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The binding energy of excitons in narrow quantum wells

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Abstract. The effects of discontinuities of the material parameters at the interfaces of narrow GaAs-based quantum wells as well as of the nonparabolicity of the conduction band on the exciton binding energy are studied. These effects are taken into account in the three-dimensional Schrödinger equation for the exciton. Accurate exciton ground state energies are obtained using the improved numerical scheme of Khramtsov *et al.* (2016 *J. Appl. Phys.* **119** 184301). The enhancement of the exciton binding energy for narrow quantum wells is observed and the contribution of each effect is estimated.

1. Introduction

Exciton states in quantum wells (QWs) have been experimentally and theoretically studied for several decades. The theoretical studies [1–12] were generally ahead of experimental ones due to the well-developed quantum theory and an obvious lag between a technological design and the practical realization of experimental techniques [13–19] for growth of high-quality heterostructures. The defects and roughness of interfaces of heterostructures, the segregation effect [20, 21] especially for narrow QWs, impair the accuracy of measurements of optical transitions between exciton states [22]. The high-quality heterostructures have only become available only recently and provide a good possibility for the experimental studies [23–26, 12, 27, 28]. Therefore, the precise theoretical results on the exciton states can significantly advance the ongoing experiments on heterostructures.

The binding energy of excitons in QWs is usually obtained from the solution of the two-body Schrödinger equation (SE) which is, in turn, complicated by impossibility to separate the center-of-mass motion. Furthermore, the degenerate valence band, in the simplest case, requires one to consider the diagonal terms of the Luttinger Hamiltonian [30] for the hole. These complications lead to the three-dimensional SE which can be solved numerically by the variational approach [4, 29, 5, 8, 10, 31], shooting [11] or perturbative methods [32, 33]. Recently, we proposed a new method [12] for direct numerical solution of this equation. The method is based on the finite-difference discretization and, since it is asymptotically exact, allows one to obtain accurate exciton energies for a wide range of QW widths and potential profiles [28]. Our method is superior to the variational approach because, in particular, it does not rely on a prescribed trial solution and, furthermore, makes it possible to calculate the excited exciton states.

In the present report, we extend the proposed numerical method [12] to the case when the discontinuities of the mass and dielectric constants are properly taken into account in the SE. These discontinuities considerably enhance the exciton binding energy for the narrow QWs. Moreover, we studied the effect of the conduction band nonparabolicity which results in further increase of the binding energy, but is rarely considered in theoretical calculations. We simulate the GaAs/Al_xGa_{1-x}As heterostructures with a single rectangular QW and various Al concentrations, x, in order to provide a reliable reference for future experimental studies.

2. Theoretical model

We generally describe an exciton in a rectangular QW by the SE assuming the parabolic conduction and valence bands. The latter one is defined only by the diagonal terms of the Luttinger Hamiltonian, because for narrow QWs the nondiagonal terms contribute little to the energy states [34]. In other words, the heavy-hole–light-hole coupling is negligible. The derivation of the SE without material parameter mismatch is given in Ref. [12]. Here, we briefly outline the main points of the equation for the s-wave exciton states arising from the material discontinuities at the interfaces. This equation is given as [5]

$$\left(K+V_e^{pot}(z_e)+V_h^{pot}(z_h)+V_e^{self}(z_e)+V_h^{self}(z_h)+V^C(\rho,z_e,z_h)\right)\chi(z_e,z_h,\rho)=E_x\chi(z_e,z_h,\rho), \quad (1)$$

where the kinetic term K reads

$$K=-\frac{\hbar^2}{2}\frac{\partial}{\partial z_e}\frac{1}{m_{ez}(z_e)}\frac{\partial}{\partial z_e}-\frac{\hbar^2}{2}\frac{\partial}{\partial z_h}\frac{1}{m_{hz}(z_h)}\frac{\partial}{\partial z_h}-\frac{\hbar^2}{2}\frac{1}{\mu_{xy}}\left(\frac{\partial^2}{\partial\rho^2}-\frac{1}{\rho}\frac{\partial}{\partial\rho}+\frac{1}{\rho^2}\right).$$

In Eq. (1), indices e and h denote the electron and the hole, respectively. The potentials $V_{e,h}^{pot}(z_{e,h})$ are the finite rectangular confinement QW potentials. The self-interaction potentials $V_{e,h}^{self}(z_{e,h})$ come from the interaction of carriers with their images appeared in the materials with different dielectric constants [35, 36]. The Coulomb potential $V^C(\rho,z_e,z_h)$ also takes into account the dielectric constant mismatch. This mismatch gives rise to the electrostatic perturbation expansions over a small parameter $(\epsilon_w-\epsilon_b)/(\epsilon_w+\epsilon_b)$, where $\epsilon_{w,b}$ are the dielectric constants in the QW and in the barrier. These expansions, special corrections to avoid divergences at the interfaces, and results of variational calculations can be found in Refs. [29, 5, 10, 37]. In Eq. (1), the term $\mu_{xy}=m_{exy}m_{hxy}/\left[m_{exy}+m_{hxy}\right]$ is the reduced effective mass in the (x,y)-plane. We assume that the center-of-mass motion in the (x,y)-plane is separated. Other mass terms depend on the coordinate z of the confinement potential. In the case of nonparabolicity of the conduction band, the electron effective mass becomes dependent also on the confinement energy [38–41]. We use the linear nonparabolicity approximation: the electron mass linearly increases with the confinement energy.

The three-dimensional equation (1) cannot be solved analytically even for narrow QWs because of finite confinement potentials and complicated conditions at the QW interfaces. In our study, Eq. (1) is solved numerically and the binding energies for excitons in QWs of various widths and compositions of the QW layer and barriers are found.

3. Numerical method

Following Ref. [12], we numerically solved the boundary value problem for Eq. (1) and accurately obtained the exciton ground state energy, E_x. The exciton binding energy, E_b, is defined with respect to the quantum confinement energy of the electron, E_e, and the hole, E_h, in QW by the formula

$$E_b=E_e(\Delta m,\Delta\varepsilon)+E_h(\Delta m,\Delta\varepsilon)-E_x(\Delta m,\Delta\varepsilon).$$

Energies $E_e(\Delta m, \Delta\varepsilon)$ and $E_h(\Delta m, \Delta\varepsilon)$ are obtained from the solution of the corresponding one-dimensional SEs for the electron and the hole in QWs. These one-dimensional problems take into account the mass discontinuities, self-interaction potentials $V_{e,h}^{self}(z_{e,h})$ as well as the nonparabolicity of the conduction band for E_e. In the perturbation series of the electrostatic contributions to the potentials we kept the terms up to ten order of the expansion parameter.

The exponential decrease of the exciton wave function at large values of variables allows us to impose zero boundary conditions for the function χ at the boundary of some rectangular domain. For the discretization, we employ the second-order finite-difference (FD) approximation [42] of the partial derivatives in Eq. (1) on the equidistant grids over three variables. We use the central second-order FD formula for approximation of the terms with the second partial derivative over z with the discontinuity at the interface [43, 44]:

$$-\frac{\hbar^2}{2\Delta_z^2}\left(\frac{2}{m_{i-1}+m_i}\chi(z_{i-1}) - \left[\frac{2}{m_{i-1}+m_i}+\frac{2}{m_i+m_{i+1}}\right]\chi(z_i) + \frac{2}{m_i+m_{i+1}}\chi(z_{i+1})\right). \quad (2)$$

The grid steps over each variable have been taken to be the same, $\Delta = \Delta_{z_e} = \Delta_{z_h} = \Delta_\rho$. Eq. (2) defines the theoretical uncertainty of the numerical solution of order of Δ^2 as $\Delta \to 0$. The discontinuities of the rectangular confinement potential are treated in a similar way.

The nonzero solution of the homogeneous equation (1) with trivial boundary conditions can be found by a diagonalization of the matrix constructed from this equation. A small part of the matrix spectrum is obtained by the Arnoldi algorithm [45]. As a result, we have calculated the lowest eigenvalue of the matrix and the corresponding eigenvector. After the extrapolation to the limit $\Delta = 0$ [12], the accurate asymptotic results are obtained. Thus, we found the energy $E_x(\Delta m, \Delta\varepsilon)$ for various QW widths.

The calculations of the binding energies as a function of the QW widths were performed for the GaAs/Al_xGa_{1-x}As model heterostructures with various concentrations of aluminum. Material and energy gap parameters used for solving the eigenvalue problem (1) are based on the data from Refs. [8, 46]. In particular, the difference of the gap energies, as a function of x, is modelled by the formula $\Delta E_g = 1087x + 438x^2$ meV. A ratio of potential barriers is taken to be $V_e/V_h = 65/35$. The Luttinger parameters used in calculations are $\gamma_1 = 6.85$, $\gamma_2 = 2.10$ for GaAs and $\gamma_1 = 3.76$, $\gamma_2 = 0.82$ for AlAs; the dielectric constants are 12.53 and 10.06, respectively. Masses and dielectric constants for the ternary alloys are obtained by a linear interpolation on x. The nonparabolicity parameters are taken to be the same for GaAs and AlAs from Ref. [41].

It is also worth noting that, despite the robustness of the theoretical model and the numerical method, the underlying envelope function approximation used for derivation of the SE becomes inappropriate for very thin QWs, namely for widths less than 2 nm that is a few monolayers of GaAs [12]. For such cases, an application of the interface short-range corrections [47] or solution of the SE in the nodal representation [48] can be considered as more reliable approaches.

4. Results of calculations

First of all, we studied the one-dimensional problems for $E_{e,h}(\Delta m \neq 0, \Delta\varepsilon \neq 0)$ with respect to $E_{e,h}$. The former defines the binding energy obtained with the material mismatches, the latter — without ones ($\Delta m = 0, \Delta\varepsilon = 0$). The differences of the studied values are shown in Figure 1. It is seen that the dominant discrepancy comes from the mass mismatch at the interfaces, whereas the difference between $E_{e,h}(\Delta m \neq 0, \Delta\varepsilon \neq 0)$ and $E_{e,h}(\Delta m \neq 0, \Delta\varepsilon = 0)$ is less than 0.06 meV, and the corresponding curves are indistinguishable in the figure. Therefore, the self-potential terms $V_{e,h}^{self}(z_{e,h})$ in such one-dimensional problems contribute little to the energy shifts. From the one-dimensional result we can conclude that the change of the confinement energy in Eq. (1) due to the mass mismatch takes place only for narrow QWs. For QWs of widths larger than 10 nm, this change is negligible. Taking into account the nonparabolicity of the conduction band

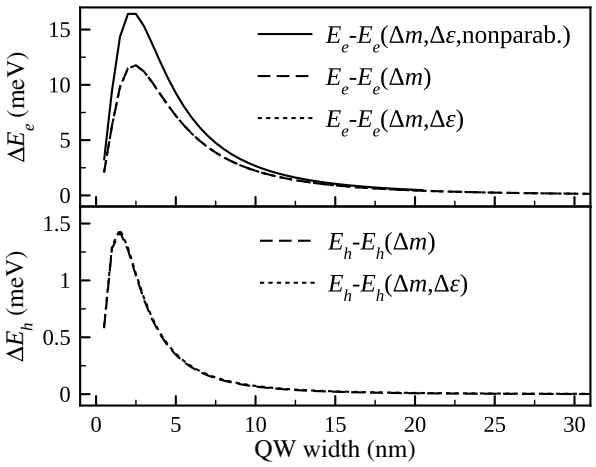

Figure 1. The difference of energies $E_{e,h}$ and $E_{e,h}(\Delta m, \Delta\varepsilon)$ for the one-dimensional problems in $GaAs/Al_{0.3}Ga_{0.7}As$ QWs. The quantity $E_{e,h}(\Delta m)$ denotes the energy $E_{e,h}(\Delta m \neq 0, \Delta\varepsilon = 0)$. The solid curve shows the energy obtained when the nonparabolicty is additionally taken into account. Dashed curves at each panel visually overlap.

results in an even greater reduction of the confinement energy for widths less than 10 nm. One can see in Figure 1 that the difference becomes more profound.

Figure 2 shows the heavy-hole exciton binding energy for three cases: without mismatches, with the mass discontinuity only, and with the mass and dielectric constant mismatches. The uncertainty of the calculated energy is 0.05 meV. One can see that the mass mismatch affects the binding energies only for QW widths of less than 10 nm. Nevertheless, this shift for narrow

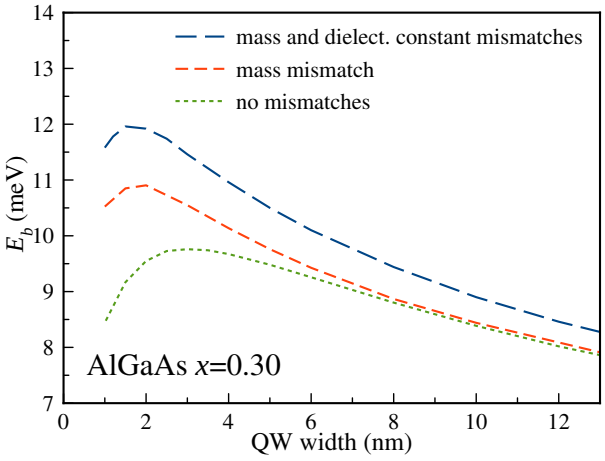

Figure 2. The binding energy of the exciton in $GaAs/Al_{0.3}Ga_{0.7}As$ QWs obtained by solving the three-dimensional problem (1). Effect of the mass mismatch, both mass and dielectric constant mismatches are shown separately.

QWs is considerable. It enhances the binding energy up to almost 25%. The dielectric constant discontinuity further increases the binding energy by about 10% for the whole studied range of QW widths. That comes from the Coulomb potential term $V^C(\rho, z_e, z_h)$ including the effect of the image charges. Our results may be compared with the calculations of Gerlach *et al.* [8]. In contrast to that paper, according to our calculations, the mass and dielectric constant mismatches both result in an increase of the binding energy. This contradiction may be caused by a truncation of the perturbative series for the electrostatic contribution after the first order term in Ref. [8].

The exciton binding energy in studied heterostructures with QW is believed to grow with increase of the Al concentration. In Figure 3 the binding energy for heavy-hole excitons in $GaAs/Al_xGa_{1-x}As$ heterostructures for various concentrations of Al, $x = 0.15, 0.3, 0.4$, is shown. The growth of the concentration increases the relative shift of the binding energy. For $x = 0.30$,

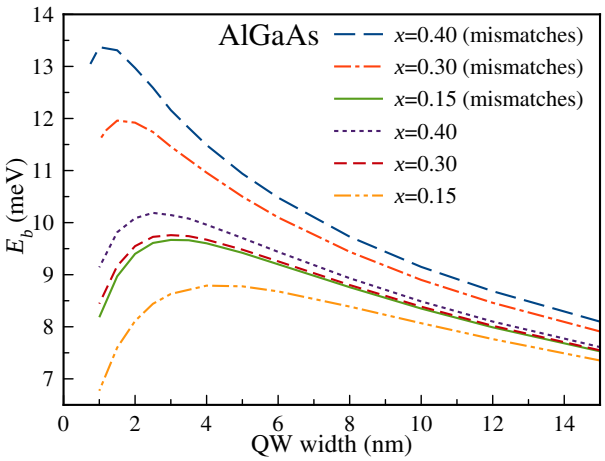

Figure 3. The binding energy of the exciton in $GaAs/Al_xGa_{1-x}As$ QWs obtained by solving the three-dimensional problem (1) for various concentrations, x, of the alloy. The results of calculations with the material parameter mismatches as well as without ones are presented.

the relative shift of about 25% is slightly larger than that estimated preliminary in Ref. [12]. The maximum of the binding energy is achieved for smaller QW widths as the concentration increases. For $x = 0.40$, the binding energy close to 13.4 meV is reached for the width of 1 nm. We compared the obtained energy for $x = 0.40$ with the results of Shuvayev *et al.* [10], who used the same Luttinger parameters, dielectric constants and the perturbation series up to several orders of the expansion parameter, but employed a different method of calculation. Both results well agree when material parameters in the QW and in the barrier are different as well as when they are identical, see Figure 4. The calculated data of Shuvayev for $GaAs/Al_{0.4}Ga_{0.6}As$ differ from our results by less than 0.2 meV. This comparison additionally confirms the correctness of our method.

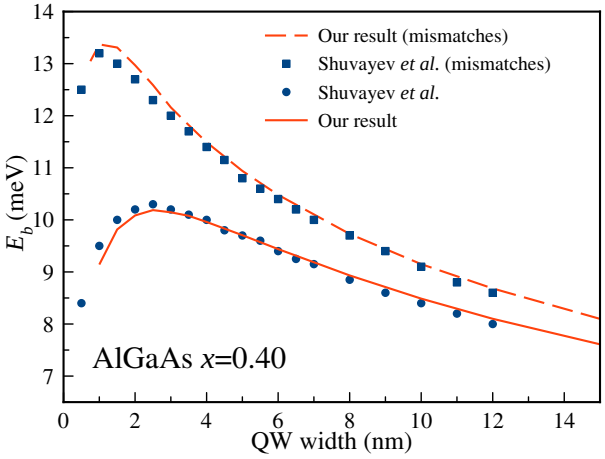

Figure 4. The comparison of the binding energy of the exciton in $GaAs/Al_{0.4}Ga_{0.6}As$ QWs obtained by our method with the results of Shuvayev *et al.* [10]. The results of calculations with the material parameter mismatches as well as without ones are shown.

The nonparabolicity of the conduction band can be taken into account by changing the electron effective masses in the z-direction and in the (x, y) plane [38–41]. We use the linear approximation of the effective mass dependence on energy, E, which is valid for QW widths larger than 3 nm. One can see in Figure 5 that due to nonparabolicity of the conduction band, the binding energy increases by about 10%, that is comparable with the case of dielectric constant mismatch accounting. The binding energy grows as the QW width diminishes. The larger concentration of the alloy causes larger enhancement of the binding energy. Therefore, a proper theoretical treatment of the binding energy in narrow QWs should take into account

not only discontinuities of material parameters but also the nonparabolicity of the conduction band.

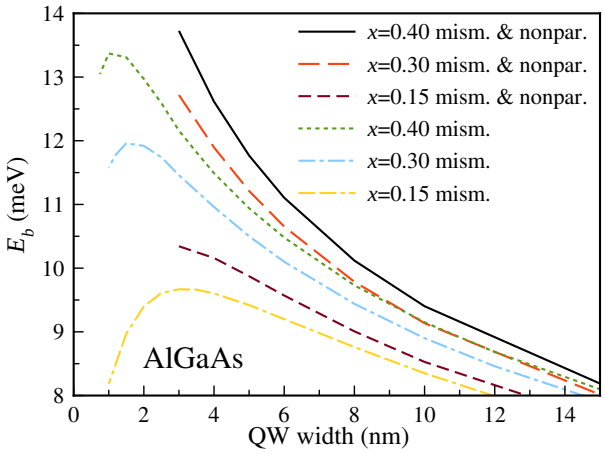

Figure 5. The binding energy of exciton in GaAs/$Al_xGa_{1-x}As$ QWs for different Al concentrations and taking into account the mismatches of the material parameters. Effect of the nonparabolicity of the conduction band is shown separately for not very narrow QWs.

5. Conclusions

In summary, we have calculated the exciton binding energies for narrow GaAs/$Al_xGa_{1-x}As$ QWs taking into account the discontinuities of the material parameters at the QW interfaces and the nonparabolicity of the conduction band. The results of calculations show that the mass and dielectric constant mismatches considerably enhance the binding energies for narrow QWs. The exciton binding energy increases up to 25% and 10%, respectively. Moreover, the nonparabolicity of the conduction band causes further increase of the binding energy, comparable with the case of dielectric constant mismatch. We hope that our results will provide a reliable reference for ongoing and future experimental studies.

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