



Excitons and quasimolecules in quasi – zero - dimensional nanostructures

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ABSTRACT

This review summarizes the results of theoretical studies of an excitons and quasimolecules (biexcitons) formed from spatially separated electrons and holes (the hole is in the semiconductor spherical quantum dot volume, and the electron is localized at the outer spherical surface of the quantum dot–dielectric matrix interface.

Keywords: excitons, biexcitons, quasimolecules, semiconductor (dielectric) quantum dots, binding energy of excitons, nanolaser.

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INTRODUCTION

The use of semiconductor nanosystems as the active region of nanolasers is prevented by the low binding energy of the quantum dot (QD) exciton [1, 2]. Therefore, studies directed toward the search for nanostructures in which a significant increase in the binding energy of QD excitons would be observed are of importance. Currently, the theory of exciton states in quasi- zero-dimensional semiconductor nanosystems has not been adequately studied, in particular, no theory exists for an exciton with a spatially separated electron and hole in quasi- zero- dimensional nanosystems.

The review analyzes the results of theoretical investigations of excitons states (electron - hole pairs states) in a quasi - zero - dimensional nanosystems consisting of spherical semiconductor nanocrystals (quantum dots) placed in transparent dielectric matrices. [1 – 5, 7, 8]. The theory of exciton states in a quantum dot under conditions of dominating polarization interaction of an electron and a hole with a spherical (quantum dot – dielectric matrix) interface are developed. An shown, that the energy spectrum of heavy hole in the valence band quantum dot is equivalent to the spectrum of hole carrying out oscillator vibrations in the adiabatic electron potential. In the framework of the dipole approximation theory are developed interband absorption of light in quasi – zero – dimensional nanosystems. An shown, that the absorption and emission edge of quantum dots is formed by two transitions of comparable intensity from different hole size – quantization levels and into a lower electron size – quantization level. Propose a theoretical prospect of using

hole transitions between equidistant series of quantum levels observed in nanocrystals for desining a nanolaser.

The theory of an exciton formed from spatially separated electron and hole (the hole is in the quantum dot volume, and the electron is localized at the outer spherical quantum dot–dielectric matrix interface) is developed [3, 4] within the modified effective mass method [5]. The effect of significantly increasing the exciton binding energy in quantum dots of zinc selenide, synthesized in a borosilicate glass matrix, relative to that in a zinc selenide single crystal is revealed. We developed the theory of a quasimolecules (biexcitons) formed from spatially separated electrons and holes (the hole is in quantum dot volume, and the electron is localized at the outer surface of the quantum dot – dielectric matrix interface) in a nanosystem that consists of zinc selenide quantum dots synthesized in a glassy matrix.

MATERIALS AND METHODS

Excitons in quasi – zero - dimensional nanosystems

Therefore, in [3, 4] we developed within the modified effective mass method [5] the theory of an exciton formed from spatially separated electron and hole (the hole is in the semiconductor spherical QD volume, and the electron is localized at the outer spherical surface of the QD–dielectric matrix interface. It was shown that the short wavelength shift of the peak of the low temperature luminescence spectrum of samples containing zinc-selenide QDs, observed under the experimental conditions of [6], is caused by quantum confinement of the ground state energy of the exciton with a spatially separated electron and hole. The effect of significantly increasing the binding energy of an exciton (with a spatially separated electron and hole) in a nanosystem containing zinc-selenide QDs in comparison with the binding energy of an exciton in a zinc-selenide single crystal (by a contribution factor of 4.1–72.6) was detected. It was shown that nanosystems consisting of zinc- selenide QDs grown in a borosilicate glass matrix can be used as the active region of semiconductor QD lasers [3, 4].

The results of variational calculation of the exciton ground-state energy $E_0(a)$ (spherical semiconductor QD of the radius a) [3, 4] are obtained for a nanosystem containing zinc-selenide QDs, synthesized in a borosilicate glass matrix, studied in the experimental work [6]. In the experimental work [6], borosilicate glass samples doped with zinc selenide with concentrations from $x = 0.003$ to 1%, obtained by the sol–gel method were studied. According to X-ray diffraction measurements, the average radii a of ZnSe QDs formed in the samples are within $a \approx 2.0$ – 4.8 nm. In this case, the values of a are comparable to the exciton Bohr radius $a_{ex} \approx 3.7$ nm in a zinc-selenide single crystal. At low QD concentrations ($x = 0.003$ and 0.06%), their interaction can be disregarded. The optical properties of such nanosystems are mainly controlled by the energy spectra of electrons and holes localized near the spherical surface of individual QDs synthesized in the borosilicate glass matrix.

In [6], a peak in the low-temperature luminescence spectrum at an energy of $E_1 \approx 2.66$ eV was observed at the temperature $T = 4.5$ K in samples with $x = 0.06\%$; this energy is lower than the band gap of a zinc-selenide single crystal ($E_g = 2.823$ eV). The shift of the peak of the low-temperature luminescence spectrum with respect to the band gap of the ZnSe single crystal to the short-wavelength region is $\Delta E_1 = (E_1 - E_g) \approx -165$ meV. The authors of [6] assumed that the shift ΔE_1 is caused by quantum confinement of the energy spectra of electrons and holes localized near the spherical surface of individual QDs and is associated with a decrease in the average radii a of zinc- selenide QDs at low concentrations ($x = 0.06\%$). In this case, the problem of the quantum confinement of which electron and hole states (the hole moving within the QD volume and the

electron localized at the outer spherical QD–dielectric matrix interface or the electron and hole localized in the QD volume) caused such a shift of the luminescence-spectrum peak remained open. Comparing the exciton ground-state energy ($E_0(a) - E_g$) [3, 4] with the energy of the shift in the luminescence-spectrum peak $\Delta E_1 \approx -165$ meV, we obtain the average zinc-selenide QD radius $a_1 \approx 4.22$ nm. The QD radius a_1 may be slightly overestimated, since variational calculation of the exciton ground-state energy can give slightly overestimated energies. The determined average QD radius a_1 lies within the range of the average radii of zinc-selenide QDs ($a \approx 2.0$ – 4.8 nm), studied under the experimental conditions of [6].

To apply semiconductor nanosystems containing zinc- selenide QDs as the active region of lasers, it is required that the exciton binding energy $|E_{ex}(a)|$ [2] in the nanosystem be on the order of several kT_0 at room temperature T_0 (k is the Boltzmann constant). Nanosystems consisting of zinc- selenide QDs grown in a borosilicate glass matrix can be used as the active region of semiconductor QD lasers [1]. In the range of zinc-selenide QD radii a , the parameter $|E_{ex}(a, \varepsilon)/kT_0|$ take significant values in the range from 3.1 to 56 [3, 4].

The effect of significantly increasing the binding energy of the exciton ground state in a nanosystem containing zinc-selenide QDs with radii a was detected; in comparison with the exciton binding energy in a zinc-selenide single crystal, the increase factor is 4.1–72.6 [3, 4]. It was shown that the effect of significantly increasing the binding energy of the exciton ground state in the nanosystem under study is controlled by two factors [3, 4]: (i) a substantial increase in the electron–hole Coulomb interaction energy and an increase in the energy of the interaction of the electron and hole with “foreign” images (the “dielectric enhancement” effect); (ii) spatial confinement of the quantization region by the QD volume; in this case, as the QD radius a increases, starting from $a \geq a_c^{(2)} \approx 29.8$ nm, the exciton becomes two-dimensional with a ground-state energy that exceeds the exciton binding energy in a zinc-selenide single crystal by almost two orders of magnitude.

In [7], one theoretically investigates the influence of a homogeneous magnetic field on the electron states localized by electrostatic image forces over the surface of an ellipsoidal germanium nanoparticle placed in vacuum. The effects of resonant interaction of light with such local electron states both in the absence and in the presence of homogeneous magnetic field were investigated for the first time. A new optical and a magneto-optical methods of diagnostics of nanostructures, allowing determination of dispersion degree of nanostructures were proposed.

Experimental and theoretical studies of physical characteristics of nanoparticles responsible for specific optical properties of dielectric nanocomposites. During theoretical description and explanation of experimental data obtained, we considered nanoparticle as a multi-particle quantum system of charges which combines elements of quantum structures of a polyatomic molecule and a bulk crystal. Considering nanocomposites with quite a low concentration of nanoparticles (fractions of a percent), we didn't take into account interparticle interaction [8,9].

The theoretical model of transmission spectrum of dielectric nanoparticles ensemble is presented. This model describes the dependence of the transmission spectrum on both of the particles size and the radiation frequency. This basis of the model is: the energy gap of the nanoparticle material contains energy levels, that is caused by the presence of a great number of defects of the nanoparticles crystal structure; the quantization of the charge carriers and excitons states. The theoretical model is compared with experimental results on the transmission spectrum [10].

Quasimolecules in quasi – zero - dimensional nanosystems

In [11] we developed the theory of a quasimolecule (biexciton) formed from spatially separated electrons and holes (the hole is in the semiconductor spherical QD volume, and the electron is localized at the outer spherical surface of the QD–dielectric matrix interface) in a nanosystem that consists of ZnSe QDs synthesized in a borosilicate glassy matrix [6]. Using the variational method, we obtained the total energy and binding energy of the quasimolecule (biexciton) singlet ground

state in such system as functions of the spacing between the QD surfaces and of the QD radius. We show that the quasimolecule (biexciton) formation is of the threshold character and possible in a nanosystem, in which the spacing between the QD surfaces exceeds a certain critical spacing [11]. It is shown that the major contribution to the quasimolecule (biexciton) binding energy is made by the energy of the exchange interaction of electrons with holes and this contribution is much more substantial than the contribution of the energy of Coulomb interaction between the electrons and holes. It is established that the spectral shift of the low-temperature luminescence peak [6] in such a nanosystems is due to quantum confinement of the energy of the biexciton ground state [11].

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