ISSN 1512-1461

UDC 539

DONOR IMPURITY IN QUANTUM DOTS CHARGE DENSITY DISTRIBUTION IN SPHERICAL NANOPARTICLE WITH CENTERED IMPURITY ATOM

T. Tchelidze, Z. S. Machavariani, B. Beradze

Department of Physics, Faculty of Exact and Natural Sciences, Ivane Javakhishvili Tbilisi State University, Chavchavadze Avenue 3,0179 Tbilisi, Georgia

e-mail: zaal.machavariani@tsu.ge

Abstract.

We investigate hydrogen impurity in spherical quantum dot. We calculated wave functions and corresponding energies of electron/hole confined in quantum dot containing centered donor/acceptor impurity. Calculations were carried out for different dot size and different materials, such as ZnO and CdSe, with different electron and hole effective masses. Electronic structure obtained by means of calculation defines as emission spectra. The calculation results can also reveal whether space confinement can alter doping (n- or p-) abilities of materials.

Keywords: Quantum dot, impurity atom, Nanoparticle

Introduction

Semiconductor quantum structures (QSs), or nanostructures, (quantum wells (QWs), quantum rods (QRs) quanrum dots (QDs) and nanowires (NWS)) are of great interest for both fundamental research and applications because of their size-dependent properties. They are very important optical materials with a wide range of potential applications due to their excellent physical and chemical properties among which is much higher brightness. Luminescence quantum yield is much improved in quantum structures, which is connected to increase overlap of carriers (electrons and holes) wave functions.

A vast number of investigations are carried out to study optical (emission) properties of low dimensional systems. In investigations, main attention is payed to size tunability of electronic spectra and size tunability of interparticle interaction [1-3].

With size reduction to nanometer donor and acceptor ionization energies change. Conductivity of samples depends on ionization energy of donors/acceptors and their compensation spices, as well as electronic band structure of the system. Therefore, the understanding of the dependence of electronic structure of Coulomb impurity on geometrical sizes of systems is crucial for its technological applications.

There has been a large interest in the study of Coulomb impurity in nanosized semiconductor structures of various shapes. A significant number of works has been devoted to the study of the behavior of Coulomb impurity in semiconductor nanoobjects, such as quantum dots, nanorods and nanowires. Obviously, the first attempt to study impurity states in a quantum well was made in [4]. In this work a variational calculation is carried out and the binding energy of donor (acceptor) has been calculated as a function of layer thickness and impurity position. This work, presumably,

ISSN 1512-1461

stimulated activity of theoreticians and in the following works the binding energies of hydrogen impurity in quantum dots [5,6], quantum wells [7,8] and quantum well wires [9-12] were calculated. The effect of applied electric and magnetic fields on binding energy was the subject of investigation in [13,14]. The common method that was employed in these works was a variational method. The calculations were carried out for both finite and infinite potential barrier. In the quoted works it has been revealed that the binding energy of impurity significantly depends on the geometry of nanosized object, location of impurity in nanoobject and external field. Main trend that is revealed is that in low dimensional structures strength of Coulomb interaction increases sharply, because of space and dielectric confinement.[15]

In the present paper, we investigate hydrogenic impurity in spherical quantum dot. We calculated wave functions and corresponding energies of electron/hole confined in quantum dot containing centered donor/acceptor impurity. Calculations were carried out for different dot size and different materials, such as ZnO and CdSe, with different electron and hole effective masses. Electronic structure obtained by means of calculation defines as emission spectra. The calculation results can also reveal whether space confinement can alter doping (n- or p-) abilities of materials.

Theoretical model

To solve the above mentioned problem the following Schrödinger equation is to solve:

$$\left[-\frac{\hbar^2}{2m^*_{e(h)}}\nabla^2-\frac{e^2}{|\mathbf{r}|}+V^{conf}_{e(h)}\left(\mathbf{r}\right)\right]\psi\left(\mathbf{r}\right)=E\psi\left(\mathbf{r}\right) \tag{1}$$

In the left-hand side of this expression the first term is kinetic energy of electron; the second term is Coulomb interaction potential energies of electron (hole) with positively (negatively) charged impurity, which is assumed to be in the center of quantum dot; $V^{conf}_{e(h)}\left(\mathbf{r}_{e(h)}\right)$ are confinement potential of electron(hole); $m^*_{e(h)}$ is an effective mass of electron (hole). For quantum dots of spherical shape these last two terms depend only radial coordinate. We consider potential well with infinitely high barrier, so confinement potential have the form:

$$V^{conf}_{e(h)}\left(r_{e(h)}\right)=\begin{cases}0, r<a\\ \infty, r>a\end{cases} \tag{2}$$

In spherical coordinates, radial and angular variables are separable in Eq (1). As in every problem of central symmetry, the solution of angular part are $Y\left(\theta,\varphi\right)$ spherical harmonics; As for radial part, it is non-zero only inside the dot ($0<r<a$), where radial equation has the form

$$\frac{d^2R\left(r\right)}{dr^2}+\frac{2}{r}\frac{dR\left(r\right)}{dr}+\frac{2m^*_e}{\hbar^2}\left[E-\frac{\hbar^2l\left(l+1\right)}{2m^*_er^2}+\frac{e^2}{r}\right]R\left(r\right)=0 \tag{3}$$

and boundary condition is $R\left(a\right)=0$. This equation is very similar to radial equation in hydrogen atom problem. The only difference is that radial wave function is zero at the boundary of spherical dot, however, the problem is very complicated. It could be solved analytically only for limiting cases, where dot radius a is very small [16] and very large [17].

ISSN 1512-1461

We solved this equation by means of shooting method [18]. This is a powerful method of solving one dimensional eigenvalue boundary value problems (BVP) of physics and mathematics, and in particular the radial Schrödinger equation of the form considered above. For this purposes we transform this equation by substituting $u(r) = rR(r)$, which gives:

$$\frac{d^2u}{dr^2} - v(r) = 0, \quad v(r) = -2\left(E - \frac{\hbar^2 l(l+1)}{2m_e^* r^2} + \frac{1}{r} \right) \tag{4}$$

where

$$u(0) = u(a) = 0. \tag{5}$$

The main idea is to reduce BVP on an initial value problem (IVP) in the following way. Equation (4) is second order differential equation, hence the two initial conditions are needed: the value of the eigenfunction $u(r)$ and its derivative du/dr at $r = 0$. The former one is given by BCs (5) to be zero and the latter may be chosen as an arbitrary nonzero number λ (the choice of this number is arbitrary in the sense that it must be not too large or small, which will be defined after all calculation by the normalization condition). Thus we have for initial conditions (ICs):

$$\begin{aligned} &u(0) = 0, \\ &\left.\frac{du}{dr}\right|_{r=0} = \lambda. \end{aligned} \tag{6}$$

Having fixed ICs, one can solve (or "shoot") the equation (4) with this conditions (by using for example Runge-Kutta 4th steps method) for some initial approximation of energy E_1, the target in $r = a$ and than check the second BC on this point

$$f(E_1) \equiv u_{E_1}(a) = 0. \tag{7}$$

After that, if this condition is not fulfilled, we must shoot one more time for some energy value $E_2 \neq E_1$ and again check the condition

$$f(E_2) \equiv u_{E_2}(a) = 0. \tag{8}$$

If E_2 does not matches the real eigenvalue, one must construct a transcendental equation for the energy eigenvalue

$$f(E_i) \equiv u_{E_i}(a) = 0, \tag{9}$$

where the new approximation in each step is found by the regula falsi method [19]:

$$E_i = E_{i-1} - f(E_{i-1}) \frac{E_{i-1} - E_{i-2}}{f(E_{i-1}) - f(E_{i-2})}. \quad i = 3, 4, 5.... \tag{10}$$

ISSN 1512-1461

It must be noticed that for each new approximation E_i of the energy one must solve the IVP (4) and (6) to find the target value of $f(E_i) \equiv u_{E_i}(a)$, then check (9) and find next approximation with (10) until (9) is fulfilled.

Results and discussion

At first glance the quantum numbers which describes the electronic configuration in quantum dot are pretty much similar to that of pure hydrogenic atom. The principle difference from hydrogenic case is that the main quantum number *n* is not determined analytically because there is no analytical solution of the corresponding radial equation which in turn provides the integrity of *n* as in case of pure hydrogenic atom. Since the equation (1) is again separable with respect of radial and angular variables the nodes of the radial wavefunctions and the orbital quantum number *l* are not related to each other. Therefore the good quantum numbers for characterizing of the electronic configuration in spherically symmetric quantum dot with centered impurity atom would be the nodes of radial wavefunction (in this paper we will denote it by *n*) and orbital quantum number *l* and its projection *m*. The modules squared of the wave functions corresponding to the first four lower states for electron bound to one charged donor in ZnO are presented in Fig.1. At the panels the total wavefunctions are presented.. In Fig. 2 Ionization energies of donor and acceptor impurities in ZnO and CdSe spherical quantum dots vs dot radius are presented. In Fig. 3 single particle energies for a few lowest states are listed for spherical quantum dot with impurity.

ISSN 1512-1461

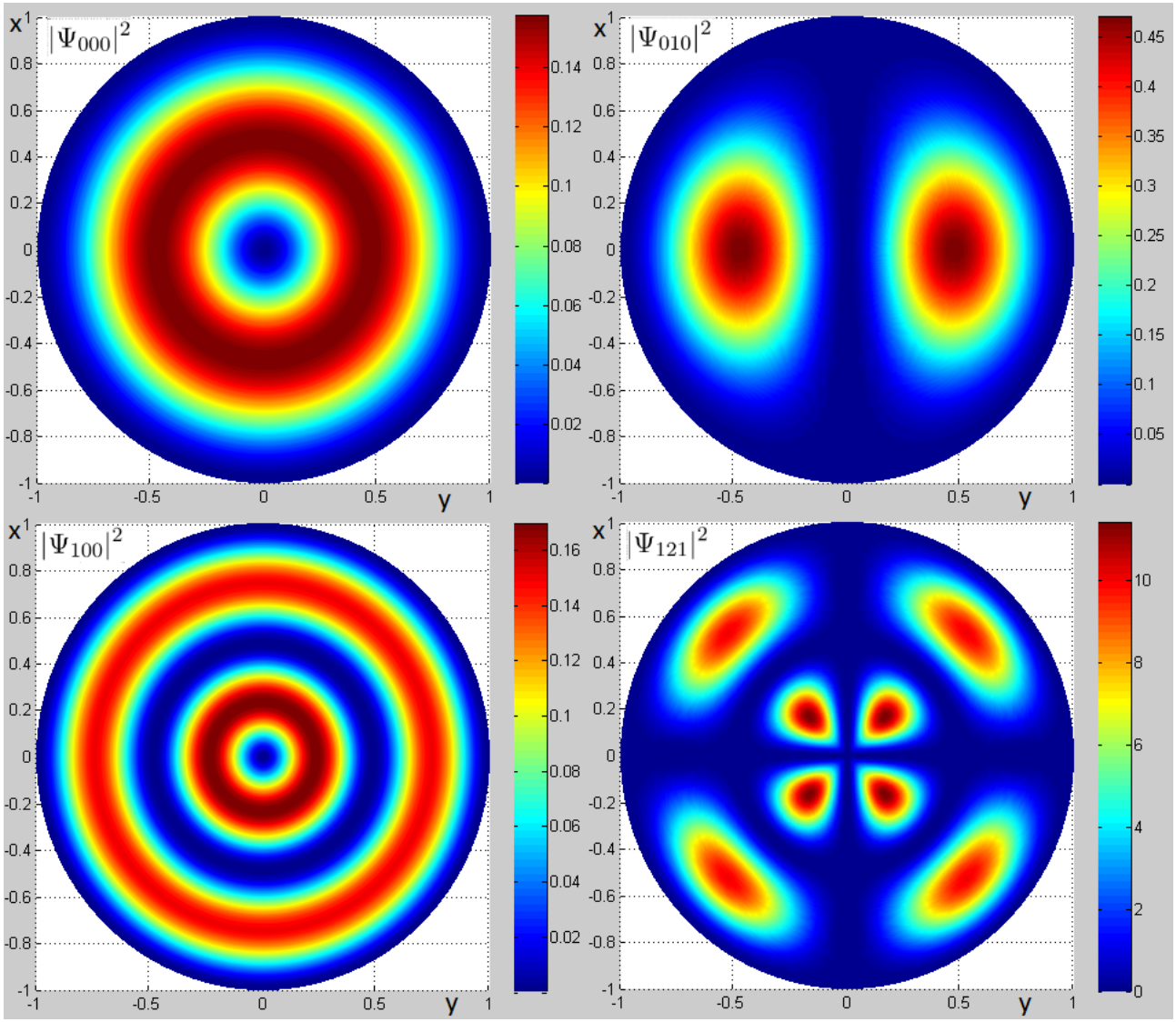

Figure 1. Wave function of the first four lower states bound to one charged donor in ZnO. The integer numbers in the subscript of the psi functions represents quantum numbers *nlm* as it is described in text.

We can see that binding energies are strongly size dependent. In CdSe, with electron and hole effective masses $0.13m_0$ and $0.45m_0$, respectively, donor and acceptor ionization energies are characterized with similar behavior with size reduction. However, in ZnO, with electron and hole effective masses $0.24m_0$ and $0.86m_0$, respectively, the situation is quite different: the ionization energy of acceptors drops to bulk value with increase of dot size much faster than ionization energy of donor. Ionization energy of acceptor in QD of 6 nm is almost the same as in bulk material, while ionization energy of donors exceeds bulk value even in QDs of 8 nm radius. This fact has got significant importance: there is a "window" (radius range 6-9 nm), where realization of p-conductivity should be easer than in bulk samples, as compensating donor becomes deeper. The fact is conditioned by big effective mass of holes in ZnO. The same picture is expected in other oxides, where obtaining of hole are problematic and holes usually have large effective masses.

ISSN 1512-1461

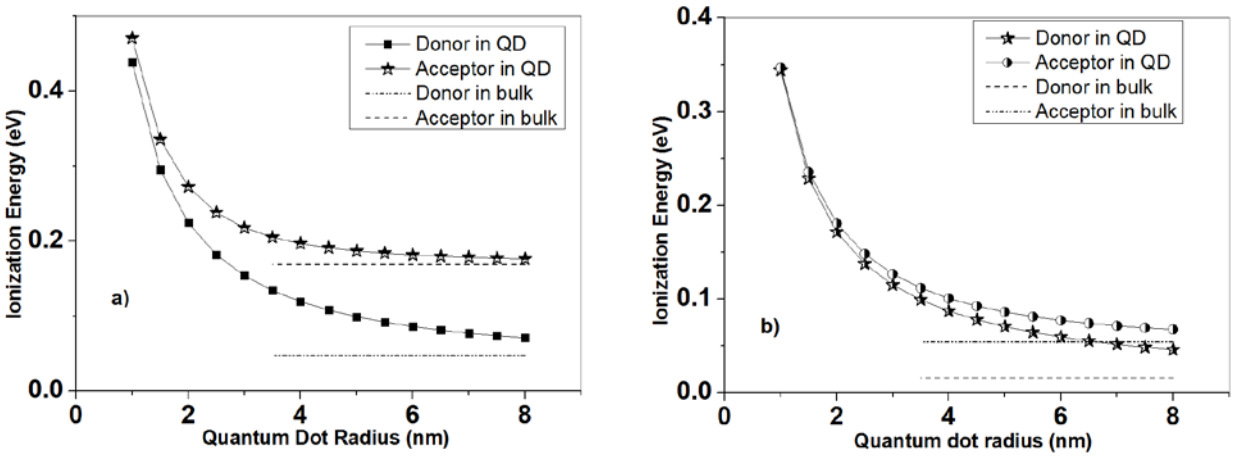

Figure 2. Ionization energies of donors and acceptors in ZnO (a) and CdSe (b).

As in any central potential problem the electron state energies are determined by use of two quantum numbers: *n* and *l*. In Fig. 3 there are expressed the dependence of first few lowest single particle energy states on spherical dot radius and on the quantum numbers. It shows that the *l*-degeneracy is cancelled in opposite to hydrogen atom, as it was expected, due to existence of strong confinement effects.

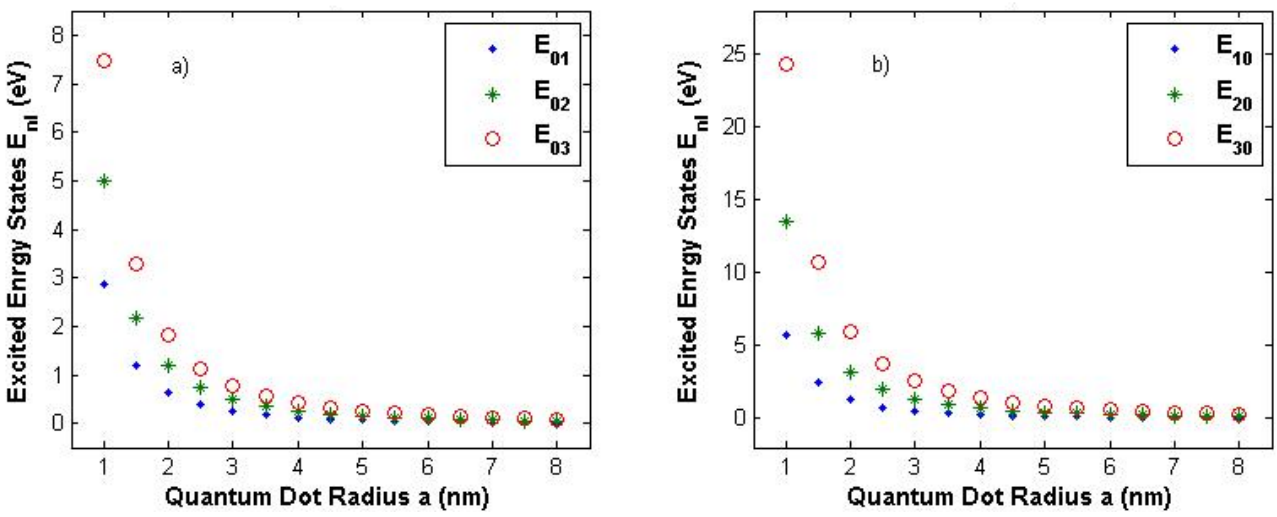

Figure 3. Energy states E_{nl} of electron bound to donor impurity in ZnO spherical quantum dots as a function of dot radius and quantum numbers a) *l* and b) *n*.

Acknowledgments

This work has been supported by joint research grant from STCU (Science and Technology Center in Ukraine, #6207) and SRNSF (Shota Rustavely National Science Foundation, Georgia, #04/04) Participation of Bachana Beradze has been supported by "Targeted research grant from TSU" ; He also wishes to thank the Professor Ramaz Bojorishvili for instructing and helpful discussion about the application of shooting method.

ISSN 1512-1461

References

1. I.Moreels, Y.Justo, B. De Geyter, K. Haustraete, J. C. Martins, and Z. Hens, *ACS Nano*, (2011) 5, pp 2004–2012
2. U. W. Pohl, A. Schliwa, R.Seguin, S. Rodt, K. Pötschke, D. Bimberg, *Advances in Solid State Physics*, (2008) 46 pp 45-58
3. R. E. Bailey and Sh. Nie *J*. Am. Chem. Soc., (2003) 125 (23), pp 7100–7106
4. G. Bastard, *Phys. Rev*. B (1981) 24, 4714
5. C. I. Mendoza, G. J. Vazquez, M. D. C. Mussot and H. Spector, *Phys. Rev.* B (2005) 71, 075330
6. J. L. Movilla, J. Planelles, *Phys. Rev.* B (2005) 71, 075319
7. S. Baskoutas and A. F. Terzis, *Physica* E (2008) 40, 1367
8. X. H. Qi, X. J. Kong and J. J. Liu, *Phys. Rev.* B (1998) 58, 10578
9. J. Lee and H. N. Spector, *J. Vac. Sci. Technol.* B (1984) 2, 16
10. G. W. Bryant, *Phys. Rev.* B (1985) 31, 7812
11. J. W. Brown and H. N. Spector, *J. Appl. Phys*., (1986) 59, 1179
12. E. Kasapoglu, H. Sari and I. Sökmen, *Physica* B (2007) 392, 213
13. M. Diarra, Y.Niquet, C. Delerue, G. Allan, *Phy. Rev*. B (2007) 75, 045301
14. H. Wang, L. Jiang, Q. Gong and S. Feng, *Physica* B (2010) 405, 3818
15. J.Giblin, F. Vietmeyer, M.P. McDonald, and M. Kuno, *Nano Lett*., (2011)11 (8), pp 3307–3311
16. W.Wilcox, O. V. Maxwell and K. Milton. Phys Rev D. 31, 1081 (1985)
17. W.Wilcox. Am. J. of Physics 57,526(1989)
18. Harrison, P. Quantum Wells, Wires and Dots. Wiley, Chichester, (2002), pp 73-76
19. J. Naghipoor, S. A. Ahmadian and Ali R. Soheili, Applied Mathematical Sciences, (2008) 2 (8), pp 381 – 386

Article received: 2017-05-25