A Computational Investigation of Electron Energy States for Vertically Coupled Semiconductor Quantum Dots

Yiming Li and Hsiao-Mei Lu*

National Nano Device Laboratories
Microelectronics and Information Systems Research Center, National Chiao Tung University
P.O. Box 25-178, Hsinchu 300, Taiwan, ymli@faculty.nctu.edu.tw
*Department of Bioengineering, University of Illinois at Chicago, Chicago, USA, hlu7@uic.edu

ABSTRACT

We study the electron energy state for single and vertically coupled quantum dots (QDs). Our realistic three-dimensional (3D) modeling for narrow gap semiconductor QDs considers: (1) the effective one electronic band Hamiltonian; (2) the energy- and position-dependent electron effective mass approximation; (3) a finite height hard-wall confinement potential; and (4) the Ben Daniel-Duke boundary conditions. A robust nonlinear iterative algorithm is applied to solve the model for disk- (DI-) and conical- (CO-) shaped QDs. For single QD, we find that the most stable against the dot size deviations (between dots of the same base radius) is the electron energy spectra of the CO-shaped QDs. For vertically coupled QDs with the fixed dot size, the energy spectra can be controlled by an inter-dot distance between two dots. Due to weak interaction of wavefunctions, electron energies of the CO-shaped coupled QDs are less dependent on the inter-distance than that of the DI-shaped coupled QDs. This investigation is related to optical spectra and useful in optoelectronics applications.

Keywords: 3D quantum dots; Artificial atom; $In_xGa_{1-x}As$; Electron energy spectra; Computer simulation.

1 INTRODUCTION

In recent years, there have been a number of experimental studies in semiconductor quantum dots (QDs), especially including coupled QDs (two or more coupled QDs form the so-called "artificial molecules") [1-10]. The electronic structures of these QDs can be controlled artificially with advanced nanotechnology [2]. These structures have strong quantum confinement and result in characteristic discrete energy spectrum and delta-liked density of states. The coupled QDs produce energy states tunneling effects, electronic entanglement, and charge transfer between dots [3,9]. Their rich properties enhance the physical interests and various applications [3-8], such as optoelectronic and information devices, cellular automata, and quantum computation. Theoretical studies in these artificial molecules have been done with 2D approximation, infinite and parabolic confinement potential models [3,4,11-15]. Unfortunately, these models have limitation in studying the geometric effects on energy structures for realistic 3D QDs. From device application point of view, it is necessary to consider 3D modeling and simulation for single and vertically coupled QDs system.

In this work, we study shape and inter-dot distance effects on electron ground and excited state energies for small single (Fig. 1) and coupled QDs. Fig. 2 shows that the different two shapes QDs, DI- and CO-shaped QDs, are separated by an inter-dot distance d in our investigation [5-9]. For the 3D single QD system, InAs is embedded into the matrix GaAs to form the InAs/GaAs QD. Similarly, the 3D vertically coupled QD system $In_xGa_{1-x}As$ QD is embedded into GaAs. Our 3D realistic model considers [16-20]: (1) an effective one electronic band Hamiltonian, (2) the energy- and position-dependent electron effective mass approximation, (3) finite hard wall confinement potential, and (4) Ben Daniel-Duke boundary conditions. The model is solved with a nonlinear iterative method. It was proposed by us for nanostructures simulations [16-18]. Our calculation shows that for single dot the electron ground and excited energy states depend on dot shape and volume significantly. For the same dot radius, we observe that energy variation of the DI-shaped QD is larger than that of the CO-shaped QD when the dot volume is increased. For vertically coupled QDs, the inter-dot distance plays an important role in varying the dot energy spectra. Due to strong wavefunctions interaction, it is easy to adjust the energy spectra of DI-shaped coupled QDs with inter-dot distance. For fixed volume and shape dots, the energy variation of DI-shaped coupled QDs is more sensitive than that of the CO-shaped coupled QDs when the inter-dot distance is reduced.

This paper is organized as follows. Sec. 2 states the 3D model and simulation technique. Sec. 3 shows the calculation results. Concluding remarks are in Sec. 4.

2 A 3D QUANTUM DOT MODEL AND COMPUTER SIMUATION TECHNIQUE

In the one-band envelop-function formalism, our unified 3D model for single and coupled quantum dots is considered in the electron Hamiltonian [19,20]:

$$H = -\frac{\hbar^2}{2}\nabla_{\mathbf{r}}\left(\frac{1}{m(E,\mathbf{r})}\right)\nabla_{\mathbf{r}} + V(\mathbf{r}), \qquad (1)$$

where $m(E, \mathbf{r})$ is the electron effective mass which depends on energy and position

$$\frac{1}{m(E,\mathbf{r})} = \frac{P^2}{\hbar^2}\left[\frac{2}{E + E_g(\mathbf{r}) - E_c(\mathbf{r})} + \frac{1}{E + E_g(\mathbf{r}) + \Delta(\mathbf{r}) - E_c(\mathbf{r})}\right] \quad (2)$$

and $V(\mathbf{r}) = E_c(\mathbf{r})$ is the confinement potential of quantum dots. $E_c(r)$, $E_g(r), \Delta(r)$, and P are the position dependent electron band edge, band gap, spin-orbit splitting in the valance band, and momentum matrix element, respectively [19,20]. For $In_xGa_{1-x}As$ systems with a sharp discontinuity of the conduction band on the interface between the quantum dots ($material_1$ and $material_2$) and the crystal matrix ($material_3$), $V(\mathbf{r}) = -V_{0x1}$, for $material_1$; $V(\mathbf{r}) = -V_{0x2}$, for $material_2$; and $V(\mathbf{r}) = 0$, for $material_3$. The dot material parameters are computed with [20]

$$V_{ox} = 0.7(E_{g3} - E_{gx}), \quad (3)$$

$$E_{gx} = xE_{gInAs} + (1-x)E_{gGaAs}, \quad (4)$$

$$\Delta_{gx} = x\Delta_{gInAs} + (1-x)\Delta_{gGaAs}, \text{ and} \quad (5)$$

$$\frac{1}{m_x(0)} = x\frac{1}{m_{InAs}(0)} + (1-x)\frac{1}{m_{GaAs}(0)}. \quad (6)$$

For material parameters, we can use Eqs. (3)-(6) for relatively small x. For large inter-dot distance x, a quadratic composition dependence should be deployed [20]. In this paper we concentrate on principal consequences of a 3D approach in modeling of electron states in artificial molecules. Further corrections will definitely result in a more accurate estimation. For example, strains in InGaAs/GaAs structures will slightly correct calculation results, but for electronic band of III-V semiconductor strains lead to relatively small changes in parameters and it can be adjusted by a proper selection of the parameters [11]. Integrating the Schrödinger equation with the Hamiltonian (1) along the direction perpendicular to the interface, we obtain the Ben Daniel-Duke boundary conditions for wavefunction $\Psi(\mathbf{r})$

$$\Psi_{material_{1,2}}(\mathbf{r}_s) = \Psi_{material_3}(\mathbf{r}_s) \text{ and} \quad (7)$$

$$\frac{\hbar^2}{2m(E,\mathbf{r}_s)}\nabla\Psi(\mathbf{r}_s) = C, \quad (8)$$

where C is an arbitrary constant and $\mathbf{r}_s$ is the system interface of $material_{1\text{-}3}$ and $material_{2\text{-}3}$.

We solve the quantum dot problem above in the cylindrical coordinate (R, ϕ, z) [17,18]. We note that the Schrödinger equation in our quantum dot model is a nonlinear equation in energy. To compute electron energy spectra and wavefunctions for single and vertically coupled quantum dots, we apply the nonlinear iteration scheme [17,18] to calculate final convergent solution of the model. The computational algorithm is outlined as follows: (i) set an initial energy E, (ii) compute effective mass $m(E,\mathbf{r})$ with Eq. (2), (iii) solve the model with Eqs. (1)-(8), and (iv) update the new energy and back to step (ii). If it converges, we stop the feedback iteration. In step (iii) the nonlinear Schrödinger equation is discretized with finite volume method, and the discretized equation forms a matrix eigenvalue problem. To handle and solve the QD models with complicated dot shapes, a novel accelerating solution scheme is further developed for the above nonlinear eigenvalue problem. It combines a balanced and shifted QR algorithm and implicitly restarted Arnoldi method. This method converges globally for all simulation cases and enables us to study very large scale 3D QD systems. For a convergent eigenenergy, the inverse iteration method is applied to calculate the corresponding eigenfunction.

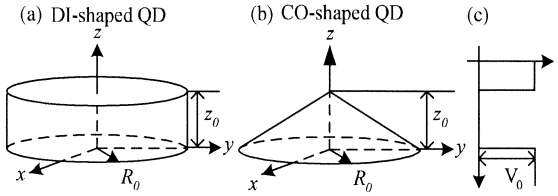

Figure 1: (a) DI-shaped and (b) CO-shaped single QDs. (c) Band offset of the single QD, where the $V_0 = 0.77$ eV.

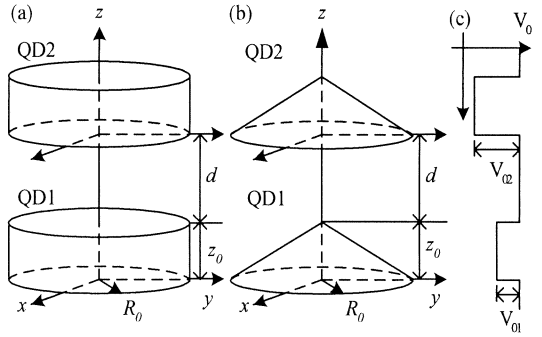

Figure 2: (a) DI-shaped and (b) CO-shaped vertically coupled QDs. (c) A plot of the system band offset, where the $V_0 = 0.77$ eV.

3 RESULTS AND DISCUSSION

Based on the nonlinear iterative method, we have successfully developed a 3D artificial molecules simulator and applied it to study QDs energy spectra. The material parameters used in our calculation are summarized briefly;

for InAs inside the ring, we have parameters E_g = 0.42 eV, Δ = 0.38 eV, and $m(0) = 0.024m_0$. For GaAs outside the ring, the material parameters are E_g = 1.52 eV, Δ = 0.34 eV, and $m(0) = 0.067m_0$. The band offset V_0 is equals to 0.77 eV.

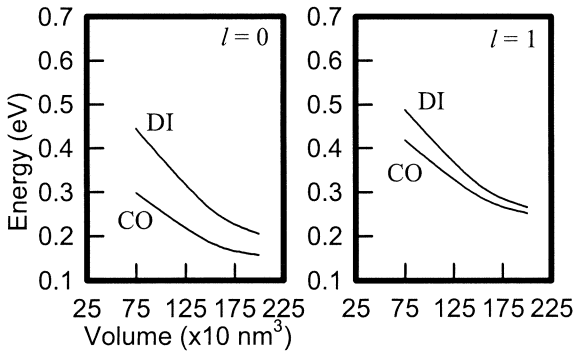

Figure 3: Electron energy states of InAs/GaAs QDs versus the dot volume. The left figure is the ground state energy and the right one is the first excited state energy.

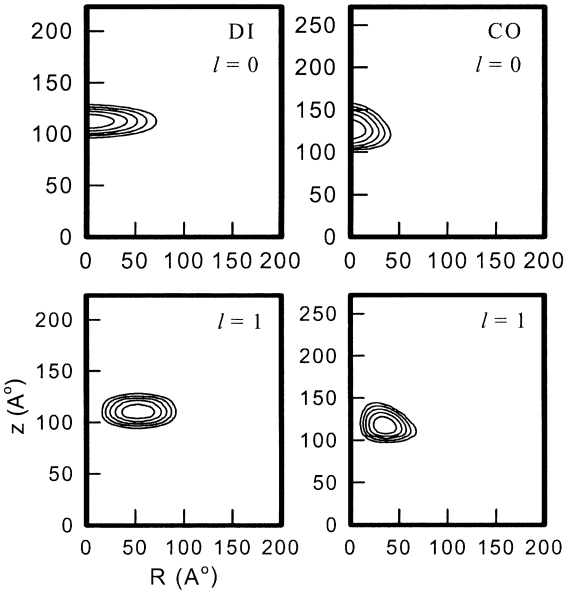

Figure 4: Contour plots of the electron ground state (upper two figures) and first excited state (lower two figures) wavefunctions.

As shown in Fig. 3, we firstly investigate the ground and excited state energy of single InAs/GaAs QD. The base radius of the dots is fixed at R_0 = 10 nm for these two shapes. Notice that the range of the dot volume and the radius of the base are taken from available experimental data [12,13]. When the dot volume increases the energy states of different shapes converge. The most sensitive to the dot volume variation is the quantum disks, and the least one is that of the conical shape dots. This is no surprise since the electron wavefunction is the best confined for the disk geometry when the volume and the radius are fixed. The electron ground state wavefunctions with a fixed 750 nm^3 dot volume for all shapes are plotted in Fig. 4. The wave function shape confirms weaker confinement for conical shaped dots. The first excited state ($l = 1$), however, has demonstrated a weaker sensitivity to the dot shape and volume [14,15] (see Fig. 3). This is because that the electron wave functions of the exited states (see Fig. 4) are less confined and, therefore, are less sensitive to the dot shape and size. We have found about 100 meV difference for the electron ground state energy in InAs/GaAs single dots of the same volume but different shapes.

The artificial molecule is complicated system than the single QD and enables us to manipulate system energy states with diverse ways. A complete picture of the energy and carrier transport in such high-density bi-modal system is still under investigation. We study here the electron energy spectra for DI- and CO-shaped vertically coupled $In_xGa_{1-x}As$ QDs by only considering adjustment of the inter-dot distance.

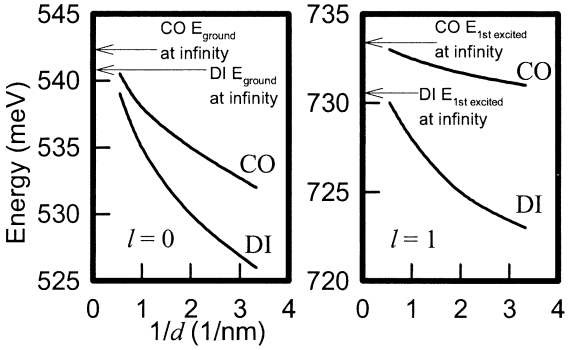

Figure 5: The electron ground (left figure) and first excited (right one) energy states versus reciprocal distance.

In coupled QDs systems, the dots are designed to have the same fixed radius R_0 = 7.8 nm and height z_0 = 1.8 nm. Firstly, when these two dots are totally separated, i.e., $d = \infty$, for both of the DI- and CO-shaped vertically coupled QDs, the QD1 is constructed with no any bounded states where x = 0.8 has been chosen. To form the QD2 that contains only few bounded states for DI- and CO-shaped coupled dots, we set x = 0 so that it has only two bounded states. Both of the DI- and CO-shaped coupled QDs have the ground state energy in the regime 540 meV. The 2nd state equals to 735 meV is near the barrier height of the system. Now, the designed two DI-shaped QDs are developed close to each other so that the system has a certain vertical distance $d < \infty$. we find that the system energy spectra can be significantly modified by the inter-dot distance d. It is dependent on not only the inter-dot distance but also QDs shape and dimension.

For DI- and CO-shaped vertically coupled $In_{0.8}Ga_{0.2}As$ dots, as shown in Fig. 5, both of the ground (left figure) and excited (right one) states are decreased when the inter-dot distance is decreased. This phenomenon is a result of the increasing electron wave function penetration into the other dot region when the inter-dot distance becomes small. The inter-dot coupling caused by wavefunctions overlapping may useful to create artificial molecules [9] that offer a different way to implement computing concepts [10]. Our result presents a relatively strong influence of the second dot potential well on the ground energy state for $d \approx 3$ nm.

Due to the stronger wave functions interaction, as shown in Fig. 6, the DI-shaped coupled QDs have larger ground state variation than that of CO-shaped QDs when the distance d is decreased. For the first excited states, we have similar but weaker interaction for both shapes. Energy levels of the coupled QDs can be further investigated experimentally with such as photoluminescence techniques

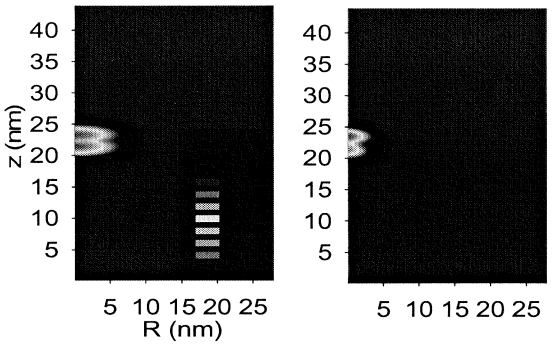

Figure 6: Ground state wave functions of the DI- (left figure) and CO-shaped (right one) vertically coupled QDs with $d = 0.8$ nm.

4 CONCLUSIONS

In conclusion, we have applied a 3D realistic model to investigate the energy states for single and vertically coupled QDs. Without any artificial fitting parameters, for a noncoupled single QD, when the dot volume increases, the energy stats of different shapes converge. We have found that the most sensitive to the dot volume variation is the ground state of the DI-shaped QD, and the least sensitive one is that of the CO-shaped QD. The first excited state shows a weaker sensitivity to the dot shape and volume. For the vertically coupled QDs system, we have also found energy spectra can be controlled and modified not only by structure shapes, sizes, and band parameters but also by the inter-distance of the dots. For DI-shaped artificial molecules, the electron energy state variation is stronger than that of the CO-shaped artificial molecules. The study is useful in studying optical properties of coupled dots and information device and system applications.

ACKNOWLEDGEMENTS

This work was partially supported by the National Science Council of Taiwan under contract number NSC 91-2112-M-317-001 and the 2002 Research Fellowship Award of the Pan Wen-Yuan Foundation in Taiwan.

REFERENCES

[1] M. Rontani, G. Goldoni, F. Manghi, and E. Molinari, Europhys. Lett. 58, 555, 2002.

[2] D. Bimberg, M. Grundmann, F. Heinrichsdorff, N. N. Ledentsov, V. M. Usitinov, A. E. Zhukov, A. R. Kovsh, M. V. Maximov, Y. M. Shernyakov, B. V. Volovik, A. F. Tsatsul'nikov, P. S. Kop'ev, Zh. I. Alferov, Thin Solid Films 367, 235, 2000.

[3] X. Hu and S. Das Sarma, Phys. Rev. A 61, 062301, 2000.

[4] J. L. Cheng and M. W. Wu, Solid State Commun. 124, 203, 2002.

[5] P. Yu, W. Langbein, K. Leosson, J. M. Hvam, N. N. Ledentsov, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, A. F. Tsatsul'nikov, and Yu. G. Musikhin, Phys. Rev. B 60, 16680, 1999.

[6] G. Sek, K. Ryczko, J. Misiewicz, M. Bayer, F. Klopf, J.P. Reithmaier, A. Forchel, Solid State Commun. 117, 401, 2001.

[7] F. Troian, U. Hohenester, and E. Molinari, Phys. Rev. B 65, 161301, 2002.

[8] Z. R. Wasilewski, S. Fafard, J.P. McCaffrey, J. Crystal Growth 201/202, 1131, 1999.

[9] R. Akis and D. K. Ferry, Phys. Rev. B 59, 7529, 1999.

[10] S. Benjamin and N. F. Johnson, Appl. Phys. Lett. 70, 2321, 1997.

[11] F. Guffarth, R. Heitz, A. Schliwa, O. Stier, N. N. Ledensov, A. R. Kovsh, V. M. Ustinov, and D. Bimberg, Phys. Rev. B 64, 085305, 2001.

[12] Y. Ebiko, S. Muto, D. Suzuki, S. Itoh, K. Shiramine, T. Haga, Y. Nakata, Y. Sugiyama, N. Yokoyama, J. Crystal Growth 201/202, 1150, 1999.

[13] P. M. Petroff, K. H. Schmidt, G. M. Ribeiro, A. Lorke, and J. Kotthaus, Jpn. J. Appl. Phys. 36, 4068, 1997.

[14] R. Leon, S. Fafard, P. G. Piva, S. Rusimov, and Z. Liliental-Weber, Phys. Rev. B 58, R4262, 1998.

[15] F. Heinrichsdorff, M. Grundmann, O. Stier, A Krost, and D. Bimberg, J. Crystal Growth 195, 540, 1998.

[16] Yiming Li, O. Voskoboynikov, C. P. Lee, and S. M. Sze, Solid State Commun. 120, 79, 2001.

[17] Yiming Li, O. Voskoboynikov, C. P. Lee, and S. M. Sze, Comput. Phys. Commun. 141, 66, 2001.

[18] Yiming Li, O. Voskoboynikov, C. P. Lee, and S. M. Sze, J. Comput. Elec. 1, 227, 2002.

[19] G. Bastard, "Wave Mechanics Applied to Semiconductor Heterostructures," Les Edition de Physique, Les Ulis, 1988.

[20] S. L. Chuang, "Physics of Optoelectronic Devices," John Wiley & Sons, New York, 1995.