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Original Research Article

The density matrix approach for determination of influence of temperature on photoluminescence spectrum of a single quantum dot

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ABSTRACT
The semi-classical density matrix method has been used in this study to examine how temperature affects the optical characteristics of semiconductor quantum dots. It is appropriate to consider the excitonic and biexcitonic energy levels when describing the optical characteristics of the quantum dot. The optical transitions happen between the exciton state $|e\rangle$ and the biexciton state $|b\rangle$ as well as the exciton state $|e\rangle$ and the crystal ground state $|o\rangle$. By closely following the Varshni contribution, the band gap shrinkage in III-V semiconductors can be roughly integrated. The hydrostatic (δE_H) and shear strain modify the hole energy levels as a result of the tensile strain (δE_S). The photoluminescence spectra of $In_xGa_{1-x}As/GaAs$ quantum dots have thus been plotted at various temperatures while accounting for strain effect and including thermal effects. The full width at half maximum (FWHM) of the photoluminescence peaks is also displayed as a function of temperature to critically assess how the photoluminescence spectra have widened.

1. Introduction

Low dimensional semiconductor nanostructures can now be due to contractual to the recent, rapid advancements in semiconductor technology (quantum dots, quantum wires etc.). By altering their size and shape, nanostructures, in particular quantum dots (QDs), can have their properties changed in a controlled way. The QD's smaller dimensions result in more spatial overleap between the electrons and the holes. As a result, biexcitons are produced in addition to excitons. Biexciton thus impart an important contribution in determining the optical characteristics of semiconductor QDs.

The device engineering demands a thorough understanding of the temperature dependency of photoluminescence spectra of the quantum dots since an energy separation of the excitonic and bi-excitonic levels is thermally sensitive. Numerous research [1-4] have been conducted to examine how temperature affects the photoluminescence characteristics of quantum dots. The photoluminescence peak in InGaAs–GaAs QDs has been found to redshift with temperature by Jiang et al. [3] and Mazur [2]. According to one theory, the lateral and vertical excitation transfer causes the red shift in an arranged matrix of QDs [2].

The heat suppression of emission spectra in quantum dots is believed to be caused by thermal activation of charge transfer from restricted well into barrier followed by an effective non-radiative recombination [1-3]. In recent years, single quantum dot structures have been isolated using high spatial resolution techniques, enabling the determination of Stark-shiftung, linewidth, and bi-exciton energy levels. The majority of that kind of research use photoluminescence as a probe. The influence of temperature on the photoluminescence spectra of single QD has been investigated in current research.

2. Theoretical formulations

The impact of temperature on the optical characteristics of semiconductor QDs has been studied using a semi-classical density matrix technique. It is appropriate to consider the excitonic and bi-excitonic energy levels when describing the photoluminescence characteristics of a single QD. The radiative transformation happen among the excitonic state represented by $|e\rangle$ and the bi-excitonic state represented by $|b\rangle$ as well as the excitonic state represented by $|e\rangle$ and the crystal ground state represented by $|o\rangle$. By closely following the Varshni contribution, the band gap narrowing in $A^{III}B^{V}$ type semiconducting materials could be approximately integrated. The hydrostatic (δE_H) and shear strain modify the hole energy levels as a result of the tensile strain (δE_S). The energy of excitonic state E_e in the $In_xGa_{1-x}As$ quantum dot is therefore obtained by accounting for the strain effect and including thermal factors as follows:

$$
\begin{aligned}
&E_e(=\eta(\omega_{el}-\omega_{hh}))\\
&=1.519-1.47x+0.375x^2-\frac{\alpha T^2}{(T+\beta)}\\
&+\int_0^z\int_{-y}^{y}\int_{-x}^{x}\psi_{el}^{*}(H_{el}+V_{el}(x,y,z)\psi_{el}dx\,dy\,dz\\
&-\int_0^z\int_{-y}^{y}\int_{-x}^{x}\psi_{hh}^{*}(H_{hh}+V_{h}(x,y,z)\psi_{hh}dx\,dy\,dz-\delta E_H-\frac{1}{2}\delta E_S. \quad (1)
\end{aligned}
$$

Here, x denotes the indium concentration, ε denotes the material medium's dielectric constant, and, α, β denote the Varshni parameters. The author determined the exciton energy

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E_e = 1.23 eV for an $In_xGa_{1-x}As/GaAs$ QD with dimensions of 20×20×5 nm^3, l = 0 and conc. level x = 0.4 at a fixed temperature of T = 100K.

Biexciton energy E_b is slightly less than the energy of two excitons because of the Coulomb's exchanging influence among excitonic pairs, and is defined as:

$$E_b = 2E_e - B_{xx}\,. \tag{2}$$

The biexciton binding energy in this case, B_{xx}, is dependent upon the dimensions of QD [9]. The author discovered the biexciton energy E_b = 2.4 eV for the $In_{0.4}Ga_{0.6}As/GaAs$ quantum dot possessing identical set of parameters as previously stated.

The lowest (n = 1,1s) excitonic state $|e\rangle$, the bi-excitonic state $|b\rangle$, and the crystal's ground state $|o\rangle$ are regarded to be the three levels between which the optical transitions are thought to occur.

Accordingly, the unperturbed Hamiltonian can be defined as follows using a 3×3 matrix:

$$H_o = \begin{bmatrix} E_o & 0 & 0 \\ 0 & E_e & 0 \\ 0 & 0 & E_b \end{bmatrix} \tag{3}$$

$E_{o,e,b} = \eta\omega_{o,e,b}$ represents the energies of the ground state, the excitonic state, and the bi-excitonic state, respectively.

It is believed that the QD's interaction with electromagnetic wave $\vec{E}(\omega) = \frac{1}{2}\vec{E}_0 e^{i\omega t} + c.c.$ is of the dipole type because the interaction Hamiltonian is provided by

$$H_I = \begin{bmatrix} 0 & \hat{\mu}_{0e}\vec{E}_0 & 0 \\ \hat{\mu}_{e0}\vec{E}_0^* & 0 & \hat{\mu}_{eb}\vec{E}_0^* \\ 0 & \hat{\mu}_{0e}\vec{E} & 0 \end{bmatrix}. \tag{4}$$

Here, $\hat{\mu}_{eb}$, $\hat{\mu}_{0e}$ stand for transition dipole moments for the transitions between the excitonic state $|e\rangle$ and the bi-excitonic state $|b\rangle$ and the ground state $|o\rangle$ and the excitonic state $|e\rangle$, respectively. Here, the important point to be noted is that $\vec{E}_0 \parallel \hat{\mu}_{ij}$, where $\vec{E}_0$ represents the electric field and $\hat{\mu}_{ij}$ represents the transition dipole moments with $i, j = e, b$, o.

We have taken into account the losses resulting from both radiative and non-radiative decay mechanisms in the current computation. $In_xGa_{1-x}As$ quantum dot has shown uniform broadening in the region of 50 eV at low temperatures, which is due to pure dephasing [5]. Through H_r, these relaxing techniques are implemented as:

$$H_r = \eta\begin{bmatrix} \Gamma_0 & 0 & 0 \\ 0 & \Gamma_e & 0 \\ 0 & 0 & \Gamma_b \end{bmatrix}. \tag{5}$$

The relaxation terms are substantially thermally sensitive, which may be expressed as [5, 6]:

$$\Gamma_0 \approx 0 \tag{6a}$$

$$\Gamma_e = \Gamma_{00} + pT + \frac{q}{\exp(E_a / k_B T) - 1} \tag{6b}$$

$$\Gamma_0 = \Gamma_{11} + p_1 T + \frac{q}{\exp(E_a / k_B T) - 1}\,. \tag{6c}$$

In Eqs. (6), Γ_0, Γ_e and Γ_b represent the dephasing coefficients accordingly to the ground state, the excitonic state, and the bi-excitonic state, respectively. Here, E_a, Γ_{00}, Γ_{11}, a, a_1 and b are taken from the experimental observation of Bori and co-workers [5, 6] as:

$E_a = 16\,\text{meV}$, $\Gamma_{00} = 0.67\,\mu\text{eV}$, $\Gamma_{00} = 2\,\mu\text{eV}$,
$p = 0.22\mu\text{eV/K}$, $q = 1.1\,\text{meV}$, and
$p_1 = 0.37\mu\text{eV/K}$.

The Schrodinger's equation of motion gives the following representation of the QD- radiation interaction:

$$\dot{\rho} = -\frac{i}{\eta}[H,\rho] - \frac{1}{\eta}\{H_r,\rho\}, \tag{7}$$

where the density matrix is represented by

$$\rho = \begin{pmatrix} \rho_{00} & \rho_{0e} & \rho_{0b} \\ \rho_{e0} & \rho_{ee} & \rho_{eb} \\ \rho_{b0} & \rho_{be} & \rho_{bb} \end{pmatrix}. \tag{8}$$

The 0th order thermally sensitive density distribution functions corresponding to the ground state, excitonic state, and the bi-exciton state are defined as:

$$\rho_{00}^0 = 1 - \rho_{ee}^0 - \rho_{bb}^0, \tag{9a}$$

$$\rho_{ee}^0 = \exp\left(\frac{-\eta\Delta\omega_x}{k_B T}\right), \tag{9b}$$

$$\rho_{bb}^0 = \exp\left(\frac{-\eta\Delta\omega_{xx}}{k_B T}\right). \tag{9c}$$

In Eqs. (9a) – (9c), $k_B T$ stands for the thermal energy.

Using Eqs. (3)–(9), several ordering of the density matrix elements are determined, such as:

$$\rho^{(1)} = \begin{pmatrix} 0 & \frac{2\Omega_e(\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{0e}^+} & 0 \\ \frac{-2\Omega_e(\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{0e}^-} & 0 & \frac{2\Omega_b(\rho_{ee}^0 - \rho_{bb}^0)}{\Delta_{eb}^+} \\ 0 & \frac{-2\Omega_b(\rho_{ee}^0 - \rho_{bb}^0)}{\Delta_{eb}^-} & 0 \end{pmatrix} \tag{10}$$

$$\rho^{(2)} = \begin{pmatrix} A & 0 & B \\ 0 & C & 0 \\ D & 0 & F \end{pmatrix}. \quad (11)$$

In Eqs. (10) and (11), $\Delta_{ij}^{\pm} = \omega \pm \omega_{ij} + i\Gamma_{ij}$ represents the damping incorporated detuning parameter; $\Omega_{ij} = \frac{\mu_{ij} E_0}{2\eta}$ is the Rabi frequency. Also the author obtained

$$A = \frac{4\Omega_e^2}{\omega}\left[\frac{1}{\Delta_{0e}^{+}} + \frac{1}{\Delta_{0e}^{-}}\right](\rho_{00}^0 - \rho_{ee}^0) \quad (12a)$$

$$B = \frac{4\Omega_e^2}{\omega}\left[\frac{-(\rho_{ee}^0 - \rho_{bb}^0)}{\Delta_{0b}^{+}} + \frac{(\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{0e}^{+}}\right] \quad (12b)$$

$$C = \frac{4}{(\omega - 2i\Gamma_e)} \times \left[\left(\frac{1}{\Delta_{0e}^{+}} + \frac{1}{\Delta_{0e}^{-}}\right)(\rho_{00}^0 - \rho_{ee}^0) + \left(\frac{1}{\Delta_{eb}^{+}} + \frac{1}{\Delta_{0b}^{-}}\right)(\rho_{ee}^0 - \rho_{bb}^0)\Omega_b^2\right] \quad (12c)$$

$$D = \frac{4\Omega_e\Omega_b}{\omega}\left[\frac{(\rho_{00}^0 - \rho_{ee}^0)}{\Delta_{0b}^{+}} - \frac{(\rho_{ee}^0 - \rho_{bb}^0)}{\Delta_{eb}^{+}}\right] \quad (12d)$$

$$F = \frac{-4\Omega_b^2}{(\omega - 2i\Gamma_b)}\left[\frac{1}{\Delta_{eb}^{+}} + \frac{1}{\Delta_{eb}^{-}}\right](\rho_{ee}^0 - \rho_{bb}^0). \quad (12e)$$

The photoluminescence emission corresponding to an excitonic state has a population density $|C|^2$ that is directly proportional to it. It should be clear that (i) electrons moving from the ground state to the excitonic state, which results in the production of bound *e-h* pairs at the 1s excitonic state, and (ii) electrons moving from the bi-excitonic state to the excitonic state, lead to the populated excitonic state. In Eq. (12c), the 1st term proportional to $(\rho_{00}^0 - \rho_{ee}^0)$ stands in for the earlier contribution, whereas the 2nd term, which relies on $(\rho_{ee}^0 - \rho_{bb}^0)$, stands in for the later contribution.

3. Results and discussion

We analyse the temperature dependence on $\chi^{(3)}$ nonlinearity in an $In_xGa_{1-x}As/GaAs$ QD exposed to cw coherent radiation in this part by numerically analysing the theoretical formulations mentioned before. The author has examined the photoluminescence spectra of $In_xGa_{1-x}As/GaAs$ single QDs using our technique. $m_e = 0.05m_0$ and $m_{hh} = 0.377\ m_0$ are the material characteristics that were chosen for the numerical analysis [4, 7, 8]. The input field strength $E_0 = 10^7$ Vm^{-1} has been chosen. The bi-excitonic binding energy is determined to be 4.084 meV for the $In_{0.4}Ga_{0.6}As/GaAs$ QD with dimensions of 20×20×5 nm^3.

Eq. (12c) has been used to plot the photoluminescence spectrum of $In_{0.4}Ga_{0.6}As/GaAs$ QD at different temperatures. Figure 1 shows that photoluminescence intensity drops off as temperature rises. This graphic also shows the line broadening with rising temperature and the red-shifting of the photoluminescence peak. S. Sanguinetti et al. [4], who are researching the temperature dependency of the photoluminescence spectrum of an $In_xGa_{1-x}As/GaAs$ QDs rather than wetting layer, also make experimental observations of a related kind of nature.

It has been shown that FWHM of the peaks in the photoluminescence spectrum as a function of temperature in Figure 2 to critically analyse the broadening of the photoluminescence spectrum. This graph demonstrates the FWHM's abnormal character at low temperatures.

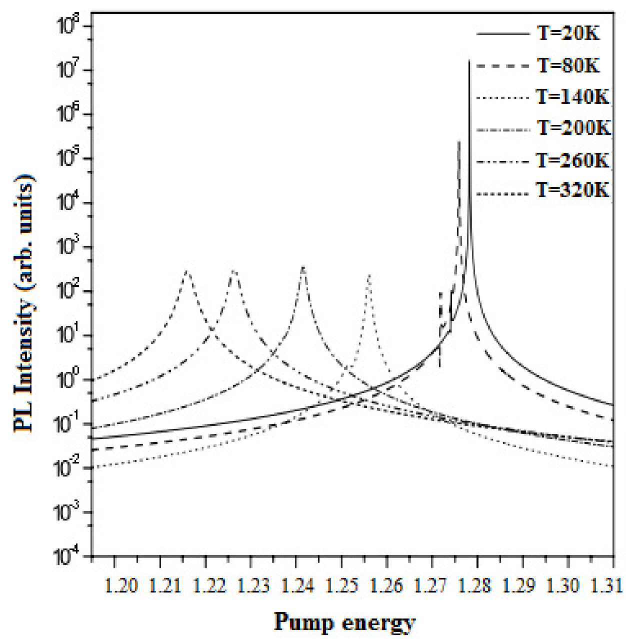

Figure 1. Photoluminescence spectrum of $In_{0.4}Ga_{0.6}As/GaAs$ QD (self assembled) at different temperatures.

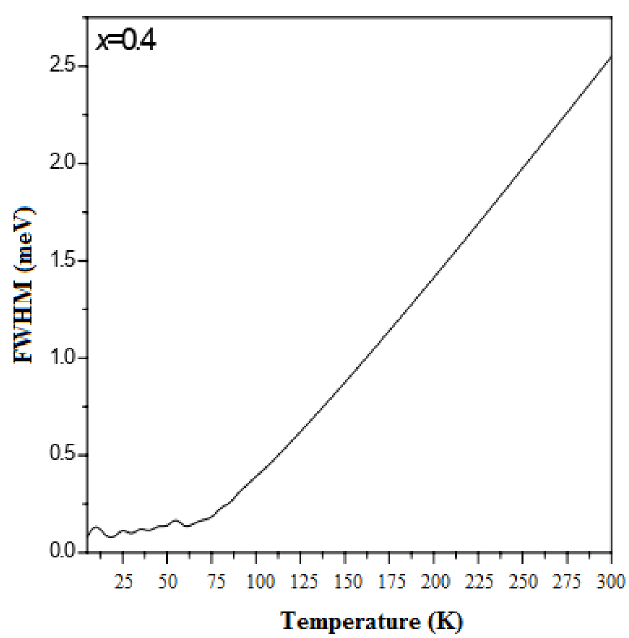

Figure 2. Photoluminescence spectrum's FWHM of $In_{0.4}Ga_{0.6}As/GaAs$ QD (self assembled) as a function of temperature.

In conclusion, FWHM of peaks in the photoluminescence spectrum of $In_xGa_{1-x}As/GaAs$ QDs demonstrate abnormal change with rising temperature whereas the photoluminescence peak undergoes a red-shift with temperature.

4. Conclusions

This study uses a semi-classical density matrix technique to examine how temperature affects the optical characteristics of $In_{0.4}Ga_{0.6}As/GaAs$ QD (self assembled). One can come to the following conclusions thanks to the analysis:

1. As the temperature rises, the photoluminescence intensity drops.
2. At low temperatures, the photoluminescence peaks' FWHM displays an anomalous nature.

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