INTERNAL OPTICAL BISTABILITY OF QUASI-TWO-DIMENSIONAL SEMICONDUCTOR NANOHETEROSTRUCTURES

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Abstract: *We represent the results of numerical computations of the frequency and temperature domains of possible realization of internal optical bistability in flat quasi-two-dimensional semiconductor nanoheterostructures with a single quantum well (i.e., nanofilms). Particular computations have been made for a nanofilm of layered semiconductor PbI_2 embedded in dielectric medium, i.e. ethylene-methacrylic acid (E-MAA) copolymer. It has been shown that increasing thickness of a nanofilm results in the long-wavelength shift of the frequency interval of realization of bistability, increasing size of the hysteresis loop, and broadening temperature interval of realization of this phenomenon.*

KEYWORDS*: OPTICAL BISTABILITY, EXCITON, NANOSYSTEM, NANOFILM, EXCITON-PHONON INTERACTION.*

1. Introduction

Along three last decades, attention of many researchers is attracted boy searching for the materials for development of pure optical systems for registration, storing, retrieval and transmission of information as well as for development of the devices for data processing (optical computers) [1, 2]. The advantages of the use of an optical radiation for the mentioned purposes consist in exploiting the carriers much higher than ones for radio waves that results of considerable broadening the frequency range and, as a consequence, data transmission rate. Optical systems are applicable for implementation of parallel data processing as their ensure implementation not only the temporal light modulation but the space modulation, including independent modulation of the part of a signal. Besides, such systems provide high density of data recording using miniature optical storage devices. One can, in principle, to design on the base of the single technological process such integrated-optics devices as laser sources, modulators, thin-film waveguides, beam splitters, logical storing device, what opens the perspectives for design of an optical computer.

Creation of a pure optical information system presumes the solving of two practically important problems, viz., development of the optical techniques for data transmission and for data storage and processing. The first of these problems (replacing the transmission of EM pulses in wire-, radio- and TV nets by the transmission of optical signals into fiber-optical systems) has found up to now comprehensive technological solution. Technology of fiber-optical systems with low optical losses and high-efficient techniques of inputing the light pulses from laser sources into the processor has stipulated impetuous progress of fiber optics telecommunications of high information capacity.

Emergence of optical computers (in form and in time) depends on the choice of physical processes on the base of which the optical logical device will be created. The prospect of design of such device is connected with the use of the phenomenon of optical bistability (OB) [4] revealing the feasibilities for control of optical radiation and ensuring implementation in the optical domain of the analogues of various electronic devices, such as storing cells, transistor, discriminator, switching etc. [4-6].

Bistability is the phenomenon inherent in wide class of matters and devices, based on the nonlinearity of the system's response on intense action of external EM field. Under interaction of EM wave with any resonant nonlinear system, this system (under certain conditions) occurs in one of two steady (dynamically stable) states differing in the magnitude of absorption of the exciting field energy. The systems where this phenomenon takes place are referred to as bistable ones.

OB is one of the manifestations of the mentioned phenomenon being a more general property of a nonlinear optical system with the feedback. Observability of this phenomenon has been predicted in [6] and firstly demonstrated by McCall et al. observed the presence of the hysteresis anomaly in transmittance of laser radiation of a resonator filled by sodium vapor under low pressure [7]. The essence and variety of this phenomenon have been studied in numerous theoretical and experimental papers (see, for example, [4, 8-15]). Searching for the media where OB is realized with the minimal energy consumption and the rate of switching between two states, investigation of the mechanisms of nonlinearity favorable for this, as well as influence of external factors on realization of this phenomenon are continuing [16-23]. Following to the report on design of an optical processor [24], the interest to OB increased considerably, and the problem of searching for new physical mechanisms for realization of an OB and media for implementing it was actualized.

Peculiar feature of optical absorption in semiconductors is caused by peculiarities of the structure of the energy spectrum and the mechanisms of excitation of electron and phonon sub-systems. Just these peculiarities are responsible for the nonlinear optical properties governing OB in semiconductors. Such nonlinearities result from the dependence of the absorption coefficient or the refraction coefficient on the intensity of illumination of a crystal. The physical processes, such as dynamical Burshtein-Moss effect, screening of the electron-hole Coulomb interaction by nonequilibrium carriers, saturation of absorption due to the Franz-Keldysh effect (cf. [9]), saturation of absorption at the exciton resonance lines [12], temperature dependences of the fundamental absorption edge [25] and the relaxation time of free carriers of charge [17], as well as the shift of the energy levels due to applying the electrical field of high-power laser radiation [22]. Revealing of new mechanisms of realization of OB in semiconductors as well as determination of influence on the range of manifestations of it of electric [22] and magnetic [26] fields argues topicality and prospects of further elaboration of this problem showing that the theory of the phenomena of interest is far from completion.

In this paper, we represent the results of the theoretical study of peculiarities of the phenomenon of inner (resonatorless) OB [14] into semiconductor nanofilm (NF), viz. into quasi-two-dimensional nanoheterostructure with a single quantum well (QW). Topicality of this investigation follows from that the results of it provide the feasibilities of the expedient searching for the conditions of experimental observation of internal OB within the domain of the exaction absorption of a NF. The main difficulty of observation of OB in bulk semiconductors is that the frequency range where this phenomenon is realized is too narrow. Its position and extension are determined by the structure of the energy spectra of the electron and phonon systems of a semiconductor, as well as by the efficiency of interaction of them. For that, the problem arises concerning to the choice of a laser with the radiation frequency corresponding to the possible realization of OB. Dependence of the energy of transition into the exciton state and the efficiency of its interaction with phonons on a NF thickness provides the feasibilities for matching of the frequency of laser radiation with the frequency domain of the OB realization. Besides, increasing energy of coupling of exciton into nanostructures in comparison with the one into massive crystals

helps to observe this phenomenon due to higher stability in respect to thermal dissociation.

2. Statement of the problem

To solve this problem, we have computed the coefficient of exciton absorption of a flat semiconductor NF illuminated by powerful laser radiation providing the densities of exciton states which are enough for arising the nonlinearities into absorption spectrum [14]. Computation have been performed for the model of a NF of layered semiconductor 2*H*-PbI_2 introduced into dielectric medium, viz. ethylene-methacrylic acid (E-MAA) copolymer.

Microcrystallites shaped as the hexagonal plates of several layers of PbI_2 synthesized using the chemical-thermal technique in organic medium бреде E-MAA have been obtained and investigated in paper [27]. As thickness of a microcrystallite is much less of its transverse sizes, such structure can be considered as quasi-two-dimensional. By modeling it as a flat nanoheterostructure with a QW of infinite depth, we have calculated the energy of transition into the ground exciton state and the spectral dependence of the exciton absorption into SF of various thicknesses, for various temperatures [28, 29]. It has been shown that decreasing of the NF thickness causes increasing energy of coupling of exciton and the energy of its ground state.

Using the technique described in papers [28, 29], one can calculate the spectral dependence of the coefficient of exciton absorption of a NF

$$\alpha(\omega,T) = 2\pi |D_0|^2 S(\omega,T), \quad (1)$$

where D_0 is the dipole momentum of the direct exciton transition;

$$S(\omega,T) = \frac{\Gamma(\omega,T)}{[\omega-\omega_0-\Delta(\omega,T)]^2+\Gamma(\omega,T)^2} \quad (2)$$

– the dependent on the transition frequency ω and temperature *T* function of the form of the absorption band, determined by the real and imaginary parts of the mass operator of the exciton-phonon interaction $M(\omega,\vec{k}) = \Delta(\omega,\vec{k}) - i\Gamma(\omega,\vec{k})$; ω_0 – frequency of the exciton transition in a NF calculated neglecting the exciton-phonon interaction [28].

The mass operator of the exciton-phonon system, under strong absorption associated with the transition only in the ground exciton state, can be represented in the form

$$M(\vec{k}_\perp,\omega,T) = \sum_{n,\alpha,\vec{q}_\perp} |\Phi_{1n}^{(\alpha)}(\vec{q}_\perp)|^2 [\frac{1+\nu_\alpha(\vec{q}_\perp,T)+N(\vec{q}_\perp,T)}{\omega-\omega_0(\vec{k}_\perp-\vec{q}_\perp)-\Omega_\alpha(\vec{q}_\perp)} + \frac{\nu_\alpha(\vec{q}_\perp,T)-N(\vec{q}_\perp,T)}{\omega-\omega_0(\vec{k}_\perp+\vec{q}_\perp)+\Omega_\alpha(\vec{q}_\perp)}], \quad (3)$$

where

$$\Phi_{nn'}^{(\alpha)}(\vec{q}_\perp) = F_{nn'}^{\alpha}(\vec{q}_\perp)[\frac{1}{1+(m_{h\perp}a_{ex}q_\perp/4m_{ex})^2} - \frac{1}{1+(m_{e\perp}a_{ex}q_\perp/4m_{ex})^2}]; \quad (4)$$

$F_{nn'}^{\alpha}(\vec{q}_\perp)$ – the function of coupling of an electron from the *n*-th level within a QW with a phonon (the component that is transversal in respect to the direction of the NF growth equaling $\vec{q}_\perp$), involving the higher (*n*') states of electron spectrum;

$$\nu_\alpha(\vec{q}_\perp,T) = [\exp\frac{\hbar\Omega_\alpha(\vec{q}_\perp)}{k_BT})-1]^{-1}$$

and

$$N(\vec{k}_\perp,T) = [\exp\frac{\hbar\omega(\vec{k}_\perp)-\mu}{k_BT})-1]^{-1}$$

– the numbers of filling of the phonon and exciton states, respectively; μ – the chemical potential of excitons; α – the index determining the kind of a photon and the longitudinal component of its wave vector $\vec{q} = (\vec{q}_\perp, q_\parallel)$; Ω_α – the phonon frequency; $m_{e_\perp}$ and $m_{h_\perp}$ – the transversal components of the tensor of effective masses of electron and hole, respectively; m_{ex} and a_{ex} – the mass and radius of an exciton at the ground state [28, 29].

One considers the direct transitions in the region of the bottom of the exciton zone ($\vec{k}_\perp = 0$), that is why at low intensity of illumination of an NF (I_0) the exciton concentration is also low, so that the mass operator (3), the form function of the absorption band (2) and the absorption coefficient (1) depend only on the frequency of an exciting wave and temperature. For that, absorbing capacity of a nanosystem is controlled only by the processes of phonon relaxation of excitation of excitons, and the dependence of intensity

$$I(\omega) = I_0 e^{-\alpha(\omega)d} \quad (5)$$

at the output of a NF of thickness *d* on I_0 becomes linear.

Increasing intensity I_0 causes increasing density of the exciton gas N_{ex}. In this case, the magnitudes of Δ, Γ and, as a consequence, the absorption coefficient α are determined not only by frequency ω and temperature *T*, but also by intensity of absorption at crystal at the specified frequency. Indeed, due to existence of coupling between the chemical potential of excitons and their concentration into nanosystem, the numbers of filling of the exciton states are dependent on N_{ex}, whose magnitude is determined by intensity of illumination I_0. As so, the dependence (5) of *I* on I_0 becomes nonlinear. It leads to the possibility for existence of some frequency and temperature ranges where two different magnitudes of *I* correspond to one magnitude of intensity I_0, so that OB is realized [14].

Using the explicit form of the functions of coupling [28], one can calculate the spectral dependence of the absorption coefficient for a fixed temperature, and using the technique described in paper [30] one can find the dependence of the absorption coefficient, α, or transmittance, $\tau = I/I_0$, on I_0 at fixed magnitudes of frequency and temperature *T*. Due to nonlinearity of absorption, the range of changing I_0 can exist, where α and τ become two-valued functions of intensity of illumination I_0. Extension of the region of OB can change with changing frequency, temperature and NF thickness.

3. Results and discussion

Computation has been performed for the nanoheterostructure E-MAA/PbI_2/E-MAA, as the example, the parameters of which as well as the technique of computation of the energy of the exciton transition $E_{ex}^{(0)}$ are represented in paper [31]. Using the magnitude of an optical density measured in [9] for ultrathin microcrystallites of 7 layer-thickness 2*H*-PbI_2 (thickness of one layer is 6.979 Å) equaling 0.387, one can find that the maximal magnitude of the absorption coefficient equals $\alpha = 1.84\cdot10^6$ cm^{-1}, and the corresponding magnitude of the transmittance coefficient equals $\tau = 0.41$.

The altitude of the function of the absorption band, calculated for a NF of 7 layer-thickness 2*H*-PbI_2 accounting interaction only with the bounded phonons, is $S_{max} = 0.3536$. On these data, we have determine the coefficient of proportionality of α and *S*, that provides making further calculations of the spectral dependence of the absorption coefficient for various magnitudes of intensity and temperature.

The results of computation are represented in Table 1 and in Fig. 1, where we use dimensionless energy units applying the normalizing on the width of exciton energy band in 2*H*-PbI_2.

Table 1. The limits of the frequency (for $T = 40$ K) and temperature (for $w = w_s$) intervals of OB at an *N* layer-thick NF of 2*H*-PbI_2

N	w_1	w_2	w_s	T_1, K	T_2, K
5	-0.013952	-0.013774	-0.013815	32	114
7	-0.018160	-0.017908	-0.018034	33	117
10	-0.027696	-0.027276	-0.027485	35	120

One can see that increasing of NF thickness leads to (*i*) long-wavelength shift and broadening of the frequency range of realization of OB, $\Delta\omega = \omega_2 - \omega_1$, (*ii*) broadening of the temperature interval of realization of OB, $\Delta T = T_2 - T_1$. Increasing temperature of a 10 layer-thickness NF of 2*H*-PbI_2 results in decreasing of the

reference frequency w_s of observability of OB and in increasing the hysteresis loop, cf. Fig.1.

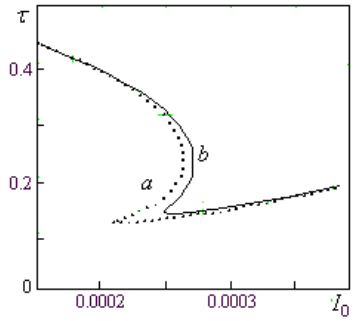

Fig. 1. Dependence of transmittance τ of a 10-layer-thick NF of 2*H*-PbI_2 on intensity of illumination I_0 (in arbitrary units) calculated for the reference frequency w_s = –0.027432 and temperature: *a*) 100 K; *b*) 50 K.

The mentioned facts are explained by that increasing both NF thickness and temperature amplifies the exciton-phonon interaction with the bounded phonons.

4. Conclusions

The represented results of numerical calculation carried out for a NF of 2*H*-PbI_2 introduced into polymer E-MAA show that increasing of its thickness results in long-wavelength shift of the frequency range of realization of OB, increasing size of the hysteresis loop and broadening temperature range of observability of this phenomenon.

In contrast to bulk crystal, the conditions of experimental observation of internal OB in NF are more favorable. Firstly, the dependence of locus of the exciton maximum and the frequency range of possible realization of OB on thickness of a NF ensures to match the frequencies within this range with the exciting source. Secondly, due to essentially increasing of the energy of coupling of an exciton in respect to one in bulk semiconductors, the probability of destroying of the bistable state resulting from the thermal dissociation of excitons decreases.

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