q}} = \Omega_2 q^2)$ and dispersionless optical phonons $(\Omega_{\vec{q}} = \Omega_1)$. Here $\Omega_{\vec{q}}$ and $\vec{q}$ mean the phonon frequency and wave vector, respect-ively. The first interaction is nonlinear, with the coupling function [16,19]

$V_2(q,-q) =$

$$= \pm f_0 \Omega_2 m_0 \left(\frac{1}{m_h(1+\alpha_h q^2)^2} - \frac{1}{m_e(1+\alpha_e q^2)^2} \right) q \equiv (1)$$

$$\equiv f_0 \Omega_2 F_2(q) \ q$$

The second one is linear and we have [20]

$$V_1^2(q) = F_0 \Omega_1 \left[\frac{1-\alpha_e q^2}{(1+\alpha_e q^2)^2} - \frac{1-\alpha_h q^2}{(1+\alpha_h q^2)^2} \right]^2$$

$$\equiv F_0 \Omega_1 F_1(q). \qquad (2)$$

Here $\alpha_i = \dfrac{(\alpha_{ex} m_i)^2}{(2 \cdot (m_e + m_h))^2}$ ($i = e$, h and a_{ex} is the exciton radius), and f_0 and F_0 represent the constants of the interactions. The signs plus and minus in Eq. (1) correspond respectively to the cases of light propagation within the layer plane and perpendicular to it (see [16]).

Assuming a weak exciton-phonon coupling and excitation of the lowest exciton state only, we may express the light absorption coefficient at the frequency ω as

$$\alpha(\omega) = 2\pi \cdot |D_0|^2 \frac{\Gamma}{(\omega - \omega_0 - \Delta)^2 + \Gamma^2}. \qquad (3)$$

$$= 2\pi |D_0|^2 S(\omega)$$

Here D_0 is the exciton-photon interaction parameter, and Δ and Γ the real and imaginary parts of the Green's function mass operator [19]:

$$M(\omega,T) = M_1(\omega,T) + M_2(T),$$

where

$$M_1(\omega,T) =$$

$$= \frac{1}{N\hbar^2} \sum_{\vec{q}} V_1^2(\vec{q}) \left(\frac{1 + n_{\vec{q}} + N_{\vec{q}}}{\omega - \omega_{\vec{q}} - M_2(T) - \Omega_{\vec{q}} + i\eta} + \right. \qquad (4)$$

$$\left. + \frac{n_{\vec{q}} - N_{\vec{q}}}{\omega - \omega_{\vec{q}} - M_2(T) + \Omega_{\vec{q}} + i\eta} \right)$$

$$M_2(T) = \frac{1}{N\hbar} \sum_{\vec{q}} V_2(\vec{q}, -\vec{q})(1 + 2n_{\vec{q}}), \qquad (5)$$

$$\eta \to +0, \text{ and}$$

$$n_{\vec{q}} = \left[\exp\left(\frac{\hbar\Omega_{\vec{q}}}{k_B T} \right) - 1 \right]^{-1},$$

$$N_{\vec{k}} = \left[\exp\left(\frac{\hbar\omega_{\vec{k}} - \mu}{k_B T} \right) - 1 \right]^{-1}$$

are respectively the occupation numbers of the phonon and exciton states. The chemical potential μ of the excitons defines their concentration in the crystal (the volume V and the temperature T):

$$N = \frac{1}{V} \sum_{\vec{k}} N_{\vec{k}}. \qquad (6)$$

Eqs. (4) and (5) represent the contributions of the linear interactions of excitons with optical phonons and acoustic-type BW [21]. Replacing the sum over $\vec{q}$ by the integral over dimensionless variable $y = aq/\pi$ (a being the lattice constant), we obtain the real and imaginary parts of frequency-temperature dependences for the mass operator:

$$\Delta(\omega,T) = \Delta_1(\omega,T) + \Delta_2(T), \qquad (7)$$

$$\Gamma(\omega,T) = \Gamma_0 + \frac{\pi}{2} F_0 \Omega_1 [F_1(q_+)(n_1(T) - N_{q_+})$$

$$+ F_1(q_-)(1 + n_1(T) + N_{q_-})]. \qquad (8)$$

Here Γ_0 is the radiation width of the exciton level:

$$\Delta_1(\omega,T) = F_0 \Omega_1 [(n_1(T) - N_{q_+}) I(\omega + \Omega_1)$$

$$+ (1 + n_1(T) + N_{q_-}) I(\omega - \Omega_1)], \qquad (9)$$

$$\Delta_2(T) = f_0 \Omega_2 \int_0^1 [1 + 2n_2(y,T)] F_2(y) y^2 dy, \qquad (10)$$

$$q_\pm \equiv \sqrt{\omega - \omega_0 - M_2(T) \pm \Omega_1},$$

$$I(z) \equiv \int_0^1 \frac{F_1(y) y \, dy}{z - \omega_0 - \Delta_2 - L y^2},$$

$L = \pi^2 \hbar /(2a^2 m_{ex})$ is the exciton band width, $n_{1,2}$ and $N_{q\pm}$ the phonon (optical or BW) and the exciton ($k = q_\pm$) occupation numbers.

Thus, assigning definite values to the above parameters, one can calculate the absorption coefficient spectrum at arbitrary temperatures of crystal. In our case the parameters Δ, Γ and the absorption coefficient α are determined not only by the frequency ω and the temperature T, but also the absorption intensity of crystal at the given frequency. The correlation between the chemical potential μ of the exciton and the exciton concentration N in crystal causes N-dependence of the occupation numbers $N_{q\pm}$.

The occupation number itself is determined by the input light intensity I_0. Because of this fact, dependence of the output light intensity I on I_0 becomes nonlinear. This brings about the existence of certain frequency and temperature regions, where two different I values match one intensity value I_0, i.e., the OB would take place [10].

When the exciton, phonon and the radiation systems are in thermodynamically equilibrium state, the balance equation for the exciton concentration may be used [10]:

$$I_0\alpha(\omega, T, N) - N = 0 . \qquad (11)$$

By applying Buguer-Lambert law, one can enclose the system of Eqs. (1)–(11) and define the $I(I_0)$ dependence.

Assuming the exciton concentration to be variable (which is achieved by changing I_0), we can derive the absorption coefficient for the fixed frequency and temperature on the basis of equation system (3) and (7)–(11). Using then the Buguer-Lambert law, we find the output radiation intensity (or the transmission $\tau = I/I_0$) for the crystal of thickness d as a function of I_0. The nonlinear absorption may cause appearance of I_0 region (at the fixed ω and T), in which these parameters acquire two values [7,8]. The sizes of those OB regions may change with changing frequency, temperature and exciton-phonon interaction parameters.

3. Results of calculation and discussion

In order to study the influence of nonlinear exciton-phonon interaction with the BW on the position and size of frequency and temperature OB regions arising in LC, we now perform the calculations according to the model mentioned above.

As an example, we consider the most commonly studied 2H-polytype of PbI_2 [15], in which nonlinear exciton-phonon interaction with the BW has been detected [17]. The temperature dependence of the absorption band form function is presented in Fig.1. It has been calculated when taking the values $a = 4.557$Å, $\Omega_1 = 120\,\text{cm}^{-1}$, $\Omega_2 = 60$ cm^{-1}, $\varepsilon = 11.1$ [15], $m_e^{\parallel}$ = $1.12m_0$ and $m_h^{\parallel} = 0.22m_0$ [18] typical for this crystal in the case of light propagation along the layer plane and putting the parameters of the model to be equal to $L = 10^4$ cm^{-1}, $\Gamma_0 = 4.5$ cm^{-1}, F_0 = 0.35 and f_0 = 0.002. An anomalous absorption band shift towards higher energies is

clearly seen in the low-temperature region (up to 40 K). It takes place for the reason of nonlinear exciton-phonon interaction with the BW. According to Eq. (10), the shift in this region is proportional to the interaction constant f_0.
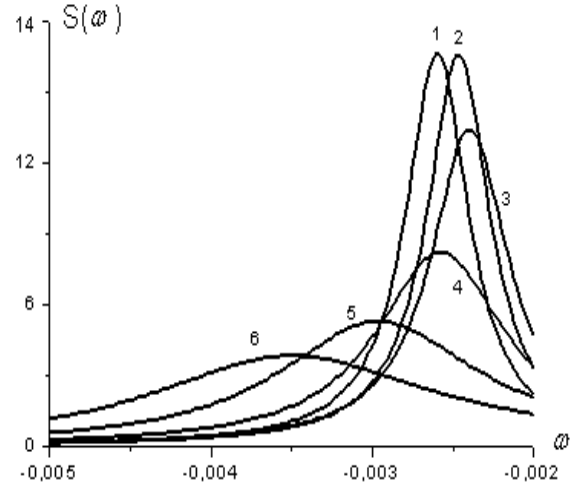

Fig. 1. Dependences of the function of exciton absorption band form S on the frequency ω represented in dimensionless units $\hbar(\omega - \omega_0)/L$ for different temperatures: 1 – 0 K, 2 – 20 K, 3 – 40 K, 4 – 60 K, 5 – 80 K and 6 – 100 K.

We have also analyzed the results of calculations performed at different f_0 values in the region of frequency changes (i.e., at fixed temperatures) and the temperatures, for which the OB takes place (i.e., at some fixed reference frequency ω_s chosen in the middle of the OB region, whose value depends on f_0). It has been found that the increase in the BW coupling function leads to:

- a short-wave shift of the frequency OB region $\Delta\omega = \omega_2 - \omega_1$ and a decrease of its size (see Table 1);

- an increase in the temperature OB region $\Delta T = T_2 - T_1$ (see Table 1);

- a shift of the hysteresis loop towards the region of higher intensities, as well as a decrease in its height and width (see Fig. 2).

The increase in the reference frequency of OB with increasing f_0 constant (Table 1) is explained by the shift of exciton band maximum towards the short-wave region, as a result of

exciton-BW interaction. In a similar manner, the short-wave shift of the frequency OB region and the decrease of its size in isotropic crystals cause a decrease in the linear exciton-phonon interaction constant F_0 [10]. From this standpoint, we can assert that the influence of BW on the conditions for observation of OB is opposite to the influence of optical phonons.

Interactions with the BW are also responsible for decrease in the minimum temperature, for which the OB appears. Linear mechanism of exciton-phonon interaction, as a channel for dissipation of exciton energy in 2H-PbI$_2$ that promotes the appearance of OB, becomes actual at $T \geq 40$ K [17]. But the low-energy oscillations of the BW-type get excited at lower temperatures. Therefore the strongerthe interaction of excitons with these oscillations, the lower the temperature of OB appearance is. We notice that the frequency region where OB is possible in 2H-PbI$_2$ crystal is rather narrow (1.7–3.5 cm^{-1}), like in the case of HgI$_2$ crystal. The linear exciton-phonon interaction in these crystals is characterized by a similar value of F_0 constant [9], though the OB in HgI$_2$ has been observed at lower temperature (25 K; its decrease produces decreasing $\Delta\omega$ [7]). Nonetheless, this temperature is still higher than the excitation point of optical phonons in this crystal (12 K). This indicates that the nonlinear exciton-phonon interaction in HgI$_2$ is weaker than that in PbI$_2$, whose structure is "more layered" [17].
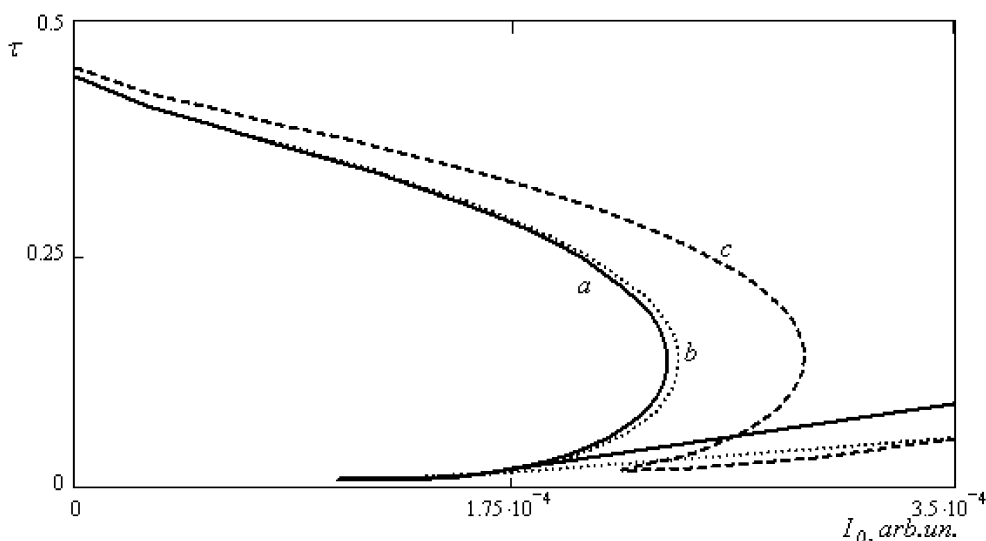

Fig.2. Dependence of optical transmission at the corresponding reference frequency (see Table 1) on the input light intensity for different values of nonlinear exciton-phonon interaction parameter f_0: 0.0003 (*a*), 0.003 (*b*) and 0.01 (*c*).

Table 1. The limits for the frequency and temperature OB regions in PbI$_2$ ($T = 45$ K).

f_0	w_1	w_2	w_s	T_1, K	T_2, K
0.0003	-0.00348	-0.00313	-0.00335	39	110
0.003	-0.00345	-0.00310	-0.00331	37	115
0.01	-0.00318	-0.00301	-0.003085	35	117

4. Conclusions

The temperature region where the OB appears in the LC with efficient exciton scattering by the BW is notably wider, when compare with the corresponding region for isotropic crystals. This allows one to suggest that the cavityless intrinsic OB should be more accessible for the former crystals, though at the cost of narrower frequency region.

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