THE INFLUENCE OF THE NANOHETEROSTRUCTURE GROWTH TEMPERATURE ON THE OPTICAL GAP OF A QUANTUM DOT

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In the framework of the deformation potential model, the influence of the growth temperature of a heterostructure with quantum dots (QDs) and of the QD dimensions on the width of the QD optical gap has been studied. The optical gap has been found to become narrower as the growth temperature of the heterostructure increases.

1. Introduction

Nowadays, significant interest is attracted by heterostructures with (In, Ga)As QDs in a GaAs matrix [1]. It is connected with an opportunity to expand the optical range of emission produced by structures grown on GaAs substrates up to 1.3 μm, and stimulated by modern optoelectronics needs in effective solid-state emitters (lasers) [2].

The process of formation of InAs QDs on singular and vicinal GaAs surfaces by the method of molecular beam epitaxy in the InAs/GaAs system within the temperature interval 350—700 °C is composed of two stages [2, 3]. In the first stage, the growth of an InAs pseudomorphic strained layer occurs. After the critical thickness of 1.5—1.7 ML (monolayer) has been reached, the second stage begins. The latter, according to the Stranski-Krastanow growth model, comprises a spontaneous decay of the pseudomorphic layer into a system of crystal islands (QDs) and a wetting InAs layer about 1 ML in thickness. Up to the thickness of 3—4 ML, the quasi-three-dimensional islands remain coherently strained, i.e. dislocation-free. This process is accompanied by the increase of the surface density and the average size of the ODs. The further growth of InAs results in emerging mesoscopic clusters that contain dislocations.

The strained state of the InAs growing layer is caused by different constants of InAs and GaAs lattices ($\alpha^{\rm InAs} = 6.08$ Å and $\alpha^{\rm GaAs} = 5.65$ Å [4]) as well as by different factors of thermal expansion of contacting materials ($\alpha_T^{\rm InAs} = 4.5 \times 10^{-6}$ K^{-1} and $\alpha_T^{\rm GaAs} = 5.9 \times 10^{-6}$ K^{-1} [5]). The lattice deformation of the growing layer depends on growing conditions, e.g., the growth temperature of the heterostructure with QDs (350—700 °C [6]), the concentration and the chemical nature of dopants [7]. In particular, varying the growth temperature, one can affect the degree of deformation of an epitaxial layer deposited onto a substrate. Ultimately, it will cause a modification of the energy spectrum of current carriers in the QD, i.e. a variation of optical properties of heterostructures with QDs.

This work aims at calculating the energy of the basic optical transition within a coherent-strained QD and analyzing the dependence of this energy (or the relevant frequency) on the growth temperature and the dimensions of a QD.

2. The Model of a Heterostructure with Coherent-strained QDs, which Takes Into Account Thermal Deformations

Consider the system of strained islands (QDs), which has three sources of elastic stress field [8]: a mismatch between the lattice parameters of a substrate and a material, which is epitaxially deposited onto it ($f = (a^{\rm InAs} - a^{\rm GaAs})/a^{\rm InAs} \approx 7\%$), a mismatch between their

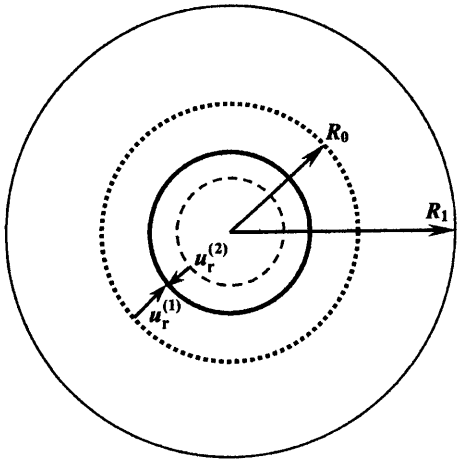


Fig. 1. Scheme of a spherical quantum dot

thermal expansion factors, and a discontinuity of the surface tension tensor at the edges of islands.

We consider QDs that possess no well-defined crystallographic facet, in particular, QDs, the form of which is approximately spherical. For example, such QDs are formed in a InAs/GaAs(001) heterosystem provided the thickness of the InAs growing layer is about 2 ML [2]. Therefore, a contribution of the island edges to the energy of elastic relaxation will be neglected below.

To reduce a problem with plenty of QDs to a problem with a single QD, the following approximation is adopted: the energy of pair-wise elastic interaction between QDs is replaced by the energy of interaction of every QD with a mean field of elastic deformation of all other QDs $\sigma_{\text{eff}}\,(N-1)$.

We represent a QD (see Fig. 1) as a spherical elastic dilatation microinclusion with radius R_0 (the dotted curve) inserted into a spherical cavity in a GaAs matrix (the dashed curve), the volume of the cavity being smaller than that of the microinclusion by ΔV. For such a spherical microinclusion to be inserted into the matrix, the former must be squeezed, while the surrounding GaAs matrix to be stretched, both in radial directions. The result of simultaneous actions of distortions caused by mismatch of lattice constants and thermal factors of contacting nanomaterials is described by the variation of volume ΔV expressed in terms of the parameter $f(R_0, R_1, T)$:

$$\Delta V\left(R_0, R_1, T\right) = f\left(R_0, R_1, T\right) 4\pi R_0^3. \tag{1}$$

Contrary to works [9, 10], we consider the mismatch parameter $f(R_0, R_1, T)$, which stems from both the mismatch of lattice constants a_i, where $i = 1$ for InAs and 2 for GaAs, and different thermal factors of a QD and a surrounding matrix, as a function of the QD dimension R_0, the radius of the cavity R_1 in a surrounding matrix, and the QD growth temperature T:

$$f\left(R_0, R_1, T\right) = f_1\left(R_0, R_1\right) + f_2\left(R_0, R_1\right) + f_T\left(T\right), \tag{2}$$

where $f_1(R_0, R_1)$ and $f_2(R_0, R_1)$ are the relative variations of the lattice parameters of the QD and the surrounding matrix materials, respectively, caused by difference between the radial, $a_r^{(i)}$, and angular, $a_\theta^{(i)}$ and $a_\varphi^{(i)}$, components of the lattice parameters both in the QD and the surrounding matrix materials, with respect to their values in unstrained bulk InAs and GaAs materials;

$$f_i\left(R_0, R_1\right) = \frac{\mathrm{Sp}\varepsilon^{(i)}\left(R_0, R_1\right)}{3},$$

$$\mathrm{Sp}\varepsilon^{(i)}\left(R_0, R_1\right) = \frac{1}{a_i}\left(2a_\theta + a_r^{(i)}\right) - 3, \tag{3}$$

$$a_\theta = a_\varphi = \frac{a_1 G_1 R_0 + a_2 G_2\left(R_1 - R_0\right)}{G_1 R_0 + G_2\left(R_1 - R_0\right)},$$

$$a_r^{(i)} = a_i\left(1 - D_{001}^{(i)} \cdot \left(\frac{a_\theta}{a_i} - 1\right)\right); \quad D_{001}^{(i)} = 2\frac{C_{12}^{(i)}}{C_{11}^{(i)}},$$

$C_{11}^{(i)}$ and $C_{12}^{(i)}$ are the elastic constants of InAs ($i = 1$) and GaAs ($i = 2$) materials. Since $R_1 \gg R_0$, then $f_2 \ll f_1$.

The third summand $f_T(T)$ in expression (2) describes a contribution made by a mismatch between the InAs and GaAs lattice parameters, resulted from different values of the QD, $a_T^{(1)}$, and the matrix, $a_T^{(2)}$, thermal factors (thermal stresses [11]),

$$f_T\left(T\right) = \left(\alpha_T^{(2)} - \alpha_T^{(1)}\right)\left(T_k - T\right), \tag{4}$$

where T is the epitaxial growth temperature ($T = 350 \div 700$ K), and T_k is the temperature, to which the heterosystem was cooled. For a heterosystem planned to operate at room temperature, $T_k = 300$ K, and in a cryoelectron device, $T_k = 77$ or 4.2 K.

Mechanical stresses $\sigma_{rr}^{(1)}$ and $\sigma_{rr}^{(2)}$ in InAs and GaAs materials are determined by the expression [12]

$$\sigma_{rr}^{(i)} = \frac{E_i}{(1+\nu_i)(1-2\nu_i)}\left[(1+\nu_i)\varepsilon_{rr}^{(i)} + \nu_i\left(\varepsilon_{\varphi\varphi}^{(i)} + \varepsilon_{\theta\theta}^{(i)}\right)\right], \tag{5}$$

where ν_i and E_i are Poisson's ratios and Young's moduli, respectively, of the QD and the surrounding matrix

materials, which are expressed in the known way [13] in terms of corresponding elastic constants $C_{11}^{(i)}$ and $C_{12}^{(i)}$.

To define the components of the strain tensor, it is necessary to find explicit forms of atom displacements $u_r^{(1)}$ and $u_r^{(2)}$ in InAs and GaAs materials, respectively. With this purpose in view, we write down the equations of balance [14]

$$\vec{\nabla}\mathrm{div}\vec{u} = 0 \tag{6}$$

with the following boundary conditions for a spherical QD:

$$\begin{cases} 4\pi R_0^2 \left(u_r^{(2)} \mid_{r=R_0} - u_r^{(1)} \mid_{r=R_0} \right) = \Delta V, \\ \sigma_{rr}^{(1)} \mid_{r=R_0} = \sigma_{rr}^{(2)} \mid_{r=R_0} + P_{\mathrm{L}}, \quad P_{\mathrm{L}} = \frac{2\alpha\left(\varepsilon^{(1)}\right)}{R_0}, \\ \sigma_{rr}^{(2)} \mid_{r=R_1} = -\sigma_{\mathrm{eff}}(N-1). \end{cases} \tag{7}$$

Here, R_1 is the radius of the cavity in the GaAs matrix, P_{L} is the Laplace pressure, $\alpha(\varepsilon^{(1)})$ is the QD (InAs) surface energy, which is the function of the QD surface stress, $\sigma_{ij}^{(1)}$, and deformation, $\varepsilon_{ij}^{(1)}$, tensors [15]:

$$\alpha\left(\varepsilon^{(1)}\right) = \alpha(0) + \sum_{i,j} \sigma_{ij}^{(1)} \varepsilon_{ij}^{(1)} +$$

$$+ \frac{1}{2} \sum_{i,j,k,l} \varepsilon_{ij}^{(1)}(1) \cdot s_{ijkl}^{(1)} \cdot \varepsilon_{kl}^{(1)} + ...,$$

and $s_{ijkl}^{(1)}$ is the stress tensor of the second order. The left hand side of the first equation of system (7) is equal to a geometrical difference ΔV of the microinclusion and the cavity volumes in the GaAs matrix, depicted in Fig. 1.

In case of spherical QDs, the solution of Eq. (6) looks like

$$u_r^{(1)} = C_1 r + \frac{C_2}{r^2}, \quad 0 \le r \le R_0; \tag{8}$$

$$u_r^{(2)} = C_3 r + \frac{C_4}{r^2}, \quad R_0 \le r \le R_1. \tag{9}$$

Since the displacement at the point $r = 0$ has to be finite, C_2 in solution (8) should be assigned zero.

The displacement field defines the following components of the strain tensor:

$$\varepsilon_{rr}^{(1)} = C_1, \tag{10}$$

$$\varepsilon_{\varphi\varphi}^{(1)} = \varepsilon_{\theta\theta}^{(1)} = C_1, \tag{11}$$

$$\varepsilon_{rr}^{(2)} = C_3 - \frac{2C_4}{r^3}, \tag{12}$$

$$\varepsilon_{\varphi\varphi}^{(2)} = \varepsilon_{\theta\theta}^{(2)} = C_3 + \frac{C_4}{r^3}. \tag{13}$$

Coefficients C_1, C_3, and C_4 are determined by solving system (7) taking into account Eqs. (2)—(5) and (8)—(13).

3. Calculation of the Energy of the Basic Optical Transition

Knowing the components of the strain tensor, let us find the potential energy of electrons and holes in a stressed heterostructure with QDs. The energy is reckoned from the bottom of the corresponding band in a stressed nanocrystal InAs:

$$U_e(r) = \begin{cases} 0, \quad 0 \le r \le R_0 \\ \Delta V_c(0) - a_c^{(1)} \varepsilon^{(1)}(R_0, R_1, T) + \\ + a_c^{(2)} \varepsilon^{(2)}(R_0, R_1, T), \quad R_0 \le r \le R_1, \end{cases} \tag{14}$$

$$U_h(r) = \begin{cases} 0, \quad 0 \le r \le R_0, \\ \Delta V_h(0) + a_v^{(1)} \varepsilon^{(1)}(R_0, R_1, T) - \\ - a_v^{(2)} \varepsilon^{(2)}(R_0, R_1, T), \quad R_0 \le r \le R_1. \end{cases} \tag{15}$$

Here, $\Delta V_c(0)$ and $\Delta V_h(0)$ are the depths of the potential well for an electron and a hole, respectively, in a QD in a non-deformed heterostructure, $\varepsilon^{(i)}(R_0, R_1, T) = \mathrm{Sp}\ \varepsilon^{(i)}$ is the trace of the strain tensor; and $a_c^{(i)}$ and $a_v^{(i)}$ are the constants of the hydrostatic deformation potential of the conduction and valence bands, respectively.

The energy of transition into the ground state in a stressed QD is defined as follows:

$$E(\varepsilon, T) = E_{00}^{(e)} + E_{00}^{(h)} + E_g^{(1)}, \tag{16}$$

where $E_{00}^{(e,h)}$ is the ground state energy of an electron or a hole in a stressed QD,

$$E_g^{(1)} = E_g^{(1)}(0) + \varepsilon^{(1)}(R_0, R_1, T)\left(a_c^{(1)} - a_v^{(1)}\right), \tag{17}$$

is the width of the energy gap in stressed QD material, and $E_g^{(1)}(0)$ is the width of the energy gap in QD material provided no deformation effects.

The calculation of the energy spectra of an electron and a hole will be carried out in the effective mass approximation. So, the physical condition that the geometrical dimensions of a QD and a space between two neighbor QDs should considerably exceed the sizes of elementary cells in QD and matrix crystals [16], i.e. $R_0 \gg a_1, a_2$, have to be satisfied.

We are to solve the Schrödinger equation

$$H_{e,h}\Psi_{e,h}(\vec{r}) = E^{(e,h)}\Psi_{e,h}(\vec{r}) \tag{18}$$

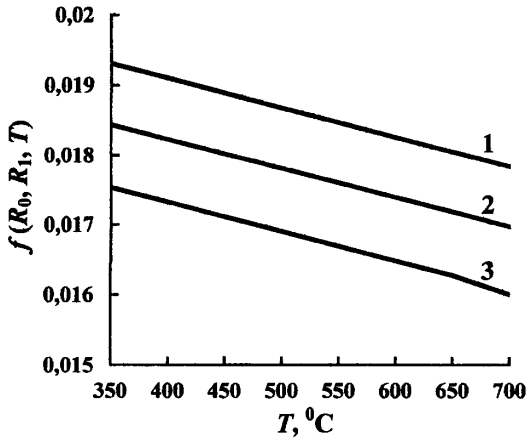


Fig. 2. Dependences of the mismatch parameter $f(R_0, R_1, T)$ on the InAs QD growth temperature for various QD radii R_0 = 30 (*1*), 60 (*2*), and 90 Å(*3*)

with the Hamiltonian

$$H_{e,h} = -\frac{\hbar^2}{2}\vec{\nabla}\frac{1}{m^*_{e,h}(\vec{r})}\vec{\nabla} + U_{e,h}(r, R_0). \tag{19}$$

The electron and hole effective masses, $m^*_{1,2_e}$ and $m^*_{1,2_h}$, respectively, are supposed to be known both in the QD and in the matrix; namely, they are taken to be equal to corresponding values in the relevant bulk crystals.

The solution of the Schrödinger equation (18) in the spherical coordinate system looks like

$$\Psi_{nlm}(r, \Theta, \varphi) = R_{nl}(r) \cdot Y_{lm}(\theta, \varphi). \tag{20}$$

Here. $Y_{lm}(\theta, \varphi)$ are Legendre spherical functions [17]. The radial functions $R_{nl}(r)$ are expressed in terms of Bessel spherical functions [17]:

$$R_{1_{nl}}(r) = Aj_l(k_{e,h}r) + Bn_l(k_{e,h}r), 0 \le r \le R_0, \tag{21}$$

$$R_{2_{nl}}(r) = Ch^{(1)}_l(i\chi_{e,h}r) + Dh^{(2)}_l(i\chi_{e,h}r),$$

$$R_0 \le r \le R_1, \tag{22}$$

where

$$k^2_{e,h} = \frac{2m^*_{1_{e,h}}}{\hbar^2}E^{(e,h)}_{nl},$$

$$\chi^2_{e,h} = \frac{2m^*_{2_{e,h}}}{\hbar^2}\left(U^{(2)}_{e,h}(r) - E^{(e,h)}_{nl}\right), \tag{23}$$

and the potential energies $U_{e,h}(r)$ of electron and hole are determined according to formulae (14) and (15), respectively.

The continuity conditions for the wave functions and the density of the probability stream at the QD–matrix interface

$$\begin{cases} R_{1_{nl}}(r)_{|r=R_0} = R_{2_{nl}}(r)_{|r=R_0}, \\ \frac{1}{m^*_{1_{e,h}}}\frac{dR_{1_{nl}}(r)}{dr}\Big|_{r=R_0} = \frac{1}{m^*_{2_{e,h}}}\frac{dR_{2_{nl}}(r)}{dr}\Big|_{r=R_0}, \end{cases} \tag{24}$$

together with the condition of function regularity for $R_{nl}(r)$ at $r \to 0$ and $r \to R_1$, as well as normalization consideration, determine the spectrum E_{nl} and the wave functions of electron and hole in the heterosystem InAs/GaAs with InAs QDs.

Therefore, the ground state energies $E^{(e,h)}_{00}$ of electron and hole in a QD are determined from the following transcendental equation:

$$\frac{m^*_{2e,h}}{m^*_{1e,h}}[1 - k_{e,h}R_0 \operatorname{ctg}(k_{e,h}R_0)] =$$

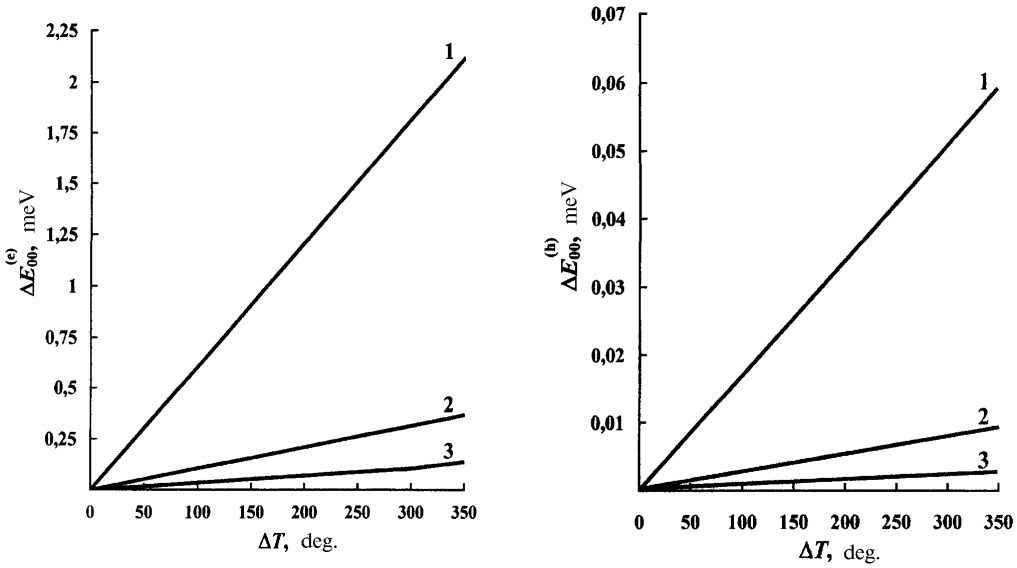
$$= \frac{1 + \chi_{e,h}R_0 + e^{2\chi_{e,h}(R_0 - R_1)}(\chi_{e,h}R_0 - 1)}{1 - e^{2\chi_{e,h}(R_0 - R_1)}}. \tag{25}$$

4. Numerical Calculations and Discussion of Results

The numerical calculations of the energy of the basic optical transition, considered as a function of QD dimensions and its growth temperatures, were carried out for a InAs/GaAs nanoheterosystem with InAs QDs, the parameters of which are quoted in the table.

Fig. 2 demonstrates that the mismatch parameter $f(R_0, R_1, T)$ decreases linearly as the temperature of the QD growth increases. The increase of the growth temperature shifts the thermal component of the mismatch parameter $f_T(T)$ towards larger by modulus negative values (see formula (4)). In so doing, such a temperature rise brings about stress relaxation at the QD–matrix interface. Fig. 2 testifies that an even greater effect of the diminishing of the mismatch parameter is observed if the QD radius R_0 increases. This circumstance substantially reduces stresses, which are caused by a mismatch between lattice parameters of contacting materials and described by the component $f_1(R_0, R_1)$, because the QD material becomes more massive.

The energy $E(\varepsilon, T)$ of the basic optical transition (see formula (16)) is determined by a sum of three energy components: the energies of the ground state of an electron $E^{(e)}_{00}$ and a hole $E^{(h)}_{00}$, and the width $E^{(1)}_g$ of the energy gap of QD material. So, the character of the dependence of the transition energy $E(\varepsilon, T)$ on the QD



a b

Fig. 3. Temperature dependences of the energy shift of an electron (a) and hole (b) levels in a quantum dot with various radii R_0. Notations are the same as in Fig. 2

growth temperature T is governed by the T-behavior of its components $E_{00}^{(e,h)}(\varepsilon, T)$ and $E_g^{(1)}(\varepsilon, T)$. The increase of the growth temperature stimulates the raise of electron and hole energy levels in the QD and a reduction of its energy gap width (Figs. 3 and 4). Figs. 3 and 4 allow the contributions of the components $E_{00}^{(e,h)}(\varepsilon, T)$ and $E_g^{(1)}(\varepsilon, T)$ to the formation of the QD optical gap to be estimated. Fig. 3 provides a quantitative estimation for the increase of the electron, $\Delta E_{00}^{(e)} = E_{00}^{(e)}(T) - E_{00}^{(e)}(T_0)$ (Fig. 3,a), and the hole, $\Delta E_{00}^{(h)} = E_{00}^{(h)}(T) - E_{00}^{(h)}(T_0)$ (Fig. 3,b), ground states in a QD with radius R_0, if the growth temperature rises from $T_0 = 350°\mathrm{C}$ to T ($\Delta T = T - T_0$). Fig. 4 demonstrates the magnitude $\Delta E_g = E_g^{(1)}(T) - E_g^{(1)}(T_0)$ (see formula (17)) of the reduction of the energy gap width in the material of a QD with radius R_0 for the same variations of the growth temperature ΔT. Having evaluated, as the tangents of the slope angles of corresponding straight lines in Figs. 3 and 4, the rates of variation of components to the energy of transition into the ground state, we can draw a conclusion that the rate, at which the energy gap diminishes, substantially exceeds the rates, at which the electron and the hole ground state energies in the QD raise. In particular, an increase of the growth temperature for a QD with radius $R_0 = 30$ Å from $T_0 = 350°\mathrm{C}$ to $T = 400°\mathrm{C}$ increases the total energy of the electron and the hole ground states by 0.31 meV and reduces the energy gap width of QD material by 1.64 meV. It means that the rate, at which the electron and the hole ground state energies increase, amounts to 0.006 meV/K, whereas the rate of the energy gap reduction constitutes 0.033 meV/K and is 5.5 times as large as the growth rate of two other contributors to the optical gap width. Therefore, it is the variations of the energy gap width in QD material that determine the character of the dependence of the transition energy into the ground state on the variation of the QD growth temperature.

Fig. 5 shows how QD dimensions and temperature conditions, under which the QD grows, affect the energy of the basic optical transition $E(\varepsilon, T)$. From this figure, it is seen that the increase of the QD growth temperature stimulates a practically uniform narrowing of its optical gap $E(\varepsilon, T)$. Variation of the QD size does not change the character of the $E(\varepsilon, T)$ decline with growing T. An increase of the QD radius R_0 monotonously shifts the energy of the basic optical transition towards lower energies, with the energy shift being larger for smaller QDs (20—40 Å).

Parameters of InAs and GaAs crystals [4, 5, 15]

	a, Å	C_{11}, Mbar	C_{12}, Mbar	a_c, eV	a_ν, eV	α_T, K^{-1}	E_g, eV	$\frac{m_e^*}{m_0}$	$\frac{m_h^*}{m_0}$	$\alpha(0)$, N/m
InAs	6.08	0.833	0.453	−5.08	1	4.5×10^{-6}	0.36	0.057	0.41	0.657
GaAs	5.65	1.23	0.571	−7.17	1.16	5.9×10^{-6}	1.452	0.065	0.45	

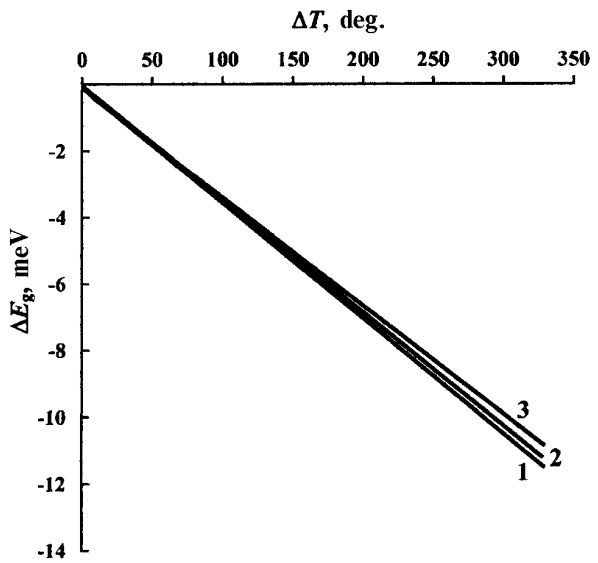


Fig. 4. Temperature dependences of the energy gap variation in material of a stressed QD for various QD radii. Notations are the same as in Fig. 2

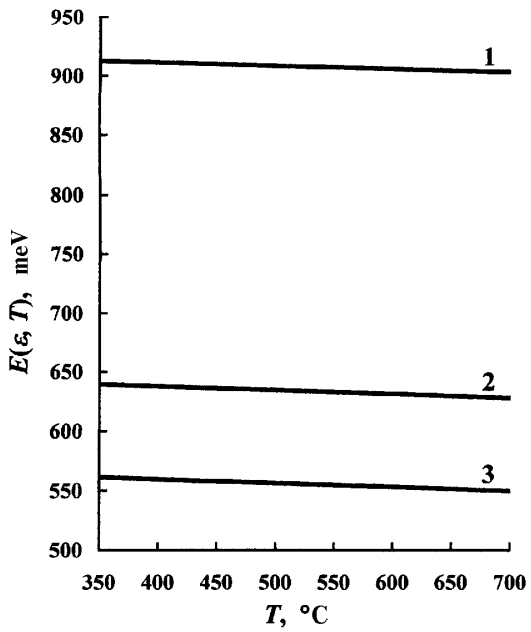


Fig. 5. Dependences of the energy of transition into the ground state on the growth temperature of a heterostructure with QDs for various QD radii. Notations are the same as in Fig. 2

On the basis of the obtained temperature dependence of the energy of basic optical transition $E(\varepsilon, T)$, one can indirectly estimate the growth temperature of a heterostructure with QDs. In particular, knowing from experimental data the spectral position $\lambda_{\max}$ of the maximum of the photoluminescence curve, we can estimate the growth temperature making use of the equality $hc/\lambda_{\max} = E(\varepsilon, T)$.

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ВПЛИВ ТЕМПЕРАТУРИ РОСТУ НАНОГЕТЕРОСИСТЕМИ НА ШИРИНУ ОПТИЧНОЇ ЩІЛИНИ КВАНТОВОЇ ТОЧКИ

Р.М. Пелещак, О.О. Даньків

Р е з ю м е

В рамках моделі деформаційного потенціалу досліджено вплив температури росту гетероструктури з квантовими точками (КТ) та розмірів КТ на енергетичну ширину оптичної щілини КТ. Встановлено, що із збільшенням і температури росту гетероструктури, і розмірів КТ оптична щілина звужується.