Optically active energy states of the exciton in quantum wells of various widths

P A Belov
Spin Optics Laboratory, St. Petersburg State University, Ulyanovskaya 1, 198504 St. Petersburg, Russia
E-mail: pavelbelov@gmail.com

Abstract. The optically active energy states of the exciton in GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum wells of various widths are calculated by the direct solution of the three-dimensional Schrödinger equation. A dependence of energies on the quantum well width as a parameter is obtained for widths up to 200 nm. The results are compared with the model of quantization of the exciton as a whole in wide quantum wells. The radiative decay rates for several lowest exciton states are also found.

1. Introduction
The quantum states of excitons in GaAs-based heterostructures, their energies and radiative properties have been a focus of intensive studies for several decades [1-9]. The numerical study of excitons in these heterostructures has mainly been based on solving the Schrödinger equation by the variational method, which allowed one to describe the exciton ground state [2,10-13]. Only very recently has the focus of study been extended to the excited exciton states [14-18]. This is largely motivated by the new experimental data obtained for high-quality heterostructures [19-22]. Reflectance spectra of these structures clearly demonstrate peculiarities from the bound and quasibound electron-hole states even for relatively wide quantum wells ($L \geq 100$ nm) [23, 24].

In this report, we present the results of numerical modeling of the optically active electron-hole bound states in GaAs-based quantum wells (QWs) of various widths. The obtained numerical results expand the findings presented in reference [17] by the data for wide QWs. We simulate heterostructures with GaAs/Al$_{0.3}$Ga$_{0.7}$As single QWs of widths $L = 5 - 200$ nm. The modeling is performed by the developed finite-difference method of numerical solution of the Schrödinger equation for the exciton in a QW [12, 16, 17, 25]. Our method is superior to the standard variational approach [2, 6] because it makes it possible to calculate not only the ground state but also the excited states of excitons [26]. It also allows one to obtain accurate exciton energies for a wide range of QW widths and potential profiles [27]. Here, we calculate the energy levels of the 1s-like states of the heavy-hole exciton in a single QW and observe a dependence of the obtained energies on a QW width as a parameter. The calculated energy levels correspond to different quantum-confined hole-states: from the ground state up to the 13th one. For wide QWs, the calculated energies are also compared to the energies obtained within the model of quantization of the exciton as a whole [6]. By our numerical method, we also obtain the wave functions of the quantum states. As a result, we compute the radiative decay rates [28] of several
lowest optically active exciton states. We observe a significant increase in the radiative decay rate of the first excited quantum-confined exciton state in wide QWs.

The calculated energy levels are important for optical studies of the semiconductor heterostructures [29], namely, for identifying resonances in the measured reflectance spectra [8, 22, 24, 30]. The obtained radiative decay rates may also be essential for theoretical studies of the exciton-polaritons in wide QWs [31, 32].

2. Theory

For calculation of the energy levels, we consider the time-independent three-dimensional Schrödinger equation for the exciton in a QW. This equation is derived assuming the parabolic dispersion of the conduction and valence bands. The latter one is defined by the diagonal terms of the Luttinger Hamiltonian [33, 34]. It means that we disregard the heavy-hole–light-hole dispersion of the conduction and valence bands. The latter one is defined by the diagonal terms of the Luttinger Hamiltonian [33, 34]. It means that we disregard the heavy-hole–light-hole dispersion of the conduction and valence bands. The latter one is defined by the diagonal terms of the Luttinger Hamiltonian [33, 34]. It means that we disregard the heavy-hole–light-hole dispersion of the conduction and valence bands. The latter one is defined by the diagonal terms of the Luttinger Hamiltonian [33, 34]. It means that we disregard the heavy-hole–light-hole dispersion of the conduction and valence bands. 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Figure 1. The theoretical scheme of energy levels of the exciton in a QW. There are bound states below the energy level $E_{e1} + E_{h1}$ and the quasibound ones above.

where $m'$ is a value of the mass parameter at the grid point $z'$. A simplified form of formula (2) is used for the second partial derivative over $\rho$. The finite-difference formula for the first partial derivative over $\rho$ can be easily found in reference [39]. The grid steps over each variable have been taken to be the same, $\Delta = \Delta_z = \Delta_h = \Delta_r$. Equation (2) defines the theoretical uncertainty of the numerical solution of order of $\Delta^2$ as $\Delta \to 0$. A discretization of equation (1) leads to a large, block-tridiagonal matrix with sparse blocks [42]. A small part of the matrix spectrum is obtained by the Arnoldi algorithm [43]. As a result, we calculate several lowest eigenvalues of the matrix and the corresponding eigenvectors. After extrapolation to the limit $\Delta = 0$ [12], the accurate results are obtained.

Having the wave function $\psi(z_e, z_h, \rho)$ calculated, we can obtain the radiative decay rate by the formula [6]:

$$\Gamma_0 = \frac{2\pi q}{\hbar c} \left( \frac{|p_{cv}|}{m_0 \omega_0} \right)^2 \left| \int_{-\infty}^{\infty} \psi(z_e = z_h = z, \rho = 0) \exp(iqz)dz \right|^2,$$

(3)

where $q = \sqrt{\epsilon \omega/c}$ is the light wave vector, $\omega_0$ is the exciton frequency, $|p_{cv}|$ is the matrix element of the momentum operator between the single-electron conduction- and valence-band states. Equation (3) does not take into account the coupling of the exciton states due to the light-wave for wide QWs. Such a coupling is taken into account in a more advanced model described in reference [44].

The calculations were performed for a heterostructure with the single GaAs/Al$_{0.3}$Ga$_{0.7}$As QWs of various $L$, up to 200 nm. The difference of the gap energies, as a function of $x$, is modeled by the formula $\Delta E_g = 1087x + 438x^2$ meV. The ratio of potential barriers is taken to be $V_e/V_h = 65/35$ [12, 45]. The Luttinger parameters used in the calculations are $\gamma_1 = 6.85$, $\gamma_2 = 2.10$; the dielectric constant is $\epsilon = 12.53$.

3. Results of calculations
3.1. Energy levels
We calculated the 1s-like energy levels of the heavy-hole exciton in the single GaAs/Al$_{0.3}$Ga$_{0.7}$As QW for different widths $L = 5 - 200$ nm. A dependence of the calculated energy levels on the
Figure 2. 1s-like energy levels of the heavy-hole exciton in GaAs/Al_{0.3}Ga_{0.7}As QW as a function of the QW width. The energy levels are shown with respect to the lower boundary of the continuous spectrum that is defined by a sum of the lowest quantum-confinement energies \( E_{e1} + E_{h1} \). This boundary is denoted by the zero binding energy.

QW width is shown in figure 2. The energy levels of the exciton states are shown with respect to the lower boundary of the continuous spectrum which is marked as the zero baseline. As it is described in reference [17], this boundary is defined by a sum of the lowest quantum-confinement electron and hole energies \( E_{e1} + E_{h1} \) which can be easily calculated for a QW of arbitrary \( L \). The energy levels were also classified based on the types of their in-plane relative and quantum-confinement motions. The levels are labeled as \((e1, h_j, 1s)\), where index \( j \) is the number of hole quantum-confined state. The indices in \( e1 \) and \( 1s \) correspond to the first electron quantum-confined state and the first principal quantum number, \( N = 1 \), of the s-like 2D Coulomb state \(-1/\rho\), respectively. Such a classification is precise for narrow QWs. For wider QWs, it is an approximate one. It represents the dominant pure state \(|e1, h_j, 1s\rangle\) of the exciton in a very narrow QW, inherent in the calculated state for a given \( L \). These pure states are defined by equation (1) with the 2D Coulomb potential instead of the 3D Coulomb one, since for \( L \rightarrow 0 \) the 3D Coulomb potential degenerates to the 2D one. The wave functions of the pure states are factorized into a product of three one-dimensional functions \( \psi_{e1}(z_e)\psi_{hj}(z_h)\varphi_1(\rho) \), where \( \psi_{e1}(z_e) \), \( \psi_{hj}(z_h) \) are quantum-confinement carrier’s ones and \( \varphi_1(\rho) \) is the 1s-like 2D Coulomb ground state [49].

In figure 2, the lowest dark blue curve corresponds to the energy of the exciton ground state. In particular, this curve demonstrates the crossover from the model of the exciton in a narrow quantum well to the quantization of the exciton as a whole in a wide quantum well. The binding energy is varied from about 16 meV (if the barriers are rigid) down to about 4 meV, respectively.

As the QW width increases, more excited \( e-h \) bound states appear in the spectrum. They correspond to the upper quantum-confinement hole states \( \psi_{hj}(z_h) \) where \( j > 1 \). The energies in the continuum part of the spectrum, gradually decreasing with a growth of \( L \), correspond to the resonant states [46, 47, 38, 48]. They, then, descent below the continuum and are ranged in the
discrete part of the spectrum by an order of the quantum-confined hole states \( h_j, j = 2, 3, \ldots \). For wide QWs \( (L \sim 150 \, \text{nm}) \), there are many closely adjacent energy levels. For example, for \( L = 200 \, \text{nm} \), the energy difference between the ground and first excited energy level is of about 0.1 meV.

For wide QWs, the calculated energies are compared with the data from the model of quantization of the exciton as a whole. This model is based on the Coulomb interaction of the electron and the hole in bulk GaAs and additionally complicated by the quantization of the exciton center-of-mass motion along the growth direction. It leads to the model wave function [6]

\[
\psi_{k,1}(z_e, z_h, \rho) = \psi_k(Z)\phi_1(r),
\]

where

\[
\psi_k(Z) = \sqrt{\frac{2}{L}} \begin{cases} 
\cos(\pi k Z/L) & \text{if } k = 1, 3, 5, \ldots \\
\sin(\pi k Z/L) & \text{if } k = 2, 4, 6, \ldots 
\end{cases}
\]

describes the quantization across the QW plane and \( \phi_1(r) \) is the radial 1s Coulomb function [38]. The variables in equation (4) are \( Z = (m_e z_e + m_h z_h)/(m_e + m_h) \), \( z = z_e - z_h \), and \( r = \sqrt{\rho^2 + z^2} \) is the relative \( e-h \) distance. The comparison of energies is shown in figure 3. The upper panel shows the calculated exciton energies confronted with the data from the model of quantization of the center-of-mass motion over the \( Z \) direction. The quantization energies are defined by the model in a standard way:

\[
E_k = \frac{\hbar^2}{2(m_e + m_h)} \left( \frac{\pi k}{L} \right)^2.
\]

It is seen in figure 3 (a) that the qualitative behaviour of energy levels is reasonably well described by the model. However, the calculated energy levels are higher than the energies from the model. It means that, in addition to the quantization over the \( Z \) direction, the wave function

![Figure 3](image-url)
Figure 4. The radiative decay rates in energy units, $\hbar \Gamma_0$, for the 1s-like quantum states of the heavy-hole exciton in the GaAs/Al$_{0.3}$Ga$_{0.7}$As QW as a function of the QW width.

of the exciton is squeezed by the QW potentials $V_{e,h}(z_{e,h})$, see reference [17] for details. Such a squeezing is phenomenologically simulated by the so-called dead layer [50]. Therefore, in order to achieve better quantitative agreement of the energies, we employ a dead layer $L_d = 10$ nm over the quantization coordinate. Redefining $L$ as $L^* = L - 2L_d$, we obtain the energies by equation (5). The comparison with the model using the dead layer is shown in the lower panel. A good agreement of the results is apparent.

The upper Coulomb states (2s, 3s-like) are not shown in figure 2 because they are optically less active [6]. It is worth noting that the shown energy levels have several crossing and, especially, anticrossings with such optically inactive states [17]. Therefore, the shown energy levels are unsmooth in the figure, in particular in the energy range of the continuum.

3.2. Radiative decay rates
Using the obtained wavefunctions, we calculated the radiative decay rates of the four lowest 1s-like states by equation (3). The radiative decay rates in energy units, $\hbar \Gamma_0$, for various QW widths are shown in figure 4. The dark blue curve corresponds to the exciton ground state $(e1, h1, 1s)$. For very narrow QW, $L < 5$ nm, $\hbar \Gamma_0$ for this state is about 48 $\mu$eV. As the QW width increases, $\hbar \Gamma_0$ firstly decreases down to 36 $\mu$eV for $L = 30$ nm and then grows up to about 60 $\mu$eV for $L = 140$ nm. For wider QWs, $\hbar \Gamma_0$ decreases. In contrast, the radiative decay rate of the first excited state, $(e1, h2, 1s)$, is almost zero for narrow QWs, but for wider ones it increases significantly. For $L = 200$ nm, the first excited state has $\hbar \Gamma_0 \sim 110$ $\mu$eV and it seems to be increasing for larger $L$.

In order to explain this significant increase in the radiative decay rate of the first excited state, we compared the slices of the wave functions $\psi(z_{e}, z_{h}, \rho)$ of the ground state $(e1, h1, 1s)$ and the first excited state $(e1, h2, 1s)$ as $\rho = 0$ nm for three different QW widths: $L = 50, 100, 200$ nm. These slices are shown in figure 5. One can see that, as $L$ increases, the wave functions become localized close to the line $z = z_{e} - z_{h} = 0$. This indicates the quantization of the exciton.
Figure 5. The slices of the wave functions of the ground state \((e_1, h_1, 1s)\) and the first excited state \((e_1, h_2, 1s)\) as \(\rho = 0\) nm for three different QW widths: (a), (b) \(L = 50\) nm; (c), (d) \(L = 100\) nm; (e), (f) \(L = 200\) nm. The line \(z_e = z_h\) is additionally indicated in plots (c), (d).

as a whole in wide QWs [6]. The ground state has the even wave function, whereas the first excited one has the odd wave function: it exhibits a clearly visible maximum and minimum. The minimums and maximums of the \((e_1, h_2, 1s)\) are both located along the line \(z_e = z_h\) for all shown QWs. For QWs of \(L \sim 200\) nm, the characteristic wavelength of the exciton first excited state is close to the wavelength \(\lambda \sim 230\) nm of the light wave at the exciton resonance frequency. In this case, the product \(\psi(z_e = z_h = z, \rho = 0) \sin qz\), where \(\psi\) is the first excited state wave function, becomes the dominant contribution to the integral in equation (3), because it is a product of two functions of the same parity and the similar supports. Instead, in contrast to narrow QWs, the product \(\psi(z_e = z_h = z, \rho = 0) \cos qz\) includes two functions of a similar scale, but of different parity. Therefore, the value of this product is negligibly small. As a result, the radiative decay rate of the state \((e_1, h_2, 1s)\) dominates due to such a resonance-like condition. We can also expect that the maximum \(\hbar \Gamma_0\) is achieved for a QW width of about \(L = 230\) nm. Our preliminary calculations, however, show that the state \((e_1, h_2, 1s)\) has the maximal radiative decay rate of about 140 \(\mu\)eV for \(L = 250\) nm.
4. Conclusion
In summary, we calculated several optically active (1s-like) states of the exciton in the GaAs/Al$_{0.3}$Ga$_{0.7}$As single quantum well for various widths by the direct numerical solution of the three-dimensional Schrödinger equation. A dependence of energies on the quantum well width as a parameter was observed for widths up to 200 nm. The results were compared with the model of the quantization of the exciton as a whole in wide quantum wells. The radiative decay rates for several lowest exciton states were also found.

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