Excitonic quasimolecule consisting of two semiconductor quantum dots

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The effect of a significant increase in the binding energy of the singlet ground state of an excitonic quasimolecule consisting of two CdS quantum dots, in comparison with the binding energy of a biexciton in a single crystal of CdS (almost by two orders of magnitude), has been found.

1. Introduction

The idea of superatom was fruitful for the development of nanophysics [1–3]. The superatom (quasiatomic nanoheterostructure) consists of a spherical quantum dot (QD) with the radius *a*, the volume of which contains the semiconductor (or dielectric) material. QD is surrounded by a dielectric (semiconductor) matrix [2, 3]. A hole is localized in the volume of QD, and an electron is localized over the spherical interface (QD–matrix). In this nanosystem, the lowest electronic level is located in the dielectric matrix, and the lowest hole level is within the volume of QD. A large shift of the valence band (700 meV) generates the localization of holes in the volume of QD. A significant shift of the conduction band (about 400 meV) produces a potential barrier for electrons [4]. The electrons moving in the matrix do not penetrate into the volume of QD [2–4]. The energy of the Coulomb interaction of the electron with a hole and the energy of the polarization interaction of the electron is localized over the surface of the QD. Certain orbitals, localized around the QD, correspond to electrons in the superatom [2, 3].

The optical characteristics of nanosystems with CdS, ZnSe, Al_2O_3 and Ge QDs published in experimental papers [4–9] has been revauled that the electron can be localized above the surface of the QD while the hole moves in the volume of the QD.

Keywords: spatially separated electron and holes, quantum dots, binding energy, Coulomb and exchange interaction.

In [4–9] the appearance of superatoms located in dielectric matrices as the cores containing CdS, ZnSe, Al_2O_3 and Ge QDs was apparently established experimentally for the first time. A substantial increase in the binding energy of the ground state of an electron in a superatom in comparison with the binding energy of an exciton in CdS, ZnSe and Al_2O_3 and single crystals was detected in [2, 3, 10].

In [5–9] the optical characteristics of samples of borosilicate glasses doped with CdS, ZnSe and Al₂O₃ at concentrations $x \approx 0.003\%$ to 1% were investigated. The average radii \overline{a} of CdS and ZnSe QDs were in the range of about 2.0 to 20 nm. In the low-temperature absorption spectra the maximum was detected when there were large concentration x os CdS QDs in the samples (from about 0.6% to 1%). This experimantal findings has been interpreted as the apperance of the bounded QD states. In order to explain the optical characteristics of such nanosystems, we proposed a model of a quasimolecule representing two ZnSe QDs that form an exciton quasimolecule as a result of the interaction of electrons and holes [11].

It was noted [6] that, if the samples contain such QDs, one must take into account the interaction between charge carriers localized above the QD surfaces. Therefore, in [11], we develop the theory of a biexciton (or an exciton quasimolecule), formed from spatially separated electrons and holes, in a nanosystem that consists of ZnSe QDs synthesized in a borosilicate glassy matrix. Using the variational method, we obtain the total energy and the binding energy of the biexciton singlet ground state in such a system as functions of the spacing between the QD surfaces and the QD radius. We show that the biexciton formation is of the threshold character and demonstrate the existence of biexciton states in nanosystems, in which the spacing between the QD surfaces exceeds a certain critical spacing. It is established that the spectral shift of the low-temperature luminescence peak [6] in such a nanosystem is due to quantum confinement of the energy of the biexciton ground state.

The convergence of two (or more) QDs up to a certain critical value D_c between surfaces of QDs leads to overlapping of electron orbitals of superatoms and the emergence of exchange interactions [3, 11, 12]. In this case, the overlap integral of the electron wave functions takes a significant value. As a result, the conditions for the formation of quasimolecules from QDs can be created [3, 11]. One can also assume that the above conditions of formation of quasimolecules can be provided by external physical fields. This assumption is evidenced by results presented in [13, 14], in which the occurrence of the effective interaction between QDs at considerable distances under conditions of electromagnetic field was observed experimentally. In [15] energies of the ground state of "vertical" and "horizontal" located pair of interacting QDs ("molecules" from two QDs) were determined as a function of the steepness of the confining potential and the magnetic field strength. The quantum part of a nanocomputer, which was implemented on a pair of QDs ("molecules" from two QDs) with charge states is *n* qubits [16]. The first smoothly working quantum computer has been proposed on QDs with two electron orbital states as qubits. In the present work (unlike [11]), the dependences of the total energy and also of the binding energy of the singlet ground state of a quasimolecule consisting of two CdS QDs were obtained by the variational method as functions of the distance between the surfaces of the QDs and also the radius of the QD. A substantial increase in the binding energy of the singlet ground state of the exciton quasimolecule (by almost two orders of magnitude) in comparison with the bond energy of the biexciton in a single crystal of CdS was detected. It was shown that the maximum observed in the absorption spectrum of the nanosystem is due to the formation of an exciton quasimolecule. In the present work, we are studying the possibility of formation of an exciton quasimolecule consisting of two CdS QDs. A theory of a "molecular" quasicrystal will then be developed on the basis of such a model of an exciton quasimolecule.

2. Binding energy of a singlet ground state of an excitonic quasimolecule

Consider the model of nanosystems, containing two superatoms. In this model, the superatoms consist of the spherical semiconductor QDs, A and B, synthesized in a matrix of borosilicate glass with dielectric constant ε_1 . The QDs radius is a, the spacing between the spherical QD surfaces is D. Each QD is formed from a semiconductor material with dielectric constant ε_2 . For simplicity and without loss of generality, we assume that the holes h(A) and h(B) with effective masses m_h are localized in the centers of QD(A) and QD(B), and electrons e(1) and e(2) with effective masses $m_e^{(1)}$ are localized near the spherical surfaces of QD(A) and QD(B), respectively. The above assumption is reasonable, since the ratio of the effective masses of the electron and the hole in the nanosystem is much smaller than unity ($m_e^{(1)}/m_h \ll 1$). Let us assume that there is an infinitely high potential barrier on the spherical QD–matrix interface. Therefore, in the nanosystem, the holes do not leave the volume of QD, whereas the electrons do not penetrate into QDs.

In this model nanosystem, we study the possibility of the formation of an excitonic quasimolecule consisting of spatially separated electrons and holes. The holes are localized at the centers of QD(A) and QD(B), and the electrons are localized on the spherical surfaces QD(A) and QD(B). Within the framework of the adiabatic approximation, as well as in the effective mass approximation, the Hamiltonian of a quasimolecule can be written as

$$\hat{H} = \hat{H}_{A(1)} + \hat{H}_{B(2)} + \hat{H}_{\text{int}}$$
(1)

where $\hat{H}_{A(1)}$ and $\hat{H}_{B(2)}$ are Hamiltonians of superatoms. The hole h(A) is localized in the center of QD(A), while the electron e(1) is localized on the surface of QD(A), the hole h(B) is localized in the center of QD(B), and the electron e(2) is localized on the surface of QD(B). The contribution of energy of the polarization interaction of electrons and holes with the surface of QD into Hamiltonians $\hat{H}_{A(1)}$ and $\hat{H}_{B(2)}$ of superatoms can be ignored at the first approximation [3]. Therefore the superatom Hamiltonian $\hat{H}_{A(1)}$ takes the following form

$$\hat{H}_{A(1)} = -\frac{\hbar^2}{2\mu} \Delta_{(1)} + V_{e(1)h(A)}(r_{A(1)}) + E_g$$
(2a)

where the first term is the operator of the kinetic energy of electrons and holes, $\mu = m_e^{(1)} m_h / (m_e^{(1)} + m_h)$ is the reduced mass of the electron and the hole, $V_{e(1)h(A)}$ is the Coulomb interaction energy between the electron e(1) and the hole h(A),

$$V_{e(1)h(A)} = -\frac{1}{2} \left(\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} \right) \frac{e^2}{r_{A(1)}}$$
(2b)

and $r_{A(1)}$ is the distance of electron e(1) from the center of QD(A), E_g is the band gap of CdS bulk. The form of the Hamiltonian $\hat{H}_{B(2)}$ is similar to the Hamiltonian $\hat{H}_{A(1)}$. In the first approximation, we can neglect the contribution of the energy of interaction of the electrons e(1) and e(2) and the holes h(A) and h(B) with the fields of polarization, induced by these carriers on the surfaces of QD(A) and QD(B) in the Hamiltonian \hat{H}_{int} . Therefore, the Hamiltonian \hat{H}_{int} includes only the energy of the Coulomb interaction between electron e(1) and hole h(B), electron e(2) and hole h(A), electrons e(1) and e(2), as well as holes h(A) and h(B).

On the assumption that the spins of electrons e(1) and e(2) are antiparallel, we write the normalized wave function of the singlet ground state of the exciton quasimolecule in the form of a linear combination of wave functions $\Psi_1(r_{A(1)}, r_{B(2)})$ and $\Psi_2(r_{A(2)}, r_{B(1)})$ [11]

$$\Psi_{S}(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)}) =$$

$$= \left\{ 2\left[1 + S^{2}(D, a)\right] \right\}^{-1/2} \left[\Psi_{1}(r_{A(1)}, r_{B(2)}) + \Psi_{2}(r_{A(2)}, r_{B(1)}) \right]$$
(3)

where S(D, a) is the overlap integral of one-electron wave functions. Considering that electrons e(1) and e(2) are moving independently of each other, wave functions $\Psi_1(r_{A(1)}, r_{B(2)})$ and $\Psi_2(r_{A(2)}, r_{B(1)})$ (see Eq. (3)) can be represented as one-electron variational wave functions $\varphi_{A(1)}(r_{A(1)})$ and $\varphi_{B(2)}(r_{B(2)})$, $\varphi_{A(2)}(r_{A(2)})$ and $\varphi_{B(1)}(r_{B(1)})$, respectively [11]:

$$\Psi_1(r_{A(1)}, r_{B(2)}) = \varphi_{A(1)}(r_{A(1)})\varphi_{B(2)}(r_{B(2)})$$
(4)

$$\Psi_2(r_{A(2)}, r_{B(1)}) = \varphi_{A(2)}(r_{A(2)})\varphi_{B(1)}(r_{B(1)})$$
(5)

We represent the one-electron wave functions $\varphi_{A(1)}(r_{A(1)})$ and $\varphi_{B(2)}(r_{B(2)})$ that describe, correspondingly, the electron e(1) localized above the QD(A) surface and the electron e(2) localized above the QD(B) surface and the wave functions $\varphi_{A(2)}(r_{A(2)})$

and $\varphi_{B(1)}(r_{B(1)})$ that describe, correspondingly, the electron e(2) localized above the QD(A) surface and the electron e(1) localized above the QD(B) surface as variational Coulomb-shaped wave functions [11]:

$$\varphi_{A(1)}(r_{A(1)}) = \tilde{A} \exp\left[-\overline{\mu}(\tilde{a}) \frac{r_{A(1)}}{a_{\text{ex}}^0}\right]$$
(6)

$$\varphi_{B(2)}(r_{B(2)}) = \tilde{A} \exp\left[-\overline{\mu}(\tilde{a}) \frac{r_{B(2)}}{a_{\text{ex}}^0}\right]$$
(7)

$$\varphi_{A(2)}(r_{A(2)}) = \tilde{A} \exp\left[-\overline{\mu}(\tilde{a}) \frac{r_{A(2)}}{a_{\text{ex}}^0}\right]$$
(8)

$$\varphi_{B(1)}(r_{B(1)}) = \tilde{A} \exp\left[-\bar{\mu}(\tilde{a}) \frac{r_{B(1)}}{a_{\text{ex}}^0}\right]$$
(9)

where $\tilde{A} = \pi^{-1/2} (a_{ex}^0)^{-3/2} \mu^{-3/2} \exp(\bar{\mu}\tilde{a}) [2(\bar{\mu}\tilde{a})^2 + 2(\bar{\mu}\tilde{a}) + 1]^{-1/2}$ is the normalization constant, $\bar{\mu}(\tilde{a}) = \tilde{\mu}(a)/m_0$ is the variational parameter ($\tilde{\mu}(a)$ is the reduced exciton effective mass, m_0 is the electron mass in vacuum), $\tilde{a} = a/a_{ex}^0$ is the dimensionless QD radius, $a_{ex}^0 = [2\varepsilon_1\varepsilon_2/(\varepsilon_1 + \varepsilon_2)][\hbar^2/\mu_0e^2]$ is the Bohr radius of a 2D exciton localized above the planar interface between the semiconductor with the permittivity ε_2 and the matrix with the permittivity ε_1 (the hole is in motion within the semiconductor, whereas the electron is in the matrix), e is the electron charge, $\mu_0 = m_e^{(1)} m_h/(m_e^{(1)} + m_h)$ is the reduced 2D exciton effective mass.

Because of the identity of the electrons, the wave function $\Psi_2(r_{A(2)}, r_{B(1)})$ (Eq. (5)) is equivalent to the wave function $\Psi_1(r_{A(1)}, r_{B(2)})$ (Eq. (4)). In Eq. (3), the integral S(D, a) is determined by the following formula:

$$S(D, a) = \int d\tau_1 \varphi_{A(1)}(r_{A(1)}) \varphi_{B(1)}(r_{B(1)})$$

where $d\tau_1$ is the volume element of the electron e(1).

In this case, the spins of electrons e(1) and e(2) are parallel and, similarly, to the theory of the chemical bond of the hydrogen molecule [17], the excitonic quasimolecule, consisting of two QDs, is not formed. Therefore, we did not consider this case.

Within the framework of the variational method in the first approximation, the total energy of the ground singlet state of the excitonic quasimolecule is determined by the average value of the Hamiltonian $\hat{H}(1)$ for states which are described by wave functions of the zero approximation $\Psi_{S}(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)})$ (see Eq. (3)) [11]:

$$E_0(D, a) = 2E_{ex}(a) + E_b(D, a)$$
(10)

where $E_b(D, a)$ is the binding energy of the singlet ground state of the excitonic quasimolecule and $E_{ex}(a)$ is the binding energy of the ground state of an electron in the superatom, found in [12]. The wave function $\Psi_S(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)})$ (see Eq. (3)) contains wave functions (see Eqs. (4)–(9)). The results of a numerical variational calculation of the binding energy $E_b(D, a)$ of the ground singlet state of the excitonic quasimolecule, containing two CdS QDs with average radii of $\overline{a}_1 = 4$ nm and $\overline{a}_2 = 4.4$ nm ($\varepsilon_2 = 9.3$, and effective hole mass in QD $m_h/m_0 = 5$), grown in a matrix of borosilicate glass ($\varepsilon_1 = 2$, and the electron effective mass in the matrix $m_e^{(1)}/m_0 = 0.537$), which was investigated in experimental work [5, 6], are shown in Tables 1 and 2.

In [5, 6] we studied the optical properties of the samples of borosilicate glass, doped with CdS with concentrations from $x \approx 0.003\%$ to 1%. The average radius \bar{a} of CdS QDs was ranged from about 2.0 nm to 20 nm. At concentrations of CdS QDs in the range of $x \approx 0.6\%$ to 1%, the peak ($E - E_g \approx -712$ meV) was found in the absorption spectra of the samples at the temperature of 4 K (where E_g is the band gap of CdS QD) [6]. The variational method, used for estimation of the binding energy $E_b(D, a)$ of the ground singlet state of the excitonic quasimolecule, will be valid if the binding energy

T a ble 1. The dependence of the binding energy $E_b(D, \bar{a}_1)$, and the total energy $E_0(D, \bar{a}_1)$ of ground singlet state of excitonic quasimolecule consisting of two CdS QDs with average radii of $\bar{a}_1 = 4$ nm, on the distance D between surfaces of QDs. In this case, the binding energies of an electron in superatoms are $E_{ex}(\bar{a}_1) \cong -320$ meV.

<i>D</i> [nm]	$E_{\rm b}(D, \overline{a}_1) [{\rm meV}]$	$E_0(D, \overline{a}_1) \text{ [meV]}$
1.8	0	-640
2.2	-27	-667
2.6	-28.1	-668.1
3	-27.8	-667.8
4	-23.2	-663.2
6	-9.5	-649.5
8.4	0	-640

T a ble 2. The dependence of the binding energy $E_b(D, \bar{a}_2)$, and the total energy $E_0(D, \bar{a}_2)$ of ground singlet state of excitonic quasimolecule, consisting of two CdS QDs with average radii of $\bar{a}_2 = 4.4$ nm, on the distance D between surfaces of QDs. In this case, the binding energies of an electron in superatoms are $E_{ex}(\bar{a}_2) \cong -344$ meV.

D [nm]	$E_{\rm b}(D, \overline{a}_2) [{\rm meV}]$	$E_0(D, \overline{a}_2)$ [meV]
1.6	0	-688
2.2	-29.4	-717.4
2.48	-32.8	-720.8
2.6	-31.9	-719.9
3	-28.9	-716.9
4	-24.4	-712.4
6	-13.2	-701.2
8	-3.4	-691.4
9.8	0	-688

 $E_{\rm b}(D, a)$ of the quasimolecule is small, compared with the binding energy $E_{\rm ex}(a)$ of the ground state of the superatom [11]

$$\frac{E_{\rm b}(D,a)}{E_{\rm ex}(a)} \ll 1 \tag{11}$$

Binding energies $E_b(D, a)$ of the ground singlet state of the excitonic quasimolecule, containing two CdS QDs with average radii of $\overline{a}_1 = 4$ nm and $\overline{a}_2 = 4.4$ nm have a minimum $E_b^{(1)}(D_1, \overline{a}_1) \approx -28.1$ meV for a distance $D_1 \cong 2.6$ nm and $E_b^{(2)}(D_2, \overline{a}_2) \approx -32.8$ meV for a distance $D_2 \cong 2.48$ nm ($E_b^{(1)}$ and $E_b^{(2)}$, see Tables 1 and 2, correspond to critical temperatures $T_{c(1)} \approx 326$ K and $T_{c(2)} \approx 380$ K). Binding energies of the ground state of electron of superatoms are $E_{ex}(\overline{a}_1) \cong -320$ meV and $E_{ex}(\overline{a}_2) \cong -344$ meV [12]. In this case, the energies of the singlet ground state (Eq. (4)) of the excitonic quasimolecule are $E_0(D_1, \overline{a}_1) \approx -668.1$ meV and $E_0(D_2, \overline{a}_2) \approx -720.8$ meV (see Tables 1 and 2). From the comparison of the total energy $E_0(D, \overline{a}_2)$ of the quasimolecule with a maximum ($E - E_g$) ≈ -712 meV, we get the value of the distance $D_3 \approx 4.0$ nm between QDs. The criterion (11) of the applicability of used variational estimation of the binding energy $E_b(D, a)$ of the quasimolecule is fulfiled $E_b^{(1)}(D_1, \overline{a}_1)/E_{ex}(\overline{a}_1) \cong 0.09$, and $E_b^{(2)}(D_2, \overline{a}_2)/E_{ex}(\overline{a}_2) \cong 0.09$.

With increasing QDs radius *a*, the binding energy $E_{ex}(a)$ of the ground state of the electron in the superatom increases [12]. The average size of the state of electron in the superatom decreases. Therefore, the distance, for which the square of the overlapping integral S(D, a) of the one-electron wave functions, takes on a maximum value. Also it decreases with increasing QD radius *a*, and so the distance between the surfaces of QD D_2 is less than D_1 . As a result, with increasing QD radius *a* also the maximum value of the binding energy of the singlet ground state of the excitonic quasimolecule $|E_b(D, a)|$ increases (*i.e.*, $|E_b^{(2)}| > |E_b^{(1)}|$). In this case, the criterion (11) of the applicability of used variational estimation of the binding energy $E_b(D, a)$ of the quasimolecule is fulfiled.

Thus, the proposed model of the excitonic quasimolecule enables us to explain the optical properties of nanosystems, consisting of CdS QDs, grown in a borosilicate glass matrix [5, 6], in particular, the appearance of a peak $(E - E_g) \approx -712$ meV in the absorption spectra of the samples at 4 K.

It should be noted that the binding energy $E_{ex}(a)$ of the ground state of an electron in the superatom [12] can be applied only for values of the binding energy $E_{ex}(a)$, for which the inequality

$$E_{\rm ex}(a) - E_g \ll \Delta V \tag{12}$$

is correct (ΔV is the depth of the potential well for an electron and hole in a QD). In the experimental work [5] it has been found that for CdS QDs with radii of $a \le 20$ nm, $\Delta V = 2.5$ eV. Condition (12) makes it possible to consider the motion of an electron

and hole in the superatom, using QD model as an infinitely deep potential well [12]. In the Hamiltonian (1) of the quasimolecule, the terms, causing fluctuations of QD in the quasimolecule, were not considered. The obtained quasimolecule binding energies (see Tables 1 and 2) are significantly higher than the typical energy of QD fluctuations.

It follows from Tables 1 and 2 that the excitonic quasimolecule, consisting of two QDs occurs for the distance between the surfaces of QDs $D \ge D_c^{(1)} \cong 1.8 \text{ nm}$ for QD with a radius of $\overline{a}_1 = 4 \text{ nm}$ and $D \ge D_c^{(1)} \cong 1.6 \text{ nm}$ for QD with a radius of $\overline{a}_2 = 4.4$ nm. The formation of such quasimolecule has a threshold character. It is possible only for nanosystems containing QDs with average radii of \bar{a}_1 and \bar{a}_2 , in which the distance D between the surfaces of QDs exceeds certain critical distance $D_{c}^{(1)}$. The existence of such critical distance $D_c^{(1)}$ is associated with a dimensional quantum effect, for which decreasing of the distance D between the surfaces of QDs leads to a decrease in the interaction energies of electrons and holes in the Hamiltonian (1) of the quasimolecule, which cannot be compensated by the increase in the kinetic energy of electrons. With the increase of the distance D between the surfaces of QDs, starting from the values $D \ge D_c^{(1)} \cong 8.4 \text{ nm}$ for QD with a radius of $\overline{a}_1 = 4 \text{ nm}$ and $D_c^{(2)} \cong 9.8 \text{ nm}$ for QD with a radius of $\overline{a}_2 = 4.4 \text{ nm}$, the excitonic quasimolecule splits into two superatoms. Thus, the excitonic quasimolecule of nanosystem may occur when the condition $D_c^{(1)} \le D \le D_c^{(2)}$ is realized (see Tables 1 and 2). In addition, the quasimolecule can exist only at temperatures below a certain critical temperature $T_{c(1)} \approx 326$ K and $T_{c(2)} \approx 380$ K. Biexcitons arose in CdS single-crystal with a binding energy $E_b = 0.59$ meV [11]. The binding energies $E_b^{(1)}$ and $E_b^{(2)}$ of the excitonic quasimolecule significantly exceed $E_{\rm b}$, almost by two orders of magnitude. Apparently the latter fact opens the possibility of observing such excitonic quasimolecules at room temperature. The energy of the exchange interaction between the electrons and the holes mainly contributes to the binding energy of the excitonic quasimolecule, which is significantly greater than the energy of the Coulomb interaction between electrons and holes (*i.e.*, their ratio ≤ 0.11). The estimations of the binding energy $|E_b(D, a)|$ of the singlet ground state of a quasimolecule are variational and may give low values of the binding energy $|E_{\rm b}(D, a)|$ and $|E_{\rm b}^{(1)}|$.

3. Conclusion

The binding energy of the singlet ground state of the excitonic quasimolecule, consisting of two QDs, has significantly large value, larger than the binding energy of the biexciton in a single CdS crystal, almost by two orders of magnitude. This effect opens the possibility to use the excitonic quasimolecule as an active medium of nanolasers, radiating in the infrared range of the spectrum and working on the excitonic transitions at room temperature, as well as a nanoelements base in a quantum computer [10]. These results demonstrate the fundamental possibility of creating new quasiatomic nanosystems – excitonic quasimolecules, with new physical properties. On this basis, it is possible

to construct new quantum nanosystems – pseudocrystals, managed by symmetry and the lattice period, which will allow to realize the unique physical effects and phenomena, and to establish new principles in material science.

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