TEMPERATURE DEPENDENCE OF THE EXCITONIC TRANSITION ENERGY IN FLAT SEMICONDUCTOR NANOFILMS

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PACS 71.35.CC, 72.20.Mf
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The Green's function method has been applied to study the temperature dependence of the exciton ground state energy in a flat semiconductor nanofilm. The binding energy of an exciton and its temperature dependence have been evaluated within the Bethe method. Numerical calculations were carried out in the framework of the finite-depth rectangular quantum well model, by considering flat nanofilms on the basis of double heterojunctions β-HgS/β-CdS and GaAS/$Al_xGa_{1-x}As$ as examples. The exciton binding energy in the studied nanosystems with weak electron-phonon coupling was found to be practically independent of the temperature T. The energy of the excitonic transition into the ground state decreases nonlinearly with increase of T owing to the renormalization of the charge carrier energy by the electron-phonon interaction. The rate of temperature-induced exciton energy change depends on the nanofilm thickness a, being the largest at $a \leq 10 \div 25$ nm.

1. Introduction

Many results of experimental and theoretical researches concerning the formation and decay of exciton states in nanostructures with quantum wells (QWs) and their interaction with other quasiparticles and external fields have been published for last 10–15 years. The reason for those studies is a hope to observe collective phenomena, such as the condensation and superfluidity of excitons, in them [1] and the fact that the binding energy (BE), the oscillator strength, and the lifetime of excitons grow in such systems as a result of the quasiparticle motion confinement in QWs. That is why the excitons in nanoheterostructures with QWs can be observed at rather high temperatures, up to room one, which increases the possibility to use the processes with their participation for the creation of novel optoelectronic devices [2–4].

The manifestations of exciton effects in absorption, reflection, and photoluminescence spectra produced by flat semiconductor nanoheterostructures with QWs were observed in works [5–14]. The domination of phonon-free lines of excitonic transitions in photoluminescence spectra [15] and the dependence of the exciton energy on the QW width, the external field, and the temperature [8, 16] have been established experimentally.

The problem of the determination of the energy and the BE of an exciton in nanostructures with QWs was considered in plenty of works, an incomplete list of which is given in work [17]. The excitonic transition energy in such structures is governed by the energies of an electron and a hole in the QW and by the energy of their interaction. Theoretical researches of the charge carrier spectra in QWs, which take the interaction with phonons into account, are carried out, as a rule, by the theory of perturbation [18] or the Lee–Low–Pines [19] method, and the BE either is supposed to be independent of the QW width [20, 21] or is calculated within the Ritz variational method and making or not allowance for the interaction with phonons [17, 22–25]. However, the analytic dependence of the BE on the QW width cannot be obtained even in the simplest version of this method [17].

Moreover, the temperature-induced variations of the exciton energy were studied neither in the works indicated above nor in other theoretical works. Provided a low concentration of quasiparticles, such a problem can be solved in the framework of the Green's function method and making use of the Feynman–Pines diagram technique [26]. For this purpose, we found the Hamiltonian of the electron-phonon system in the second-quantization representation for all variables for a nanofilm (NF)–a flat semiconductor nanoheterostruc-

ture with a single QW [27]. This gave us an opportunity to study, on the basis of the universal Green's function method, the interaction of electrons with phonons of all polarization branches in NFs and, in particular, the renormalization of the position of the electron ground-state band bottom due to this interaction [28].

As soon as the energies of an electron and a hole in NFs of various thicknesses renormalized by the interaction with phonons are known, the calculation of the exciton state energies becomes feasible. This work aims at studying the origin and the character of temperature-induced variations of the exciton energy in NFs with various thicknesses and various strengths of the electron-phonon coupling. The specific calculations were carried out for NFs based on double heterojunctions β-HgS/β-CdS and GaAS/$Al_{0.3}Ga_{0.7}As$. The exciton BE was found to be practically independent of the temperature T in the studied NFs with a weak electron-phonon interaction. At the same time, the energy of the transition into the ground exciton state decreases nonlinearly as T increases. The rate of temperature-induced variations of the exciton energy in the NF depends on its thickness, being the most in thin $(10-25$ nm) films.

2. Binding Energy and Exciton-phonon Interaction in a Flat Semiconductor NF

In this section, a short review of the technique applied in theoretical researches of the temperature dependence of the exciton energy in a flat semiconductor NF is made. The method is based on our previous works [27–29], where the necessary relations were given in the explicit form, and the detailed analysis of the electron-phonon and electron-hole interaction in NFs was carried out.

Leaving aside the free motion of the center of masses of an electron and a hole in the directions parallel to the film plane, the Hamiltonian for an exciton in the NF can be taken in the form

$$\hat{H}_{\rm ex} = \hat{H}_e^{\perp} + \hat{H}_h^{\perp} + \hat{H}_{SS} + \hat{H}_p = \hat{H}_0 + \hat{H}_p, \tag{1}$$

where the following notations are used: the terms

$$\hat{H}_j^{\perp} = -\frac{\hbar^2}{2}\frac{\partial}{\partial z_j}\frac{1}{m_j(z_j)}\frac{\partial}{\partial z_j} + V_j(z_j)\ (j=e,h) \tag{2}$$

describe the motion of the j-th carrier confined by the potential V_j along the axis OZ which is perpendicular to the NF surface;

$$\hat{H}_{SS} = -\frac{\hbar^2\nabla_{\boldsymbol{\rho}}^2}{2\mu} - \frac{\beta e^2}{\varepsilon\rho} \tag{3}$$

is the Shinada–Sugano Hamiltonian [30] which describes a two-dimensional (2D) exciton with the reduced mass $\mu = m_e m_h/(m_e+m_h)$ and is modified by introducing the Bethe variational parameter β [29];

$$\hat{H}_p = \frac{\beta e^2}{\varepsilon\rho} - \frac{e^2}{\varepsilon|\mathbf{r}_e - \mathbf{r}_h|} \tag{4}$$

is the perturbation operator which makes allowance for the difference between the motions of three-dimensional (3D) and 2D excitons in the NF; and $\boldsymbol{\rho} = \boldsymbol{\rho}_e - \boldsymbol{\rho}_h$.

We suppose $\hat{H}_p$ to be a small perturbation of the system with the Hamiltonian $\hat{H}_0$ and seek the exciton energy in the NF in the form

$$E_{n_e,n_h,N}(a) = E_g^{(0)} + E_{n_e}^{(e)}(a) +$$

$$+E_{n_h}^{(h)}(a) + E_N + \Delta E_{n_e,n_h,N}, \tag{5}$$

where $E_N = -\beta^2 R_{\rm ex}/(N+1/2)^2$ is a 2D exciton energy in the state with the principal quantum number N ($N = 0,1,2\ldots$) [30], $R_{\rm ex}$ the effective Rydberg constant for the NF material, $E_g^{(0)}$ the energy gap width in the NF material, $E_{n_j}^{(j)}(a)$ the quasiparticle energy in the n_j-th band in the QW, and a the QW width [18]. The last term on the left-hand side of formula (5) is a correction to the eigenvalue of the Hamiltonian $\hat{H}_0$ which arises due to a perturbation defined by the operator $\hat{H}_p$. The explicit form of its dependence on the NF thickness a for the ground state of an exciton ($n_e = 1, n_h = 1, N = 0$) was found in work [29]. It allows the transition energy

$$E_{\rm ex}(a) = E_g^{(0)} + E_1^{(e)}(a) + E_1^{(h)}(a) - E_b(a) \tag{6}$$

and the BE

$$E_b(a) = 4R_{\rm ex}\beta_0 - \Delta E(a,\beta_0) \tag{7}$$

to be expressed as functions of a (here, $\Delta E(a,\beta) = \Delta E_{1,1,0}$, and β_0 is the value of a variational parameter that minimizes the energy of the exciton ground state). For every fixed a, the quantity $\Delta E(a,\beta)$ and, therefore, the exciton BE are determined by the nanostructure parameters: the lattice constants and the dielectric permittivities of the media (the NF itself and the barrier region), the effective masses, and the transverse components of the wave vectors of an electron and a hole in each medium.

The Hamiltonian of the exciton-phonon system in a NF looks like

$$\hat{H} = \hat{H}_{\rm ex} + \hat{H}_{\rm ph} + \hat{H}_{\rm int}, \tag{8}$$

where

$$\hat{H}_{\rm ph} = \hat{H}_{L0} + \hat{H}_{L1} + \hat{H}_I \tag{9}$$

is the Hamiltonian of the system of longitudinal optical polarization phonons: confined in the NF ($L0$), semiconfined in the barrier medium ($L1$), and interface ones (I) [18]. Provided that the exciton radius considerably exceeds the polaron one, we may assume that exciton-phonon coupling is realized through the phonon interaction with the electron and the hole. Then, the Hamiltonian of the exciton-phonon interaction looks like

$$\hat{H}_{\rm int} = \hat{H}_{{\rm ex}-L0} + \hat{H}_{{\rm ex}-L1} + \hat{H}_{{\rm ex}-I} =$$

$$= \sum_{j=e,h} \eta_j \left[\sum_{n_j,n'_j,\mathbf{k}_\parallel} \sum_{\lambda,\mathbf{q}_\parallel} F^{\lambda}_{n_j n'_j}(\mathbf{q}_\parallel) \hat{a}^+_{n'_j \mathbf{k}_\parallel + \mathbf{q}_\parallel} \hat{a}_{n_j \mathbf{k}_\parallel} \hat{B}_{\lambda q_\parallel} + \right.$$

$$+ \sum_{q_\perp,\mathbf{q}_\parallel} F^{q_\perp}_{n_j n'_j}(\mathbf{q}_\parallel) \hat{a}^+_{n'_j \mathbf{k}_\parallel + \mathbf{q}_\parallel} \hat{a}_{n_j \mathbf{k}_\parallel} \hat{B}_{q_\perp q_\parallel} +$$

$$\left. + \sum_{\sigma,p,\mathbf{q}_\parallel} F^{\sigma p}_{n_j n'_j}(\mathbf{q}_\parallel) \hat{a}^+_{n'_j \mathbf{k}_\parallel + \mathbf{q}_\parallel} \hat{a}_{n_j \mathbf{k}_\parallel} \hat{B}_{\sigma p q_\parallel} \right], \tag{10}$$

where $\eta_e = 1$; $\eta_h = -1$; $\hat{a}^+_{n_j \mathbf{k}}$ and $\hat{a}_{n_j \mathbf{k}}$ are the operators of creation and annihilation, respectively, of an electron (hole) with the wave vector $\mathbf{k}$ in the n_j-th band; $\hat{B}_{\Lambda \mathbf{q}_\parallel} = \hat{b}_{\Lambda,\mathbf{q}_\parallel} + \hat{b}^+_{\Lambda,-\mathbf{q}_\parallel}$ ($\Lambda = q_\lambda, q_\perp, (\sigma,p)$ is the subscript that distinguishes the phonon type and, simultaneously, determines the transverse component $q_\perp$ of its wave vector $\mathbf{q}$ [18]); and $F^{\Lambda}_{n_j n'_j}(\mathbf{q}_\parallel)$ are the functions of coupling with the corresponding phonon state. The explicit forms for the dependences of coupling functions on the NF thickness a and the longitudinal component $\mathbf{q}_\parallel$ of the wave vectors of $L0$- and $L1$-phonons, as well as the symmetric ($\sigma = S$) and antisymmetric ($\sigma = A$) branches of I-phonons, are given in work [27].

The interaction with phonons shifts the ground-state band bottom of the j-th carrier in the QW by Δ_j and, by varying the transverse components of its wave vector $k^{(j)}_{ln}$ (here, n is the band index, and l the medium index), changes the exciton BE. Then, the energy of the exciton transition into the ground state renormalized by the interaction with phonons in the NF with the thickness a is determined by the expression

$$E_{\rm ex}(a) = E^{(0)}_g + E^{(e)}_1(a) + \Delta_e(a) +$$

$$+ E^{(h)}_1(a) + \Delta_h(a) - E_b(a). \tag{11}$$

If the electron-phonon coupling is weak, the shift E_1 of the electron ground-state band bottom in the QW at any temperature T is determined by the magnitude of the one-phonon mass operator of Green's function [26]

$$M(\mathbf{k}_\parallel, E, T) = \sum_{\Lambda,\mathbf{q}_\parallel} \left[\sum_n |F^{\Lambda(d)}_{1n}(\mathbf{q}_\parallel)|^2 \times \right.$$

$$\times \left(\frac{1 + \nu_\Lambda(\mathbf{q}_\parallel, T)}{E - E_n(\mathbf{k}_\parallel - \mathbf{q}_\parallel) - \Omega_\Lambda(\mathbf{q}_\parallel)} + \right.$$

$$\left. + \frac{\nu_\Lambda(\mathbf{q}_\parallel, T)}{E - E_n(\mathbf{k}_\parallel + \mathbf{q}_\parallel) + \Omega_\Lambda(\mathbf{q}_\parallel)} \right) +$$

$$+ \int_0^{\pi/a} dk_\perp |F^{\Lambda(c)}_{1k_\perp}(\mathbf{q}_\parallel)|^2 \left(\frac{1 + \nu_\Lambda(\mathbf{q}_\parallel, T)}{E - E(k_\perp, \mathbf{k}_\parallel - \mathbf{q}_\parallel) - \Omega_\Lambda(\mathbf{q}_\parallel)} + \right.$$

$$\left. \left. + \frac{\nu_\Lambda(\mathbf{q}_\parallel, T)}{E - E(k_\perp, \mathbf{k}_\parallel + \mathbf{q}_\parallel) + \Omega_\Lambda(\mathbf{q}_\parallel)} \right) \right], \tag{12}$$

where $\nu(\mathbf{q}_\parallel, T) = [\exp(\Omega_\Lambda(\mathbf{q}_\parallel)/k_{\rm B}T) - 1]^{-1}$ are the occupation numbers of phonon states, $\mathbf{k}_\parallel$ is the longitudinal component of the electron wave vector, $k_{\rm B}$ is the Boltzmann constant, and the superscripts d and c distinguish between the states in the discrete and continual parts of the spectrum. If the electron-phonon coupling is weak, the mass operator weakly depends on the energy E and the quasimomentum $\mathbf{k} = (\mathbf{k}_\parallel, k_\perp)$ of electron; therefore, the shift of the electron ground-state band bottom in the QW

$$\Delta(a, T) \equiv M(\mathbf{k}_\parallel = 0, E = E_1, T) =$$

$$= \Delta_{L0}(a, T) + \Delta_{L1}(a, T) + \Delta_I(a, T) \tag{13}$$

becomes a sum of partial shifts which are dependent on the NF thickness a and the temperature T and caused by the interaction with $L0$-, $L1$-, and I-phonons, respectively. Each of the partial shifts is formed by mechanisms of electron-phonon interaction through the states in the discrete and continual parts of the spectrum. The explicit forms for the shift dependences caused by each

indicated mechanism on the NF thickness and the temperature were published in work [28] and, since they are cumbersome, are not given here.

The interaction with phonons makes the shifts of the energy band edges and, therefore, the exciton BE (through the variation of $k_{l1}^{(j)}$-values), as well as the energy of the exciton transition into the ground state in the NF dependent on a and T:

$$E_{\rm ex}(a,T) = E_g^{(0)} + E_1^{(e)}(a) + \Delta_e(a,T) +$$

$$+ E_1^{(h)}(a) + \Delta_h(a,T) - E_b(a,T). \qquad (14)$$

3. Analysis of the Results Obtained

The results of our calculations evidence for a weak influence of the exciton-phonon interaction on the exciton BE magnitude in the studied NFs [29]. Accordingly, the temperature dependence of the exciton BE turned out insignificant. Therefore, temperature-induced variations in the energy of the exciton ground state in the NF take place as a result of the electron and hole ground state energy renormalization owing to their interaction, mainly, with confined and symmetric interface phonons [27]. In Figs. 1 and 2, the temperature dependences of the partial (Δ_I, Δ_{L0}) and total (Δ) shifts of the bottoms of the electron and hole ground-state bands, which arise owing to the interaction with interface and confined phonons, are plotted for various NF thicknesses. One can see that Δ_{L0} in studied NFs grows, when the temperature increases above 100 K. In thick enough NFs, the shift becomes substantial, reaching–at $T \approx 300$ K– 5 (7) meV for electrons (holes) in CdS/HgS/CdS and 3.5 (11) meV in $Al_{0.3}Ga_{0.7}As/GaAS/Al_{0.3}Ga_{0.7}As$ heterosystems.

Temperature changes of Δ_I manifest themselves substantially at $T > 100$ K, as it was in the case of the interaction with confined phonons, but in thin NFs. In thin enough NFs, the shift becomes considerable, reaching – at $T \approx 300$ K–16 (18) meV for electrons (holes) in CdS/HgS/CdS and 5 (15) meV in $Al_{0.3}Ga_{0.7}As/GaAS/Al_{0.3}Ga_{0.7}As$.

In contrast to $\Delta_{L0}(T)$, the dependence $\Delta_I(T)$ is more complicated: the temperature growth can either increase or diminish the shift of the electron (hole) ground-state band bottom, depending on the NF thickness. This fact originates from the feature in the dependence of the function of the electron-phonon coupling with I-phonons on the quasimomentum: its maximum can fall in the range of small or large quasimomenta, depending on the NF thickness and the band number in the QW. Respectively, the interaction with phonons will be actual for carriers with small or large (in comparison with the phonon energy) values of kinetic energy. The manifestations of this interaction are different; it is most probably that low-energy quasiparticles will get energy from the phonon system, whereas the high-energy ones will give it back. Each of those processes favors the band bottom shift to opposite directions. The number of levels in the QW, their positions, and the distances between them change with the NF thickness variation. Accordingly, the influence of processes with absorption and emission of I-phonons of both types–low- ($p = -$) and high-energy ($p = +$) ones–changes as well. The nonmonotonic behavior of those effects at the NF thickness variation is the factor that is responsible for the indicated differences between the temperature dependences $\Delta_I(T)$.

The total shift Δ turns out to be a growing function of the temperature. However, its magnitude and the rate of temperature-induced growth are different for different numbers N of monolayers in the NF. It can be explained by the fact that, in thin films, the main role is played by the interaction of charge carriers with interface phonons (in extremely thin films, with semiconfined phonons as well), whereas, in thick enough films, with confined phonons, which is substantially weaker. The difference between the temperature-induced variations of occupation numbers for different types of phonons is also important.

The dependences of the partial and total shifts on the NF thickness a at various temperatures are shown in Fig. 3. As is seen, the long-wave shift of the electron and hole ground level in thin NFs (less than 20–25 nm for β-HgS and 10–15 nm for GaAs) is mainly caused by the interaction with interface phonons, and–for larger NF thickness–with confined ones. If the NF is thick enough, the influence of interface phonons vanishes, and Δ_{L0} approaches the value characteristic of the barrier medium. It can be explained by the fact that the probability of finding an electron at the heterojunction interface decreases – in the QW region, on the contrary, increases – as the NF thickness grows.

In Fig. 4, the plots of the temperature dependences of the excitonic transition energy $E_{\rm ex}(a)$ in the NF under investigation, calculated taking the exciton-phonon interaction into account, are presented for two values of the NF thickness: 2 and 20 monolayers. One can see that the temperature growth brings about a reduction of the excitonic transition energy owing to the renormalization of the energy spectrum of the electron and the hole.

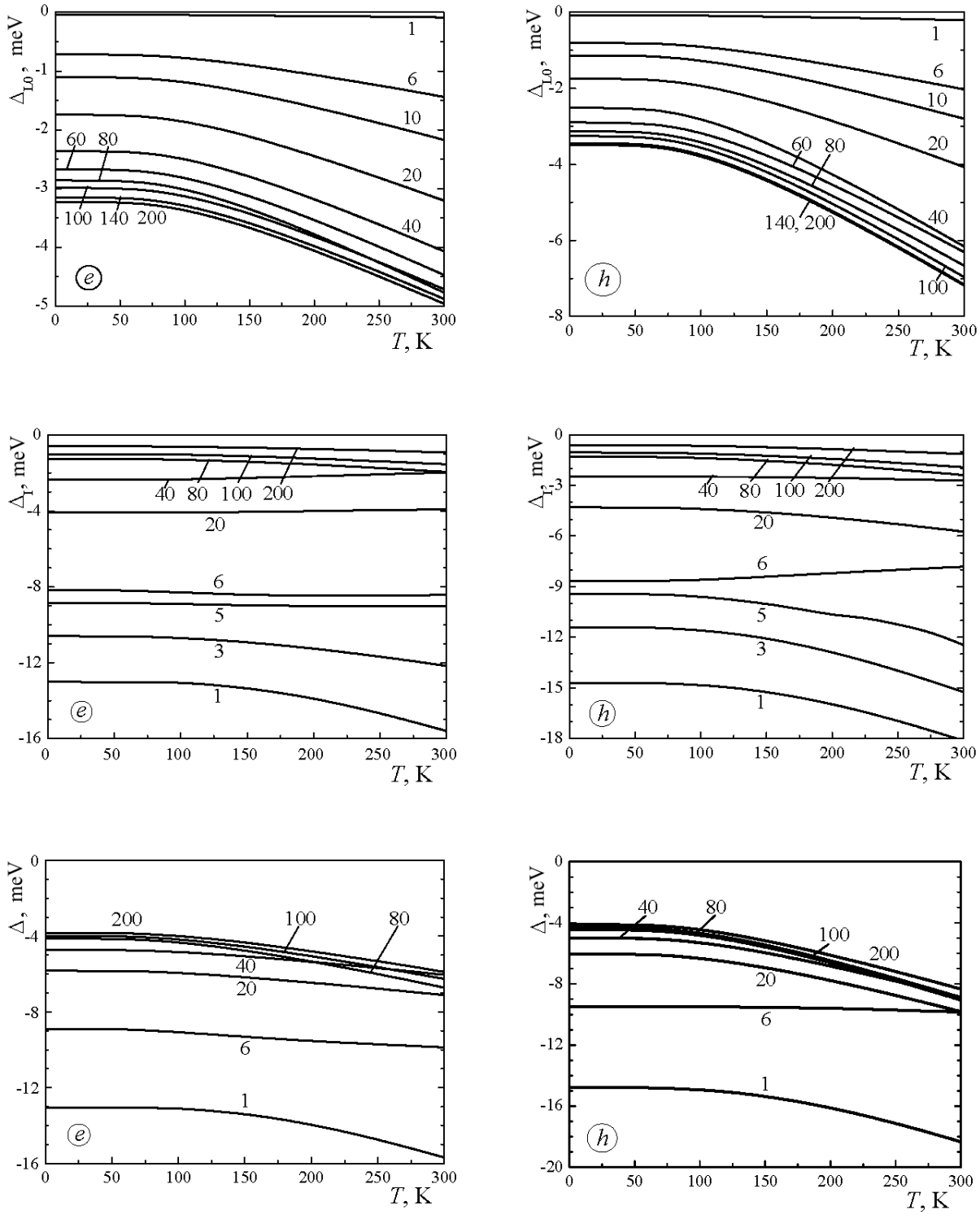

Fig. 1. Temperature dependences of the partial (Δ_{L0}, Δ_I) and total ($\Delta = \Delta_{L0} + \Delta_I$) shifts of the electron (e) and hole (h) ground state energy in the NF having a thickness of N β-HgS monolayers (indicated by the numbers near the curves) surrounded by massive β-CdS

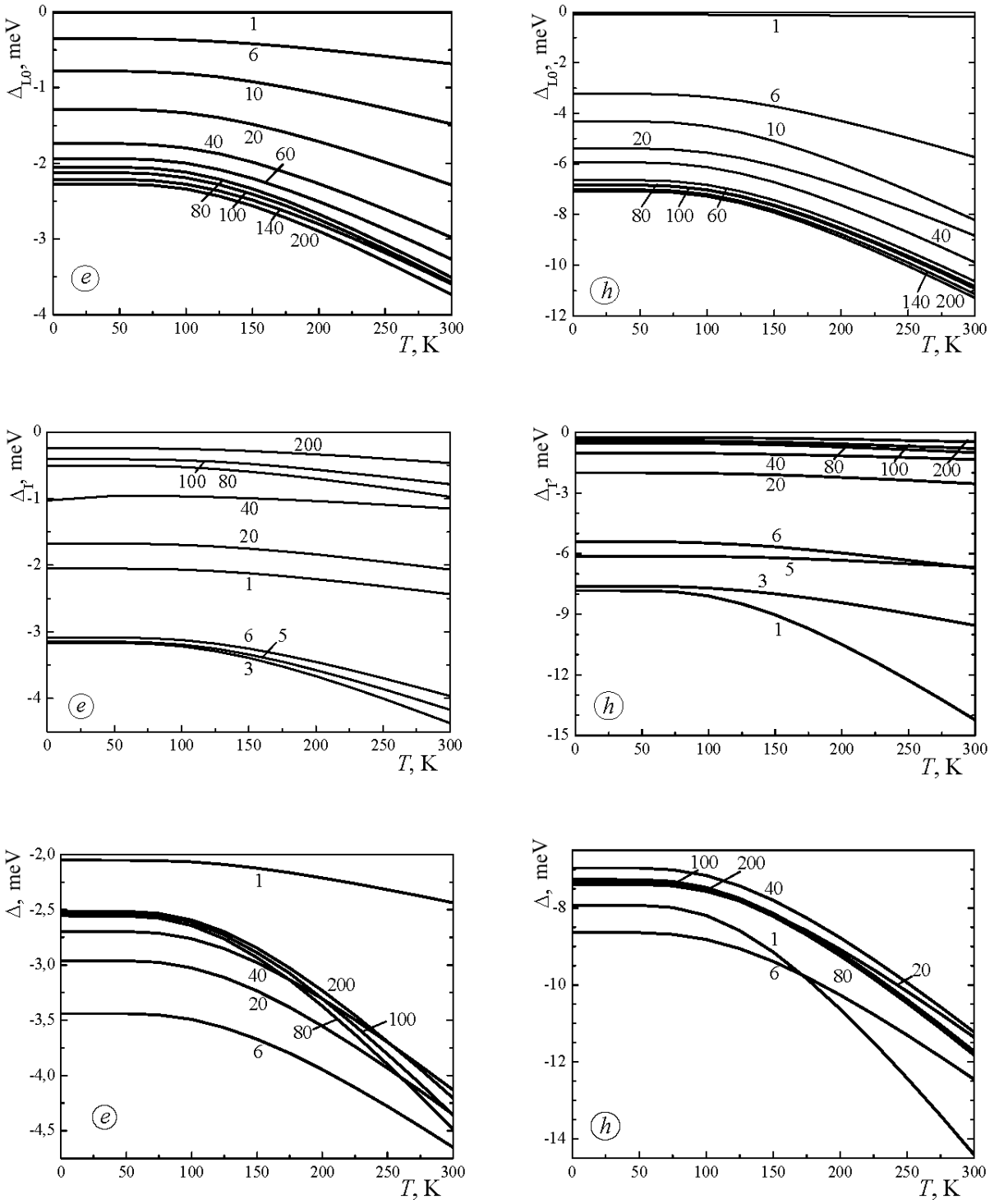

Fig. 2. Temperature dependences of the partial and total shifts of the electron and hole ground state energy in the NF having a thickness of N GaAs monolayers surrounded by massive $Al_{0.3}Ga_{0.7}As$

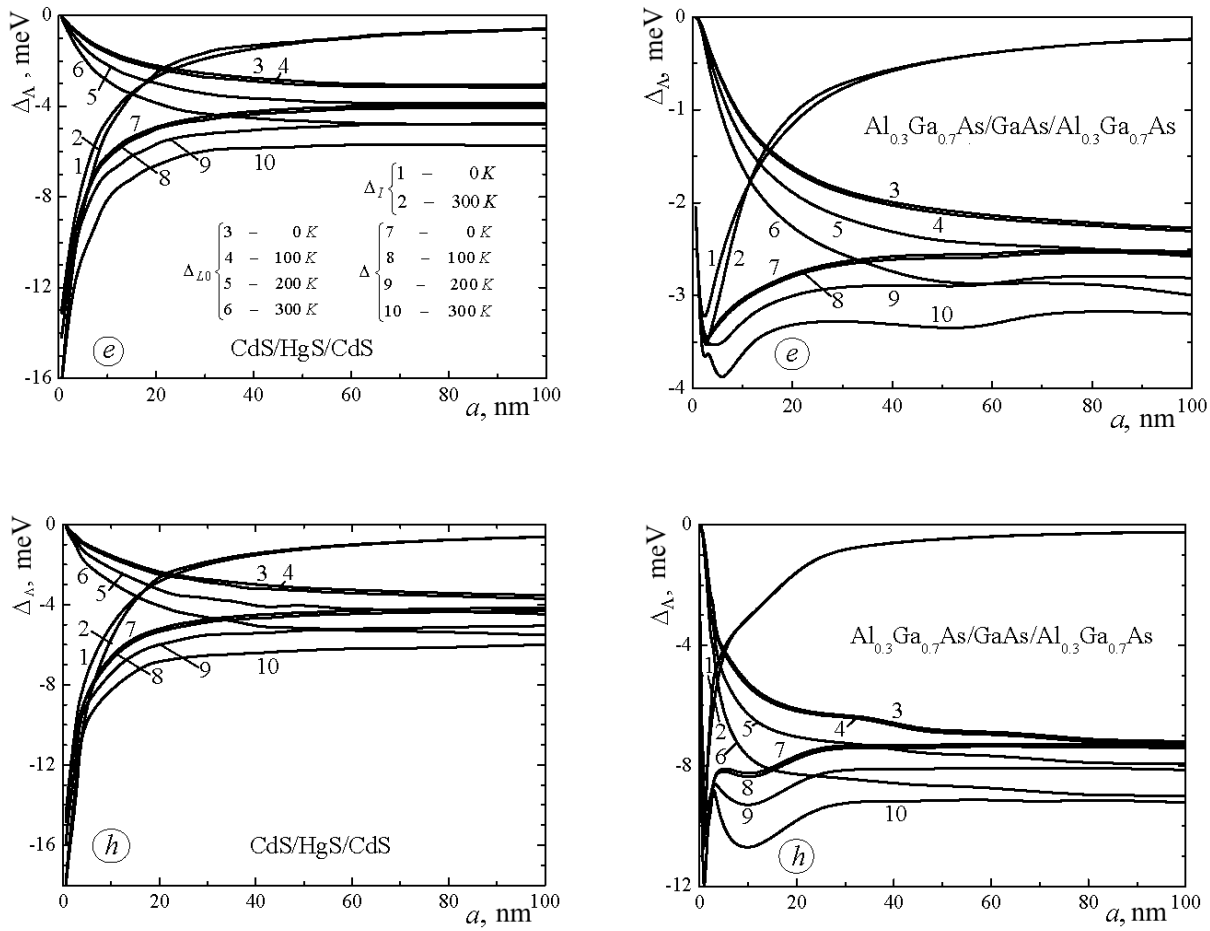

Fig. 3. Dependences of the partial, Δ_Λ ($\Lambda = I, L0$), and total, $\Delta = \Delta_{L0} + \Delta_I$, shifts on the NF thickness a at various temperatures

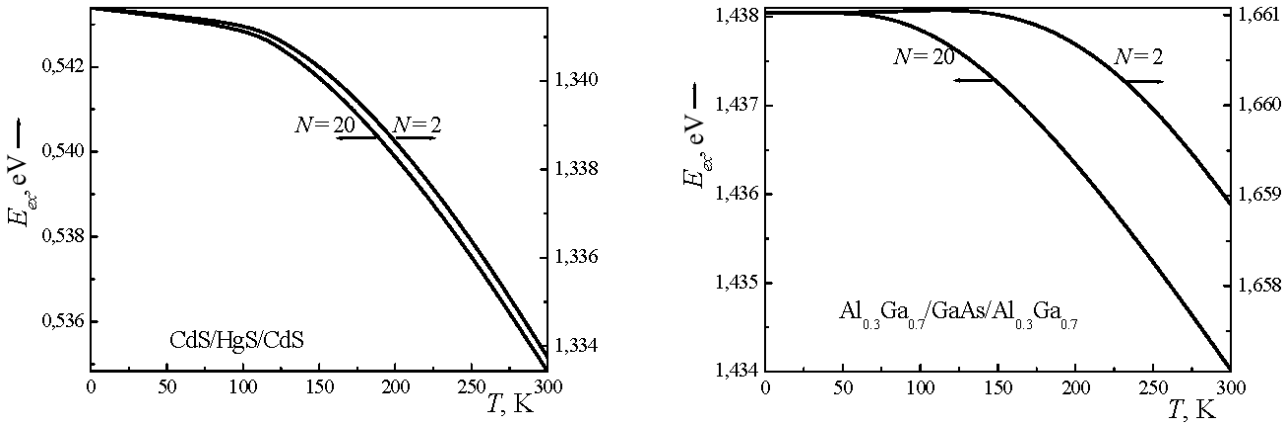

Fig. 4. Temperature dependences of the exciton energy $E_{\rm ex}$ in the NF having a thickness of N monolayers

Similar temperature changes of the excitonic transition energy were observed experimentally in work [8], where the photoconductivity spectra in $Ga_xIn_{1-x}As/InP$ nanoheterostructures with numerous QWs were studied. However, the quantitative comparison between the results of our calculations and

the experimental ones is impossible. At the same time, unfortunately, there are no experimental data concerning the temperature dependence of the excitonic transition energy in flat NFs with a single QW.

4. Conclusions

In the framework of the single rectangular finite-depth QW model and making use of the Green's function method, the theoretical research of the temperature dependence of the exciton energy in a flat semiconductor nanofilm has been carried out for the first time. The results of calculations executed for CdS/HgS/CdS and $Al_{0.3}Ga_{0.7}As/GaAS/Al_{0.3}Ga_{0.7}As$ nanostructures as examples and taking the interaction of excitons with longitudinal optical phonons (confined in the NF, semiconfined in the barrier medium, and interface ones) testify to a nonlinear reduction of the excitonic transition energy with the temperature growth as a result of temperature-induced changes in the energy spectrum of the electron and the hole.

It was found that the temperature dependence of the exciton energy in thin NFs is a result of the interaction of charge carriers mainly with the symmetric branch of interface phonons, whereas in thick ones, it is interaction with confined LO-phonons. Such a conclusion agrees with the results of theoretical and experimental researches of the influence of the electron-phonon interaction on the electron mobility in flat nanostructures with QWs [9].

The rate and the amplitude of temperature-induced variations depend on the NF thickness. They are the largest in thin films, where the interaction with interface phonons is strong.

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Received 04.02.09.
Translated from Ukrainian by O.I. Voitenko

ТЕМПЕРАТУРНА ЗАЛЕЖНІСТЬ ЕНЕРГІЇ ЕКСИТОННОГО ПЕРЕХОДУ У ПЛОСКИХ НАПІВПРОВІДНИКОВИХ НАНОПЛІВКАХ

В.М. Крамар

Р е з ю м е

Методом функцій Гріна досліджено температурну залежність енергії переходу в основний стан екситону у плоскій напівпровідниковій наноплівці. Варіаційним методом Бете оцінено величину і досліджено температурну залежність енергії зв'язку екситону. Розрахунки виконано у моделі прямокутної квантової ями скінченної глибини на прикладі плоских наноплівок, утворених на основі подвійних гетеропереходів β-HgS/β-CdS та GaAS/$Al_xGa_{1-x}As$. Встановлено, що у досліджуваних наносистемах зі слабкою електрон-фононною взаємодією енергія зв'язку екситону практично не залежить від температури T. Енергія переходу в основний екситонний стан нелінійно зменшується при зростанні T внаслідок перенормування енергії носіїв електрон-фононною взаємодією. Швидкість температурних змін енергії екситону залежить від товщини наноплівки a і є найбільшою при $a \leq$ 10–25 нм.