Effect of temperature on non-linear optical properties of InGaAs/GaAs single quantum dot

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Based upon the semi-classical density matrix approach, a detailed theoretical investigation has been made to analyze the effect of temperature on non-linear optical properties of InGaAs/GaAs single quantum dot (QD). The temperature effects have been incorporated via temperature dependent (i) dephasing mechanism, (ii) band gap energy and (iii) population density. The semiconductor QD has been chosen to be small enough such that the confinement effect dominates over the Coulombic contribution. Detailed numerical estimates of the non-linear refraction and absorption properties have been made. Redshifts of the non-linear refraction and absorption peaks and reduction in the induced polarization are found to occur with increasing temperature. The present analysis further reveals that the non-linear gain reduces at higher temperature.

Keywords: Semiconductor quantum dot, Temperature dependent non-linear absorption, Exciton, Biexciton, Third-order susceptibility.

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1 Introduction

The zero dimensional structures are finding applications in advanced optoelectronic devices such as lasers, detectors and optical memory elements as they prove advantageous in terms of lower threshold current densities, higher gain and higher quantum efficiencies. The properties of the nanostructures, in particular quantum dots (QDs) can be modified in a controlled manner by changing their size and shape. The reduced dimensions of QD cause increased spatial overlap between electron and holes. As a result, along with excitons, biexcitons are also formed. Thus in the determination of optical properties of semiconductor QDs, biexciton also play an important role. The energy separation of the excitonic and biexcitonic levels is temperature sensitive and hence the device engineering requires a full understanding of the temperature dependence of the photoluminescence spectra (PL) in the QDs. Extensive studies have been carried out to discuss the effect of temperature on the photoluminescence properties of quantum dot. Jiang et al. and Mazur have reported a red shift in the PL peak in InGaAs/GaAs QDs with increasing temperature. The red shift in an ordered array of quantum dot is suggested to occur due to the lateral and vertical transfer of excitations. Thermal quenching of photoluminescence in QDs can be attributed to the thermal activation of charge carriers from confined well into barrier followed by an effective non-radiative recombination. The unusual temperature behaviour like reduction in linewidth with increasing temperature as well as faster redshift in energy implies interesting interactions in dot systems such as thermionic emission and carrier repopulation among QDs. In recent years, high spatial resolution techniques have been used to isolate single QD structure allowing the measurement of linewidth, Stark-shift, biexcitonic energy levels etc. Most of these studies are based on PL as a probe. In the present paper, we have studied the effect of temperature on the PL spectra of a single QD.

2 Theoretical Formulations

Semiclassical density matrix approach has been employed to analyze the effect of temperature on the optical properties of semiconductor QDs. We consider single semiconductor quantum dot of spherical shape in a strong confinement regime. It is appropriate to describe the optical properties of the quantum dot by taking into account the excitonic and biexcitonic energy levels. The optical transitions occur between the crystal ground state \( |o> \) to exciton state \( |e> \) as well as exciton \( |e> \) to the biexciton state \( |b> \). The
exciton energy level in this confinement regime is defined as:
\[ \hbar \omega_e = \hbar \omega_{g}^{\text{eff}} + \frac{h^2 \kappa^2_n}{2m_e R^2} - \left( \frac{3.572 a_B}{R} + 0.248 \right) E_g; \quad \ldots (1) \]
and
\[ \hbar \omega_{g}^{\text{eff}} = \hbar \omega_{g} - \left( \frac{\alpha T^2}{T + \beta} \right). \]

In Eq. (1), \( \hbar \omega_{g}^{\text{eff}} \) is the bulk crystal band gap energy at temperature \( T \) (Ref. 11); \( m_e \) and \( E_g \) are effective reduced mass and exciton Rydberg energy, respectively while \( \alpha \) and \( \beta \) are Varshni parameters. The second term on the R.H.S. of Eq. (1) represents the confinement energy. \( \kappa \) is the \( n \)th root of the \( l \)th order Bessel function with \( n \) and \( l \) corresponding to the 1s, 1p and 1d levels of the electrons and holes, respectively. The third term represents the Coulombic interaction energy of electron-hole pairs where \( \alpha \) and \( \beta \) are Varshni parameters.

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The unperturbed Hamiltonian can be defined as,
\[ H_0 = \hbar \begin{bmatrix} \omega_0 & 0 & 0 \\ 0 & \omega_e & 0 \\ 0 & 0 & \omega_b \end{bmatrix}. \quad \ldots (3) \]

The interaction of QD with the electromagnetic radiation \( \mathcal{E}(t) = \frac{1}{2} \mathcal{E}_0 e^{i \omega t} + c.c. \) is considered to be dipole type. The interaction Hamiltonian is represented by
\[ H_I = \begin{bmatrix} 0 & \mu_{oe} \mathcal{E} & 0 \\ \mu_{oe}^* \mathcal{E}^* & 0 & \mu_{eb} \mathcal{E}^* \\ 0 & \mu_{be} \mathcal{E} & 0 \end{bmatrix}, \quad \ldots (4) \]

where the electric field is taken to be parallel to the transition dipole moment operators \( \mu_{ij} \), which is related to the transition momentum \( p \) as
\[ \mu_{ij} = \frac{e}{m_0 \omega_{ij}} | \langle j | p | i \rangle |. \quad \ldots (5) \]

Here, \( m_0 \) is the electron rest mass, and \( \hbar \omega_{ij} \) is the transition energy. In the present calculation, we have incorporated the losses occurring due to both radiative and non-radiative decay processes. The homogenous broadening at low temperature is reported in the range of 50 \( \mu \text{eV} \), in In\(_x\)Ga\(_{1-x}\)As QD and is attributed to pure dephasing\(^{14}\). These relaxation processes are incorporated via \( H_r \) as:
\[ H_r = \hbar \begin{bmatrix} \gamma_0 & 0 & 0 \\ 0 & \gamma_e & 0 \\ 0 & 0 & \gamma_b \end{bmatrix} \quad \ldots (6) \]

The relaxation parameters are strongly temperature dependent and are given by
\[ \gamma_e = \gamma_{e0} + aT + \frac{b}{\exp(E_a / k_B T) - 1} \quad \ldots (7a) \]
and
\[ \gamma_b = \gamma_{b1} + a_1 T + \frac{b}{\exp(E_a / k_B T) - 1}. \quad \ldots (7b) \]

Here, \( E_a, \gamma_{e0}, \gamma_{b1}, a, a_1 \), and \( b \) are taken from the experimental observation of Borri and Co-Workers\(^{14,15}\). The QD-radiation interaction is represented by the Schrodinger’s equation of motion
\[ \frac{d\rho}{dt} = -\frac{i}{\hbar} [H, \rho] - \frac{1}{\hbar} [H_r, \rho], \quad \ldots (8) \]
with density matrix taken as
\[ \rho = \begin{bmatrix} \rho_{oo} & \rho_{oe} & \rho_{ob} \\ \rho_{eo} & \rho_{ee} & \rho_{eb} \\ \rho_{bo} & \rho_{be} & \rho_{bb} \end{bmatrix}. \quad \ldots (9) \]

The density matrix equations are expanded for various orders of the density matrix elements \( \rho = \rho^{(1)} + \rho^{(2)} + \rho^{(3)} + \ldots \), where the zeroth order temperature dependent density distribution function of crystal ground, exciton and biexciton states are defined as
\[ \rho_{00}^0 = 1 - \rho_{ee}^0 - \rho_{bb}^0, \quad \cdots (10a) \]

\[ \rho_{ee}^0 = \exp\left(-\frac{\hbar \omega_{ee} - \hbar \omega_{g}^{\text{eff}} - (\hbar^2 \kappa_{i}^2 / 2m, R^2)}{k_T} \right), \quad \cdots (10b) \]

and

\[ \rho_{bb}^0 = \exp\left(-\frac{\hbar \Delta \omega_{ee}^{\text{ex}}}{k_T} \right). \quad \cdots (10c) \]

Here, \( k_T \) is the thermal energy.

Various orders of the density matrix elements are calculated by using Eqs (4)-(10) as

\[ \rho^{(1)} = \begin{pmatrix} 0 & \frac{2\Omega_e (\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{ee}} & 0 \\ -\frac{2\Omega_e (\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{ee}} & 0 & \frac{2\Omega_e (\rho_{00}^0 - \rho_{bb}^0)}{\Delta_{ee}} \\ 0 & -\frac{2\Omega_e (\rho_{00}^0 - \rho_{bb}^0)}{\Delta_{bb}} & 0 \end{pmatrix}, \quad \cdots (11) \]

\[ \rho^{(2)} = \begin{pmatrix} A & 0 & B \\ 0 & C & 0 \\ D & 0 & E \end{pmatrix}, \quad \cdots (12) \]

and

\[ \rho^{(3)} = \begin{pmatrix} 0 & \frac{2\Omega_e (\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{ee}} & 0 \\ -\frac{2\Omega_e (\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{ee}} & 0 & \frac{2\Omega_e (\rho_{00}^0 - \rho_{bb}^0)}{\Delta_{ee}} \\ 0 & -\frac{2\Omega_e (\rho_{00}^0 - \rho_{bb}^0)}{\Delta_{bb}} & 0 \end{pmatrix}, \quad \cdots (13) \]

with

\[ A = \frac{4\Omega_e}{\omega} \left[ \frac{1}{\Delta_{ee}} + \frac{1}{\Delta_{cc}} \right] (\rho_{00}^0 - \rho_{ee}^0), \]

\[ B = \frac{4\Omega_e \omega}{\Delta_{cc}} \left[ \frac{-(\rho_{ee}^0 - \rho_{bb}^0)}{\Delta_{cc}} + (\rho_{00}^0 - \rho_{ee}^0) \right] \]

\[ C = \frac{4}{\omega - 2i\gamma_e} \left[ -\left( \frac{1}{\Delta_{ee}^*} + \frac{1}{\Delta_{cc}^*} \right) (\rho_{00}^0 - \rho_{ee}^0) \Omega_{ee}^2 + \left( \frac{1}{\Delta_{ee}^*} + \frac{1}{\Delta_{cc}^*} \right) (\rho_{ee}^0 - \rho_{bb}^0) \Omega_{bb}^2 \right] \]

\[ D = \frac{4\Omega_e \Omega_{bb}}{\omega} \left[ \frac{(\rho_{00}^0 - \rho_{ee}^0) - (\rho_{ee}^0 - \rho_{bb}^0)}{\Delta_{ee}^* - \Delta_{bb}^*} \right], \]

and

\[ E = \frac{-4\Omega_e^2}{\omega - 2i\gamma_b} \left[ \frac{1}{\Delta_{bb}^*} + \frac{1}{\Delta_{bb}^*} \right] (\rho_{ee}^0 - \rho_{bb}^0) \]

where, \( \Delta^{\pm}_{ij} = \omega \pm \omega_j + i\gamma_j \) is the damping incorporated detuning parameter with suffixes 1 and 2 corresponding to (+)ive and (–)ive signs, respectively. Also \( \Omega_{ij} = \mu_i E_o / 2h \) is the Rabi frequency. The damping constant represented by \( \gamma_j \) corresponds to the phase relaxation terms of exciton and biexciton, respectively. The total induced polarization \( P = P^{(1)} + P^{(2)} + P^{(3)} + \ldots \); has been obtained.

The various orders of the induced polarization are obtained are:

\[ P^{(1)} = \frac{2\mu_e^2 E (\rho_{00}^0 - \rho_{ee}^0)}{(\omega - \omega_{ee} - i\gamma_e)} + \frac{2\mu_b^2 E (\rho_{00}^0 - \rho_{bb}^0)}{(\omega - \omega_{cc} - i\gamma_b)}, \quad \cdots (14) \]

\[ P^{(2)} = 0, \quad \cdots (15) \]

\[ P^{(3)} = \frac{-16\mu_e^4 E^3 (\rho_{00}^0 - \rho_{ee}^0)(\omega - i\gamma_e) + 8(\mu_e)^2 (\mu_b)^2 E^2 (\rho_{00}^0 - \rho_{ee}^0)}{\hbar^3 \omega(\omega - 2i\gamma_e)(\omega - \omega_{ee} - i\gamma_e)^2} + \]

\[ \frac{8(\mu_e)^2 (\mu_b)^2 E^2 (\rho_{00}^0 - \rho_{ee}^0)}{\hbar^3} \times \frac{\omega(\omega - \omega_{ee}) - \omega \Delta \omega_{ee} - 2\gamma_e^2 + 2\gamma_e \gamma_{bb}}{\omega(\omega - 2i\gamma_e)(\omega - \omega_{ee} - i\gamma_e)^2(\omega - \omega_{ee} + \Delta \omega_{ee} - i\gamma_{bb})} \]

\[ \frac{-i(\omega - \omega_{ee} + 2\omega_{ee} + \omega_{ee} \gamma_e - 2\gamma_e \Delta \omega_{ee})}{\omega(\omega - 2i\gamma_e)(\omega - 2i\gamma_e)(\omega - \omega_{ee} + \omega_{ee} + \Delta \omega_{ee} - i\gamma_{bb})} \]
from the non-linear polarization by as

\[ \frac{8\mu_n^2\mu_b^2E^3(\rho_{ee}^0 - \rho_{bb}^0)}{\hbar^3} \]

\times \left[ \frac{(\omega^2 - \omega_{ee}^0 + 2\gamma_{ee}^0 + 2\gamma_{ee} - 2\omega\Delta\omega_{ee})}{\omega(2\gamma_{ee} - \omega_{ee} + 2\gamma_{ee} - \omega\Delta\omega_{ee})} \right]

\[ = \frac{-i(2\gamma_{ee} - \omega_{ee} + 2\gamma_{ee} - \omega\Delta\omega_{ee})}{\omega(2\gamma_{ee} - \omega_{ee} - i\gamma_{ee})(\omega - \omega_{ee} + \Delta\omega_{ee} - i\gamma_{ee})^2} \]

\[ - \frac{16\mu_n^4\epsilon_{ee}^0E^3(\rho_{ee}^0 - \rho_{bb}^0)(\omega - i\gamma_{ee})}{\hbar^3(\omega - 2\gamma_{ee})(\omega - \omega_{ee} + \Delta\omega_{ee} - i\gamma_{ee})^2} \] ...

In obtaining Eqs (14-15), the off resonant term \( \Delta \gamma \) has been neglected. Equation (14) distinctly yields the first order-induced polarization \( P^{(1)} \). The first and the second term correspond to the \( |\alpha>, |\alpha> \) and \( |\beta>, |\beta> \) transitions via first and the second term, respectively. At low temperatures where the exciton population levels are empty, the \( |\alpha> \) will be absent. For a symmetric quantum dot, as given by Eq. (15) the second-order polarization term vanishes. In realistic situation, the asymmetry in the quantum dots due to their shape and size is always present and we have not given due attention to the asymmetry property of the QD. The third-order induced polarization as represented by Eq. (16) comprises of the contributions from both \( |\alpha>, |\alpha> \) and \( |\beta>, |\beta> \) transitions. Also the third-order induced polarization can be directly related to the third-order non-linear optical susceptibility via the well-known relation \( \chi^{(3)} = \left( \frac{P^{(3)}}{\epsilon_0|E|^2E} \right) \). The first term in Eq. (16) represents the third-order optical non-linearity arising due to \( |\alpha>, |\alpha> \) transitions only and depends strongly on the resonance of incident photon energy with the excitonic energy. The second and third terms have their origins in the contributions from both the excitonic \( |\alpha>, |\alpha> \) and biexcitonic \( |\beta>, |\beta> \) transitions.

3 Results and Discussion

The numerical analysis to examine the temperature dependence of third-order non-linear optical susceptibility in a semiconductor QD subjected to a \( \text{cw} \) coherent radiation is presented. We have chosen InGaAs/GaAs QD duly irradiated by a \( \text{cw Ti: Sapphire laser} \). The material parameters selected for the present analysis are \( h\omega_0 = 1.23 \text{ eV}, m_e = 0.05 m_0, m_{bh} = 0.384 m_0 \) with \( m_0 \) being the free electron mass and quantum dot diameter = 15 nm. In order to calculate the exciton (\( \gamma_e \)) and biexciton (\( \gamma_b \)) dephasing times, we use Eq. (7) with the numerical values of the various parameters from the experimental observations of Borri \textit{et al.} \cite{14,15} as \( E_a = 16 \text{ meV}, \gamma_0 = 0.67 \text{ \( \mu \) eV}, \gamma_1 = 2 \text{ \( \mu \) eV}, a = 0.22 \text{ \( \mu \) eV/K}, \alpha_1 = 0.37 \text{ \( \mu \) eV/K} \) and \( b = 1.1 \text{ meV} \) for specific values of the temperature \( T \). We have selected the input field strength \( E_0 = 10^7 \text{ Vm}^{-1} \) in the present computation. In Fig. 1, the total induced polarization as a function of the pump photon energy at 50, 100, 150 and 200 K has been plotted. From Fig. 1, it can be seen that two distinct peaks appear at 50 K. These peaks correspond to the exciton and biexciton energies (biexciton peak being at lower energy). With increase in temperature, the thermal broadening also increases such that the biexciton peak becomes almost insignificant. This is quite obvious due to the well-known fact that biexcitonic effect plays important role only at very low temperatures. The peak positions of the induced polarization further reveals that the increasing temperature causes significant redshift in the peak value of the polarization. The reduction in both exciton and biexciton energies with increasing temperature is clearly manifested from the same figure, such red shift in the PL peak was also reported by Jiang \cite{9} and Mazur \cite{8} independently.

We have calculated the non-linear absorption coefficient \( \Delta \alpha \) from the non-linear polarization by

defining it as \( \Delta \alpha = k \text{ Im} \left( \frac{P^{(3)}}{\epsilon_0|E|^2E} \right) \); \( k \) being

the wave vector of the pump photon. In Fig. 2, \( \Delta \alpha \) as

\[ \text{Fig. 1 — Characteristic temperature dependence of the total induced polarization in In}_{x}\text{Ga}_{1-x}\text{As/GaAs single quantum dot} \]
a function of pump photon energy has been plotted at 140, 145 and 150 K. A negative value of \(\chi^{(3)}\) near the resonance energy manifests the occurrence of non-linear gain. From Fig. 2, a red shift in resonance energy with increasing temperature can be noticed. The magnitude of the non-linear gain is also found to decrease with increase in temperature. Further, the effect of temperature on non-linear refraction (non-linear refractive index) \(n\) is examined and is found to be directly proportional to the real part of the third-order non-linear optical susceptibility. In Fig. 3, \(n\) has been plotted as function of the pump photon energy at 140, 145 and 150 K. It can be clearly observed that \(n\) becomes significant only near the resonance frequency. From Fig. 3, we find that apart from redshift in resonance frequency, the magnitude of non-linear refraction increases at lower temperatures.

4 Conclusion
The effect of temperature on non-linear optical properties gain in an InGaAs/GaAs single quantum dot is studied. It is found that the biexcitons play an important role only at low temperature. Moreover, the lowering of temperature enhances both non-linear refraction and absorption properties of the single quantum dot.

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References