
Polarizable Optical Bistability of Frenkel Excitons

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Abstract

Optical spectra of molecular crystals with two molecules in their elementary cell are investigated. Depending on the polarization angle one or two exciton excitation bands can be realized in the spectrum. The bands are separated by the terminal frequency interval. The exciton absorption bands genesis and the peculiarities of the function of their form at the change of the laser radiation polarization have been analyzed and the regularity of arising bistable states in organic benzol-type crystals has been established.

Keywords: exciton, molecular crystal, optical bistability

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1. Introduction

One of the most prospective ways of creating optical information memory elements is the realization of the optical bistability (OB) phenomenon [1]. The principal possibility of obtaining a hysteresis loop in the incoming – outgoing light intensity dependence for the semiconductor exciton frequency region is of no doubt [2]. The mechanism of arising of two or more bistable states in a crystal can vary, but the most important thing is to determine the mechanism of effecting both the width and the height of the hysteresis loop, that is the influence of OB formation on the nonlinear process. Both intrinsic and external factors are possible, such as temperature, magnetic field, semiconductors with different exciton frequency range, laser wave polarization and others. We shall investigate the passage of the light wave through organic semiconductors of the molecular crystal type. The special feature of such crystals is the realization of the neutral electron excitation – the Frenkel exciton. In

semiconductors, which contain σ molecules in their elementary crystal cells, the electron excitation of one isolated molecule generates σ bands (Davydov-splitting) [3]. Each of these bands (which is very important) is characterized by a certain direction of the exciton transition dipole moment and is realized only at a certain polarization of the external light excitation wave. Obviously if there is a strict polarized radiation of a given direction only one specific isolated band of exciton excitations will be realized in the absorption spectra. The depolarization of the external excitation wave causes a shift of all available exciton bands and the realization of the structured absorption band with a complex dynamics of its form function. The aim of our investigation is to calculate the exciton absorption polarization genesis, its influence on the formation and concrete parameter of the hysteresis loop in the height of the pumping level and exciton density state dependence and the arising of the OB phenomenon itself.

2. Exciton-Phonon Interaction in Molecular Crystals

The Frenkel Exciton can be best investigated in molecular crystals, in which the forces of interaction between the combining molecules are much smaller than the forces of the intermolecular interaction. A number of solids, formed by big molecules of organic substances – anthracene, pentacene, naphthalene, benzol and others belong to such crystals.

We shall consider a molecular crystal, which has two molecules in its elementary cell with different dipole moments of transforming into an excited state. In this case the crystal exciton excitation energy will consist of two energy bands

$$H_{ex} = \sum \sum E_{\alpha}(\vec{k}) B_{\alpha k}^{+} B_{\alpha k}, \quad (1)$$

where B_k^{+}, B_k - are operators of creation and annihilation of excitons with the quasi-momentum \vec{k} . So two exciton excitation bands correspond to one non-degenerate excited state of a free molecule in a crystal. The energy of these excitons in the case of two identical molecules in a crystal elementary cell will be [3]:

$$E_1(\vec{k}) = \Delta\varepsilon + D + L_{11}(\vec{k}) + L_{12}(\vec{k}) \quad (2)$$

$$E_2(\vec{k}) = \Delta\varepsilon + D + L_{11}(\vec{k}) - L_{12}(\vec{k}) \quad (3)$$

Here $\Delta\varepsilon$ – is the energy of molecular excitation, D – is the variation of intermolecular interaction under molecular excitation, $L_{\alpha\beta}$ is the matrix of the resonance interaction, and $\Delta E = 2L_{12}(0)$ – is the energy of the resonance exciton band splitting. These energy bands can be correlated with corresponding specific transition dipole moments in crystals \vec{d}_1 and d_2 . In spite of the fact that exciton excitations in a crystal with two molecules in an elementary cell belong to one energy level in a free molecule, they differ not only in their energy, but in their polarization as well. Their polarization is perpendicular in its direction, that is $\vec{d}_1 \perp \vec{d}_2$. The value of Davydov splitting ΔE is different for

different crystals and varies between 575 cm^{-1} in the anthracene crystal and 40 cm^{-1} in the benzol crystal.

The energy of the crystal exciton excitation (1) is the same as when the crystal molecules are in a steady-state condition in the crystal lattice. In order to find the energy of interaction excitons and crystal lattice oscillations it is necessary to expand the operator of the exciton excitation energy as a series in little shifts of the molecules from their equilibrium state. Then for the full excitation energy of the crystal in which excitons and phonons interact at the same time we obtain:

$$H = H_{ex} + H_{ph} + H_{int}, \quad (4)$$

$$H_{int} = (N)^{-1/2} \sum \sum \sum F_s(\vec{k}, \vec{q}) B_{k+q,s}^{+} B_{rs} (b_{qs} + b_{-qs}^{+}) \quad (5)$$

Here b_{qs}^{+}, b_{qs} are operators of production and destruction of phonons of the s -branch with the energy $\Omega_s(q)$ and quasi-momentum \vec{q} , F_s – is the function, which shows the interaction of the phonons of the s -branch with the crystal exciton excitation. H_{int} operator describes the elementary excitations of the crystal, which are characterized by a certain number of independent excitons and phonons. The operator of interaction (5) does not change the total number of excitons in the system, but only causes the transition of excitons from one sublevel of the zone to another one, absorbing or irradiating phonons at the same time. Thus, this interaction leads to the process of exciton scattering on phonons employing the laws of conserving energy and quasi-momentum.

The calculation of the function of exciton phonon bond in the general case of three-dimensional crystals involves great mathematical difficulties. In simple cubic isotropic crystals, when taking into consideration the interaction only between the neighboring molecules, we use approximate expressions. In particular for interaction with an optical dispersion less phonon [3]:

$$F_{opt} = f_0 \cos(ka + \frac{qa}{2}) \cos(\frac{qa}{2}). \quad (6)$$

2. The Form Function of the Exciton Absorption Band

It is customary that the theory of exciton optical spectra is based on such a simple model scheme: when a light quantum is absorbed a new exciton is born (a direct phototransition), and then the exciton-phonon interaction is dealt with (linear with respect to the phonon operator), which determines the finite life of an exciton with a given energy and quasi-momentum. In this model the light absorption coefficient is determined by the probability of crystal transition from the ground state to the exciton state, and is related to the correlation exciton function [4]:

$$K(N, \omega, \varphi) = \sum D_\alpha^2 \int \exp(i\omega t) \langle B_Q(t) B_Q^\dagger(t) \rangle dt = 2\pi \sum D_\alpha^2 S_\alpha(N, \omega, \varphi) \quad (7)$$

Here the time dependence is put down in the Heisenberg Presentation, and it is averaged over the full Hamiltonian of the crystal (4). The value $S(\omega)$ is called the isolated exciton absorption band form function. D_α – is the matrix element of the interaction of light and crystal, whose value depends on the angle (φ) between the polarization wave \vec{e}_Q and the dipole moment \vec{d}_α of the crystal transition into the given exciton state

$$D_\alpha = D_0(\vec{e}_Q \vec{d}_\alpha) = D_{0\alpha} \cos \varphi_\alpha. \quad (8)$$

Hence it follows that the interaction of light with crystal is maximum, when the photon polarization vector is parallel to the given dipole moment of the quantum transition ($\varphi_\alpha = 0$). The Green function method allows us to figure out in detail the exciton light absorption coefficient on the basis of the deduced exciton-phonon system mass operator:

$$S_\alpha(\omega, Q) = \Gamma_\alpha(\omega, Q) \{ [\omega - E_\alpha(Q) - \Delta_\alpha(\omega, Q)]^2 + \Gamma_\alpha^2(\omega, Q) \}^{-1}, \quad (9)$$

where $M_\alpha(\omega, Q) = \Delta_\alpha(\omega, Q) - i\Gamma_\alpha(\omega, Q)$ – is the mass operator, which in the general case is the complex frequency function.

Thus, the outline (circuit) of the exciton absorption curve with a weak exciton-phonon bond is described by a quasi-Lorentz type function with a width determined by the imaginary part of the mass operator. The calculation of the mass operator is performed in detail in [4]:

$$M(\omega, Q) = \sum \sum |F(Q, q)|^2 \{ (n_{qs} - N_{Q+q}) \times [\omega - E_\alpha(Q+q) + \Omega_{qs} + i\tau]^{-1} + (1 + n_{qs} + N_{Q+q}) \times [\omega - E_\alpha(Q+q) - \Omega_{qs} + i\tau]^{-1} \} \quad (10)$$

Here $N_{\alpha Q}$ and n_{qs} are the number of exciton of the α -zone and phonons of the s -branch at a given crystal temperature T:

$$N_{\alpha Q} = \left\{ \exp\left(\frac{E_\alpha(Q) - \mu}{k_B T}\right) - 1 \right\}^{-1}, \quad (11)$$

$$n_{qs} = \left[\exp\left(\frac{\Omega_{qs}}{k_B T}\right) - 1 \right]^{-1}, \quad (12)$$

where k_B – is the Boltzmann constant; T – is the crystal temperature in Kelvin units; μ – is the exciton chemical potential, whose value is conditioned by the exciton concentration N_{ex} in a crystal at a given temperature:

$$N_{ex} = \frac{1}{V} \sum N_{\alpha Q}. \quad (13)$$

3. Light Absorption in a Benzol Crystal

The experimental evidence of arising exciton states in molecular crystals as the availability of two or more exciton absorption bands for different polarization's of the incident light wave.

Thus, for investigating Frenkel excitons we must take polarization spectral measurements at first. The splitting of non-degenerated molecular terms in polarized light absorption spectra is registered in a number of experimental works and for different molecular crystals. These are crystals formed by the aromatic benzol molecule (C_6H_6), anthracene ($C_{14}H_{10}$), naphtacene ($C_{18}H_{12}$), pentacene ($C_{22}H_{14}$) naphtalene and others. The form and the nature of exciton absorption bands

in molecular crystals is different. Anthracene and naphthalene spectra have wide intensive bands, and benzol zones have much narrower and weaker intensive bands. Then we shall dwell on experimental results devoted to exciton absorption in benzol crystal [5].

The availability of four electron excitation levels is typical for benzol molecules. The first $\Delta\varepsilon_1 = 4.722$ eV – is a pure electron transition. The second $\Delta\varepsilon_2 = 4.837$ eV, the third $\Delta\varepsilon_3 = 4.952$ eV and the fourth $\Delta\varepsilon_4 = 5.507$ eV – are one-fold, two-fold and three-fold combinations of totally symmetric intramolecular vibrations (0.115 eV) with an electron transition. In crystal absorption spectra four bands in one polarization are obtained accordingly, and four bands in the perpendicular polarization are obtained. They are with the following spectral characteristics: the first one with the energy $E_1 = 4.687$ eV and with the value of the resonant polarizable splitting band $\Delta E_1 = 4$ cm⁻¹; the second one $E_2 = 4.401$ eV and $\Delta E_2 = 44$ cm⁻¹; the third one $E_3 = 4.918$ eV, $\Delta E_3 = 23$ cm⁻¹, the fourth $E_4 = 5.033$ and $\Delta E_4 = 15$ cm⁻¹.

For the sake of theoretical treatment of the experimentally obtained results we choose one of the bands, namely the first one, for which the polarization splitting is 40 cm⁻¹, and in whose spectrum there are two absorption bands (a- and c- components) with orthogonal polarization. Thus the crystal electron excitation will have two exciton excitation bands, which according to (2) and (3) are characterized by the energies:

$$\begin{aligned} E_1 &= E_{01} + L_1(1 - \cos y) = E_0 + Ly^2 \\ E_2 &= E_{02} + L_2(1 - \cos y) = E_0 + \delta + \eta Ly^2 \end{aligned} \quad (13)$$

Here we introduced the following designations: E_0 – is the bottom of the exciton band from which we start the reckoning of the crystal excitation energy, L – is the width of the first exciton excitation bands, δ – is the exciton resonant splitting value, ηL – is the width of the

second exciton excitation band, $y = ka/\pi$ ($0 < y < 1$). Taking into consideration that for the visual and ultra-violet light (it is in this spectral region that exciton excitation energy is present) the product $Qa \ll 1$, hereinafter we shall neglect the photon wave vector and take it as zero ($Q = 0$).

Two different transition dipole moments correspond to two-exciton excitation bands, thus the interaction of light with the exciton excitation of each band, according to Eq. (8), will be different:

$$\begin{aligned} D_1 &= D_{01} \cos \varphi = D_0 \cos \varphi ; \\ D_2 &= D_{02} \cos(\varphi - \frac{\pi}{2}) = \xi D_0 \sin \varphi, \end{aligned} \quad (14)$$

where D_0 stands for the force value of the oscillator of crystal transiting into the first exciton state, and ξD_0 – for the force value of the oscillator of crystal transiting into the second exciton state, and the polarization angle $0 < \varphi < \pi/2$.

For the introduced designations the crystal absorption light coefficient on the frequency ω will be

$$\begin{aligned} K(N, \omega, \varphi) &= 2\pi \sum D_\alpha^2 S_\alpha(N, \omega) = \\ &= 2\pi D_0^2 [\cos \varphi S_1(N, \omega) + \xi \sin \varphi S_2(N, \varphi)] = \\ &= 2\pi D_0^2 [\alpha_1(N, \omega, \varphi) + \alpha_2(N, \omega, \varphi)] \end{aligned} \quad (15)$$

As it is evident from Eq. (15), the exciton band consists of two components – α_1 and α_2 , whose ultimate form function of the absorption curve will depend on the polarization angle of the incident wave. We can but argue that at the value of the angle $\varphi = 0$ there will be only the first band in the spectrum (the one with the smaller exciton energy), and when $\varphi = \pi/2$ only the second exciton absorption band will appear.

In order to investigate this particular view of the anthracene crystal absorption spectrum, we shall consider the case of weak exciton densities ($N \ll 1$) are calculate the absorption coefficient (15) for certain values of the parameters. It is evident that the most effective way of exciton energy relaxation is achieved by interaction with

optical phonons. Thus, hereinafter we shall restrict our consideration only to processes of excitons scattering from optical phonons of the Einstein model $\Omega_s(q) = \Omega_0$. We pass on in Eq. (10) from the summation of the quasi-momentum of the phonon q to the integrating over the immeasurable parameter $y = qa/\pi$, and put down all energy values in the width units of the first exciton band (e.g. $\Omega = \Omega_0/L$, $f_0 = f_{opt}^2/L$, $w = (\omega - E_0)/L$, $\delta = \delta/L$ and so on). Using the equation

$$\lim_{\tau \rightarrow 0} (x + i\tau)^{-1} = P \int \frac{1}{x} - i\pi\delta(x),$$

and also equations (6), (10) and (13) we obtain for the imaginary part of the mass operator the following expressions:

$$\Gamma_1(0, w) = \pi f_0 \left[J(q_{1+}) \frac{n_0}{2q_{1+}} + J(q_{1-}) \frac{1+n_0}{2q_{1-}} \right] \quad (16)$$

$$\Gamma_2(0, w) = \frac{\pi f_0}{\eta} \left[J(q_{2+}) \frac{n_0}{2q_{2+}} + J(q_{2-}) \frac{1+n_0}{2q_{2-}} \right] \quad (17)$$

In these expressions we introduced $q_{1\pm} = (w \pm \Omega)^{-1/2}$, $q_{2\pm} = [(w - \delta \pm \Omega)/\eta]^{-1/2}$. Having performed similar changes for the real part of the mass operator as well we get the following expressions:

$$\Delta_1(0, w) = f_0 \{ n_0 I(w + \Omega) + (1 + n_0) I(w - \Omega) \} \quad (18)$$

$$\Delta_2(0, w) = \frac{f_0}{\eta} \left\{ n_0 I \left(\frac{w - \delta + \Omega}{\eta} \right) + (1 + n_0) I \left(\frac{w - \delta - \Omega}{\eta} \right) \right\} \quad (19)$$

$$n_0 = \left[\exp \left(\frac{\Omega}{\theta} \right) - 1 \right]^{-1}, \quad (20)$$

where we introduced the following abbreviated designations for the crystal temperature $\theta = k_B T/L$ and for the integrals

$$I(x) = P \int J(y) (x - y^2)^{-1} dy \quad (21)$$

$$J(y) = (1 + \cos y)^2 y^2 \quad (22)$$

In the expression (21) the sign P shows that the integral is taken in its principal meaning, i.e. the numerical value of the integral is determined not by the area under the subintegral function, but by its meanings in the integrating circles ($y=0, y=1$). Hence it follows that in spite of the occurrence of subintegral function poles, all integrals have an ultimate numerical value. In order to carry out the final calculations of the absorption band we use the following parameters, unique to the benzol crystal: $L=10^3 \text{ cm}^{-1}$, $\Omega_0 = 120 \text{ cm}^{-1}$, $\delta = 40 \text{ cm}^{-1}$, $\eta=0.7$, $\xi = 0.83$, $f_0 = 0.02$. The crystal temperature is chosen to

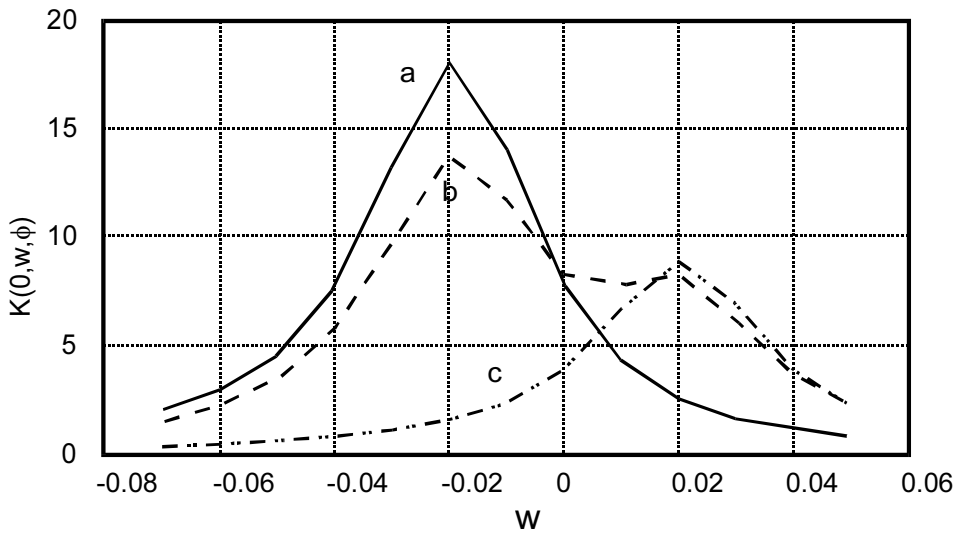


Fig.1. The exciton absorption spectra at temperature $T=180 \text{ K}$ for three polarization angles: a) $\varphi = 0$; b) $\varphi = \pi/4$; c) $\varphi = \pi/2$

be equal to 77 K ($\theta=0.054$) and 180K ($\theta=0.12$), we choose three values of polarization angles φ ($0, \pi/4, \pi/2$). Fig. 1 and Fig.2 show the calculation results of the immeasurable absorption coefficient $K_{im} = 2\pi D_0^2 / f_0 L^2$ for different angles and temperatures.

The analysis of the calculations shows that there is a considerable variation in the vector direction dependence of the light wave polarization in exciton optical spectra. If the polarization angle $\varphi = 0$ (the curve a in Fig.1), i.e. the polarization vector coincides with the direction of the dipole moment \vec{d}_1 the exciton absorption band is clearly realized at the frequency equal to the energy of the lower exciton zone. In this case the band shape has a traditional short-wave asymmetry. For the polarization angle $\varphi = \pi/2$ (the curve c on Fig. 1) with the light wave polarization vector directed along the dipole moment \vec{d}_2 , there is only one band in the absorption spectrum at the frequency E_2 . This band has a lower intensity of the absorption peak and a changed asymmetry sign (the long-wave wing of the absorption curve is increased). In the case of an intermediate polarization the exciton band becomes weakly structured (the curve b in Fig. 1) and has no well-defined outlines of individual exciton lines. As is shown by the results of our calculations (Fig. 2) a significant temperature decrease allows us to get rid of this drawback, and then in absorption spectra at arbitrary intermediate polarization angles both components of Davydov exciton doublet are obtained simultaneously. It is worth noting that the size of Davydov splitting is also temperature-dependent. It increases ($\delta(180K) = 0.036$, $\delta(77K) = 0.038$) with the decrease of temperature and, obviously, at the temperature of an absolute zero reaches its peak ($\delta = 0.04$).

4. Bistable States of Frenkel Exciton

At a weak intensity of the excitation light wave the exciton density can be ignored and the absorptivity will be only be the function of temperature, frequency, and incident wave polarization. In this case the absorptive power of the medium is formed only by the processes of phonon relaxation of the exciton excitation. The increases of the excitation wave intensity leads to the increase of the exciton states density. As this takes place the response of the system to the external irradiation will also depend upon the occupation numbers of exciton states (7) whose value, in its turn, will be determined by the external irradiation intensity.

When an intense light wave interacts with a crystal the imaginary part of the mass operator will be determined by the expression:

$$\Gamma_1(N, w) = \pi f_0 \left\{ J(q_{1+}) \frac{n_0 - N_{q_{1+}}}{2q_{1+}} + J(q_{1-}) \frac{1 + n_0 + N_{q_{1-}}}{2q_{1-}} \right\}, \quad (23)$$

$$\Gamma_2(N, w) = \pi \frac{f_0}{\eta} \left\{ J(q_{2+}) \frac{n_0 - N_{q_{2+}}}{2q_{2+}} + J(q_{2-}) \frac{1 + n_0 + N_{q_{2-}}}{2q_{2-}} \right\}. \quad (24)$$

And for the real part of the mass operator, in contrast to Eq. (18) and Eq. (19), we obtain the following expressions:

$$\Delta_1(N, w) = \Delta_1(0, w) + f_0 \left[-I_N(w + \Omega) + I_N(w - \Omega) \right], \quad (25)$$

$$\Delta_2(N, w) = \Delta_2(0, w) + \frac{f_0}{\eta} \left[-I_N\left(\frac{w - \delta + \Omega}{\eta}\right) + I_N\left(\frac{w - \delta - \Omega}{\eta}\right) \right] \quad (26)$$

in which the value of the integral is defined as

$$I_N(X) = P \int J(y) N_y (X - y^2)^{-1} dy. \quad (27)$$

The occupation numbers of excitons are calculated, using Eq. (11) and Eq. (13)

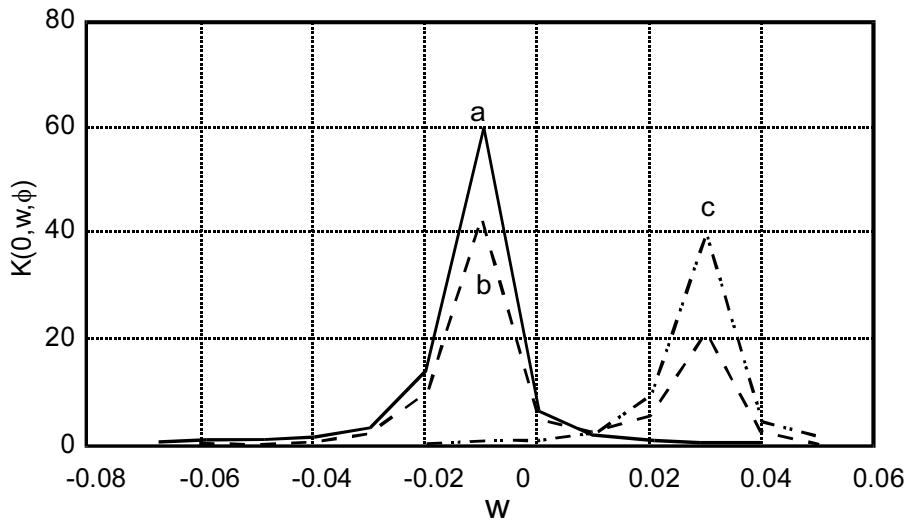


Fig.2. The exciton absorption spectra at temperature $T = 77$ K for three polarization angles: a) – $\varphi = 0$; b) – $\varphi = \pi/4$; c) – $\varphi = \pi/2$.

$$N_y = \left[A \exp\left(\frac{y^2}{\theta}\right) - 1 \right]^{-1}, \quad (28)$$

$$N_{ex} = \left[\pi \theta^{3/2} \nu^{-1} \right] \sum A^{-n} n^{-3/2}.$$

In order to calculate the exciton chemical potential we take into consideration that the level of the chemical potential cannot be higher than the bottom of the exciton band ($A \geq 1$). This means that the maximum allowable number of excitons at the given temperature will be $N_{ex}^{max} = N_{ex}$ ($A = 1$). Thus to determine the value A we may write down the following equation

$$N_{ex} = N \cdot N_{ex}^{max} \quad (29)$$

where the value N varies from 0 to 1, and for each of its significant figures there is only one particular value of the chemical potential and of the value A .

It is evident that in the case of an external exciton pumping their real density will depend on the laser radiation power. In this case the distribution of exciton states will differ from the condition of thermodynamic equilibrium, and their density will be the function of temperature and the excitation wave intensity. In order to take into consideration this factor when investigating the absorptive power of a crystal

we must put down the energy balance equation as well, which determines the amount of the absorbed energy for creating the desirable density of the exciton states

$$I \cdot K(N, w, \varphi) = N \quad (30)$$

In this expression $I = I_0 / RN_{ex}^{max}$ is the value, related to the laser radiation intensity I_0 [W/m^2] and the coefficient of exciton recombination R [W]. The exciton absorption coefficient is the exciton density function, the function of crystal temperature and laser wave polarization function. Having fixed three of them, for instance, w, θ, φ , we determine the exciton dependence of the absorption. It turns out, that there is an exciton frequency region w_s , for which the function $K(N)$ has a bell-like character and in this case Equation (30) may have three solutions [6]. It should be noted that for pure phonon mechanisms of exciton energy relaxation the frequency region w_s is very narrow. Fig.3 presents the excitation wave intensity dependence of the exciton density states on the frequencies of the first exciton band $w_{1s} = 0.01275$ and of the second exciton band $w_{2s} = 0.02725$. We see that there is a hysteresis loop, which causes the arising of two stationary states with an assigned exciton density at the same laser radiation intensity. The

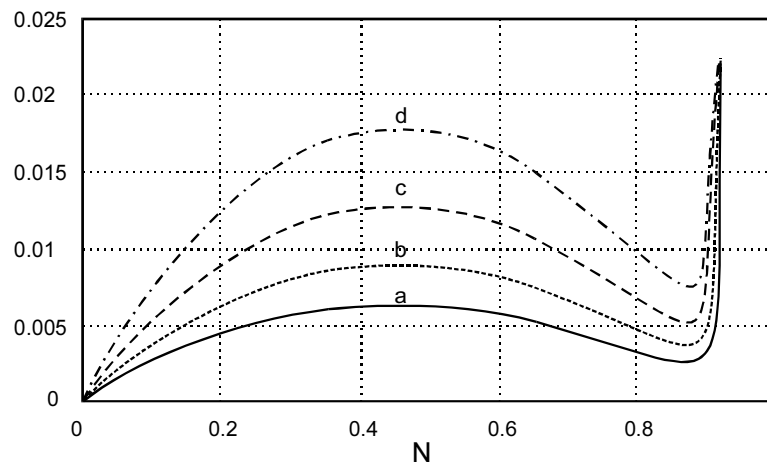


Fig.3. The excitation wave intensity dependence of the exciton density states for different polarization angles: $\varphi = 0$ (a) i $\varphi = \pi/4$ (b) at the frequency $\omega = -0,01275 \tau$ a $\varphi=\pi/2$ (c) and $\varphi=\pi/4$ (d) at the frequency $\omega = 0,02725$

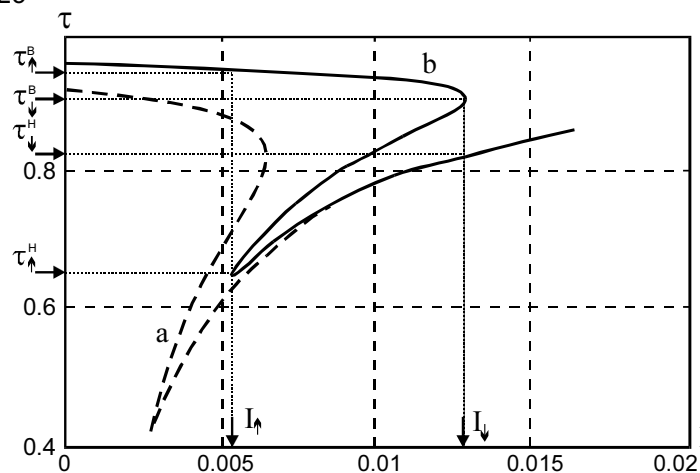


Fig.4. The excitation wave intensity dependence of the light transmission coefficient at frequency $\omega = -0,01275$ for the polarization angle $\varphi = 0$ (a) and at frequency $\omega = 0,02725$ for the angle $\varphi=\pi/2$ (b).

depolarization of the external laser wave at the lower exciton frequency (the increase of the angle φ from 0 to $\pi/2$), which causes the decrease of the absorption peak height, results in the shift of the hysteresis loop into the direction of the high intensities and to the enlarging of the hysteresis loop, and thus causes the increase of OB regions on the intensity scale. In conclusion it may be said that the possibility of obtaining bistable states with the decrease of the crystal absorptive power requires the increase of the incident laser wave intensity. It is also confirmed by the shift of the hysteresis loop when converting the laser wave to the orthogonal polarized absorption at the frequency of the upper exciton band (Fig. 3b) (whose absorption peak height is much smaller). It

should be noticed that the decrease of the excitation wave frequency causes the decrease of the bistable states regions on the scale of the exciton density states N .

Having obtained the expression $N(I)$ we can count a new absorption coefficient and determine the incident wave intensity dependence $K(I)$ of the absorption. Using the calculated data as the base we find also the absorption coefficient (or the ration of the output light intensity to the input light intensity) for the crystal with the thickness d :

$$\tau = \exp[-K(I) \cdot d]. \quad (31)$$

The results of the calculations by Eq. (31) for the given parameters (the thickness of the crystal $d=0.5 \times 10^{-4} \text{ cm}^{-1}$), are shown in Fig.4. We see that the realization of the OB

Table 1. The characteristic parameters of the hysteresis loop in the dependence $\tau(I)$

| The lower exciton band $w = -0,01275$ | | | | | | |
|---------------------------------------|-----------------------|------------------|-----------------------|------------------|--------|--------------|
| φ | τ_{-}^{B} | $\Delta\tau_{-}$ | τ_{-}^{B} | $\Delta\tau_{-}$ | I- | ΔI_s |
| 0 | 0,910 | 0,491 | 0,825 | 0,155 | 0,0026 | 0,0037 |
| $\pi/6$ | 0,918 | 0,446 | 0,867 | 0,158 | 0,0031 | 0,0043 |
| $\pi/4$ | 0,932 | 0,390 | 0,941 | 0,190 | 0,0038 | 0,0052 |
| The upper exciton band $w = 0,02725$ | | | | | | |
| φ | τ_{-}^{B} | $\Delta\tau_{-}$ | τ_{-}^{B} | $\Delta\tau_{-}$ | I- | ΔI_s |
| $\pi/2$ | 0,955 | 0,308 | 0,921 | 0,09 | 0,0053 | 0,0087 |
| $\pi/4$ | 0,965 | 0,230 | 0,940 | 0,07 | 0,0075 | 0,0109 |
| $\pi/6$ | 0,971 | 0,167 | 0,955 | 0,05 | 0,0108 | 0,0151 |

phenomenon is possible in both orthogonal polarization's of the excitation laser wave. It is also confirmed that with the decrease of the crystal absorptive power at the upper exciton wave frequency the OB realization is only possible with the increase of the laser wave intensity.

The hysteresis loop transformation regularities in the dependence $\tau(I)$ with the change of the polarization angle are tabulated in the table 1. Basically, they amount to the following. The depolarization of optical bistable bands, irrespective of their excitation frequency, results in the decrease of the hysteresis loop height $\Delta\tau_{\uparrow}$, and the decrease of the interval values of the weak-strong absorption switching, it shifts the hysteresis to the higher intensity region I_{\uparrow} and enlarges its loop (ΔI_s), which causes the increase of the region of OB realization at the cost of the increase of the excitation wave intensity. However there are variations in the dynamics of the non-linear absorption development in different polarization's. The change of the weak-strong absorption-switching interval $\Delta\tau_{\downarrow}$ has different signs: for the lower exciton band the switching interval increases with the increase of the depolarization and for the upper exciton band it decreases. Clearly, it involves different ratios of decreasing the height of the peak in different polarization's.

5. Conclusions

1. Under high levels of pumping the process of exciton absorption becomes nonlinear: with the change of the excitation wave intensity threshold values of the exciton density emerge at which a jump-like transition from the weak absorption state into the strong-absorption state takes place.
2. A hysteresis loop arises in the excitation wave intensity dependence of the crystal transmission power which results in the realization of optical bistability in the given crystal on the given excitation frequency.
3. The width of the optical hysteresis (the distance in intensity between the transitions of weak-strong and strong-weak absorption) is influenced only by the outer laser wave – the width of the hysteresis range decreases sharply with the frequency reaching the absorption band maximum.
4. The height of the hysteresis loop (the difference between the strong and weak absorption is essentially influenced by the depolarization value – the height is maximum at strictly polarized absorption.

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