Neighboring Levels Statistics and Shape of Quantum Dots: Si/SiO_2

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ABSTRACT

Spherical shaped Si quantum dots (QDs) embedded into the SiO_2 substrate are considered within the single sub-band effective mass approach. The electron and heavy hole sub-bands are taken into account. The energy dependence of electron effective mass is applied especially for small size QD. Calculations of low-lying single electron and hole energy levels are performed. For small sizes QD (diameter $D \leq 6$ nm) there is a strong confinement regime when the number of energy levels is restricted to several levels. First order perturbation theory is used to calculate neutral exciton recombination energy taking into account the Coulomb force between electron and heavy hole. The PL exciton data are reproduced well by our model calculations. For weak confinement regime (size $D \geq 10$ nm), when the number of confinement levels is limited by several hundred, we considered the statistical properties of the electron confinement. To determine the type of the nearest neighbor spacing (NNS) statistics, the distribution function is calculated. The influence of the QD shape on the NNS distribution is investigated.

Keywords: quantum dots and rings, single carrier levels, optical properties, excitons, Coulomb shifts

1 INTRODUCTION

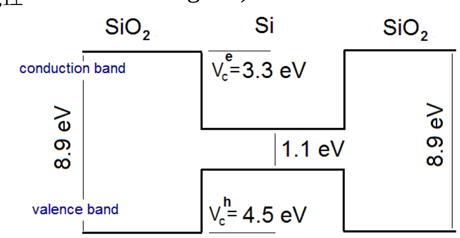
The Si/SiO_2 heterostructure has wide perspectives in the applications to the various optoelectronic nanodevices [1]. Theoretical study of the energy spectra properties of carriers confined in the Si/SiO_2 QDs is important for understanding the related electronic processes. A number of works devoted to the evaluation of the electron/hole energy levels [2] have been performed under the effective mass approximation using the envelope functions. In the present work we also use this method. However our approach is supplemented by energy dependence of electron effective mass [3] which is important for small size nanocrystals (diameter $D \leq 6$ nm). We calculate low-lying single electron and hole energy levels and neutral exciton recombination energy. The last results are compared with the PL exciton data [4-7]. Another application of the model is the study of the chaotic properties of quantum confinement. The chaotic properties of the quantum heterostructures play an important role in the various phenomena connected with the transport properties (i.e. conductance, Hall resistance, phonon scattering, optical properties etc.) The problem has not only theoretical interest but also technological importance due to the role of the shape of the heterostructures (e.g. hyperbolic versus rectangular or circular shapes of quantum dots) for the onset of Chaos. In Quantum Mechanics the chaos exhibits itself, for instance, in the NSS statistics with the shape of the GOE (Gaussian Orthogonal Ensemble). Since Quantum Dots are intrinsically quantum objects, the natural question arises concerning the statistics of the neighboring levels in QD. If these objects are not extremely small (> 10 nm for Si QDs) there is sufficiently enough number of levels to provide a good statistical analysis. In such analysis one can answer the question about importance of the QD shape, from the point of view of the onset of Chaos. To determine the type of NNS statistics, the distribution function is calculated. We study the influence of the QD shape on the NNS distribution using the Brody formula [8]. Change of the Brody parameter is related to different QD shapes. Obtained results are discussed.

2 MODEL

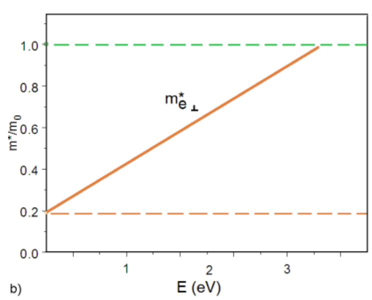
A Si/SiO_2 heterostructure is modeled utilizing a kp-perturbation single sub-band approach with an energy dependent quasi-particle effective mass [3]. The energies and wave functions of a single carrier in a semiconductor structure are solutions of nonlinear Schrödinger equation:

$$\left(-(\nabla, \frac{\hbar^2}{2m^*(r,E)} \nabla) + V(r) - E \right) \psi = 0, \qquad (1)$$

where $V(r)$ is the band gap potential, proportional to the energy misalignment of the conduction (valence) band edges of the Si QD (index 1) and the SiO_2 substrate (index 2). $V(r) = V_c$ (see below) is the potential inside the substrate, and $V(r) = 0$ inside the quantum dot. The electron effective mass $m^* = m^*(x, y, z, E)$ is linearly dependent on energy for $0 < E < V_c$ and varies within the limits of the QD/substrate bulk effective mass values [3]. V_c is defined as $V_c = \kappa\, (E_{g,2} - E_{g,1})$, where E_g is the band gap and the coefficient $\kappa < 1$ is different for conduction (CB) and valence (VB) bands. We use κ^{CB} =0.42 and κ^{VB} =0.58, Ref. [2]. For experimental values $E_{g,1}$=1.1 eV and $E_{g,2}$=8.9 eV, the band gap potential for the conduction band (valence band) is V_c=3.276 eV (V_c=4.524 eV). Bulk effective masses of Si and SiO_2 are $m^*_{0,1}$=($m_{0,1\perp}$=0.19 m_0, $m_{0,1\parallel}$=0.91 m_0) and $m^*_{0,2}$=1.0 m_0 respectively, where m_0 is the free electron mass. The value of $m^*_{0,1}$=0.34 m_0 is used for the effective mass of the heavy hole for Si QD and $m^*_{0,2}$=5.0 m_0 for substrate. The presented values applied as initial parameters for the Si/SiO_2 QD may change due to the effective mass dependence [3]. The band structure of the model is shown in Fig. 1a). The energy dependence of electron effective mass $m_{0,1\perp}$ is shown in Fig. 1b).



a)



b)

Fig. 1 a) Band structure for Si/SiO_2 QDs. b) Energy dependence of electron effective mass $m_{0,1\perp}$.

3 COMPARISONS WITH EXCITON PL DATA

In this section a comparison of results given by our model and PL experimental data is presented. For the Si/SiO_2 heterostructure the PL experimental data are available from [4-7]. We used first order perturbation theory to calculate neutral exciton recombination energy taking into account the Coulomb force between electron and heavy hole. The results of our calculation are shown along with the experimental data [4-7] in Fig. 2. The PL exciton data are reproduced well by our model. We also compare these results with those obtained within the model given in [2]. One can see that our model gives better description for the PL data than [2]. The difference of both results, presented in Fig. 2 is due to the non-parabolic effect that is taken into account in our calculations. In the asymptotical area when D>6 nm (when the non-parabolicity can be neglected) the results of both calculations coincide.

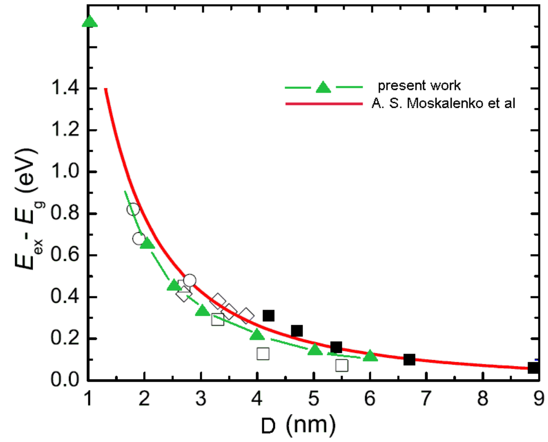
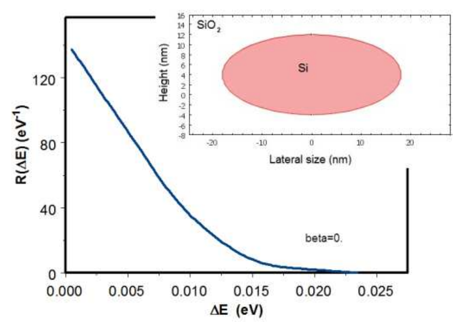


Fig. 2 Neutral exciton recombination energy E_{ex} of Si/SiO_2 spherical QDs. Calculated values are shown by triangles (which are connected by solid lines). The red solid curve is result of [2]. Experimental data are taken from [4-7] and are shown by various symbols.



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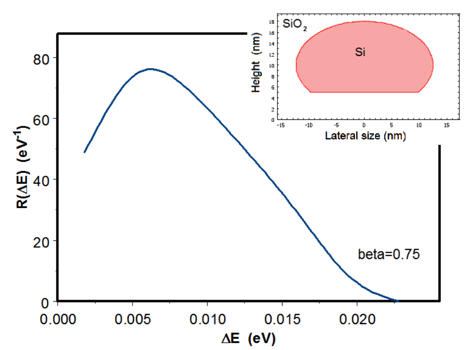

b)

Fig. 3 Distribution functions for electron NNS in Si/SiO_2 QD for different shapes a) and b). In the inset the QD cross section is presented. The Brody parameter (beta) is shown.

4 ELECTRON LEVEL STATISTICS

For the weak confinement regime (Si/SiO_2 QD diameter $D \geq 10$ nm), when the number of confinement levels is of the order of several hundred, we studied NNS statistics of the electron spectrum. The low-lying single electron levels are marked by E_i, $i = 0,1,...N$. One can obtain the set $\Delta E_i = E_i - E_{i-1}$, $i = 1,...N$ of energy differences between neighboring levels. We need to evaluate the distribution function $R(\Delta E)$, distribution of the differences of the neighboring levels. The function is normalized by $\int R(\Delta E) d\Delta E = 1$. For numerical calculation we define a finite-difference analog of the distribution function:

$$R_j = N_j / H_{\Delta E} / N \,,\; j = 1,...M \,, \qquad (2)$$

where $\sum N_j = N$ represents total number of levels considered, $H_{\Delta E} = ((\Delta E)_1 - (\Delta E)_N)/M$ is the energy interval which we obtained by dividing the total region of energy differences by M bins. N_j ($j = 1,...M$) is the number of energy differences which are located in the j -th bin.

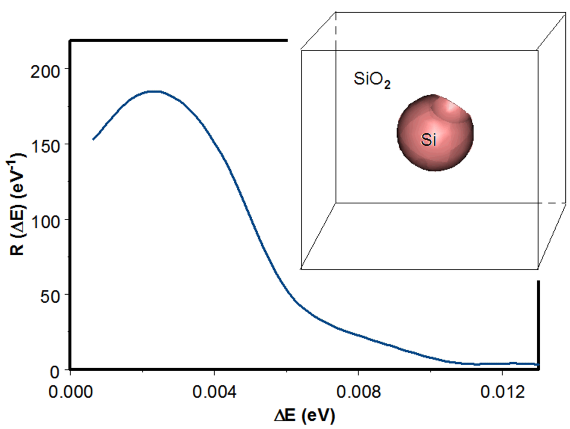

Fig. 4 Distribution functions for electron neighboring levels of the Si/SiO$_2$ spherical shaped QD with defect (a cavity). In insets the QD is shown in 3D. The Brody parameter β for this distribution is equal to 1.

We used the Brody distribution [10] to obtain fit for $R(\Delta E)$:

$$R(s) = (1+\beta) b s^{\beta} \exp(-b s^{1+\beta}), \qquad (3)$$

with the Brody parameter β and $b = (\Gamma[(2+\beta)/(1+\beta)]/D)^{1+\beta}$, D is the averaged level spacing. The Poisson distribution is realized when in (3) the Brody parameter is equal to zero.

The role of QD geometry with different shapes of boundaries has been studied in details previously (see e.g. [9]) within quasiclassical approach in 2D. Our results are related to the Si/SiO$_2$ QD considered in 3D. The distribution function calculated for ellipsoidal shaped QD is resented in Fig. 3a. The curve obtained by fitting the $R(\Delta E)$ calculated points gives the Poisson-like distribution. The curve of Fig. 3a (and all figures below) is obtained by the spline smoothing method for calculated values R_j (2).

For the case of QD with the shape that includes the break of the ellipsoidal symmetry (Fig. 3b), because of the cut below the major axis, we obtained a non-Poisson distribution. Figures 3 and 4 are examples of non-Poisson distribution for the QD with shapes that imply breaking of the discrete symmetry. In Fig. 4 the spherical symmetry (implies the Poisson statistics) is broken by semi-spherical cavity. The change in the statistics is indicated by the neighboring levels distribution function presented here.

For QD with more complex geometry the type of the QD levels statistics depends on existing "defects" which violate the discrete symmetry. In Fig. 5 presented are results of calculations for the distribution functions of NNS in QD having the form of quantum ring (QR) (cross section is shown in the inset) (Fig. 5a-b). The non- Poisson statistic that appears for QR with shape defect is shown in Fig. 5b.

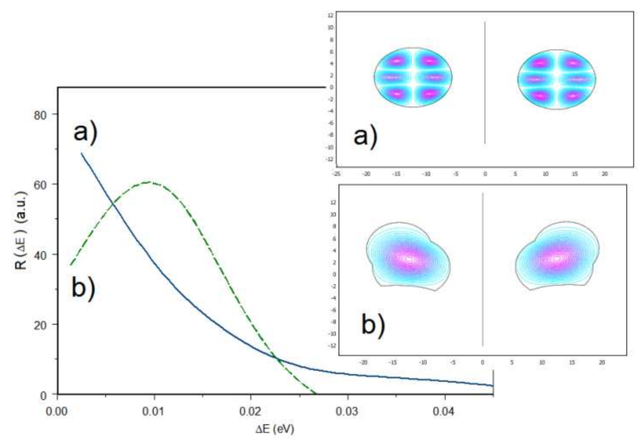

Fig. 5 Distribution functions for electron neighboring levels in Si/SiO$_2$ QRs. In insets the QR cross sections are shown (a) - "regular" QR, (b) - QR with defects. Electron wave function for 11-th exited state (a) and ground state (b) are shown.

It is reasonable to assume that these "chaotic" properties could affect electric and magnetic properties of QR.

The violated "regular" Poisson statistic can be reinstated by restoring the QD symmetry without fixing "defects". In Fig. 6 the distribution functions of QDs are shown for the non-symmetric and symmetric defects. In the first case the NNS statistics is found to be non-Poisson type, for the second it follows the Poisson distribution. This result is in agreement with the Ref. [10], in which the electron transport through non-symmetric and restored symmetry objects has been considered. In the last case the transport properties are improved essentially, compared with the first one. This could be considered as an evidence of change of the type of the statistics, from non-Poisson (chaotic) to Poisson (regular) statistics.

The double QD gives another example for regular statistics properties of the system, including QD pairs as an

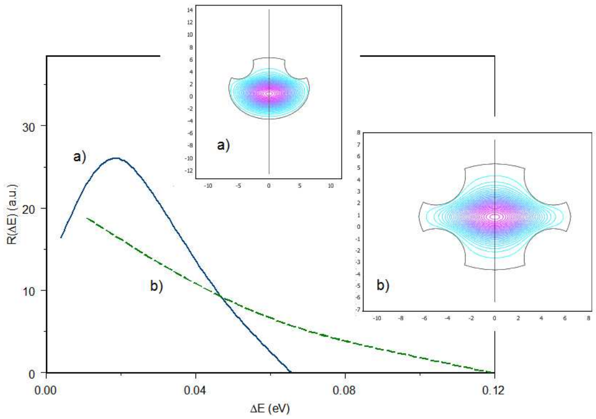

Fig. 6 Distribution functions (a-b) for electron NNS in Si/SiO_2 QDs. In insets the QD cross sections (a-b) and electron wave functions of ground state are shown.

isolated object, non-symmetric and with “chaotic properties”. This object could be constructed from two semi-spherical shaped QDs (with cuts such as shown in Fig. 3b) connected by thin bridge, as in Fig. 7 (in the inset). The level distribution of each independent QDs is non-Poissonian as it was in Fig. 3b. The connection by the bridge leads to the levels interference. In the case of the identical QDs we have global symmetry of the double QD shape. Corresponding NNS statistics is found to be the Poisson-like (Fig. 7a). Reducing the shape of one of the QDs violates the symmetry and the statistics of the double QD becomes non-Poissonian (see Fig. 7b). The repulsion of the levels can reflect the chaos in this system, which can have an impact on the transport properties of this system.

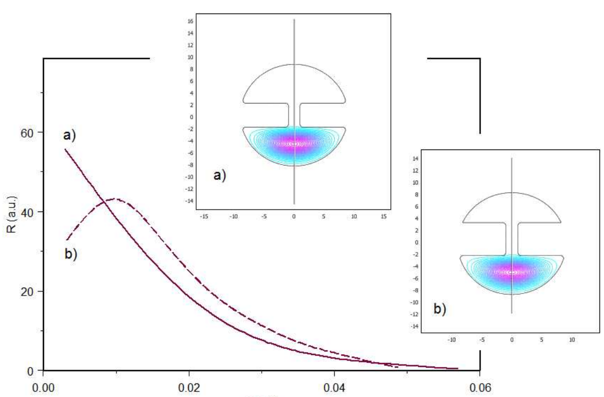

Fig. 7 Distribution functions (a-b) for electron NNS in double Si/SiO_2 QD. In insets (a-b) the QD cross sections and electron wave functions are shown.

5 SUMMARY

We found that the PL data of neutral exciton recombination energy of Si/SiO_2 spherical QDs are well reproduced by our model. The effect of non-parabolicity is important for the small size QDs.

We found that the deformations of shape of QD strongly affect the statistical properties of QD energy levels. The deviation of the shapes from the symmetric ones leads to the non-Poisson statistics, which may be a sign of the quantum chaos. This phenomenon, particularly, will influence the conductance and other transport properties of the QDs, and it may have technological implications.

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