

Excitonic Transition Energy Renormalization in Lead Diiodide Nanofilm Caused by Self-Polarization Effects and Electron-Phonon Interaction

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Results of the theoretical study of the contributions of self-polarization effect and exciton-phonon interaction to the exciton transition energy in a flat semiconductor nanofilm are presented here. Calculations of the binding and exciton ground state energies are performed using 2H-PbI₂ nanofilm embedded in ethylene-methacrylic acid (E-MAA) copolymer. It has been revealed that in ultrathin nanofilms the self-polarization effect contribution to the exciton energy is more than from the interaction with polarizing optical phonons.

Keywords: Nanoheterostructure, Quantum well, Exciton, Lead diiodide.

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1. INTRODUCTION

Thin films of lead diiodide and low-dimensional nanostructures on its basis are considered to be a promising material for the creation of a number of quantum optoelectronic devices [1-5]. Large values of charge numbers of Pb and I atoms, band gap width, and exciton binding energy in these structures make it impossible to observe the exciton effects in their optical spectra [6-8].

Position of exciton lines in the optical spectra of low-dimensional semiconductor crystal structures is defined by the conditions of spatial constraints (SC) of the electron and phonon systems [9, 10], as well as self-polarization (SP) effect, i.e. interaction of carriers with polarized charges on the boundaries of heterotransitions [11].

A great number of experimental and theoretical works is devoted to the investigation of the laws of the formation and dynamics of exciton states in such structures. Exciton spectra of heterostructures with single quantum wells (QW) were observed in the experimental papers [6, 8, 12-16]. In particular, the authors of [6, 8, 15, 16] have obtained the hybrid – organic-inorganic – heterostructures of this type based on lead diiodide, which are interesting by the combination of the properties of both materials, in whose spectra exciton lines were observed. Theoretical investigations of the exciton states in single QW are performed by a number of authors by different methods in different approximations (see, for example, [17-21]), but nanostructures based on layered semiconductors were not considered.

Sets of peaks on the long-wavelength edge of the low-temperature ($T = 2$ K) band of fundamental absorption of ultrathin PbI₂ microcrystallites (hexagon plates with the transverse diameter of 2-80 nm) in the E-MAA matrix were experimentally observed in the work [6]. Their appearance was connected by authors with the realization of exciton transitions and difference in the position – with the dispersion of the plate thickness. It is established that with the increase in the plate thickness the maximum of the exciton band is shifted to the long-wave spectral region.

Under the condition that plate thickness is significantly less than its transverse sizes, it can be considered as quasi-two-dimensional structure. In this paper we present the results of the theoretical investigation

of the influence of the SC and SP effects and exciton-phonon interaction (EPI) on the values of the exciton ground state energies in the structure E-MAA / PbI₂ / E-MAA nanofilm (NF).

Within the dielectric continuum model and using the effective-mass approximation and methods of the Green function theory, we have obtained the analytical expressions which allow to calculate the exciton ground state energy in NF and its binding energy. Calculated dependence of the exciton energy on the NF thickness is found to be nonlinearly decaying and determined, mainly, by the SC influence. Contributions of the SP and EFI effects are considerably less, and their ratio is changed with the NF thickness. In ultrathin NF influence of the SP effect (shift of the exciton ground state toward the larger energies) exceeds the EFI influence (shift toward the opposite direction). At the increase in the NF thickness influence of the SP effect decreases, and of the EFI effects – increases.

2. INVESTIGATED MODEL AND HAMILTONIAN OF THE PROBLEM

We simulate NF by a plane double heterostructure of the I type whose well material is a layered semiconductor placed into bulk dielectric medium. Peculiarities of the crystal structure of layered semiconductors condition the atomic smoothness of their surfaces with a small amount of dangling bonds [22]. Thus, NF thickness a is found to be multiple to the thickness of the ball packet c ($a = Nc$, where N is the natural number). Heterotransition of such structure can be considered unloaded, QW – rectangular and, in the presence of dielectric barrier medium, infinitely deep.

Under these conditions Hamiltonian of the exciton-phonon system is given as a sum

$$\hat{H} = \hat{H}_{\text{ex}} + \hat{H}_{\text{ph}} + \hat{H}_{\text{int}}$$

of the single-particle Hamiltonians of free exciton (\hat{H}_{ex}) and phonon (\hat{H}_{ph}) contained in NF and operator of their interaction (\hat{H}_{int}).

Placing the origin of the coordinate system into NF

center, we direct the OZ -axis along the crystallographic axis C of the layered crystal and use the Wannier-Mott exciton model. Then, in the system of the mass center of exciton

$$\hat{H}_{\text{ex}} = -\frac{\hbar^2 \nabla_{\rho}^2}{2\mu} + \sum_{j=e,h} \hat{H}_j(z_j) + V_{\text{e-h}}(\bar{\rho}, z_e, z_h), \quad (1)$$

where

$$\hat{H}_j(z_j) = -\frac{\hbar^2}{2m_j^{\parallel}} \frac{\partial^2}{\partial z_j^2} + V(z_j) + V_{\text{sp}}(z_j) \quad (2)$$

is the single particle Hamiltonian which describes the electron ($j=e$) or hole ($j=h$) motion in the corresponding QW specified by the confining potential

$$V(z_j) = \begin{cases} 0, & |z_j| < a/2; \\ \infty, & |z_j| \geq a/2; \end{cases}$$

$$V_{\text{sp}}(z_j) = \frac{e^2}{2\epsilon_{\infty}^{(0)}} \sum_{n \neq 0} \frac{\xi^{|n|}}{|z_j - (-1)^n z_j + na|}$$

is the interaction potential of a carrier with its electrostatic image in the planes of heterojunctions (SP potential [11]);

$$V_{\text{e-h}}(\bar{\rho}, z_e, z_h) = -\frac{e^2}{\epsilon_{\infty \perp}^{(0)}} \sum_{n=-\infty}^{\infty} \frac{\xi^{|n|}}{\sqrt{\rho^2 + \{\epsilon_{\infty \parallel}^{(0)} [z_e - (-1)^n z_h + na] / \epsilon_{\infty \perp}^{(0)}\}^2}}$$

is the Coulomb interaction potential of electron and hole with each other and electrostatic images of both quasiparticles in the planes of heterojunction [23] adapted for the case of layered semiconductor; m_j^{\parallel} (m_j^{\perp}) and $\epsilon_{\infty \parallel}^{(l)}$ ($\epsilon_{\infty \perp}^{(l)}$) are the carrier effective mass and dynamical permittivity of environment (well at $l=0$ or barrier at $l=1$) typical for the direction along (across) the C axis; vector $\bar{\rho} = (x_e - x_h, y_e - y_h)$ defines the mutual arrangement of electron and hole in NF in the XOY plane;

$$\xi = \frac{\epsilon_{\infty}^{(0)} - \epsilon_{\infty}^{(1)}}{\epsilon_{\infty}^{(0)} + \epsilon_{\infty}^{(1)}}.$$

Hamiltonians of free phonons and EPI (in the coordinate representation by electron variables) are obtained in the works [24, 25]. Adapted for the case of the investigated here system, they have the following form:

$$\hat{H}_{\text{ph}} = \hat{H}_{\text{LO}} + \hat{H}_1 = \sum_{\lambda, \vec{q}_{\perp}} \Omega_{\text{LO}} (\hat{b}_{\lambda \vec{q}_{\perp}}^{\dagger} \hat{b}_{\lambda \vec{q}_{\perp}} + 1/2) + \sum_{\sigma=S,A} \sum_{\vec{q}_{\perp}} \Omega_{\sigma}(\vec{q}_{\perp}) (\hat{b}_{\sigma \vec{q}_{\perp}}^{\dagger} \hat{b}_{\sigma \vec{q}_{\perp}} + 1/2), \quad (3)$$

where Ω_{LO} is the energy of the longitudinal optical polarization (LO) phonons limited in the QW medium; $\hat{b}_{\lambda \vec{q}_{\perp}}^{\dagger}$ and $\hat{b}_{\lambda \vec{q}_{\perp}}$ are, respectively, the production and annihilation operators 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 $\lambda \pi / a$

($\lambda = 1, 2, \dots, N$);

$$\Omega_{\sigma}(\vec{q}_{\perp}) = \sqrt{\frac{\epsilon_{\sigma}^{(0)}(\vec{q}_{\perp}) \Omega_{\text{LO}}^2 + \epsilon_{\sigma}^{(1)}(\vec{q}_{\perp}) \Omega_{\text{TO}}^2}{\epsilon_{\sigma}^{(0)}(\vec{q}_{\perp}) + \epsilon_{\sigma}^{(1)}(\vec{q}_{\perp})}}$$

are the energies of interface (I) phonons of symmetric ($\sigma=S$) or antisymmetric ($\sigma=A$) modes. $\Omega_{\text{TO}} = \Omega_{\text{LO}} \sqrt{\epsilon_{\infty} / \epsilon_0}$ is the energy of the transverse optical vibrations;

$$\epsilon_{\sigma}^{(0)} \Big|_{\substack{S \\ A}}(\vec{q}_{\perp}) = \epsilon_{\infty \perp}^{(0)} [1 \mp \exp(-q_{\perp} a)],$$

$$\epsilon_{\sigma}^{(1)} \Big|_{\substack{S \\ A}}(\vec{q}_{\perp}) = \epsilon_{\infty}^{(1)} [1 \pm \exp(-q_{\perp} a)];$$

$\hat{b}_{\sigma \vec{q}_{\perp}}^{\dagger}$ ($\hat{b}_{\sigma \vec{q}_{\perp}}$) is the production (annihilation) operator of the corresponding I-phonon state.

Hamiltonian of the EPI [24, 25] reduced by the authors of [20] to the representation of the secondary quantization by all variables of the electron-phonon system,

$$\begin{aligned} \hat{H}_{\text{int}} &= \hat{H}_{\text{e-LO}} + \hat{H}_{\text{e-I}} = \\ &= \sum_{j=e,h} \eta_j \left\{ \sum_{n, n', \vec{k}_{\perp}} \left[\sum_{\lambda, \vec{q}_{\perp}} F_{n, n'}^{\lambda}(\vec{q}_{\perp}) \hat{a}_{n', \vec{k}_{\perp} + \vec{q}_{\perp}}^{\dagger} \hat{a}_{n, \vec{k}_{\perp}} \hat{B}_{\lambda \vec{q}_{\perp}} + \right. \right. \\ &\left. \left. + \sum_{\sigma, \vec{q}_{\perp}} F_{n, n'}^{\sigma}(\vec{q}_{\perp}) \hat{a}_{n', \vec{k}_{\perp} + \vec{q}_{\perp}}^{\dagger} \hat{a}_{n, \vec{k}_{\perp}} \hat{B}_{\sigma \vec{q}_{\perp}} \right] \right\}, \end{aligned} \quad (4)$$

is defined by the functions of the electron-phonon coupling $F_{mn}^{\alpha}(\vec{q}_{\perp})$, whose explicit form for QW of a finite depth is given in [20], and of an infinite depth – in [26]. Here $\eta_e = 1$, $\eta_h = -1$; n and n' are the numbers of the electron sub-bands in the state (n, \vec{k}_{\perp}) ; a is the index of the phonon branch which is equal to λ in the case of LO – or σ – in the case of I-phonons; $\hat{B}_{\alpha \vec{q}_{\perp}} = \hat{b}_{\alpha \vec{q}_{\perp}} + \hat{b}_{\alpha \vec{q}_{\perp}}^{\dagger}$ is the phonon and $\hat{a}_{n \vec{k}_{\perp}}$, $\hat{a}_{n \vec{k}_{\perp}}^{\dagger}$ are the electron operators of the secondary quantization.

Hamiltonian (3) is obtained in the assumption that EPI is realized through the personal interaction of electron and hole, which form exciton, with phonons. Such interaction renormalizes the energy spectrum of quasiparticles inducing shift of their states along the energy scale. In particular, in the temperatures range close to the absolute zero, only virtual phonons can exist in the system, therefore, interaction with phonons is exhibited exclusively in the processes with their radiation that results in the shift of electron states toward the long-wave energy region.

The shift value of the bottom of the corresponding minizone of a carrier in this case is determined by the positions of the Fourier transform poles of the corresponding 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 electron-phonon system [27] determined on the eigenfunctions

of Hamiltonian (2). Explicit form of MO in the single electron approximation is given in [26]. Its calculation allows to define the shift value of the bottom of the corresponding minizone of a carrier in QW:

$$\Delta \equiv -M_n(\vec{k}_\perp = 0, \hbar\omega = E_n). \quad (5)$$

Correspondingly, exciton energy is shifted toward long-wavelength region by the value equal to the sum of energy shifts of the corresponding states of electron ($\Delta^{(e)}$) and hole ($\Delta^{(h)}$).

The aim of the present work is the creation of the algorithm and calculation of the exciton ground state energy and its binding energy in QW formed by placing of the NF of layered semiconductor of the PbI₂ type into non-crystal isotropic dielectric medium, and establishment of their dependence on the NF thickness as well.

3. CALCULATION METHOD AND OBTAINED RESULTS

Exact solution of the stationary Schrödinger equation with Hamiltonian (1) is impossible, since its eigenfunctions are unknown. For a search of the approximate solution we will use, similarly to [20, 28], the Bethe variational method. To this end, we will transform (1) to the following sum:

$$\hat{H}_{\text{ex}} = \sum_{j=e,h} \hat{H}_j(z_j) + \hat{H}_{SS} + U_\beta(\vec{\rho}, z_e, z_h) - \frac{e^2}{\varepsilon_{\infty\perp}^{(0)}} \sum_{n \neq 0} \frac{\xi^{|n|}}{\sqrt{\rho^2 + \{\varepsilon_{\infty\parallel}^{(0)} [z_e - (-1)^n z_h + na] / \varepsilon_{\infty\perp}^{(0)}\}^2}}, \quad (5)$$

where

$$\hat{H}_{SS} = -\frac{\hbar^2 \nabla_\rho^2}{2\mu} - \frac{e^{*2}}{\varepsilon_{\infty\perp}^{(0)} \rho} \quad (6)$$

is the Shinada-Sugano Hamiltonian [29] which describes the ground state of 2D-exciton whose reduced mass is μ ; $e^* = \beta e$; β is the variational parameter, and

$$U_\beta(\vec{\rho}, z_e, z_h) = \frac{e^{*2}}{\varepsilon_{\infty\perp}^{(0)} \rho} - \frac{e^2}{\varepsilon_{\infty\perp}^{(0)} \sqrt{\rho^2 + [\varepsilon_{\infty\parallel}^{(0)} (z_e - z_h) / \varepsilon_{\infty\perp}^{(0)}]^2}} \quad (7)$$

is the function dependent on this parameter, which defines perturbation induced by the difference of exciton motion in NF in contrast to two-dimensional motion. The last term in (5) describes the polarization interaction of exciton with planes of heterotransitions [17, 23].

Eigenfunctions $\varphi(z_j)$ and eigenvalues $E^{(j)}$ of Hamiltonian (2), which correspond to the ground state of a carrier in NF with taking into account the SP effect, are given in [26], and of Hamiltonian (6), respectively, $\varphi(\vec{\rho})$ and E_{2D} – in [29]. Therefore, considering two first terms of Hamiltonian (5) its main part, in the first approximation of the perturbation theory, eigenfunction of this operator can be given in the form of the product

$$\psi(\vec{\rho}, z_e, z_h) = \phi(z_e) \phi(z_h) \varphi(\vec{\rho}), \quad (8)$$

and exciton ground state energy in NF (without taking into account the EPI)

$$E_{\text{ex}}^{(0)} = \min_\beta [\langle \psi | \hat{H}_{\text{ex}} | \psi \rangle / \langle \psi | \psi \rangle]$$

– in the form of the sum

$$E_{\text{ex}}^{(0)} = E_g + E^{(e)} + E^{(h)} - E_b, \quad (9)$$

where E_b is the exciton binding energy; E_g is the band gap width of the well material.

Using expressions for the energy of 2D-exciton (E_{2D}) and corrections for perturbations which are determined by the averaging of two last terms of Hamiltonian (5) on functions (8), we have found the analytical expression for the calculation of the exciton binding energy

$$E_b = 4R_{\text{ex}} \beta_0^4 \left\{ 1 - \frac{64a}{\pi a_{\text{ex}}} [I_0(a, \beta_0) + \frac{\varepsilon_{0\parallel}^{(0)}}{\varepsilon_{0\perp}^{(0)}} \sum_{n=1}^{\infty} \xi^n I_n(a, \beta_0)] \right\}. \quad (10)$$

Here

$$I_n(a, \beta) = \int_0^{2\pi} \cos nx \cdot \sin^2 \frac{x}{2} \left[\frac{1}{x} - \frac{x}{x^2 + 4\pi^2} \right]^2 \frac{\ln\left(\frac{a_{\text{ex}} x}{4a\beta^2}\right)}{x^2 - \left(\frac{4a\beta^2}{a_{\text{ex}}}\right)^2} dx$$

is the function which defines at $n=0$ correction for the exciton energy due to the perturbations (7) and at $n \geq 1$ – due to the SP effect; a_{ex} and R_{ex} are, respectively, the exciton radius and the Rydberg constant in the well material, and β_0 is the parameter which minimizes the value of $E_{\text{ex}}^{(0)}$.

Formal analysis of the dependence of the second term in (10) on the variable a shows that remaining positive it tends to zero in the borderline cases of the infinitely small and infinitely large a . The second case is outside the framework of the studied model, but such analysis allows to state that with the increase in the variable a binding energy decreases from the value of $E_b = 4R_{\text{ex}}$ typical for 2D-exciton.

Calculation technique of the carrier energy $E^{(j)}$ with taking account the SP effect and determination of its shift value $\Delta^{(j)}$ due to the interaction with phonons in NF of different thickness is given in [26]. Its use gives the possibility to study the change of the exciton energy (9) with the change in the SC conditions and estimate the contribution of the SP and EPI effects by numerical analysis of the a -dependent function

$$E_{\text{ex}} = E_{\text{ex}}^{(0)} - \Delta^{(e)} - \Delta^{(h)}. \quad (11)$$

4. ANALYSIS OF THE OBTAINED RESULTS AND CONCLUSIONS

Specific calculations are performed on the example of the NF of layered semiconductor of the 2H-PbI₂-type (parameters of the crystal lattice are the following: $a_\perp = 4,557 \text{ \AA}$, $a_\parallel \equiv c = 6,979 \text{ \AA}$; energy of the LO-phonon $\Omega_{LO} = 110 \text{ cm}^{-1}$ [30]; permittivities $\varepsilon_{0\parallel} = 7,5$; $\varepsilon_{0\perp} = 8,31$; $\varepsilon_{\infty\parallel} = 6,0$; $\varepsilon_{\infty\perp} = 6,25$ [32]; effective masses of an electron $m_\parallel = 1,25$ and $m_\perp = 0,25$ [33]) embedded into E-MAA polymer ($\varepsilon_0 = \varepsilon_\infty = 2,38$ [34]).

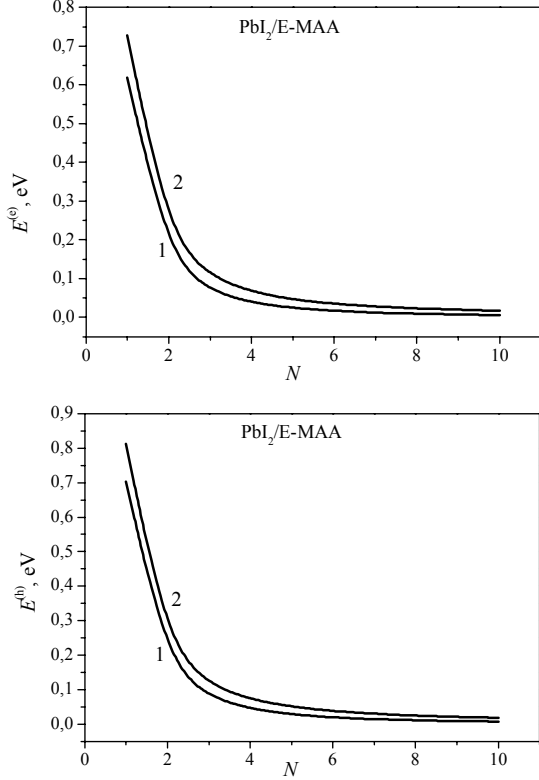


Fig. 1 – Dependence of the energy of the bottom of main minizone of an electron (above) and a hole (below) on the NF thickness calculated without (curves 1) and with taking into account (curves 2) the SP influence

Results of the numerical calculation of the dependence on the NF thickness ($\alpha = Nc$) of the energy of the bottom of main minizone for an electron (hole) $E^{(e)}$ ($E^{(h)}$) and of the binding energy E_b are illustrated in Fig. 1. With the decrease in the NF thickness from 10 to 2 layers of lead diiodide the bottom of main minizone of an electron (hole) in QW increases by the non-linear law from 6 to 620 (from 7 to 704) meV at $N \leq 2$ (curves 1). Contribution of the SP effect also increases (curves 2) taking the values of 50-100 meV in ultrathin ($N \leq 2$) NF. However, for all values of the NF thicknesses, SC are the dominant factor which determines the carrier energy in QW; influence of the SP effect is significant in ultrathin ($N \leq 2$) films.

Dependence of the exciton ground state binding energy on the NF thickness is shown in Fig. 2. As seen, account of the SP effects increases binding energy. The value of correction for the energy increases with the decrease in the NF thickness exceeding 50 meV at $N \leq 2$.

Energy of the transition to the exciton ground state (Fig. 3) increases in this case that is explained by dominant increase in the carrier energy ($E^{(j)}$) due to both SC and SP effect.

Influence of the EPI on the exciton energy is investigated within the single-phonon approximation using analytical expressions for the calculation of the bottom shift of the electron (hole) minizone $\Delta^{(e)}$ ($\Delta^{(h)}$) presented in [26]. In this approximation, interaction of carriers with LO- and I-phonons gives additive contributions to the value of the long-wavelength shift of the bottom of main exciton minizone

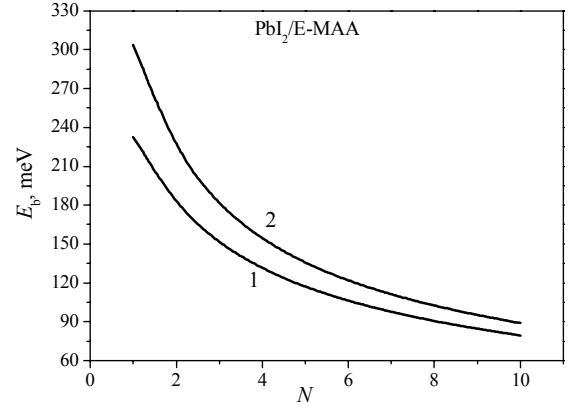


Fig. 2 – Binding energy of the exciton ground state without (curve 1) and with taking into account (curve 2) the SP influence as function of the NF thickness (N is the number of PbI₂ layers in NF)

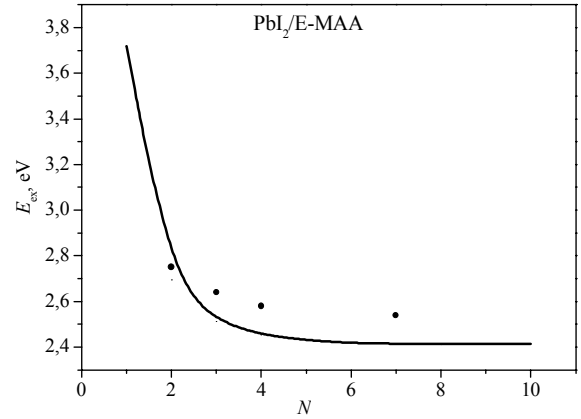


Fig. 3 – Change in the transition energy to the exciton ground state at the NF change in the thickness (dots denote the experimental results [6])

$$\Delta = \Delta^{(e)} + \Delta^{(h)} = (\Delta_{LO}^{(e)} + \Delta_I^{(e)}) + (\Delta_{LO}^{(h)} + \Delta_I^{(h)}) = \Delta_{LO} + \Delta_I.$$

As the analysis of the calculation results shows, the value of the shift induced by the interaction with LO-phonons (Δ_{LO}) non-linearly increases with the increase in the NF thickness (Fig. 4, curve 1). In this case, contribution of higher ($\lambda \geq 2$) states is considerably less than contribution of the phonon ground state ($\lambda = 1$). Contribution of higher electron states ($n \geq 2$) to the value of Δ_{LO} increases with the increase in the NF thickness that is explained by the decrease in the distance between levels of dimensional quantization and, therefore, the increase in the mutual influence of electron levels in the interaction process with phonons.

Character of the influence of I-phonons is another one: at the increase in the NF thickness the value of shift Δ_I decreases (Fig. 4, curve 2). Role of higher ($n \geq 2$) electron states also increases by the same reason that in the case of the interaction with LO-phonons, however, their influence here is less since $\Omega_1 < \Omega_{LO}$.

Dependence of the total shift of the bottom of main exciton zone on the NF thickness is found to be a non-monotonous (see Fig. 4, curve 3) that is explained by the specified differences between EPI and LO- and I-phonons. In ultrathin (up to $N = 8$) NF the value of the long-wave-

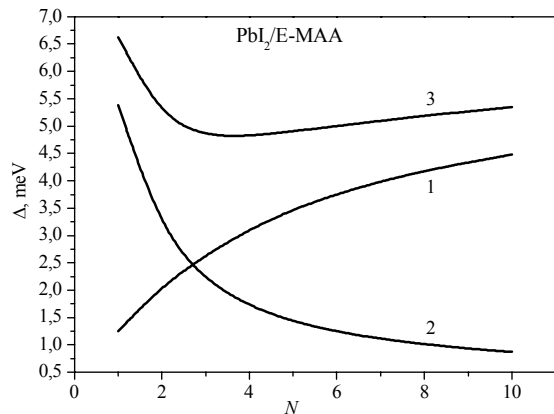


Fig. 4 – Dependence on the NF thickness of the shift value of the bottom of main exciton zone induced by the interaction with phonons: 1 – LO; 2 – I, 3 – both types

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