Temperature dependence of the emission spectra of individual self-assembled quantum dots

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Abstract. We have developed a quantum-mechanical theory for the interaction of light and electron-hole excitations in semiconductor quantum dots. Our theoretical analysis results in an expression for the photoluminescence intensity in the non-linear regime. The validity of the theoretical results is tested analyzing experimental data reported for the temperature dependence of the emission spectra of an individual lens-shaped In\textsubscript{0.4}Ga\textsubscript{0.6}As self-assembled quantum dot in a wide temperature range up to 300 K. Our theoretical predictions for the redshift of the emission peak with increasing temperature, in the range 2-300 K, agree with the experiment.

1. Introduction

Semiconductor quantum dots (QDs) are intensively studied due to their wide optoelectronic applications such as low threshold lasers and optoelectronic detectors. Single or double QDs are excellent candidates for the implementation of quantum bits [1], as single or entangled photons emitters [2, 3], as well as for biological imaging, detection and targeting [4]. Therefore, a detailed understanding of the electronic and optical properties of single QDs and their temperature dependence is of importance.

In a previous work [5] we studied the photoluminescence intensity of a single quantum dot at room temperature and interpreted the experimental findings reported by Matsuda et al. [6]. For the single-particle distribution functions of the single quantum dot we used Fermi-Dirac statistics, as electrons and holes in all the quantum dots of the assembly were at the same equilibrium state, with a uniform Fermi level in the middle of the band gap (for low excitation conditions) and energetic separation of two successive levels lower than the thermal energy. In this case, the calculated sum of the single-particle distribution functions was negligible \(1 - f^\mu - f^\nu \simeq 1\), i.e. linear regime. \(f^\mu\) (\(f^\nu\)) denotes the different electron (hole) states.

In this short article, we present our theoretical findings for the temperature dependence of the photoluminescence intensity of an individual self-assembled quantum dot in the non-linear response regime (where the sum of the single-particle distribution functions \(f^\mu + f^\nu\) obtained using Gibbs statistics, is not negligible compared to unity), and we compare our results with experiment [7].
2. Theory
The total Hamiltonian describing the physical system comprises four terms: the Hamiltonian of the non-interacting electrons and holes, the free-photon Hamiltonian, the electron-hole interaction Hamiltonian, and the interaction of carriers with the background photon-field [8]. Based on the semiclassical density formalism and applying the Heisenberg equation of motion to the photon number expectation values, to the field-matter correlation term, and to the electron (hole) distribution functions \( f^\mu \) (\( f^{\mu*} \)), we obtain a set of four luminescence equations in the non-linear regime, as we did before in the linear regime [5]. Under quasi-equilibrium conditions, this set is reduced to a closed set of two photoluminescence equations, the analytical solution of which leads to the theoretical expression of the PL intensity in the non-linear regime [9].

Within our theoretical framework, (i) we calculate the single-particle energies and wave functions within the effective mass and envelope function approximations [10]. For each temperature, using an expansion -within a periodicity box- of the electron (or hole) envelope functions into the orthonormal plane-wave basis \( \langle k|\varphi^\mu \rangle \), we obtain the eigenenergies \( E^\mu \) as well as the expansion coefficients \( C^\mu_k \) by solving the following eigenvalue problem
\[
\sum_{k'} \langle k|\left(\frac{\hbar^2}{2m^*} + V(r)\right)|k'\rangle - E^\mu \delta_{k,k'} \langle k'|\varphi^\mu \rangle = 0.
\]

(ii) We compute the Coulomb matrix elements between the carriers \( V_{\kappa_1\kappa_2\kappa_3\kappa_4}^{\kappa_1\kappa_2\kappa_3\kappa_4} \) where \( \kappa_i \) denote the different electron or hole states, as in [10].

(iii) We evaluate the eigenenergies \( E^\lambda \) and eigenfunctions \( \Phi^\lambda(r, r) \) of the non Hermitian -in the non-linear regime matrix [9]
\[
S_{\ell\ell'} = E^{ee}_{\mu\mu'} \delta_{\nu\nu'} + E^{hh}_{\mu\mu'} \delta_{\nu\nu'} - V_{\nu\nu'}^{ee}(1 - f^\mu - f^{\nu'}).
\]

Here, we have determined the single-particle renormalized energies as
\[
E^{ee, renorm}_{\mu\mu'} \equiv E^e \delta_{\mu\mu'} - \sum_{\mu_1} V_{\mu\mu_1\mu_1\mu_1}^{ee} f^{\mu_1},
\]
\[
E^{hh, renorm}_{\nu\nu'} \equiv E^h \delta_{\nu\nu'} - \sum_{\nu_1} V_{\nu\nu_1\nu_1\nu_1}^{hh} f^{\nu_1}.
\]

(iv) Finally, we calculate the PL spectra according to:
\[
I_{em}(\omega_q) \sim Im\left[ \sum_{\lambda} \int d^n r \Phi^\lambda(r, r) \int d^n r' \sum_{\mu} \sum_{\nu} C^\lambda_{\mu\nu} \varphi^\mu(\mathbf{r})\varphi^\nu(\mathbf{r}') f^\mu f^{\nu'} E^\lambda - i\gamma - \hbar \omega_q \right].
\]
\( C^\lambda_{\mu\nu} \) are the components of the expansion of the excitonic eigenfunction \( \Phi^\lambda \) in terms of the non-interacting single-particle basis [5], \( \hbar \omega_q \) is the photon energy, \( \ell \equiv \mu \nu \), and \( \gamma \) is a small damping constant that accounts for the dephasing due to phonon scattering [5].

3. Results and discussion
We use our theoretical results to interpret the experimentally observed PL peak positions as a function of temperature, reported by Bayer and Forchel [7]. The QD under study is an individual lens-shaped In\(_{0.4}\)Ga\(_{0.6}\)As self-assembled quantum dot, with diameter at the base 24 nm, height 3.8 nm, and thickness of the wetting-layer 0.5 nm, containing one electron-hole pair (low excitation conditions). The material parameters used for In\(_{0.4}\)Ga\(_{0.6}\)As quantum dot at 4 K and 300 K are listed in Table 1. The In\(_{0.4}\)Ga\(_{0.6}\)As and GaAs band gaps shrinkage can be roughly described by the Varshni law
\[
E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta}
\]
Table 1. Material parameters used for the In_{0.4}Ga_{0.6}As quantum dot. The effective masses are expressed in units of the free-electron mass $m_0$. $\varepsilon$ is the static dielectric constant. The low and high temperature values of the effective masses and of the dielectric constant are obtained by Ref. [11].

| $T$ (K) | $m_e$   | $m_{hh}$ | $\varepsilon$ |
|---------|---------|----------|--------------|
| 4       | 0.067   | 0.34     | 12.38        |
| 300     | 0.0632  | 0.5      | 13.18        |

Figure 1. Comparison of the calculated PL peak position as a function of temperature, with the experimental one [7]. The temperature dependence of the In_{0.4}Ga_{0.6}As band gap according to Varshni law is also depicted. To make the comparison clear, the values of $E_g(T)$ have been shifted so that all PL peaks for $T = 2$ K coincide.

The Varshni coefficients of GaAs and InAs were taken from Ref. [12], while for In_{0.4}Ga_{0.6}As we use the interpolated values. The temperature dependence of the conduction and valence band-offsets is obtained from the temperature dependence of the band-gap discontinuity according to $V_{BO} = Q[E_{GaAs}^g(T) - E_{InGaAs}^g(T)]$ with $Q_c = 0.67$ and $Q_v = 0.33$ for the conduction and the valence band-offset respectively. The 3D confining potential is zero inside the wetting layer and the self-assembled dot, and $V_{CBO}$ (or $V_{VBO}$) inside the barrier. We find that three confined shells exist, as it has also been reported by Ref. [7]. For the derivation of the spectra, we set $E_{InGaAs}^g(0) = 1.132$ eV, so that the theoretical and experimental PL peaks at $T = 2$ K coincide. Moreover, we fit the $\gamma$ values in order to have the experimentally observed FWHM’s.

Figure 1 depicts the theoretical PL peak energies as a function of temperature, compared with the experimentally observed. The theoretically derived PL peak energies are in agreement with the experimental ones. We find that the PL peak position energy follows the band gap temperature variation for low temperatures, while for higher temperatures, there is a small deviation, due to the single particle energies variation and the carriers occupation probabilities variation with temperature.
4. Conclusions
In summary, based on the theoretically derived expression for the photoluminescence intensity of individual quantum dots in the non-linear regime, we investigate the temperature dependence of the PL peak position energy. The theoretical results interpret the experimental data consistently. The PL peak position energy follows the band gap temperature variation for low temperatures. However, for higher temperatures, corrections should be made, due to the temperature variation of the single particle energies and of the term \((1 - f^\mu - f^\nu)\).

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