Investigated optical studies of Si quantum dot

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Abstract

Further study of the quantum dot potential for Si is presented. This potential has been calculated by means of our recent empirical model. The indirect energy gap (Γ–X) is calculated using the full potential-linearized augmented plane wave (FP-LAPW) method. The Engel–Vosko generalized gradient approximation (EV-GGA) formalism is used to optimize the corresponding potential for energetic transition and optical properties calculations of Si. The refractive index and transverse effective charge are predicted as a function of dot diameter that is in turn used to test the validity of our model. The obtained results show a reasonable agreement in comparison with experimental data and theoretical results.

Keywords: Quantum dot; Si; Optical properties

1. Introduction

Semiconductor quantum dots (QDs) have gained increasing attention of scientists and engineers of various disciplines due to their flexible processibility and unique properties (Coe et al., 2003; Bruchez et al., 1998; Landes et al., 2001). For a number of optoelectronic applications, e.g., light-emitting diodes (LED) (Peng et al., 2000; Gerion et al., 2001), strongly luminescent semiconductor nanocrystals are highly desirable. However, due to the large surface-to-volume ratio of nanoparticles, the most common reason for poor luminescence efficiency is nonradioactive recombination of light-generated charge carriers at surface-traps (Sondi et al., 2004). It is fantastic to implement QD’s in solar cells for obtaining high efficiency and green energy applications. So, band gaps, optical constants, photoemission spectra, dielectric functions depending on frequency and wave vector, alloy properties, bonding and chemical properties, etc. have been evaluated for many materials using the pseudo potential approach (Al-Douri et al., 2008; Al-Douri, 2004b).

Different quantum dot solar concentrators are set in the literature (Kennedy et al., 2009; Gallagher et al., 2007a,b). Kennedy et al. (2009) have devoted the low optical efficiencies for single-plate quantum dot solar concentrators (QDSCs) are due to low luminescent quantum yields and large overlap between quantum dot (QD) emission and absorption spectra of present commercially-available visible-emitting QDs. Also, they showed that using near infrared (NIR) emitting QDs, re-absorption of QD emitted photons can be reduced greatly, thereby diminishing escape cone losses thus improving optical efficiencies and concentration ratios. It is shown that escape cone losses using Monte-Carlo ray-trace modelling account for ~57% of incident photons absorbed in QDSCs containing...
commercially-available visible-emitting QDs. Gallagher et al. (2007a) have described a novel; non-tracking concentrator, which uses nano-scale quantum dot technology to render the concept of a fluorescent dye solar concentrator (FSC) a practical proposition. They mentioned that the quantum dot solar concentrator (QDSC) comprises quantum dots (QDs) seeded in materials suitable for incorporation into building façades, and have found that photovoltaic (PV) cells attached to the edges convert direct and diffuse solar energy collected into electricity. Meanwhile, they have fabricated small scale QDSC devices. Also, Gallagher et al. (2007b) have been undertaken spectroscopic measurements for a range of different quantum dot (QD) types and transparent host materials, and proved that high transparency in the matrix material and QDs with high quantum efficiency is essential for an efficient QDSC. They determined an optimum matrix material for a QDSC based on absorption characteristics and an optimum commercially available QD type has been chosen using steady-state absorption, photoluminescence and photoluminescence excitation spectroscopy of QDs in solution and solid matrices.

Udipi et al. (1996) presented semiclassical simulation results for the potential energy profile and electron density distribution in 200 nm silicon quantum dot. For the solution of the continuity equation, the efficient difference approximations, proposed by Scharfetter and Gummel (1969) extended to three dimensions. In essence, they followed the two-dimensional approach due to Selberherr et al. (1980) extend two to three dimensions. Zhong and Liu (2009) have presented theoretical calculations of the scattering intensity for the Electron Raman Scattering (ERS) process associated with the bulk-like longitudinal optical (LO) and interface optical (IO) phonon modes in GaAs/Ga1−xAlxAs quantum dots (QDs), additionally to their study the selection rules for these processes and the singularities in the Raman spectra for various concentrations (x). Hasanean et al. (2004) had been described a model to simulate the electrical characteristics of nonvolatile floating gate quantum dot memory cells. They computed the tunneling rate of electrons using the transition-Hamiltonian of Bardeen and calculated the wave functions and potential energies of the quantum well channel and quantum dot gate using a self-consistent numerical solution of Schrödinger and Poisson equations. They showed that by changing the quantum dot charge, the resistor values can be changed by 40%. Experimentally, Schmidt et al. (1994) had been produced effectively buried quantum well dots on the basis of InGaAs/GaAs single quantum wells using high resolution electron beam lithography and selective wet etching of the top barrier. They generated dot structures with diameters down to 20 nm and maintained high luminescence efficiencies down to the smallest sizes. Also, they observed a blue shift of up to 9 meV for 23 nm quantum dots.

The investigation of new materials research is interesting when one tries to gain some information about the diameter dependence of the compounds. It seems more fundamental to relate the diameter dependence behaviour to the bonds between nearest atoms. By controlling the evolution with diameter dependence of the compound, it could attempt to link the effect of dot diameter to the quantum dot potential. In this context, we have used this procedure for testing the validity of our model (Al-Douri, 2009) of QD’s potential. The aim of this paper is to extend this method for calculating the diameter dependence on QD’s potential for dot diameters down to 52 nm using the full potential linearized augmented plane wave (FP-LAPW) and to investigate the optical properties of Si.

2. Calculation method

Quantum dot concentrator is presented by Chatten et al. (2004) experimentally and theoretically, and then their developing a self-consistent thermodynamic model for planar concentrators to find the three-dimensional flux model shows excellent agreement with experiment; was an impetus to use FP-LAPW method. It will be benefit to discuss DFT methods to generate the theoretical electronic properties as Al-Douri et al. (2010) and Al-Douri (2004a) have preformed. The calculations were carried out using the full potential linearized augmented plane wave (FP-LAPW) method as implemented in WIEN2K code (Blaha et al., 2001). It is one among the most accurate schemes for band structure calculations. In the FP-LAPW method, the unit cell is partitioned into non-overlapping muffin-tin spheres around the atomic sites and an interstitial region. Among these two types of regions different basis sets are used, the Kohn–Sham equation which is based on the density functional theory (DFT) (Hohenberg and Kohn, 1964; Kohn and Sham, 1965) is solved in a self consistent scheme. The exchange correlation potential was treated using the generalized gradient approximation (GGA) (Perdew et al., 1996) in which the orbital of Si (3s23p3) is treated as valence electrons for the total energy calculations. Moreover, the Engel and Vosko’s (EVGGA) formalism (Engel and Vosko, 1993) is used for electronic and optical properties calculations.

3. Results and discussion

Normally, the covalent semiconductors are fourfold coordinated. The reason that the density is so low and the nearest neighbors are bound by overlapping hybridized orbitals, which are the well-known sp3 hybrids with tetrahedral direction. Hence, it is possible to tune the band gaps using dot diameter. The calculated values of the direct (Γ → Γ) and the indirect (Γ → X) and (Γ → L) band gaps within EVGGA of the investigated Si-element at different diameters are listed in Table 1 along with the experimental data (Tsidilkovski, 1982) and other previous theoretical calculation (Humphreys et al., 1978). Our calculated value of the (Γ → X) band gap is slightly overestimated com-
The calculated principal energy band gaps for Si (in eV) at different diameters (in nm) compared to other theoretical results and experimental data.

<table>
<thead>
<tr>
<th>Dot diameter</th>
<th>$E_g(\Gamma-\Gamma)$</th>
<th>$E_g(\Gamma-X)$</th>
<th>$E_g(\Gamma-L)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>54.3</td>
<td>2.742</td>
<td>1.436</td>
<td>2.028</td>
</tr>
<tr>
<td>54</td>
<td>2.747</td>
<td>1.396</td>
<td>2.094</td>
</tr>
<tr>
<td>53.6</td>
<td>2.751</td>
<td>1.352</td>
<td>2.164</td>
</tr>
<tr>
<td>53.3</td>
<td>2.757</td>
<td>1.272</td>
<td>2.279</td>
</tr>
<tr>
<td>53</td>
<td>2.752</td>
<td>1.345</td>
<td>2.174</td>
</tr>
<tr>
<td>52.7</td>
<td>2.759</td>
<td>1.233</td>
<td>2.332</td>
</tr>
</tbody>
</table>

* Kohn and Sham (1965) Expt.
* Perdew et al. (1996).

The band gaps between the valence band maximum (VBM) at point $\Gamma$ and the conduction band minimum (CBM) at point $X$ are computed on the basis of the FP-LAPW. By means of our recent model (Al-Douri, 2009), the quantum dot potential has evaluated, according to the formula:

$$P_{QD} = \frac{b}{a}E_{gX} \cdot 10^{-3} \cdot \lambda$$

where $\frac{b}{a}$ is constant (in eV$^{-1}$) [see Table 4 in Al-Douri (2009)] and $\lambda$ is an appropriate parameter for group-IV ($\lambda = 6$), III–V ($\lambda = 4$) and II–VI ($\lambda = 2$) semiconductors (in V).

If diameter is decreased, the strong sp$^3$ covalent bonding that characterizes the covalent structure is affected. From our view point, this discrepancy at diameter dependence is an immediate consequence of the difference in the corresponding quantum dot potential. In Table 2, the calculated quantum dot potential at diameter dependence is computed. The critical dot diameter is the value that separates the decrease and the increase of the QD’s potential. The diameter dependence correlates with transition pressure ($P_t$) that is important to be computed from difference in molar free energies of compounds. The Gibbs free-energy difference $\Delta G_t$ between compounds which has the tetrahedral coordination at diameter dependence is nearly given by $P_t \Delta V/V_i$ (in kJ/mol). Most of energies are larger for smaller bond lengths. Decreasing the QD’s potential with dot diameter is confirmed by the change of the positions of the energy bands at principal points as shown in Table 1.

The QD’s potential increases as diameter increases until a critical value is reached, characterized by random behaviour (Table 2) and confirmed by Fig. 2. As a consequence, fluctuations of the QD’s potential appear. Our calculated QD’s values are in accordance with other (Udipi et al., 1996). Therefore, we may reach to the fact that the sudden variation of the QD’s potential is an indication of the control of tunneling electron across the quantum dot.

The refractive index $n$ is a very important physical parameter related to the microscopic atomic interactions. From theoretical view point, there are basically two different approaches of viewing this subject: the refractive index will be related to the density and the local polarizability of these entities (Balzaretti and da Jornada, 1996). Consequently, many attempts have been made in order to relate the refractive index and the energy gap $E_g$ through simple relationships (Moss, 1950; Gupta and Ravindra, 1980; Al-Douri, 2003; Al-Douri et al., 2008; Herve and Vandamme, 1993; Ravindra et al., 1979). However, these relations of $n$ are independent of temperature and incident photon energy. Here the various relations between $n$ and $E_g$ will be reviewed. Ravindra et al. (1979) had presented a linear form of $n$ as a function of $E_g$:

$$n = \alpha + \beta E_g$$

where $\alpha = 4.048$ and $\beta = -0.62$ eV$^{-1}$. Light refraction and dispersion will be inspired. Herve and Vandamme (1995) proposed an empirical relation as follows:

<table>
<thead>
<tr>
<th>Dot diameter</th>
<th>$P_{QD}$ Cal.</th>
<th>$P_{QD}$ (Udipi et al., 1996)</th>
</tr>
</thead>
<tbody>
<tr>
<td>54.3</td>
<td>1.051</td>
<td>1.051</td>
</tr>
<tr>
<td>54</td>
<td>1.021</td>
<td>1.021</td>
</tr>
<tr>
<td>53.6</td>
<td>0.989</td>
<td>0.989</td>
</tr>
<tr>
<td>53.3</td>
<td>0.931</td>
<td>0.931</td>
</tr>
<tr>
<td>53</td>
<td>0.984</td>
<td>0.984</td>
</tr>
<tr>
<td>52.7</td>
<td>0.902</td>
<td>0.902</td>
</tr>
</tbody>
</table>

Fig. 1. Calculated direct (\(\Gamma - \Gamma\)) and indirect (\(\Gamma - X\)) and (\(\Gamma - L\)) energy band gaps of Si as a function of dot diameter.
The calculated refractive indices for Si at diameter dependence using Ravindra et al. (1979), Herve and Vandamme (1995) and Ghosh et al. (1984) models corresponding to optical dielectric constant.

<table>
<thead>
<tr>
<th>Dot diameter</th>
<th>$n$</th>
<th>$\epsilon_\infty$</th>
</tr>
</thead>
<tbody>
<tr>
<td>54</td>
<td>2.347³ 2.4297⁹ 2.4441⁹</td>
<td>5.5084⁶ 5.9034⁹ 5.9736⁹</td>
</tr>
<tr>
<td>54</td>
<td>3.882⁴</td>
<td>11.68⁴</td>
</tr>
<tr>
<td>54.3</td>
<td>2.344⁴ 2.4296⁹ 2.4422⁹</td>
<td>5.4943⁹ 5.9029⁹ 5.9643⁹</td>
</tr>
<tr>
<td>53.6</td>
<td>2.342, 2.4266⁶ 2.4406⁶</td>
<td>5.484⁶ 5.8883⁹ 5.9563⁹</td>
</tr>
<tr>
<td>53</td>
<td>2.338⁵ 2.4240⁹ 2.4383⁹</td>
<td>5.466⁹ 5.8786⁹ 5.9453⁹</td>
</tr>
<tr>
<td>53</td>
<td>2.341⁹ 2.4263⁹ 2.4402⁹</td>
<td>5.480⁹ 5.886⁹ 5.954⁹</td>
</tr>
<tr>
<td>52.7</td>
<td>2.337² 2.4240⁹ 2.4335⁹</td>
<td>5.461⁹ 5.875⁹ 5.9219⁹</td>
</tr>
</tbody>
</table>

* Ravindra et al. (1979),
* Herve and Vandamme (1995),
* Ghosh et al. (1984),

Table 3

Fig. 2. Dot diameter dependence of the quantum dot potential for Si.

Fig. 3. Dot diameter dependence of the refractive index ($n$) for Si.

$$ n = \sqrt{1 + \left( \frac{A}{E_g + B} \right)^2} $$

where $A = 13.6$ eV and $B = 3.4$ eV. For group-IV semiconductors, Ghosh et al. (1984) have published an empirical relationship based on the band structure and quantum dielectric considerations of Penn (1962) and Van Vechten (1969):

$$ n^2 - 1 = \frac{A}{(E_g + B)} $$

where $A = 8.2E_g + 134$, $B = 0.225E_g + 2.25$; and $(E_g + B)$ refers to an appropriate average energy gap of the material. Thus, using these three models the variation of $n$ with dot diameter has been calculated. The results are displayed in Fig. 3. The calculated refractive indices of the end-point compounds are listed in Table 3. This is verified by the calculation of the optical dielectric constant $\epsilon_\infty$ which depends on the refractive index. Note that $\epsilon_\infty = n^2$ (Samara, 1983).

It is clear that the calculated $n$ using the model of Ghosh et al. is in accordance with the experimental value and due to reflectivity parameter is important in enhancing the photo conversion for solar cells. Again, a linear dependence of the Si properties on the dot diameter is observed and that the refractive index for small diameter dependence tends to shift towards the blue–green. It means a high absorption and low reflection spectrum may be attributed to increase solar cells efficiency.

In conclusion, the FP-LAPW method provides a good way to calculate the electronic properties, confirm its validity, investigate optical properties of low reflectivity value for IV compounds and proved that 53.6 nm dot diameter is more suitable for technological applications, expecting new trends for other compounds and new realization for quantum dot techniques.

Acknowledgments

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References


