Coherent transient effects in a InGaAs_{1-x}/GaAs semiconductor quantum dot

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Received 20 June 2012; revised 16 January 2013; accepted 12 June 2013

We present a theoretical analysis to examine the strain effect on light and heavy hole energy levels and role of exciton and bi-excitons on the absorptive optical mutation in In_{x}Ga_{1-x}As/GaAs quantum dots (QDs) near the band edge. The analysis is based upon the strain induced Luttinger Hamiltonian and time dependent perturbation technique of the coherent radiation-semiconductor interaction model for a three-level system under near band gap resonant excitation regime. The relaxation and dephasing mechanisms phenomenological have been incorporated. Numerical estimations of refractive and absorptive optical mutation have been made for a InGaAs/GaAs quantum dot duly shined by femtosecond pulse Ti: sapphire laser. Our results are found to agree qualitatively with available experimental observations.

Keywords: Nanostructures, Quantum dots, Strain effect, Absorptive optical mutation

1 Introduction

These artificially fabricated quasi zero dimensional semiconductor structures (quantum dot) have been of tremendous technological potentiality in a wide range of spintronics devices such as lasers, detectors, optical switches, quantum logic gates and quantum computer¹ etc. The coherent transient optical processes such as Rabi oscillations, optical mutation and free induction decay have been studied in these structures usually by shining with a femtosecond near-resonant laser source². Not only do QDs possess optical properties similar to atoms³ but they also benefit from well-established semiconductor manufacturing capabilities making large-scale device integration possible. Indeed, much effort is now devoted to a possible implementation of elementary quantum computation in QD systems⁴ which was especially stimulated by the observation of Rabi oscillations (ROs) of excitonic states by several groups⁵-⁷ within the past two decades. These oscillations that the population of an excited state undergoes as a function of the input pulse area, or qubit rotations, are the hallmark of a coherent system interacting with the electromagnetic field. Exponential growth has occurred in semiconductor nano-optics research on artificially fabricated lower dimensional structures such as quantum wells (QWs), quantum wires (QWRs) and quantum dots (QDs). The entire range of coherent transients such as Rabi oscillations, optical mutation, photon echo, free induction decay, self-induced transparency, etc., occur in a time scale much shorter than the material decay time constants⁸.

Recording of ROs is also a direct way of measuring dipole moments of excitonic transitions which are much larger than their atomic counterparts. These can provide more insight about the fundamental structure of QDs which yet remains largely unexplored. Previous studies⁴ have found dipole moments typically around 20 Debye in self-assembled QDs where the confinement is strong and dimensions are small as compared to interface fluctuation QDs (IFQDs) moments close to 100 Debye have been measured⁵. An individual excitonic electric dipole transition approximates a two-level system so that many coherent phenomena such as ROs can be described by the familiar Bloch equations of atomic physics⁶.

The objective of the present analytical study is to demonstrate the applicability of optical coherent transients, specifically optical mutation to various physical mechanisms such as hot carrier dynamics and relaxation processes that limit the versatility of ultrafast optical information processing in a nanostructurers⁷. It is worth mentioning that in comparison to their bulk counterparts, the semiconductor QDs exhibit larger electro-absorption, electro-refraction and higher differential optical gain. No detailed systematic approach has yet been developed to explain the role of excitonic and
bi-excitonic non-linearities in the occurrence of the optical nutation in semiconductor QDs.

2 Theoretical Formulations

The theoretical analysis presented here comprises of (i) calculations of the exciton and bi-exciton energy levels incorporating the strain effects and (ii) calculations of the populations density in exciton and bi-exciton state. The energy levels in the quantum dot are determined by using Luttinger Hamiltonian and incorporating modifications under quantum confinement conditions. The other important parameters that affect the energies of the exciton and bi-exciton state are (i) strain effect and (ii) temperature induced band edge shrinkage/broadening effects. In case of InGaAs/GaAs QD, there is a lattice mismatch between the QD and the wetting layer giving rise to the strained system.

The exciton energy in the QD is calculated from the electron and hole energies in the conduction and valence band taking into account the Coulombic and confinement potentials. We first concentrate on the calculation of the hole energies. The valence band structure can be most appropriately described by the Luttinger Hamiltonian given by:

\[
H_L = \begin{bmatrix}
H_{hh} & c & b & 0 \\
0 & H_{lh} & 0 & -b \\
b^* & 0 & H_{hh} & c \\
0 & -b^* & c^* & H_{hh}
\end{bmatrix}
\]

where

\[H_{hh} = \frac{\hbar k_x^2}{2m_o}(\gamma_1 - 2\gamma_2) + \frac{\hbar^2}{2m_o}(k_x^2 + k_y^2)(\gamma_1 + \gamma_2)\]

\[H_{lh} = \frac{\hbar k_x^2}{2m_o}(\gamma_1 + 2\gamma_2) + \frac{\hbar^2}{2m_o}(k_x^2 + k_y^2)(\gamma_1 - \gamma_2)\]

\[b = \sqrt{\frac{\hbar^2}{m_o}}\gamma_1(k_x - ik_y)k_x\]

\[c = \sqrt{\frac{\hbar^2}{m_o}}[\gamma_2(k_y^2 - k_x^2) - 2i\gamma_1 k_x k_y]\]

where \(\gamma_1, \gamma_2,\) and \(\gamma_3\) are the Luttinger parameters and \(k_x, k_y, k_z\) are the components of the wave vector in the, \(x, y\) and \(z\) directions, respectively. Also \(m_o\) is the rest mass of the electron, \(H_{hh}\) and \(H_{lh}\) are the Hamiltonian corresponding to the heavy hole and light hole states. The hole energy levels are modified by the confinement potential. In the lattice mis-matched InGaAs/GaAs quantum dots neither the barrier height nor the confinement potentials inside the quantum dots are constant due to pseudomorphic strain. Consequently, the confinement is, generally, size dependent. The shape of the self-assembled InGaAs/GaAs QDs can be considered to be ellipsoidal. The confinement potential in an ellipsoidal QD can be described by approximating it as a parabolic potential function \(V(x, y, z)\) written as:

\[V(x, y, z) = (\alpha_x^2 x^2 + \alpha_y^2 y^2 + \alpha_z^2 z^2)\]

where \(\alpha_x, \alpha_y,\) and \(\alpha_z\) are the characteristic parameters depending upon the geometrical size of the quantum dot and the mass of the particle under consideration. The Hamiltonian of the system for valence band structure can, therefore, be written as:

\[H = H_L + V(x, y, z)\]

In the presence of the confinement potential, the eigen wave function of single particle (electron or hole) can be described as the product of the Bloch function \(U\) and the envelope function \(\chi(x, y, z)\) as:

\[\psi(x, y, z) = U \chi(x) \chi(y) \chi(z)\]

In the parabolic potential approximation, the component of the envelope function in \(x, y\) and \(z\) directions are given by:

\[\chi(x) = H(l, \alpha_x, x)e^{\frac{1}{2} \alpha_x^2 x^2}\]

\[\chi(y) = H(l, \alpha_y, y)e^{\frac{1}{2} \alpha_y^2 y^2}\]

\[\chi(z) = H(l, \alpha_z, z)e^{\frac{1}{2} \alpha_z^2 z^2}\]

where \(H(l, \alpha, i)\) is the hermite polynomial of degree \(l\) and \(\alpha, i\) are the dimensionless parameters corresponding to the dimension of the QD along \(x, y\) and \(z\) directions, respectively. The energy eigenvalue of heavy holes can be calculated by diagonalising the Hamiltonian as:

\[\varepsilon_{lh} = \hbar \omega_{lh} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \psi_{lh}^*(H_{hh} + V_h(x, y, z))\psi_{lh} dx dy dz\]
$V_{h}(x, y, z)$ represents the confinement potential experienced by heavy holes. The conduction band electron energy is also calculated in similar fashion by defining it as:

$$\varepsilon_{el}(= \hbar \omega_{el}) = \hbar \omega_{el} + \int \int \int \psi^\ast_{el}(H_{el} + V_{el}(x, y, z))\psi_{el} \, dx \, dy \, dz$$

$$- \frac{1}{2} \int \int \int \psi^\ast_{el}(H_{el} + V_{el}(x, y, z))\psi_{el} \, dx \, dy \, dz - \delta E_{H} - \frac{1}{2} \delta E_{S}$$

where $l$ is the concentration of indium. In obtaining equation, we have neglected the Coulomb potential. Such an approximation is valid for small quantum dots where confinement potential dominants over the Coulomb potential.

For an In$_{x}$Ga$_{1-x}$As/GaAs quantum dot of the lateral size $x=30$ nm, $y=30$ nm, $z=12$ nm, $l=0$ and concentration $x=0.4$ the exciton energy is found to be 1.23 eV. The magnitude of Coulomb potential was calculated to be 9.1 meV while confinement potential was found to be 15.3 meV. It is appropriate to describe the optical properties of the quantum dot by taking into account the optical transition between ground ($|o\rangle$), excitonic ($|e\rangle$) and bi-excitonic ($|b\rangle$) states while direct ground ($|o\rangle$) to bi-exciton ($|b\rangle$) states transitions are optically forbidden. In order to calculate the bi-exciton energy, the Coulomb exchange force between the exciton pairs causing bi-excitons formation should be incorporated. We consider the biexciton binding energy $B_{xx}$ in terms of these exchange forces in the Eqs (9-11) is taken over the dot size. Thus, it is clear that the bi-exciton binding energy is dot size dependent. The corresponding bi-exciton energy $E_{b}$ is given by:

$$E_{b}=2E_{e}-B_{xx}$$

For an In$_{x}$Ga$_{1-x}$As/GaAs quantum dot of the lateral size $x=30$ nm, $y=30$ nm, $z=12$ nm, $l=0$ and concentration $x=0.4$ the bi-exciton energy is found to be 2.4 eV.

Using rotating wave approximation for a three-level system, the equations of motion of the probability amplitudes of the three crystal states are expressed as:

$$\dot{a}(t) = i \sum_{k} \frac{\mu_{hk}E_{h}}{2\hbar} e^{-i(\omega_{hk}-\omega_{el})t} b(\vec{k}, t)$$

$$\dot{b}(\vec{k}, t) = i \frac{\mu_{hk}E_{h}}{2\hbar} e^{i(\omega_{hk}-\omega_{el})t} a(t)$$

and
\[ c(\vec{k}, t) = \frac{i\mu_0 E}{2\hbar} e^{-i(a_0 - \Delta a_{\text{neut})/2\hbar}} b(\vec{k}, t) - \Gamma_c c(\vec{k}, t) \] 

(13c)

where \( h\omega_{ab} \) and \( h\omega_{ba} \) are the transition energies and \( \Delta E_{\text{es}} \) (\( = h\Delta \omega_{\text{neut}} \)) is one-dimensional bi-exciton binding energy. In Eq. (1), we have considered the dephasing parameters for the excitonic and bi-excitonic states as \( \Gamma = \Gamma_{11} = 2 = T_1 \); \( T_2, \mu_{ab}, \mu_{ba} \) and \( \mu_{\text{neut}} \) are the dephasing time, transition dipole moment operators for the excitonic and bi-excitonic transitions, respectively.

The solutions of Eq. (13a) through Eq. (13c) are respectively.

\[ a(t) = u_0 p_{00} e^{i\omega t} + u_1 p_{01} e^{i\omega t} + u_2 p_{02} e^{i\omega t}, \quad \text{...(15a)} \]

\[ b(\vec{k}, t) = (u_0 p_{00} e^{i\omega t} + u_1 p_{01} e^{i\omega t} + u_2 p_{12} e^{i\omega t}) e^{i(\Delta a_{\text{neut}} - \omega)t} \quad \text{...(15b)} \]

and

\[ c(\vec{k}, t) = (u_0 p_{00} e^{i\omega t} + u_1 p_{01} e^{i\omega t} + u_2 p_{12} e^{i\omega t}) e^{i(2\omega - \Delta a_{\text{neut}} - \omega t)} \quad \text{...(15c)} \]

where \( s_{0,1,2} \) are eigenvalues and \( u_{0,1,2} \) are constants obtained from the boundary conditions at \( t=0 \) and \( p_{ij} \) \((i, j = 0, 1, 2)\) are the eigenvector for states \( |a\rangle, |b\rangle \) and \( |c\rangle \). The ensemble average of the effective transition dipole moment operators \( \hat{\mu}(\vec{k}, t) \) can be defined as sum of the contributions arising due to both excitons and bi-excitons and hence, the total induced polarization can be calculated by the following equation:

\[ P(t) = N_x (2\mu_{ab}(k)|a(t)b^*(\vec{k}, t)e^{-i(\Delta a_{\text{neut}} - \omega t)}] + 2\mu_{\text{neut}}(k)\rho(b(\vec{k}, t)c^*(\vec{k}, t)e^{i(\Delta a_{\text{neut}} - \omega t)})] \quad \text{...(16)} \]

With \( N_x \) and \( N_{\text{neut}} \) being exciton and bi-exciton densities, respectively and taken \( \rho = N_{\text{neut}}/N_x \). The complex intensity dependent optical susceptibility of the semiconductor QD can be calculated by using the definition \( P(t) = e_0 \chi^{*}(t) \) where \( \chi(= \chi_e + i\chi_i) \) includes \( \chi_e^{(1)}, \chi_e^{(3)} \), etc.

We have applied our analysis to the single InGaAs quantum dot. The material parameters for this sample are taken as follows \( m_e = 0.005m_0, m_{\text{hole}} = 0.377m_0, \gamma_1 = 11.01, \gamma_2 = 4.18, \gamma_3 = 4.84 \). For an In_{0.4}Ga_{0.6}As/GaAs quantum dot of size 30x30x12 nm^3.

Remembering that the dispersive and absorptive features of the nutating signal are manifested through the real and imaginary parts of the optical susceptibility \( \chi(= \chi_e + i\chi_i) \). The analytical behaviour of transient induced polarization in strain In_{0.4}Ga_{0.6}As/GaAs QD, for three different light hole bi-excitonic densities which is determined by \( \chi(-\chi + \chi_e) \) is shown in Fig. 1. Figure 1 shows the oscillating nature of transient induced polarization. It is clear that the rising bi-exciton density increases the transient induced polarization. The periodic decay of polarization with time is found to be in good agreement with the theoretical studies of Bafna and Sen for near band gap resonant electronic transitions in QWR. The non-linear absorption may be attributed to the photo-induced generation of excitons and bi-excitons. Transient absorptive optical nutation in strain In_{0.4}Ga_{0.6}As/GaAs QD of size 30x30x12 nm^3 for light hole and heavy hole reduced masses. Figure 2 shows the transient oscillating nature of the absorptive optical nutation for light-hole and heavy-hole effective masses at bi-exciton density \( \rho = 2 \). Similar decay characteristics of the nutating signal was also reported by Shakhmuratov et al for near band gap resonant optical transitions in QWR. The non-linear absorption may be attributed to the photo-induced generation of excitons and bi-excitons. Transient absorptive optical nutation in strain In_{0.4}Ga_{0.6}As/GaAs QD of size 30x30x12 nm^3 for light hole and heavy hole reduced masses.

![Fig. 1 — Transient induced polarization in strain In_{0.4}Ga_{0.6}As/GaAs QD of size 30x30x12 nm^3 QDs for three different light hole bi-excitonic densities](image-url)
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Conclusions
To conclude, the present analysis establishes that both transient dispersive and absorptive optical nutation in an In$_x$Ga$_{1-x}$As/GaAs QD are influenced by the presence of bi-exciton. Increased bi-exciton density enhances both transient dispersive and absorptive optical nutation. There is also a significant effect of the relaxation parameters on these coherent transient processes. The present analytical study may lead one to predict the occurrence of optical coherent in a moderately strong confined semiconductor quantum dot subjected to a not-too-strong near band gap resonant femtosecond coherent excitation.

Acknowledgement
One of the authors (Dr Manish Kumar Bafna) is grateful to Mandsaur Institute of Technology, Indore for supporting the present research work.

References