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Study of dipole moment calculations for various transitions on polarization state of output radiation

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The electron and hole wave functions have been studied in the present paper using envelope function approximation. The hole Hamiltonian in a hexagonal crystal is obtained from the Luttinger Hamiltonian. The total angular momentum F and parity will be conserved leading to the mixing of states with same F value and equal parity. The values of dipole moments calculated for various transitions are discussed in the present paper. If a given transition is allowed in the dipole approximation with linearly polarized light, it cannot be allowed with circularly polarized light and vice-versa.

Keywords: Nanostructures, Quantum dots, II-VI semiconductors, Quantum solid, Spin dynamics, Level splitting, Interactions

1 Introduction

In the present scenario of quantum information processing, quantum dots (QDs) have gain impetus due to their long spin coherence time which allows control and manipulation of electron spin. QDs have these advantages because of the discrete density of states which arises due to the confinement in all the three dimensions. They are the most promising candidates in the growing field of quantum computing, quantum information processing devices as well as in optics[1]. Mainly, the optical properties of semiconductor nanocrystals gain considerable attention due to its applications in quantum dot lasers. Among II-VI semiconductor CdSe nanocrystals have intensively been studied by several groups with different approaches and aims. Recently, Aichele *et al*[2]. have reported photoemission from single CdSe quantum dots under pulsed excitation which act as a good source for quantum cryptography.

While reading and writing of messages, the polarization state of input radiation on the optical absorption and emission is very important. On the other hand, inter sub-band optical properties are equally important and drawing attention because of the large transition dipole moment which provides flexibility in tailoring the transition energy. In this regard Pazy *et al*[3]., have given the due emphasis on the polarization property of the input radiation for quantum computing. One another group[4] has also suggested the use of circularly polarized light for the reading and writing of the spin state of the resident electron in *n*-doped InAs-GaAs quantum dot. They have suggested the quantum jump technique for the reading of the spin state. Working in the same line, very recently Bandhyopadhyay[5] has reported the advantage of using the spin polarization of the single electron to encode a binary bit of information over that of the charge devices. The motivation arises because of the large spin coherence time in solids as compared to the charge coherence time. While measuring the exciton spin relaxation time in QDs Mackowshi *et al*[6]. have emphased on the polarization property of the excited as well as the emitted radiation.

The shape and the size of the quantum dot also have large impact on the transition energies and the polarization selection rules[7-10]. The structure of most of the II-VI compounds shows anisotropy which leads to asymmetry in optical properties and hence exhibits different output polarization corresponding to emission and absorption between different energy levels. Interior bonding geometry of the lattice structure has large effect on the energy levels which results in the energy shifts. For the clear understanding of the real QD picture, one should take into account electron and hole spins into consideration.

The transient characteristics of interband transitions have been explored by keeping in mind that these will help for ultra fast optical information processing. Giving proper emphasis on the polarization properties of emitted radiation in a semiconductor QD under the strong confinement regime, the polarization dependent dipole moment for various interband transitions has been calculated. At the same time, the probable transition schemes have also been investigated.

We have neglected the Coloumbic interaction because in a strong confinement regime it just acts as a perturbation and the carriers can be treated independently, thereby neglecting the excitonic effects. The quantitative descriptions of the absorption/emission of radiation have to take into account the real band structure of the nanostructure. The proper incorporation of valence band structure is not only important from the point of view of transition energies, oscillator strength but it also accounts for the various possible polarization output relative to the semiconductor dot structure. Thus in order to get the real picture of the optical transitions we have taken the multiple valence band structure of CdSe quantum dot. The valence band which arises from p atomic orbitals has an inherent six fold degeneracy at $k = 0$ (including spin) and is split by spin-orbit coupling into a fourfold degenerate band ($J = 3/2$) which comprise of heavy hole and light hole bands and a two-fold degenerate band ($J = 1/2$). As the spin-orbit splitting is very large, so the transitions from split-off band have not been considered.

2 Theoretical Formulation

Quantum dots have discrete electronic spectra with wave functions confined by the dot boundary and delocalized within the dot. The electron and hole wave functions are described by the product of unit cell basis function and envelope function satisfying the spherical boundary condition. The hole Hamiltonian in a hexagonal crystal is obtained from the Luttinger Hamiltonian. Each state of the Hamiltonian describing the free holes in a spherical potential is characterized by the total angular momentum quantum number F (where $F = L + J$). Thus, the envelope function angular momentum (L) and the unit cell angular momentum (J) are not good quantum numbers. Only total angular momentum F and parity will be conserved leading to the mixing of states with same F value and equal parity.

The wave functions for hole have the form given by:

$$\psi_{F,M,I} = \sqrt{2F+1}\sum_{\ell_I} R_{\ell_I}(r)(-1)^{M+\ell_I \mp 3/2} \sum_{m+\mu=\mu} \begin{bmatrix} 3/2 & \ell_I & 3/2 \\ \mu & m & -M \end{bmatrix} Y_{\ell,m} u_\mu \quad , \qquad \ldots(1)$$

where ℓ_I = F–3/2, F+1/2 for odd solutions and F+3/2, F–1/2 for the even solutions, here μ = ± 3/2, ±1/2. In Eq. (1) $\begin{bmatrix} 3/2 & \ell_I & 3/2 \\ \mu & m & -\mu \end{bmatrix}$ is the Wigner 3j symbols and u_μ are the bloch functions of the degenerate valence band.

The radial part in Eq. (1) is described by :

$$R_0(r) = \frac{A}{a^{3/2}} \begin{bmatrix} j_0(kr/a) \\ -\dfrac{j_0(k)}{j_0(k\beta^{1/2})} j_0(k\beta^{1/2} r/a) \end{bmatrix}. \qquad \ldots(2)$$

where, $k = a\sqrt{2m_{11A}E_h}/\hbar$ and $\beta = m_{11B}/m_{11A}$ is the ratio of light to heavy hole masses (here m_{11A}, m_{11B} are the effective hole masses which are related to luttinger parameter) and a is the dot radius. j_0's and Y_{lm} are the spherical Bessel functions and spherical harmonics, respectively. A is the normalization constant given by:

$$A^2 = 1/\int dx\ x^2 \begin{bmatrix} \left\{ j_2(kx) + \dfrac{j_0(k)}{j_0(k\beta^{1/2})} j_2(k\beta^{1/2}x) \right\}^2 \\ + \left\{ j_0(kx) - \dfrac{j_0(k)}{j_0(k\beta^{1/2})} j_0(k\beta^{1/2}x) \right\}^2 \end{bmatrix} \qquad \ldots(3)$$

While for the particular case of CdSe nanostructure β = 0.1 <<1, which reduces the radial function given in Eq. (2) as:

$$R_0 = c[j_0(\phi_2 r/a) - j_0(\phi_2)] \qquad \ldots(4)$$

where, $c = 6.044/a^{3/2}$. The bloch functions for the heavy and light hole valence bands are represented as:

$$u_{3/2} = \frac{1}{\sqrt{2}}(X + iY)\uparrow$$

$$u_{-3/2} = \frac{i}{\sqrt{2}}(X - iY)\downarrow$$

$$u_{1/2} = \frac{1}{\sqrt{6}}[(X + iY)\downarrow -2Z\uparrow]$$

$$u_{-1/2} = \frac{i}{\sqrt{6}}[-(X - iY)\uparrow +2Z\downarrow]. \quad \ldots(5)$$

The wave function of the electron ground is given by:

$$\psi_e\alpha(r) = \left[\frac{2}{a}\right]^{1/2} \frac{\sin(\pi r/a)}{r} Y_{00} \mid S\alpha >= (r)Y_{00}S\alpha > \ldots(6)$$

where |Sα> are the Bloch functions of the conduction band and α describes the electron spin projection (α = ↑ or ↓).

The transition probability is proportional to the squared matrix element of the operator $\varepsilon\hat{p}$ between the electrons and the hole wave functions, where ε is the polarization vector which denotes the orientation of the linearly polarized electric field. The scalar product $\varepsilon\hat{p}$ can be expanded for linearly polarized light as:

$$\varepsilon\hat{p} = \varepsilon_z\hat{p}_z + \varepsilon_-\hat{p}_+ + \varepsilon_+\hat{p}_- \quad \ldots(7)$$

where z is the direction of the hexagonal axis of the crystal. The expressions of $\varepsilon_\pm$ and $p_\pm$ are given as:

$$\varepsilon_\pm = (\varepsilon_x \pm i\varepsilon_y)/\sqrt{2} \text{ and } \hat{p}_\pm = (\hat{p}_x \pm i\hat{p}_y)/\sqrt{2}. \quad \ldots(8)$$

Thus, we obtained :

$$P_{M,\alpha} = \left|\langle\psi_\mu \mid \varepsilon\hat{p} \mid \psi_e\alpha\rangle\right|^2 = k\left|\langle u_\mu \mid \varepsilon\hat{p} \mid S\alpha\rangle\right|^2 \quad \ldots(9)$$

where, K is the overlap integral squared represented as:

$$K = \left|\int drr^2 f(r)R_0(r)\right|^2. \quad \ldots(10)$$

The overlap integral describes the strength of the transitions. It is independent of the type of atom and determines the lifetime of the excited state. While the angular part determines the $\Delta\ell$ and Δm_ℓ selection rules viz for linearly polarized light $\Delta m_\ell = 0$ (π transition) while for $\Delta m_\ell = \pm 1$, the output is right /left hand circularly polarized ($\sigma_\pm$ transitions).

The transition probability has been calculated for various transitions from multiple valence band states to the conduction band. The transition for the hole state with M = 3/2 and the electron state with spin up is found to be:

$$\left|\langle S\uparrow \mid \varepsilon_+\hat{p}_- \mid 3/2\rangle\right|^2 = \frac{K(\varepsilon_x^2 + \varepsilon_y^2)}{8}$$
$$\times\left[\left\{\langle S\uparrow \mid \hat{p}_x \mid x\uparrow\rangle + \langle S\uparrow \mid \hat{p}_y \mid y\uparrow\rangle\right\}\right]$$
$$= \frac{2}{9}K(\varepsilon_x^2 + \varepsilon_y^2) = \frac{2}{9}K\sin^2\theta \quad \ldots(11)$$

where $\langle S\uparrow \mid \hat{p}_x \mid X\rangle$ is the Kane interband matrix element and θ is the angle between the light polarization and the hexagonal axis of the crystal.

The expressions for various transitions are obtained as:

$$K\left|\langle S\downarrow \mid \varepsilon_-\hat{p}_+ \mid -3/2\rangle\right|^2 = \frac{13}{288}K\sin^2\theta \quad \ldots(12a)$$

$$K\left|\langle S\uparrow \mid \varepsilon_z\hat{p}_z \mid 1/2\rangle\right|^2 = \frac{8}{27}K\cos^2\theta \quad \ldots(12b)$$

$$K\left|\langle S\downarrow \mid \varepsilon_+\hat{p}_- \mid 1/2\rangle\right|^2 = \frac{13}{216}K\sin^2\theta \quad \ldots(12c)$$

$$K\left|\langle S\downarrow \mid \varepsilon_z\hat{p}_z \mid -1/2\rangle\right|^2 = -\frac{8}{27}K\cos^2\theta \quad (12d)$$

and $K\left|\langle S\uparrow \mid \varepsilon_-\hat{p}_+ \mid -1/2\rangle\right|^2 = \frac{-4}{486}K\sin^2\theta$ (12e)

It should be noted that the direct optical excitation and recombination of $\left|S\uparrow, -3/2\right\rangle$ and $\left|S\downarrow, 3/2\right\rangle$ states are forbidden by the selection rules. These states could recombine only with the emission or absorption of phonons.

The dipole moments which is directly proportional to the probability of transition are calculated from overlap integral of wave functions for the various allowed transitions by using the relation:

$$\mu = \frac{eP_{ab}}{m\omega} \quad \ldots(13)$$

The polarization state properties of the output radiation for the various optical transitions have been investigated. For this coherent ultrafast radiation-

matter interaction model based upon the semi-classical density matrix technique is employed.

3 Results and Discussion

The present theoretical analysis is applied to the CdSe QD and the parameters of interest are chosen as: bandgap frequency (ω_g)= 2.804×10^{15} s^{-1}, light hole mass (m_{lh}=)$0.31m_0$, heavy hole mass (m_{hh})=$0.45m_0$ and dot size = 45Å.

The various values of dipole moments calculated for different transitions are presented in Table 1. This shows that the dipole allowed transitions between the bands with $J = 1/2$ to $J' = 3/2$, $J = -1/2$ to $J' = -3/2$, $J = -1/2$ to $J' = 1/2$ and $J = 1/2$ to $J' = -1/2$ are associated with the emission of circularly polarized radiation and also the circularly polarized light coupled to these transitions. While the transitions between $J = 1/2$ to $J' = 1/2$ and $J = -1/2$ to $J' = -1/2$ energy levels lead to the emission of linearly polarized radiation which can be decoupled into two opposite polarized circular field is coupled to all the transitions. It is worth mentioning that if a given transition is allowed in the dipole approximation with linearly polarized light, it cannot be allowed with circularly polarized light and vice-versa.

Leistikow *et al*[10]. in their study the size-dependent quantum efficiency and oscillator strength. It is found that the quantum efficiency decreases with increasing emission energy mostly due to an increase in non radiative decay. Similarly our study gives the information of the dipole moment calculation and the polarization states of the important class of CdSe quantum dots. The dipole moment calculations show that it vary weakly with frequency in agreement with behaviour of quantum dots in the strong confinement limit. Our results are relevant for applications of CdSe quantum dots in spontaneous emission control and cavity quantum electrodynamics.

The crystal properties of semiconductors and nanocrystals focusing on theoretical results obtained within the multi-band effective mass approximation. A comparison of the dipole moment calculations with the available experimental values shows these results are valid for nanocrystals down 40-45Å in diameter. The effect of the transition dipole moment variation on the output polarization state in the optical spectra is observed similar to the experimental. Similar calculations for the asymmetric quantum well had been carried out by Bohórquez *et al*[11]. Several recent researchers such as Sen *et al*[12]. and Flissikowski *et al*[13]. used the similar type of dipole moment calculations for the study of hyperfine splitting in CdSe semiconductor quantum dots.

Table1 — Dipole moments calculated for different transitions

Transition	Dipole moment (in cm)	Polarisation state of output radiation	Transition Frequency (ω_{ab}) s^{-1}
$S\uparrow \rightarrow 3/2$	2.477×10^{-29}	LHCP (σ_-)	1.929×10^{14}
$S\downarrow \rightarrow -3/2$	0.112×10^{-28}	RHCP (σ_+)	do
$S\uparrow \rightarrow 1/2$	0.19×10^{-28}	LP (π)	1.655×10^{14}
$S\downarrow \rightarrow 1/2$	0.138×10^{-28}	LHCP (σ_-)	do
$S\downarrow \rightarrow -1/2$	0.163×10^{-28}	LP (π)	do
$S\uparrow \rightarrow -1/2$	0.1×10^{-28}	RHCP (σ_+)	do

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