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Self-polarization Effect and Electron-Phonon Interaction Contributions in the Formation of the Electron Energy Spectrum of PbI_2 Nanofilm Embed in E-MAA Copolymer

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The paper describes the results of theoretical study of the role of self-polarization effect and electron-phonon interaction in the formation of the energy spectrum of electrons in semiconductor flat nanofilm. Concrete calculations are performed using the example of 2H-PbI2 nanofilm embedded in the methyl-acrylic acid copolymer (E-MAA). It was found that contribution of self-polarization effect to the value of the electron energy in ultrathin nanofilms is more than from the interaction with polarized optical phonons.

Keywords: Nanoheterostructure, Quantum well, Energy spectrum, Lead iodide.

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1. INTRODUCTION

Properties of lead diiodide and heterostructures on its basis are actively studied during several last decades. This is explained by their high chemical stability and application perspectiveness for the formation of quantum optoelectronic devices, such as, temperature sensors [1], photodetectors [2], biomedical sensors [3], as well as X- and γ-radiation sensors which can operate in a wide temperature range [4]. Large values of the band gap width and binding energy of exciton in these structures promote the manifestations of the exciton effects in their optical spectra [5, 6].

Spatial restrictions of quasiparticle motion in low-dimensional structures induce changes in the structure of energy spectra that was observed experimentally by many authors by the corresponding changes in their optical properties [7]. Along with this fact, theoretically dimensional and temperature dependences of the energy and optical spectra of such structures are studied insufficiently.

In this paper we present the method and calculation results of the dependence of the energy spectrum of 2H-PbI_2 nanofilm (NF) embedded in an isotropic organic dielectric medium on its thickness. Ultrathin microcrystallites obtained and experimentally investigated in the works [6, 8] are considered to be the structures of the mentioned type.

It is shown that change in the NF thickness leads to the non-linear shift of the electron energy levels in a quantum well (QW) to the long-wave spectral region. Dominance of the self-polarization (SP) effect contribution [9] to the shift value over the electro-phonon interaction (EPI) contribution [10] is established.

2. INVESTIGATED MODEL AND PROBLEM STATEMENT

We consider a plane semiconductor NF embedded in bulk medium with wide band gap. Such structure with double heterojunction forms a QW of finite or infinite depth [10, 11]. We will assume the QW profile to be rectangular and choose the coordinate system in such a way that its origin coincides with the center of NF of the thickness a and XOY area is parallel to its surface.

Theoretical investigations of semiconductor nanoheterostructures are usually performed in the effective mass approximation for the electron system and model of dielectric continuum – for the phonon system [11]. Since hereinafter we will consider a NF of layered semiconductor embedded in an amorphous organic medium, then heterojunction can be supposed as unloaded, and the electron Hamiltonian in a QW can be presented in the following form:

$$\hat{H}_e = -\frac{\hbar^2}{2m_\perp}\nabla_\perp^2 - \frac{\hbar^2}{2m_\parallel}\frac{\partial^2}{\partial z^2} + V(z) + V_s(z) = \hat{H}_0 + V_s(z). \quad (1)$$

Here $m_\parallel$ ($m_\perp$) is the electron effective mass in NF which characterizes its motion in the direction parallel (perpendicular) to the OZ-axis directed along the crystallographic C axis of layered semiconductor; $\nabla_\perp^2$ is the Laplace operator expressed through the variables (x, y); V is the limiting potential of QW;

$$V_s(z) = \frac{e^2}{2\varepsilon_\infty^{(0)}} \sum_{l=\pm1,\pm2,\ldots} \frac{\xi^{|l|}}{|z-(-1)^l z + la|} \quad (2)$$

is the SP potential [12];

$$\xi = \frac{\varepsilon_\infty^{(0)} - \varepsilon_\infty^{(1)}}{\varepsilon_\infty^{(0)} + \varepsilon_\infty^{(1)}};$$

$\varepsilon_0^{(l)}$ and $\varepsilon_\infty^{(l)}$ are the static and dynamic permittivities of the wall (l = 0) or barrier (l = 1) material.

We consider barrier medium as dielectric and take the model of an infinitely deep QW. Then, Hamiltonian eigenvalues (electron energy in a QW) are defined by the correlation

$$E_n^{(0)}(\vec{k}_\perp) = \frac{nh^2}{8m_\parallel a^2} + \frac{\hbar^2 k_\perp^2}{2m_\perp}, \quad (3)$$

where $\vec{k}_\perp$ is the wave vector of an electron from the n-th minizone in the XOY plane. Since SP potential $V_s(z)$ can significantly influence the position of energy level in QW without a considerable change in the corresponding wave function [12], then eigenfunctions of the operator (1) are the following:

$$\psi_{n\vec{k}_\perp}(\vec{\rho},z)=\sqrt{\frac{2}{a}}e^{i\vec{k}_\perp\vec{\rho}}\begin{cases}\cos(n\pi z/a), & n=1,3,5,...;\\ \sin(n\pi z/a), & n=2,4,6,...,\end{cases} \quad (4)$$

where $\vec{\rho}$ is the electron radius-vector in the *XOY* plane. System of functions (4) is complete and orthonormal that allows to determine the electron energy in a QW with taking into account the SP effect of heterojunction planes

$$E_n(\vec{k}_\perp)=\frac{nh^2}{8m_\parallel a^2}+\frac{\hbar^2k_\perp^2}{2m_\perp}+\Delta E_{ns}, \quad (5)$$

where $\Delta E_{ns}=\langle\psi_{n\vec{k}_\perp}|V_s(z)|\psi_{n\vec{k}_\perp}\rangle$.

Hamiltonian of free phonons in the studied system can be represented in the form of

$$\hat{H}_{ph}=\hat{H}_{LO}+\hat{H}_I=\sum_{\lambda,\vec{q}_\perp}\Omega_{LO}(\hat{b}^+_{\lambda\vec{q}_\perp}\hat{b}_{\lambda\vec{q}_\perp}+1/2) + \sum_{\sigma=S,A}\sum_{\vec{q}_\perp}\Omega_\sigma(\vec{q}_\perp)(\hat{b}^+_{\sigma\vec{q}_\perp}\hat{b}_{\sigma\vec{q}_\perp}+1/2), \quad (6)$$

where Ω_{LO} is the energy of the longitudinal optical polarization phonons limited in the QW medium; $\hat{b}^+_{\lambda\vec{q}_\perp}$ ($\hat{b}_{\lambda\vec{q}_\perp}$) is the initiation (annihilation) operator of the phonon state with the wave vector $\vec{q}=(q_\parallel,\vec{q}_\perp)$, whose longitudinal component ($q_\parallel$) is quantized taking discrete values from the set $\lambda\pi/a$ ($\lambda=1, 2, ..., N=\mathrm{int}(a/a_0)$ is the number of matter layers of the well medium, whose lattice constant is a_0);

$$\Omega_\sigma(\vec{q}_\perp)=\sqrt{\frac{\in^{(0)}_\sigma(\vec{q}_\perp)\Omega^2_{LO}+\in^{(1)}_\sigma(\vec{q}_\perp)\Omega^2_{TO}}{\in^{(0)}_\sigma(\vec{q}_\perp)+\in^{(1)}_\sigma(\vec{q}_\perp)}} \quad (7)$$

are the energies of interface phonons of the symmetric (σ = S) and antisymmetric (σ = A) modes [10, 11]. Here $\Omega_{TO}=\Omega_{LO}\sqrt{\varepsilon_\infty/\varepsilon_0}$ is the energy of the transverse optical vibrations;

$$\in^{(0)}_{\sigma=\begin{Bmatrix}S\\A\end{Bmatrix}}(\vec{q}_\perp)=\varepsilon^{(0)}_\infty[1\mp\exp(-q_\perp a)],$$

$$\in^{(1)}_{\sigma=\begin{Bmatrix}S\\A\end{Bmatrix}}(\vec{q}_\perp)=\varepsilon^{(1)}_\infty[1\pm\exp(-q_\perp a)],$$

$\hat{b}^+_{\sigma\vec{q}_\perp}$ ($\hat{b}_{\sigma\vec{q}_\perp}$) is the initiation (annihilation) operator of the corresponding phonon state.

Hamiltonian of the EPI in NF was obtained in the work [10]. For the study of the chosen here model, it is convenient to write Hamiltonian in the representation of the secondary quantization over all variables of the electron-phonon system.

$$\hat{H}_{e-ph}=\hat{H}_{e-LO}+\hat{H}_{e-I}=\sum_{n,n',\vec{k}_\perp}\Big[\sum_{\lambda,\vec{q}_\perp}F^\lambda_{nn'}(\vec{q}_\perp)\hat{a}^+_{n'\vec{k}_\perp+\vec{q}_\perp}\hat{a}_{n\vec{k}_\perp}\hat{B}_{\lambda\vec{q}_\perp}+ + \sum_{\sigma,\vec{q}_\perp}F^\sigma_{nn'}(\vec{q}_\perp)\hat{a}^+_{n'\vec{k}_\perp+\vec{q}_\perp}\hat{a}_{n\vec{k}_\perp}\hat{B}_{\sigma\vec{q}_\perp}\Big], \quad (8)$$

where $\hat{B}_{\Lambda\vec{q}_\perp}=\hat{b}_{K\vec{q}_\perp}+\hat{b}^+_{\Lambda\vec{q}_\perp}$ (Λ is the index of phonon branch which is equal to λ in the case of LO- or σ – in the case of I-phonons); $F^\Lambda_{nn'}(\vec{q}_\perp)$ are the functions of the electron-phonon coupling, whose explicit form (for a rectangular QW of a finite depth) was found in [13].

EPI renormalizes the electron spectrum inducing a shift of states along the energy scale on the value which can be obtained by the Green function method [14]. In particular, in the low-temperature region (formally, at T = 0 K) electron energy from the n-th minizone in QW is determined by the position of poles of the Fourier transform of the corresponding electron Green function.

$$G_n(\vec{k}_\perp,\omega)=\frac{1}{\hbar\omega-E_n(\vec{k}_\perp)-M_n(\vec{k}_\perp,\omega)},$$

where $M_n(\vec{k}_\perp,\omega)\equiv M_{nn}(\vec{k}_\perp,\omega)$ are the diagonal matrix elements of the mass operator (MO) of the investigated system [14] calculated based on the wave functions (4). In the single-electron approximation we have

$$M_n(\vec{k}_\perp,\omega)=\sum_{n',\Lambda,\vec{q}_\perp}\frac{\left|F^\Lambda_{nn'}(\vec{q}_\perp)\right|^2}{\hbar\omega-E_n(\vec{k}_\perp)-\frac{h^2}{2m_\perp}(\vec{k}_\perp+\vec{q}_\perp)^2-\Omega_\Lambda}. \quad (9)$$

Only virtual phonons can exist in the system at T = 0, therefore, the EPI is manifested solely in the processes with their radiation that induces a shift of the electron states to the long-wave energy region. The value of the bottom shift of the corresponding minizone is given by the following expression:

$$\Delta\equiv-M_n(\vec{k}_\perp=0,\hbar\omega=E_n). \quad (10)$$

The aim of the present work is the development of the algorithm and calculation of the ground-state energy of electron in QW formed by the embedding of 2H-PbI_2 NF to a non-crystalline dielectric medium and establishment of its dependence versus the HF thickness.

3. OBTAINED RESULTS AND THEIR DISCUSSION

3.1 Influence of the SP effect

Using wave functions (4), we will find a correction to the electron energy at the cost of the SF of heterojunction planes

$$\Delta E_{ns}=\langle\psi_{n\vec{k}_\perp}|V_s(z)|\psi_{n\vec{k}_\perp}\rangle=\frac{e^2}{\varepsilon^{(0)}_{\infty|}a}\sum_{l=0}^{\infty}\Big[\xi^{2l+1}\Big(\int_0^{\pi/2}\frac{\cos^2x\,dx}{x+\pi(l+1/2)}- -\int_0^{\pi/2}\frac{\cos^2x\,dx}{x-\pi(l-1/2)}\Big)+\frac{\xi^{2l+2}}{2l+2}\Big]. \quad (11)$$

Calculations of the electron energy performed for 2H-PbI_2 NF (parameters of the crystalline lattice are the following: $a_\perp\equiv a_0$ = 4,557 Å and $a_\parallel\equiv c_0$ = 6,979 Å; LO-phonon energy Ω_{LO} = 110 cm^{-1} [15]; permittivities $\varepsilon_{0\parallel}$ = 7,5; $\varepsilon_{0\perp}$ = 8,31; $\varepsilon_{\infty\parallel}$ = 6,0; $\varepsilon_{\infty\perp}$ = 6,25 [16]; effective electron masses $m_\parallel$ = 1,25, $m_\perp$ = 0,25 [17]) embedded in polymer E-MAA ($\varepsilon_0=\varepsilon_\infty$ = 2,38 [18]) has shown that the influence of SP effect is significant in ultrathin films. Account of the correction (11) increases the energy of the ground single-electron level in QW the stronger, the lesser NF thickness is (Fig. 1). In NF of the thickness of one PbI_2 layer correction is equal to 17% of the value of the ground-state energy in QW.

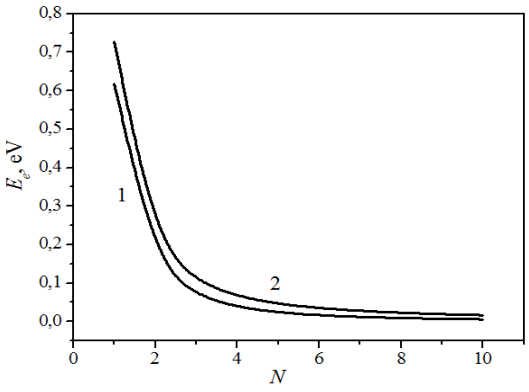

Fig. 1 – The ground-state energy of electron without (curve 1) and with taking into account (curve 2) the SP effect as the function of NF thickness (N is the number of PbI_2 layers in NF)

3.2 Influence of the EPI

Functions of the electron-phonon coupling obtained in the work [13] are easily transformed for the case of an infinitely deep QW. For the limited phonons they take the following form:

$$F^{\lambda}_{nn'}(\vec{q}_\perp) = i4\lambda\sqrt{\frac{e^2\Omega_{LO}a}{\pi\varepsilon^{(0)}\tilde{S}[(\lambda\pi)^2+(aq_\perp)^2]}}\times$$

$$\times\begin{cases}\dfrac{\cos[0.5(n-n')\pi]}{\lambda^2-(n-n')^2}-\dfrac{(-1)^n\cos[0.5(n+n')\pi]}{\lambda^2-(n+n')^2}\\ (\lambda=1,3,\ldots;n \text{ and } nn' \text{ of the same twoness});\\ \dfrac{(-1)^{n-1}\sin[0.5(n-n')\pi]}{\lambda^2-(n-n')^2}-\dfrac{\sin[0.5(n+n')\pi]}{\lambda^2-(n+n')^2}\\ (\lambda=2,4,\ldots;n \text{ and } n' \text{ of different twoness}),\end{cases}$$

and for the interface phonons –

$$F^{\sigma}_{nn'}(\vec{q}_\perp) = i\frac{2}{a}\sqrt{\frac{\pi e^2\Omega_\sigma(\vec{q}_\perp)}{\in^{(0)}_\sigma(\vec{q}_\perp)\varsigma_\sigma(\vec{q}_\perp)\tilde{S}q_\perp}}\cdot f^{\sigma}_{nn'}(\vec{q}_\perp)\ .$$

Here i is the imaginary unit; $\tilde{S}$ is the area of the main part of the NF surface;

$$\varsigma_\sigma = \frac{\varepsilon^{(0)}\Omega^2_\sigma(\vec{q}_\perp)}{\varepsilon^{(0)}_0\Omega^2_{Tl}}\left(\frac{\Omega^2_{Ll}-\Omega^2_{Tl}}{\Omega^2_{Tl}-\Omega^2_\sigma(\vec{q}_\perp)}\right)^2,\ \frac{1}{\varepsilon^{(0)}}=\frac{1}{\varepsilon^{(0)}_0}-\frac{1}{\varepsilon^{(0)}_\infty};$$

and

$$f^{\sigma}_{nn'}(\vec{q}_\perp) = a^2\sqrt{1+\exp(-q_\perp a)}\cdot\delta_{\sigma S}\cdot q_\perp\times$$
$$\times th\frac{q_\perp a}{2}\left\{\frac{\cos[0.5(n-n')\pi]}{[(n-n')\pi]^2+[q_\perp a]^2}\pm\frac{\cos[0.5(n+n')\pi]}{[(n+n')\pi]^2+[q_\perp a]^2}\right\},$$

if n and n' are of the same twoness and, similarly,

$$f^{\sigma}_{nn'}(\vec{q}_\perp) = -a^2\sqrt{1-\exp(-q_\perp a)}\cdot\delta_{\sigma A}\cdot q_\perp\times$$
$$\times\mathrm{cth}\frac{q_\perp a}{2}\left\{\frac{\sin[0.5(n-n')\pi]}{[(n-n')\pi]^2+[q_\perp a]^2}\pm\frac{\sin[0.5(n+n')\pi]}{[(n+n')\pi]^2+[q_\perp a]^2}\right\},$$

if n and n' are of different twoness (sign "plus" is taken in the case of n = 1, 3, 5, …, and for n = 2, 4, 6, … – sign "minus"); δ_{oo} is the Kronecker δ-symbol.

Coupling functions of the electron ground state in QW with LO-phonons involving the n-th state (F^{λ}_{1n}) are decay functions of the phonon wave vector in the plane of QW ($\vec{q}_\perp$) (Fig. 2). Their values parametrically depend on the quantum numbers of the electron (n, Fig. 2a) and the phonon (λ, Fig. 2b) states, they are sharply decreased with the increase of these parameters. Such behavior of the dependence allows to restrict a number of terms in (9) terminating sums over n and λ on those values at which the corresponding coupling functions become negligibly small in comparison with F^{1}_{11}.

Coupling with I-phonons also becomes weaker at the increase in the number of the combined level n, but dependence of $F^{\sigma}_{nn'}$ on $\vec{q}_\perp$, with the exception of n = 1, is not monotonous (see Fig. 3).

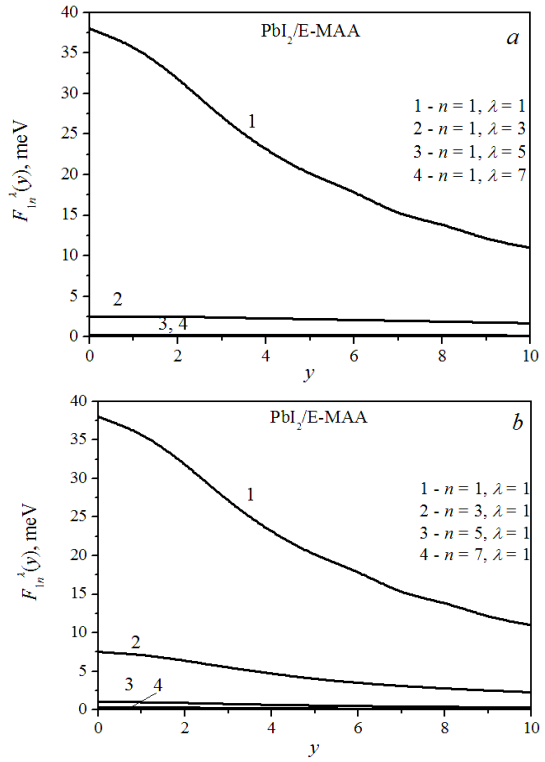

Fig. 2 – Dependence of the coupling functions with LO-phonons on the dimensionless phonon wave vector $y = aq_\perp$ at different λ (a) and n (b)

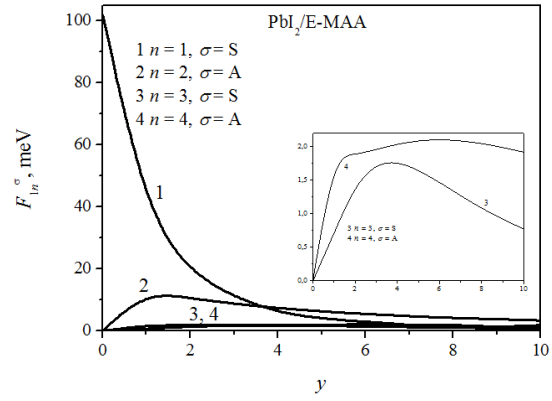

Fig. 3 – Dependence of the coupling functions with I-phonons on the dimensionless phonon wave number $y = aq_\perp$ at different values of n

The stated regularities can be used for the determination of the sums of the corresponding series in (9).

Going in (9) from the sum over $\vec{q}_\perp$ to the integral, we obtain MO of the electron-phonon system in NF

$$M_1(\vec{k}_\perp,\omega)=\frac{\tilde{S}}{4\pi^2}\int_0^{\pi/a_0} q_\perp dq_\perp \int_0^{2\pi} d\phi \times$$
$$\times\left\{\sum_n\left[\sum_\lambda \frac{\left|F_{1n}^{\lambda}(q_\perp)\right|^2}{\hbar\omega-E_n-\hbar^2/(2m)\left(k_\perp^2+2k_\perp q_\perp\cos\phi+q_\perp^2\right)-\Omega_{LO}}+\right.\right.$$
$$\left.\left.+\frac{\left|F_{1n}^{\sigma}(q_\perp)\right|^2}{\hbar\omega-E_n-\hbar^2/(2m)\left(k_\perp^2+2k_\perp q_\perp\cos\phi+q_\perp^2\right)-\Omega_\sigma(q_\perp)}\right]\right\}.$$

Using the found coupling functions, we obtain an explicit form of the dependence of the MO on the electron energy $\hbar\omega$ and NF thickness a in the region of the main minizone bottom ($n=1,\ \vec{k}_\perp=0$):

$$M_1(\omega,a)=M_{11}^{(LO)}(\omega,a)+M_{11}^{(I_S)}(\omega,a)+$$
$$+\sum_{n\geq 2}\left[M_{1n}^{(LO)}(\omega,a)+M_{1n}^{(I_\sigma)}(\omega,a)\right], \qquad (12)$$

where

$$M_{1n}^{(LO)}(\omega,a)=\frac{4e^2}{\pi^2\varepsilon^{(0)}a}\times$$
$$\times\sum_\lambda\frac{\lambda^2}{\left(\hbar\omega-E_n\right)/\Omega_{LO}+(\pi\hbar\lambda)^2/\left(2m_\perp a^2\Omega_{LO}\right)-1}\times$$
$$\times\ln\frac{1+\left(a/\lambda a_0\right)^2}{1-(\pi\hbar)^2/\left[2m_\perp a^2\left(\hbar\omega-E_n-\Omega_{LO}\right)\right]}\times$$
$$\times\begin{cases}\left(\dfrac{\cos[0.5(1-n)\pi]}{\lambda^2-(1-n)^2}+\dfrac{\cos[0.5(1+n)\pi]}{\lambda^2-(1+n)^2}\right)^2;\\ (\lambda=1,3,\ldots;\ n\text{ is odd});\\ \left(\dfrac{\sin[0.5(1-n)\pi]}{\lambda^2-(1-n)^2}-\dfrac{\sin[0.5(1+n)\pi]}{\lambda^2-(1+n)^2}\right)^2;\\ (\lambda=2,4,\ldots;\ n\text{ is even})\end{cases}$$

and

$$M_{11}^{(I_\sigma)}(\omega,a)=\frac{2e^2}{a^3}\times$$
$$\int_0^{\pi a/a_0}\frac{\Omega_\sigma(y)\left|f_{1n}^{\sigma}(y)\right|^2}{\in^{(0)}(y)\varsigma_\sigma(y)\left[\hbar\omega-E_n-\Omega_\sigma(y)-\hbar^2y^2/2m_\perp a^2\right]}dy$$

are the terms which determine the interaction contribution of the electron ground state in QW with the LO- and I-phonons involving the n-th state, respectively, ($y=aq_\perp$).

In Fig. 4 we represent the calculation result of the shift value of the electron ground level in QW induced by the interaction with LO-phonons (without participation of higher ($n\geq 2$) levels) ($\Delta_{LO}=-M_{11}^{(LO)}(E_1,a)$) for NF of different thickness. As seen, interaction with LO-phonons shifts the electron ground level to the long-wave region the stronger, the larger NF thickness is. Contribution of higher ($\lambda\geq 2$) LO-phonon states (curve 2) is small in comparison with the contribution of the ground one (curve 1).

Influence of higher electron states ($n\geq 2$) is shown in Fig. 4b. It is seen that the EPI with the participation of higher electron states (curve 2) shifts the electron ground level in QW to the long-wave region the stronger, the larger NF thickness is.

This is explained by the fact that at the increase in the NF thickness, the distance between the ground and excited levels decreases that conditions the growth of the contribution of the latter to the shift value.

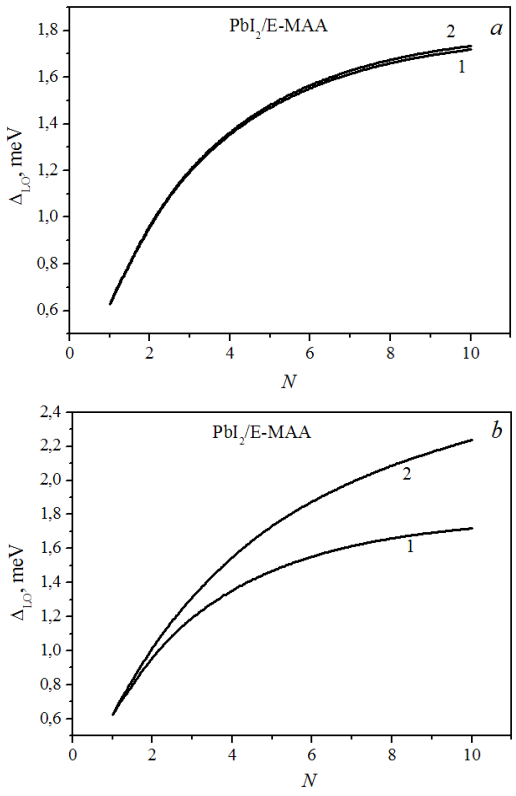

Fig. 4 – The value of shift of the electron ground level induced by the EPI with LO-phonons as the function of the NF thickness calculated in the approximation of interaction involving only ground (a) and higher (b) electron states

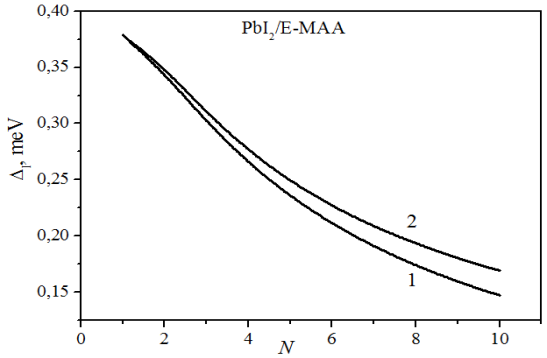

Fig. 5 – The value of shift of the electron ground level induced by the EPI with I-phonons as the function of the NF thickness N calculated in the approximation of interaction involving only ground (1) and higher (2) electron states

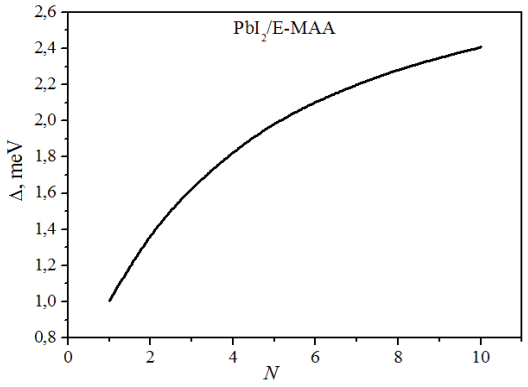

Fig. 6 – The value of the total shift Δ of the electron ground level induced by the interaction with LO- and I-phonons as the function of the NF thickness N

Results of the similar calculations performed for the case of interaction between an electron and I-phonons are represented in Fig. 5.

Interaction with I-phonons shifts the ground level to the long-wave region. Nevertheless, behavior of the dependence of the shift on the NF thickness is different here, namely, decaying. Behavior of curves 2 illustrates the influence of higher electron states on the value of shift of the ground level, in contrast to curves 1 which show the dependence $\Delta_I(N)$ obtained without taking into account higher levels. It is seen that interaction with I-phonons involving higher electron states strengthens the shift of the minizone bottom to the long-wave region the stronger, the larger NF thickness is.

As seen from expression (12), the value of the total shift Δ conditioned by both phonon types is defined by the sum of partial shifts

$$\Delta = \Delta_{LO} + \Delta_I.$$

Behavior of the dependence of the total shift of the electron main minizone bottom in QW on the NF thickness is represented in Fig, 6. As seen from this figure, the dependence is non-linear, monotonically increasing function of the argument N.

4. CONCLUSIONS

We have obtained the explicit form of the correction to SP, electron coupling functions with optical polarization phonons, and MO of the electron-phonon system in an infinitely deep QW. In the framework of the chosen model, we have investigated the influence of the EPI on the position of the main minizone bottom of a free carrier in NF.

Calculation performed on the example of 2H-PbI_2 NF embedded in the E-MAA polymer has shown that in this nanosystem the SP effect of heterojunction planes gives a considerably larger contribution (up to 100 meV) to the value of the single-electron state energy in QW than the EPI (some meV). Influence of the SP effect extremely increases with the decrease in the NF thickness, while the EPI appears stronger at its increase.

Obtained results can be used for the investigation of the optical characteristics of lead diiodide nanocrystals, in particular, the dependence of its exciton spectrum on the thickness and temperature.

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