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On the Theory of Exciton States Polarizability in Open Spherical Quantum Dot

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We analyze theoretically the polarizability features in exciton's regime for an open spherical quantum dot. Based on the polarizability features the method of self-consistent calculation of the dielectric constant for nanoparticles array is proposed.

Keywords: Quantum вот, Exciton itates, Polarizability, Dielectric constant.

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1. INTRODUCTION

Recent advances in lithography, colloidal chemistry, and epitaxial growth have made it possible to manufacture artificial metamaterials and simple functionality devices for the optical and quantum information applications using the quantum dots (QD) [1-4]. In constructing the theoretical model of these objects it is necessary to consider the broadening of the energy levels related with a finite lifetime of the electron simultaneously with the taking into account the size-quantized effects.

In view of this reasons we present a simple model of an open spherical QD taking into account the size reduction of the static polarizability. Based on the polarizability features we propose a method of self-consistent calculation of the dielectric constant for quantum dots array embedded in gelatinous matrix.

2. MODEL

Examination of the properties of an open spherical QD begins with the solution of the Schrödinger equation within the effective mass approach, in which the boundary presents by the δ-potential [5]:

$$U(r) = \Theta \cdot \delta(r - r_0), \tag{2.1}$$

where r_0 is the radius of QD and Θ is the coefficient of penetrability.

The standard boundary conditions at $r = 0$ and asymptotic behavior of the Bessel and Neumann functions lead to following expression for the radial part of the wave function in spherical coordinates:

$$R_{kl}(r) = \begin{cases} C_1 j_l(k_1 r), & r < r_0, \\ D_1 h_l^{(1)}(k_2 r), & r \geq r_0. \end{cases} \tag{2.2}$$

The continuity conditions of the wave functions, the discontinuity of the first derivatives at the boundary of QD and the normalization condition give the transcendental equation:

$$\frac{m_2}{m_1}\frac{j_{l+1}(k_1 r_0)}{j_l(k_1 r_0)} - \frac{k_1}{k_2}\frac{h_{l+1}^{(1)}(k_2 r_0)}{h_l^{(1)}(k_2 r_0)} - \frac{2m_2\Theta}{k_1\hbar^2} + \frac{l}{k_1 r_0}\left(1 - \frac{m_2}{m_1}\right) = 0. \tag{2.3}$$

Here, l – orbital quantum number, m_1 and m_2 are the effective masses of the electron (or hole) in the QD and in the environment. Solution of equation (2.3) describes the quasi-stationary electron (hole) states.

Coulomb interaction between the electron and hole can be calculated as the matrix element

$$V_c = \left\langle \varphi_{nl}(\vec{r}_e) \left| \frac{e^2}{\varepsilon |\vec{r}_e - \vec{r}_h|} \right| \chi_{n'l'}(\vec{r}_h) \right\rangle, \tag{2.4}$$

where ε is the material's dielectric constant, $\varphi_{nl}(\vec{r}_e)$ and $\chi_{n'l'}(\vec{r}_h)$ are the wave functions of the electron and hole respectively. As a result, the energy spectrum of exciton with the band gap for bulk materials E_g has the form

$$E^{ex} = \tilde{E}_{nl}^{e} + \tilde{E}_{n'l'}^{h} - V_c + E_g. \tag{2.5}$$

Using the results presented above the optical properties of the structures under consideration can be investigated. In the case of the dipole transition in direct-gap semiconductor the static polarizability of the ground state of the particle in QD is given by

$$\alpha = \frac{e^2}{\mu}\sum_n \frac{F_{n'0}}{\omega_{n'0}^2}. \tag{2.6}$$

Here, $\mu = m_1 m_2 / (m_1 + m_2)$ is the reduced mass of exciton, $\omega_{n'0} = \omega_{n'} - \omega_0$ and $F_{n'0}$ is the oscillator strength determined by the dipole matrix element:

$$F_{n'0} = \frac{2\mu\omega_{n'0}}{\hbar}\left|\langle n'|\vec{r}_e - \vec{r}_h|0\rangle\right|^2. \tag{2.7}$$

Calculations were performed for the colloidal QDs of CdS in gelatin [6] with following physical parameters: a band gap $E_g(CdS) = 2.42$ eV of the bulk materials,

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the effective masses are $m_e / m_0 = 0.205$, $m_h / m_0 = 0.7$ (m_0 – mass of the free electron), and high-frequency dielectric constant $\varepsilon_b = 5.5$. In the case of gelatin the parameters are following: $m_e = 1$ and $\varepsilon_m = 2.2$ [7].

3. RESULTS AND DISCUSSION

The performance calculations show the interesting feature of the polarizability. Namely, the dimensionless polarizability (i.e., the polarizability (2.6) normalized to the third order of the linear size, namely $\alpha_0 = \alpha / r_0^3$) linearly decreases with the decreasing of the QD's radius (Fig. 1). Such a dependence correlates with the known experimental data [8].

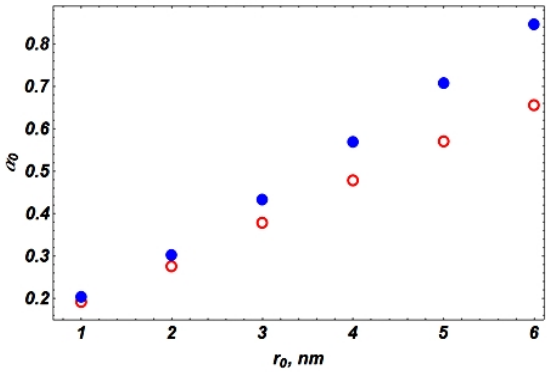

Fig. 1 – The dimensionless polarizability α_0 versus radius r_0 for ground state in open spherical QD (CdS) of single-particle electron (blue dots) (a), the exciton with $\varepsilon = 5.5$ (red circle) (b)

The obtained dependence is the result of size-quantized effect. Moreover, such a behavior of the polarizability will influence on the dielectric constant of the array of QDs.

Taking into account this effect and relationship for the effective dielectric constant

$$\varepsilon = \varepsilon_m \frac{1 + 2\vartheta\alpha_0}{1 - \vartheta\alpha_0}, \qquad (3.1)$$

(ϑ is the volume fraction of QDs) we developed a self-consistent method for calculation of the dielectric constant by the flow diagram in Fig. 2.

The initial step of iteration is the bulk dielectric constant ε_b. The iterative calculations were terminated when the variation of the dielectric constant remained to 10^{-4}. So, for the radius in range 1 – 3 nm the dielectric constant varies from 2.21 to 2.65, respectively.

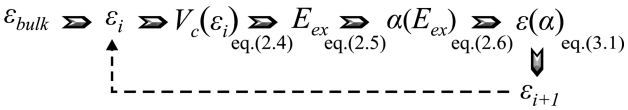

Fig. 2 – Flow diagram of the self-consistent calculation of the dielectric constant

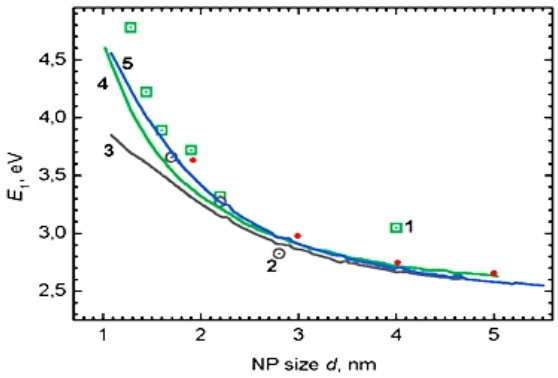

Fig. 3 – The energy of optical transition 1s-1s E_1 versus d for CdS nanoparticles in water derived from the experimental data and semiempirical models [9]

Let us note that the presented self-consistent calculation procedure allows to get the dielectric constant of an array of QDs, even when the initial step has no exact definition.

As an example, the energy of optical transition was calculated as a function of QD's size (Fig. 3, red dots). Deviation of the calculated E1 from the data of [9] is related with the different values of dielectric constant in water and gelatin.

4. CONCLUDING REMARKS

In this paper we have proposed a simple model of an open spherical quantum dot in exciton regime. Under this model we have shown that the dimensionless polarizability is proportional to the forth order of QD's radius, in contrast to the expected result, namely, that the polarizability of arbitrary quantum system is proportional to the third order of the linear size. Such a feature of the polarizability follows from the size-quantized effect.

Based on the obtained dependence, we have realized the method of self-consistent calculation of the dielectric constant. On the example of a CdS QDs array embedded in a gelatinous matrix the significant difference between the dielectric constant for nanoparticles and the bulk semiconductor material is demonstrated.

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