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# Temperature changes in the excitonic absorption band in flat double nanoheterostructures GaAs/Al$_x$Ga$_{1-x}$As

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**Abstract.** Adduced in this paper are the method and results of theoretical studying the effects of spatial confinement and exciton-phonon interaction on the position and shape of the excitonic absorption band in flat double nanoheterostructures GaAs/Al$_x$Ga$_{1-x}$As. The heterojunction has been considered as unstrained, the nanosystem is modeled as a rectangular quantum well of a finite depth. Interaction of exciton with optical polarization phonons has been taken into account. Calculated has been the temperature dependence of the energy corresponding to transition into the background excitonic state, and determined have been temperature changes in the absorption coefficient related with this transition. It has been shown that observation of these temperature changes in the energy and absorption coefficient, caused by interaction with optical phonons, is possible in the case of exciton with heavy hole at temperatures above 100 K.

## 1. Introduction

The swift development of technologies for studying and obtaining the desired physical properties of low-dimensional heterostructures, which is observed for recent decades, is related with unique features of these systems that are the base for creation of up-to-date facilities in opto- and nanoelectronics [1-4]. The most known structures used with this purpose are those based on the GaAs/Al$_x$Ga$_{1-x}$As heterostructures that are already considered as the classical ones. They serve for creation of low-threshold lasers capable to operate at room temperature, highly-efficient photodetectors, light emitting diodes, re-switchers, quantum cascade lasers and detectors [1, 5-11].

However, despite the fact that up to date their optical properties have been studied rather well (see, for instance, [12]), including also the structure of excitonic spectra in these systems [13, 14], respective investigations last till now [15-22]. In particular, using the quantitative analysis of experimental data, it was shown in [19] that the shape of spectral bands within the range of excitonic reflection inherent to heterostructures with GaAs single quantum wells (QWs) can be described in the first approximation via Lorentz oscillators. It is independent of temperature within the range 8 to 90 K.

But experimental data concerning the temperature changes in the structure of excitonic spectra corresponding to these systems within the high-temperature range are yet unknown. Theoretical investigations of these questions are also absent.

The aim of this work was to study temperature changes caused by interaction of exciton with optical phonons, which are observed in the structure of exciton absorption spectra inherent to the double nanohetero-structures – nanofilms (NF) $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ of various thicknesses and concentrations of Al atoms inside barrier material.

Assuming that interaction of exciton with phonons takes place via participation of electron and hole with them, we have calculated temperature changes in the energy of transition to the background state of excitons with heavy and light holes. It enabled us to analyze temperature changes in positions of respective excitonic peaks.

We have found the function describing the exciton-phonon coupling in the case of prevailing interaction with confined phonons inside a rectangular QW of a finite depth, which allows investigation of the spectral dependence for the coefficient of excitonic absorption and its temperature changes in these nanostructures.

## 2. Choosing the model for investigation and problem formulation

To study character of temperature changes in bands of excitonic transitions in semiconductor nanohetero-structures, usually used is approximation of effective masses for electrons as well as the model of dielectric continuum for phonons (see, for example, [23]). Modeling NF with the system of rectangular QWs (electron and hole ones) with the finite depth equal to $V_e$ and $V_h$, respectively, separated by the interval of forbidden values for energies $E_g$ (gap width for well material), the authors [24] found the explicit form of Hamiltonian for the exciton-phonon system in representation of second quantization over all the variables. Offered there is also the method for determining the binding energy of quasi-two-dimensional exciton in NFs.

We have considered the model of Wannier-Mott exciton created as a result of direct phototransition from the background sublevel $E_h$ in QW of holes to the background sublevel $E_e$ in QW of electrons with account of Coulomb interaction between them under availability of spatial confinement as well as interaction with optical polarization phonons. The energy of transition into the background excitonic state (without any account of interaction with phonons) can be written as the following sum:

$$E^{(0)} = E_g + E_e + E_h - E_b, \qquad (1)$$

where $E_b$ is the binding energy of quasi-2D exciton.

Interaction of exciton with optical branches of the phonon spectrum in a double nanoheterostructure (with confined, semi-confined and interface phonons) is realized via individual interaction of electron and hole with them. It results in re-normalization of the energy spectrum inherent to quasi-particles, i.e., the shift of levels to the side of lower energies. The shift values of the background states for electrons ($\Delta_e$) and holes ($\Delta_h$) appear to be dependent on the $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ NF thickness, concentration of aluminum atoms in barrier medium and temperature of the nanosystem [25]. In accord with (1), it leads to the change in the exciton transition energy as follows

$$E = E^{(0)} + \Delta_e + \Delta_h, \qquad (2)$$

and can be observed as a long-wave shift of the absorption band related with it.

The spectral dependence of absorption caused by exciton transition with participation of phonons is defined by the shape function of the absorption band

$$S(\omega, T) = \frac{\Gamma(\omega, T)}{\left[\hbar\omega - E^{(0)} - \Delta(\omega, T)\right]^2 + \Gamma^2(\omega, T)}, \qquad (3)$$

that depends on temperature $T$ as on parameter. Here, $\hbar\omega$ is the energy of exciting wave, $\Delta(\omega, T)$ and $\Gamma(\omega, T)$ are real and imaginary parts of the mass operator

$$M(\omega, T) = \Delta(\omega, T) - i\Gamma(\omega, T) \qquad (4)$$

corresponding to the Green function of the exciton-phonon system [24].

The main task of investigation, results of which are described in this paper, is to ascertain the character of temperature changes in exciton absorption bands observed in NF $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ of various thicknesses and concentration compositions inherent to barrier material.

## 3. Results and discussion

The mass operator (4) defines influence of exciton-phonon interaction (EPI) on the exciton energy spectrum in the system, position and width of the exciton absorption band. To find it, EPI Hamiltonian can be represented in the following form

$$\hat{H}_{int} = \sum_{v,v',\vec{k}} \sum_{\alpha,\vec{q}_\perp} \Phi_{vv'}^{(\alpha)}(\vec{q}_\perp) \hat{c}_{v''\vec{k}+\vec{q}_\perp}^+ \hat{c}_{v\vec{k}} \hat{B}_{\alpha,\vec{q}_\perp}, \qquad (5)$$

where $\hat{n}_{v\vec{k}}^+$ and $\hat{n}_{v\vec{k}}$ are operators of creation and annihilation for exciton states, while $\hat{b}_{\alpha\vec{q}_\perp}^+$ and $\hat{b}_{\alpha\vec{q}_\perp}$ – for phonon states in NF, $\hat{B}_{\alpha\vec{q}_\perp} = \hat{b}_{\alpha\vec{q}_\perp} + \hat{b}_{\alpha-\vec{q}_\perp}^+$; $\perp$ symbol distinguishes the transversal (inside QW plane) and $\alpha$ – the longitudinal component of the momentum of the phonon respective type; $v = (n_e, n_h, n, m)$, where $n$ and $m$ are quantum numbers defining the state of 2D-exciton, while $n_e$ and $n_h$ – the same for the states of electron and

hole that form it. The function of exciton-phonon coupling is as follows

$$\Phi_{\nu\nu'}^{(\alpha)}(\vec{q}_\perp) = F_{n_e n_e'}^{\alpha}(\vec{q}_\perp) I_{nm,n'm'}^{(e)}(\vec{q}_\perp) - \\ - F_{n_h n_h'}^{\alpha}(\vec{q}_\perp) I_{nm,n'm'}^{(h)}(\vec{q}_\perp), \tag{6}$$

and it is defined here for the case of interaction with confined phonons in infinitely deep QW, like to that adduced in [26]. It can be also used for QW of a finite depth if assuming a respective change in the look of the function for electron-phonon coupling $F_{nn'}^{\alpha}(\vec{q}_\perp)$; its explicit form is represented in [27].

Using the EPI function defined in this manner, in accord to [26] we find the one-phonon mass operator that takes into account interaction of the background exciton state with confined phonons and obtain the explicit form of its real

$$\Delta(w,T) = -\frac{\pi^2 e^2}{a\varepsilon_0} \sum_n \sum_{\lambda=1}^N \lambda^2 \{[1+\nu(T)] \times \\ \times [(C_1^{(e)} C_n^{(e)} X_{1n}^{(e)\lambda})^2 J_\lambda^{(e,+)}(w,a) + \\ + (C_1^{(h)} C_n^{(h)} X_{1n}^{(h)\lambda})^2 J_\lambda^{(h,+)}(w,a) - \\ - 2C_1^{(e)} C_n^{(e)} X_{1n}^{(e)\lambda} C_1^{(h)} C_n^{(h)} X_{1n}^{(h)\lambda} J_\lambda^{(eh,+)}(w,a)] + \\ + \nu(T)[(C_1^{(e)} C_n^{(e)} X_{1n}^{(e)\lambda})^2 J_\lambda^{(e,-)}(w,a) + \\ + (C_1^{(h)} C_n^{(h)} X_{1n}^{(h)\lambda})^2 J_\lambda^{(h,-)}(w,a) - \\ 2C_1^{(e)} C_n^{(e)} X_{1n}^{(e)\lambda} C_1^{(h)} C_n^{(h)} X_{1n}^{(h)\lambda} J_\lambda^{(eh,-)}(w,a)]\} \tag{7}$$

and imaginary

$$\Gamma(w,T) = \Gamma_0 + \frac{\pi^3 e^2}{a\varepsilon_0} \sum_n \sum_{\lambda=1}^N \lambda^2 \{[1+\nu(T)] \times \\ \times \frac{[(C_1^{(e)} C_n^{(e)} X_{1n}^{(e)\lambda})^2 I_0^{(e)}(q_-(w)) - (C_1^{(h)} C_n^{(h)} X_{1n}^{(h)\lambda})^2 I_0^{(h)}(q_-(w))]^2}{(a_0 q_-(w)/\pi)^2 + (\lambda/N)^2} + \\ + \frac{[(C_1^{(e)} C_n^{(e)} X_{1n}^{(e)\lambda})^2 I_0^{(e)}(q_+(w)) - (C_1^{(h)} C_n^{(h)} X_{1n}^{(h)\lambda})^2 I_0^{(h)}(q_+(w))]^2}{(a_0 q_+(w)/\pi)^2 + (\lambda/N)^2} \nu(T)\} \tag{8}$$

parts. Here, $a_0$ and $\varepsilon_0$ are the lattice parameter and static dielectric permittivity of well material, respectively; $N = a/a_0$ is the number of well material layers in NF with the thickness $a$; $C_n^{(p)}$ – normalization constant for the wave function of electron $(p = e)$ or hole $(p = h)$ in the state with quantum number $n$ in its QW; $X_{1n}^{(p)\lambda}$ – adduced in [27] value that defines the coupling function for a carrier in its background state with confined phonon in the state with the quantum number $\lambda$ with participation of the $n$-th level of carrier in QW; $\nu(T)$ are the numbers of filling the phonon states; $w = (\hbar\omega - E^{(0)})/\Omega_{LO}$ – normalized by the value of confined phonon energy (equal to the energy of longitudinal optical phonon $\Omega_{LO}$ in QW material) shift of the exciton absorption peak relatively to its position $E^{(0)}$ calculated without account of EPI; $\Gamma_0$ – phenomenological constant

taking into account relaxation of excitons caused by processes non-included to the accepted model. The rest auxiliary values and functions in Exps (7) and (8) were earlier adduced in [26].

Using the following GaAs parameters $(a_0 = 5.653$ Å, $\varepsilon_0 = 13.18$, $E_g = 1.424$ eV, $\Omega_{LO} = 36.25$ meV) and the AlAs ones $(a_0 = 5.661$ Å, $\varepsilon_0 = 10.06$, $E_g = 2.168$ eV, $\Omega_{LO} = 50.09$ meV) as well as approximation of parameters for $Al_xGa_{1-x}As$ solid solution offered in [28], we calculated temperature dependences of energies inherent to transition to the background exciton state in $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ NF of various thicknesses with aluminum concentration values 0.2, 0.3 and 0.4. As it follows from the calculation results adduced in Fig. 1, the energy of transition to the background exciton state with heavy hole $(m_h = 0.45m_0)$ is a monotonic temperature function, the character and value of which are defined by efficiency of interaction with interface, confined and semi-confined phonons [29], and therefore depend both on the NF thickness and composition of well material.

Accordingly to the results of our calculations, changes in the energy of exciton possessing heavy hole are weak in the studied nanostructures and can be observed at temperatures above 100 K. In NF with the thickness higher than 2.5 nm $(N > 4)$, increase in temperature from 100 up to 300 K causes non-linear decrease of the transition energy in dependence on the values $x$ and $a$ by the value 2 to 3 meV. In ultra-thin NF $(N < 3)$, its value (also in dependence on the values $x$ and $a$) can be lowered by the value 1 to 1.5 meV, or enhanced, or remains uncharged (see curves $N = 2$ in Fig. 1), which is explained by peculiarities of interaction with interface phonons [29].

The energy of transition to the background state of exciton possessing light hole $(m_h = 0.08m_0)$ considerably weaker depends on temperature: the value of its temperature changes does not exceed 0.5 meV.

Availability of the weak temperature dependence for the exciton energy causes the similar changes in spectra of exciton absorption. In particular, the coefficient of exciton absorption α becomes dependent on temperature. In a homogeneous absorbing system, it is related with the shape function of the absorption band via the following relation

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

where $D_0$ is the value of the moment corresponding to electro-dipole transition to the respective exciton state. Non-uniformity in the thickness of NF (roughness) causes unhomogeneous broadening of the band. Therefore, the absorption coefficient should be found as a convolution of two functions – $S(\omega,T)$ and density of normal distribution (Gauss function)
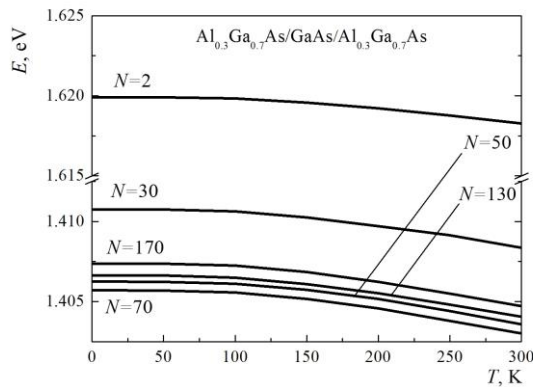
$$G(\omega) = 2\sqrt{\frac{\ln 2}{\pi}} \cdot \frac{1}{\Gamma_I} \exp\left[-4\ln 2\left(\frac{\hbar\omega - E}{\Gamma_I}\right)^2\right], \tag{9}$$

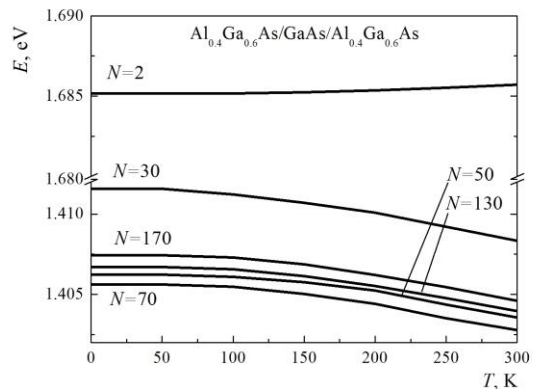where $\Gamma_I$ is the value of unhomogeneous broadening [30, 31]. Thus,

$$\alpha(\omega, T) = \frac{4\sqrt{\pi \ln 2} \cdot D_0^2}{\Gamma_I} \int S(\omega - \omega', T) G(\omega') d\omega' . \qquad (10)$$
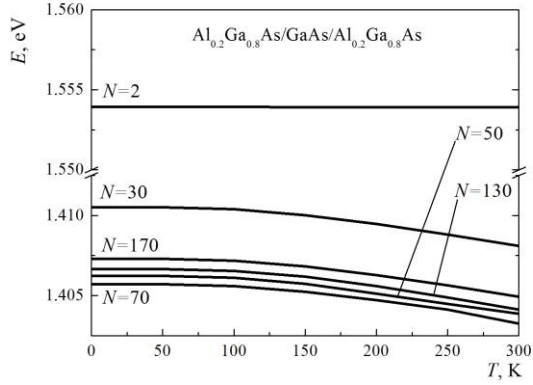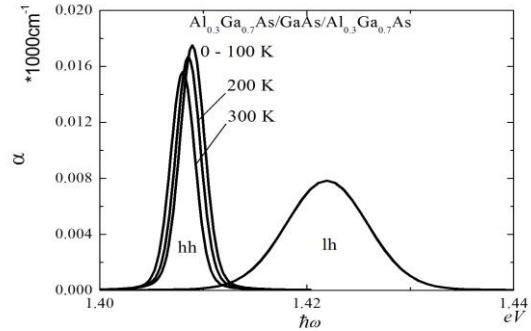


**Fig. 2.** Temperature changes in absorption bands related with transitions to the background state of exciton with heavy (hh) and light holes (lh) in the nanofilm of the thickness 73.5 nm ($N = 130$).

Using the value $\Gamma_0 = 0.33$ meV typical for bulk GaAs samples [32] and the value $D_0$ determined by comparison with experimental data for the absorption coefficient in QWs based on GaAs/Al$_{0.3}$Ga$_{0.7}$As heterojunctions [33], we calculated spectral dependences of the exciton absorption coefficient for various temperatures with account of interaction with confined phonons. In these cases, we considered random divergences in the NF thickness at the level of one GaAs layer to be possible. The results of calculations indicate that temperature changes in the exciton absorption spectrum at $T < 100$ K caused by interaction with optical polarization phonons are negligibly small. The position, height of the peak and half-width of the exciton absorption band are defined only by values of the NF thickness and composition of well material.

When the temperature increases from 100 K up to the room one in NF with the thickness 73.5 nm ($N = 130$), one can observe a weak non-linear shift of the absorption peak (related with transition to the background state of exciton possessing heavy hole) to the long-wave range. The value of this shift reaches 2 to 3 meV, and it is the larger, the higher temperature is (Fig. 2). In this case, the height of the peak is lowered, while the half-width grows from approximately 2 to almost 3 meV in all the studied nanoheterostructures. As it follows from Fig. 1, in NF with a lower thickness the temperature shift value depends both on the thickness and concentration of Al in Al$_x$Ga$_{1-x}$As solid solution.

The position and halfwidth of the band related with transition to the background state of exciton with light hole does not practically change with temperature. Like to the case of exciton with heavy hole, the band width is predominantly defined by the processes of unhomo-geneous broadening and is approximately 9 meV.



*a)*



*b)*



*c)*

**Fig. 1.** Temperature dependences of the heavy-hole-exciton energy in Al$_x$Ga$_{1-x}$As/GaAs/Al$_x$Ga$_{1-x}$As nanofilms of various thicknesses ($N$) and different concentrations of aluminum atoms ($x$): (a) 0.2, (b) 0.3 and (c) 0.4.

## 4. Conclusions

Obtained in this work have been analytical expressions allowing investigation of temperature changes in the

exciton absorption bands observed in flat nanohetero-structures with a single rectangular quantum well of a finite depth. The results of calculations performed for the nanofilms $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ ($x = 0.2, 0.3, 0.4$) have shown that noticeable temperature changes in the band position and shape, which are caused by interaction with confined, semi-confined and interface phonons in these nanosystems, are possible at temperatures above 100 K. Increasing the NF temperature causes a monotonic non-linear shift of the exciton peak to the long-wave range.

In the case of exciton with heavy hole, the temperature shift value reaches 1 to 3 meV, in dependence on the NF thickness and composition of barrier material. The band width is also slightly increased and reaches 2…3 meV. The position and half-width of the absorption band corresponding to exciton with light hole is not considerably changed. The results of our calculations agree with the experimental data described in [19, 33].

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