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EXCITONS IN ZnSe/CdSe QUANTUM DOTS MOLECULES

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ABSTRACT. We theoretically investigated electron-hole states in isolated and vertically coupled flat quantum dots. The dependence of exciton binding energy on interdot distance has been studied. The obtained results show very high value of exciton binding energy at a wide range of interdot distance.

INTRODUCTION

Semiconductor quantum dots (QDs) are solid-state nanostructures, which allow confinement of carriers in all directions within dimensions smaller than their de Broglie wavelength [1]. Quantum confinement results in a characteristic discrete energy spectrum and δ-like density of states. Confinement in nanostructures with some of the linear dimensions is small compared to the exciton radius providing a possibility of enhancement of both binding energy and the oscillator strength of exciton.

Coupling between quantum dots (QDs) is now a matter of great importance. In the "artificial molecules" formed by two or more coupled QDs interdot coupling can be tuned far out of the regimes accessible in natural molecules, and single-particle tunneling and Coulomb interactions can be varied in a controlled way.

In the present work electron-hole Coulomb interaction is calculated in the two-fold stack of ZnSe/CdSe QDs. 2D-like QDs formed by Cd fluctuation [2] are considered. In quantum structures of this type lateral sizes few times exceed the bulk exciton effective radius therefore only vertical confinement is considered. Realistic potential of Pöschl-Teller type was used as a single particle potential for describing space confinement in vertical direction. Coulomb interaction between electron and hole is defined by direct diagonalization method.

CALCULATIONS AND RESULTS

Let us first consider single sheet of QDs. In the 2D-like QDs formed by Cd fluctuation with very low height to width ratio the vertical motion of carriers is mainly governed by confinement effects while their lateral motion obeys Coulomb attraction between them.

The potential energy of interaction of two opposite elementary charges – electrons and holes placed in a thin semiconductor layers (with dialectical constant ε_1) the thickness of which is less than the radius of electron-hole pair of bulk embedded between the material with dielectric constant ε_2 is given in the formula [3]:

$$V = \frac{e^2}{\varepsilon_1}\left[\frac{1}{\sqrt{\rho^2+(Z_2-Z_1)^2}} + \frac{\varepsilon_1-\varepsilon_2}{\varepsilon_1+\varepsilon_2}\frac{1}{\sqrt{\rho^2+(Z_2+Z_1)^2}}\right].$$

Here cylindrical coordinates are used.

In our case $\varepsilon_1 \approx \varepsilon_2$, therefore the second term if the brackets in eq.1 are negligible, besides this the thickness of the embedded layer is counted to be much less than characteristic distance of in-plane motion. That is why in the first approximation may be assumed that $V_0 = -\frac{e^2}{\varepsilon_1 \rho}$. The rest of the potential, which will be accounted by means of direct diagonalization is:

$$V' = \frac{e^2}{\varepsilon_1}\left[\frac{1}{\rho} - \frac{1}{\sqrt{\rho^2+(z_e-z_h)^2}}\right].$$

In the first approximation when z-dependent Coulomb term is neglected the wave function is given by the formula:

$$\Psi_{pqnm} = \phi_p^e(z_e)\phi_q^h(z_h)\chi_{nm}(\rho,\varphi).$$

$\chi_{nm}(\rho,\varphi)$ is the solution of two dimensional Coulomb problem describing electron-hole lateral motion:

$$\chi_{nm}(\rho,\varphi) = C_{nm}e^{im\varphi}(2k_n\rho)^{|m|}e^{-k_n\rho}F(-n+|m|,2|m|+1,2k_n\rho),$$

$$k_n = \frac{e^2}{h\varepsilon_1(n+1/2)}\sqrt{2\mu},$$

where n is the main quantum number, $m = 0,\pm1...\pm n$; e and Γ are elementary charge and Planck constants, μ reduced effective mass of the electron-hole pair; F is the confluent hypergeometric function; C_{nm} normalizing constant. $\phi_{p(q)}^{e(n)}(z_{e(h)})$ are the electron (hole) single particle wave functions which describe their vertical motion. Corresponding to (3) energies are given by the formula:

$$E_{pqn} = \varepsilon_p^e + \varepsilon_q^h - \frac{e^4\mu}{2\varepsilon_1{}^2\hbar^2(n+1/2)^2},$$

where $\varepsilon_{p(q)}^{e(h)}$ are eigenvalues corresponding to $\phi_{p(q)}^{e(n)}(z_{e(h)})$. As to small dot vertical size separation between $\varepsilon_{p(q)}^{e(h)}$ shells much exceeds the Coulomb term, under each $\varepsilon_p^e + \varepsilon_q^h$ the energy levels corresponding to different main quantum number of in-plane motion are grouped. These levels are (2n+1)-fold degenerated. Taking into account z coordinate in Coulomb interaction causes their shift and splitting.

To take into account variation of Cd concentration in QDs formed by Cd segregation and interdiffusion vertical confinement is expressed by the potential (Fig.1):

$$U(z) = \frac{-U_0}{ch^2 \alpha z}$$

Here U_0 is the maximum band off set between the QD and barrier material realized in the center of QD layer. $1/\alpha$ is a measure of dot size and is defined from the emission spectra of single QDs.

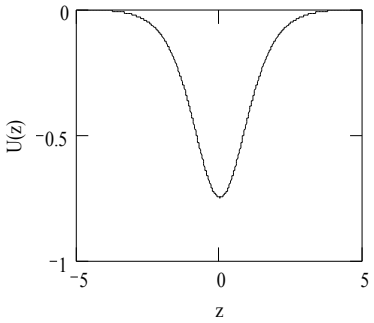


Fig.1. z-direction confinement potential in eV for single QDs

The expressions for eigenvectors $\phi^{e(h)}_{p(q)}(z_{e(h)})$ and eigenvalues $\varepsilon^{e(h)}_{p(q)}$ can be found elsewhere [4]. In our case for ZnSe/CdSe quantum dots only one electron and only one hole levels are possible inside the well. For potential barriers of electrons and holes - U_{0e} and U_{0h} 0.750eV and 0.230eV are taken; effective masses are $0.13m_0$ for electrons and $0.45m_0$ for holes.

As was mentioned we calculated z-dependent Coulomb energy by direct diagonalization of Hamiltonian matrix

$$H_{ll'} = \langle l | (H_0^e + H_0^h - H_c) | l' \rangle$$

$|l\rangle$ denotes the states of electron-hole system described by wave functions (3), $H_0^{e(h)}$ and H_c are single particle Hamiltonian and Coulomb term, respectively. In our calculations six lowest energy

states of electron-hole relative lateral motion are found to yield good convergence. In Figure.2 exciton energies in 2D ZnSe/CdSe QDs without (on the left) and with accounting (on the right) z-dependent Coulomb term are given. As is seen accounting z-dependent term in Coulomb potential causes strong shift and splitting of energy levels. Calculated value of exciton binding energy in ZnSe/CdSe QDs is increased up to 100meV for 1nm dot vertical size and significantly exceeds the binding energy of exciton in bulk CdSe crystal (15mev).

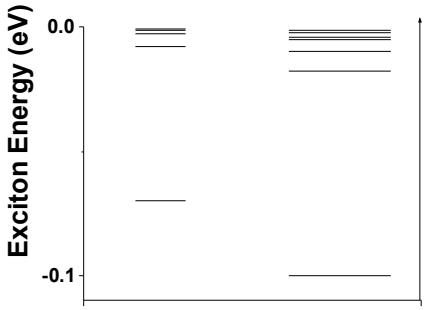


Fig. 2. Exciton energies with (right hand side) and without (left hand side) accounting z-dependent Coulomb term

For the stacks of QDs single particle potential is (Fig. 3b))

$$U(z) = \frac{-U_0}{ch^2\alpha\left(z - \frac{a+b}{2}\right)} + \frac{-U_0}{ch^2\alpha\left(z + \frac{a+b}{2}\right)}$$

z = o is taken between the centers of QDs, a and b are dots vertical size and the distance between them. If neglecting z coordinate in the Coulomb term again the wave function of electron-hole system is given as:

$$\Phi_{pqnm} = \left[\phi_p^e\left(z_e - \frac{a+b}{2}\right) \pm \phi_{ps}^e\left(z_e + \frac{a+b}{2}\right)\right] \times$$

$$\times\left[\phi_q^h\left(z_h-\frac{a+b}{2}\right)\pm\phi_q^h\left(z_h+\frac{a+b}{2}\right)\right]\chi_{nm}(\rho,\varphi),$$

Because of coupling of QDs in z direction single particle wave functions are presented as symmetric and antysymmetric combination of functions centered in neighboring dots(fig.3c)). Corresponding energies are expressed as:

$$E_{spn}=\varepsilon_p^e\pm\Delta_e+\varepsilon_q^h\pm\Delta_h-\frac{e^4\mu}{2\varepsilon_1^2\hbar^2(n+1/2)^2}$$

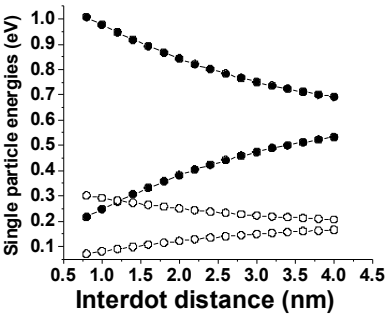
$\Delta_{e(h)}$ characterizes overlapping of wave functions centered in neighboring dots:

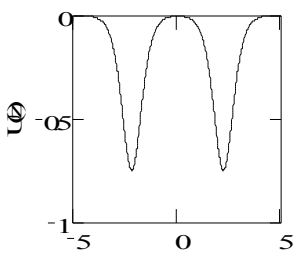
$$\Delta_{e(h)}(a,b)=$$

$$=\varepsilon_{p(q)}^{e(h)}(a)\int\phi_{p(q)}^{e(h)}\left(z_{e(h)}+\frac{\mathbf{a}+b}{2}\right)\phi_{p(q)}^{e(h)}\left(z_{e(h)}-\frac{\mathbf{a}+b}{2}\right)dz_{e(h)}$$

In Fig.3a) electron and hole single particle energies corresponding to symmetric and antisymmetric wave functions are given. The energy of electron-hole system if neglecting their interaction is the sum of electron and hole energy. So, in our case, when only one single particle energy level is possible in the single dot, there is QD molecule there are four energy levels: two with positive *z*-parity corresponding to the both carriers in symmetric or antisymmetric state; and two with negative total parity corresponding to the electron in symmetric and hole in antysymmetric state, and vice versa (Fig.3d)).

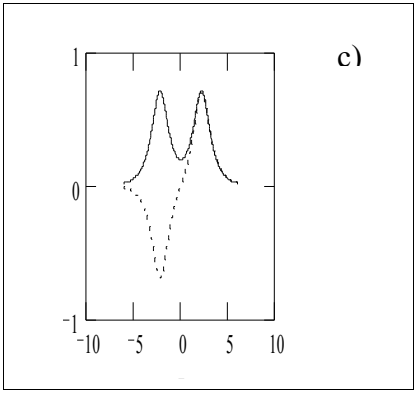
a) b)

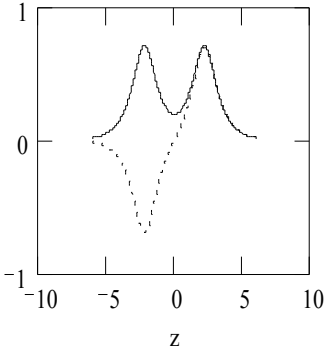

Single particle energies (eV)
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5
Interdot distance (nm)


0
-0.5
-1
-5
0
5

b)

d) c)





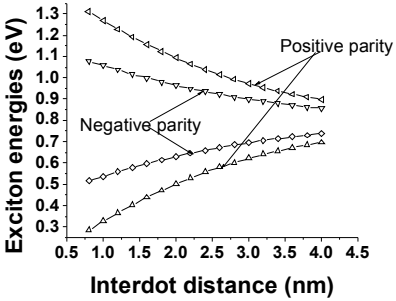


Fig.3. a) – electron (filled circles) and hole(blank circles) single particle energies as a function of interdot distance; b) - shape of confinements potential in quantum dot molecule; c) - single particle wave functions; d) energies of electron-hole pair excluding Coulomb interaction between them as a function of interdot distance

When accounting the Coulomb interaction in the first approximation, that is the last term in (10), under each of four energy levels group energy levels describing electron-hole lateral motion appear (not shown here).

We took into account z coordinate in Coulomb term by diagonalization (4) matrix with $|l\rangle$ corresponding to (6) and (5) as

single particle potential. z-dependent Coulomb interaction mixes the states with the different main quantum number of in-plane motion and owing the same total z-parity. In Fig.4.a) the obtained energies of excitons as a function of interdot distance are given. The solid lines correspond to the states with even z-parity, dotted lines to the states with odd z-parity. For simplicity only two lower energy states of "bonding" and "antybonding" behavior of positive and negative parity are presented. With increasing the distance between dots low energy level approaches to the lowest energy level of exciton confined in isolated dot (labeled by E_{exc}), the high energy levels tend to the sum of the energies of non-interacting electron and hole ($\varepsilon^e+\varepsilon^h$). Exciton binding energy is very high again (Fig.4.b)). For a = 1nm and b =1.2nm it is 0.078eV. With increasing interdot distance it decreases to 0.065eV for b = 4.2 nm, then increases again and gradually approaches the binding energy of excitons in isolated QDs.

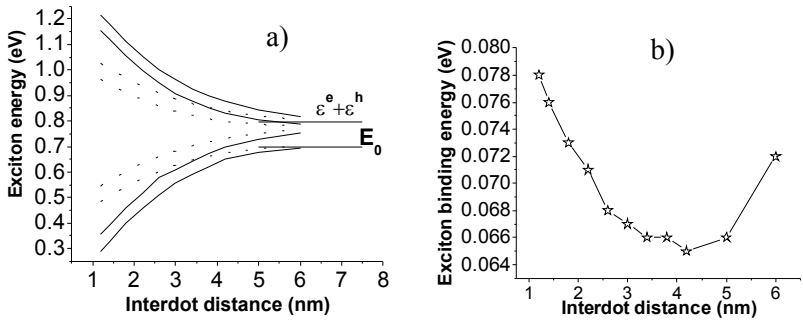


Fig.4. a) - Energies of excitons of positive total parity (solid lines labeled and negative total parity (cyrcles) as a function of interdot distance; b) - exciton binding energy as a function of interdot distance

ACKNOWLEDGMENTS

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