Optical nutation in GaAs/AlGaAs core-shell quantum dot

R Solanki¹,³, P Sen¹ and P K Sen²

¹ School of Physics, Devi Ahilya University, Khandwa Road, Indore (M.P.) 452001, India
² Department of Applied Physics, Shri G S Institute of Technology and Science, Park Road 23, Indore (M.P.) 452003, India
³ Corresponding author E-mail address: er.ravisolanki.80@gmail.com

Abstract. Optical nutation in GaAs/Al₀.₃Ga₀.₇As core-shell quantum dot (CSQD) shined by femtosecond laser has been analyzed theoretically. The nutation signal intensity, arising due to photo-induced electronic transitions between ground state and first excitonic state, has been calculated using time dependent perturbation theory and rotational wave approximation. The dependency of the properties of optical nutation signal viz. its maximum amplitude and the oscillation frequency of the nutation signal has been studied at various CSQD’s core radius and shell thickness.

1. Introduction

Semiconductor nanostructures are proving themselves as potential candidate materials for a wide range of application in nano-photonics, nano-electronics, ultrafast optical devices and logic gates in optical computing and quantum information processing devices[1]. Core-shell quantum dots (CSQDs) are the structures in which the spherical core of lower bandgap material is enclosed within a shell of relatively higher bandgap material. Of late, CSQDs have been shown to be useful in the design of low temperature nano-sensors [2]. Moreover, the properties of the structure become size dependent and have discrete energy levels if its size is of the order of or less than bulk exciton Bohr radius i.e. under strong confinement regime.

The interaction between coherent light pulse and matter on a time scale much shorter than the dipole dephasing time of the material results in coherent transient effects. Optical nutation is one of such effects in which there is a periodic change (nutation) of the population between two selected states of the system which is manifested in oscillating damped ringing behaviour of the transmitted light. It is an important tool for the study of relaxation mechanism in semiconductor materials and to calculate lifetime of excited state [3]. Analysis of optical nutation in various semiconductor nanostructures like in quantum wells [4], quantum wires [5] and core-shell quantum dots have already been reported in the recent past. One of the groups has analyzed optical nutation between the energy levels inside and outside the core of the CSQDs [6].

In the present communication, we have study the nutation signal obtained due to the transition between ground state and first excitonic state and examined the dependency of maximum amplitude and frequency of the nutation signal on core radius and shell thickness of the CSQDs under strong confinement regime.
2. Mathematical Formulation

The interaction between light and CSQD can be analyzed using semi-classical theory, representing the QD quantum mechanically and light classically. Considering the transition from crystal ground state \(|a\rangle\) to the excitonic state \(|b\rangle\), we describe the CSQD by two-level wavefunction as

\[
\Psi(t) = a(t)e^{i\omega_at} |a\rangle + b(t)e^{i\omega_bt} |b\rangle.
\]

Here, \(a(t)\) and \(b(t)\) are the probability amplitudes of the crystal ground state and the excited state, respectively. We assume the interaction Hamiltonian to be of dipole type. The equations of motion of the probability amplitudes of states can be obtained using time dependent perturbation technique [7]. The probability amplitudes have been calculated by the standard formalism as

\[
a(t) = (i \lambda \sin \theta + \beta \cos \theta) e^{-i\Delta t/2} e^{-\Gamma t/2} / \beta
\]

and

\[
b(t) = -2 i \mu E_0 / 2 \hbar \sin \theta e^{i\Delta t/2} e^{-\Gamma t/2} / \beta.
\]

Here \(\omega_k (= \omega_b - \omega_a)\) is the transition frequency. \(\Gamma = T_2^{-1}; T_2\) being the collisional dephasing time of the electrons, \(\theta = \beta t/2\), and \(\Delta (= \omega_k - \omega)\) is defined as the detuning. The transition energy is given by

\[
\hbar \omega_k = E_{g(bulk)} + E_{nl} - E_{ex},
\]

with \(E_{nl}\) as the confinement energy of the carriers confined in core region and \(E_{ex}\) as the exciton binding energy in the core material.

\(X = \Delta - i\Gamma\) represent complex detuning frequency and \(\beta = \sqrt{X^2 + 4(\mu E_0 / 2\hbar)^2}\) is the generalized complex Rabi flopping frequency in the presence of detuning and dephasing.

The corresponding induced polarization in CSQD will be

\[
P(t) = N(|eP_{ab}(0)/m_0\omega_k|) [a(t)b^*(t)e^{i\omega_k t} + b(t)a^*(t)e^{-i\omega_k t}] / 2,
\]

for \(N\) number of QDs per unit volume. Considering single CSQD we take \(N=1\). \(m_0\) is the electron rest mass and \(P_{ab}(0)\) is the transition momentum matrix element at the band edge.

The corresponding value of transmitted radiation intensity is obtained using the standard relation

\[
I_T = \frac{1}{2} n_0 \varepsilon_0 c |E_T|^2,
\]

with transmitted field \(E_T(t) (= -i\omega L P(t)/c \varepsilon_0)\) determined from Maxwell’s equations under slowly varying envelope approximation for interaction length \(L\) [8].

Now we calculate confinement energy of electron and hole.

The physical structure and energy band diagram of the CSQD under investigation is shown in figure 1. In the figure, \(r_1\) and \(r_2\) are the core and shell radius, respectively and \(d (= r_2 - r_1)\) is the thickness of the shell. The eigenenergy is calculated by solving the 3-dimensional Schrodinger equation for electrons confined in the conduction band and for the holes confined in valence band. We have restricted our analysis for 1s energy state for which \(n = 1\) and \(l = m = 0\). Using the boundary conditions of the wavefunction, we get the transcendental equation.
using which the allowed value of $E_{nl1}$ for $E_{nl1} < V_c$ can be obtained. $k = \sqrt{2m_{e1}^*E_{nl1}/\hbar^2}$ and $K = \sqrt{2m_{e2}^*(V_c - E_{nl1})/\hbar^2}$ are the propagation constants in core and shell region, respectively.

Similarly the eigenenergy $E_{nlh}$ of holes can be calculated by replacing electron effective mass by the hole effective mass and conduction band offset by valence band offset. The value of $E_{nl}$ used in equation (4) is then given by $E_{nl} = E_{nl1} + E_{nlh}$.

3. Results and discussions

We have investigated analytically the optical nutation phenomena, induced in GaAs/Al$_{0.3}$Ga$_{0.7}$As CSQDs for various core radius $r_1$ and shell thickness $d$, on application of the ultra-short coherent light pulse. The CSQD is considered to be excited by a near resonant 150-fs Ti: Sapphire pulsed laser and electric field amplitude of $5 \times 10^7$ V/m under weak excitation regime at low temperature of about 5 K. For numerical analysis, we take $\Delta = 6 \times 10^{12}$ s$^{-1}$. The dephasing time is taken as $T_2 = 200$ fs. The core and shell material parameters are tabulated in table 1. The refractive index $n_0$, exciton Bohr radius $a_{ex}$ and exciton binding energy $E_{ex}$ of GaAs core are 3.6, 12 nm and 5.7 meV, respectively.

Table 1. Material parameters of the core and shell region of CSQD used in analysis at temperature 5K.

|        | $E_r$ (eV) | $m_e^*$ ($m_0$) | $m_h^*$ ($m_0$) |
|--------|------------|-----------------|-----------------|
| GaAs   | 1.521      | 0.067           | 0.45            |
| Al$_{0.3}$Ga$_{0.7}$As | 1.923      | 0.092           | 0.585           |

We take the value of $|P_{ab}(0)|^2/m_0 = 11.44$ eV for core material. The conduction and valence band offsets are taken as 0.249 eV and 0.165 eV, respectively [9].

In this paper, we have examined the dependency of maximum intensity and frequency of the nutation signal on core radius and shell thickness. The nutation signal obtained from CSQD of various core radii at constant shell thickness is shown in figure 2 and the signal obtained from CSQD with core radius 3 nm and different shell thickness is given in figure 3. The maximum amplitude and frequency of oscillation increases with the core radius whereas the change in the above same parameters is almost insignificant on changing the shell thickness, at the core radius of 3 nm.
For a given system of pump and CSQD the electric field amplitude and detuning remains constant, and eventually the factors on which maximum amplitude and frequency of the nutation signal depend is the ensemble average of transition dipole moment $\langle \hat{\mu}(t) \rangle$ and the interaction length $L$. As we increase the core radius the electrons will move farther from the hole and thus results in increasing dipole moment.

The calculation of wavefunctions and confinement energies for different core radii and shell thicknesses reveal that the probability of electron present in the shell increases with the shell thickness keeping the core radius constant. But the penetration of holes’ wavefunction doesn’t change because of change in core radius and shell thickness due to their higher effective mass and less confinement energy and therefore they remain confined inside the core. This will also increase the separation between electron and hole and hence the dipole moment also increases with the increase in shell thickness.

Another factor responsible for change with core radius is due to the increase in the interaction length. Since the incident radiation is coherent only with the core region and the shell acts as a transparent medium for it because of its higher bandgap than the core region; the interaction length is equal to the core diameter.

The interaction length thus remains constant when the core radius is kept same and the shell thickness is varied as shown in figure 3. This is the parameter which makes the intensity of nutation signal dependents more on the core radius than the shell thickness.

4. Conclusions

We have studied the optical nutation process in GaAs/Al$_{0.3}$Ga$_{0.7}$As CSQDs using effective mass approximation and time dependent perturbation theory. From the results, one may conclude that both the maximum amplitude and Rabi frequency of the nutation signal depends on the core radius whereas its dependency on the shell thickness is insignificant for the bigger core radius CSQDs. The
dependency of nutation signal on physical parameters arises due transition dipole moment and the
interaction length dependency on physical parameters.

Acknowledgments

Authors kindly acknowledge the financial support from Science and Engineering Research Board
(SERB) research project of Department of Science and Technology (DST), New Delhi, India.

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