Magnetic-field effects on exciton–polaron in a CdSe quantum disk

K. Sellami\textsuperscript{a}, S. Jaziri\textsuperscript{a,b,*}

\textsuperscript{a}Laboratoire de Physique des Matériaux, Faculté des Sciences de Bizerte, 7021 Jarzouna, Tunisia
\textsuperscript{b}Laboratoire de Physique de la Matière Condensée, Faculté des Sciences de Tunis, Tunisia

Received 17 February 2004; received in revised form 22 June 2004; accepted 22 June 2004
Available online 3 September 2004

Abstract

We study theoretically the interaction between excitons and longitudinal optical (LO) phonons in a cylindrical disk-like semiconductor quantum dot under an applied magnetic field. Due to the intensity of the interaction in the strong coupling regime, a composite quasi-particle called exciton–polaron is formed. We focus on the effect of the disk size and an external magnetic field on the exciton–phonon interaction energy and the exciton–polaron modes. The numerical computation for a CdSe quantum disk have shown that the exciton–phonon interaction energy is very significant and is even dominant when the disk height is small, which leads to a large Rabi splitting between the exciton–polaron modes. We investigate also the effect of the temperature on the integrated photoluminescence (PL) intensity, and show that at relatively high temperature the LO phonons have a noticeable effect on it. This physical parameter also shows a great dependence on quantum disk size and on magnetic field.

© 2004 Published by Elsevier Ltd

Keywords: Semiconductor quantum dots; Exciton; Exciton–phonon interaction; Polaron

In recent years a great deal of interest has been devoted to the study and the engineering of high-quality devices of very low dimension, essentially quantum-well, wires, or quantum-dot (QD) semiconductors [1–14]. Because of their zero-dimensionality,
The quantum dots exhibit many new physical effects [15] which are extremely interesting from the point of view of fundamental physics and also for their potential applications in microelectronic device technology. Consequently much effort has lately gone into understanding and exploring the physical properties of these systems both theoretically and experimentally. The effects of an applied magnetic field on the physical properties of quantum dots have been studied with interest from the theoretical and experimental points of view. These studies have been performed with the proposal of understanding the fascinating novel phenomena and of fabricating devices with new functions or to improve the performance of the existing devices [16]. Excitons play a dominant role in their physical properties; therefore, their stability is important for possible devices requiring this characteristic [17]. The properties of confined excitons have been the subject of many theoretical studies. Bryant [18] used variational and configuration representations to study excitons in quantum boxes. Later, matrix diagonalization techniques were used to study the exciton energy in a quantum dot with a parabolic confinement potential. Song et al. [19] studied the effect of non-circular symmetric structures, and Halonen et al. [20] studied the influence of a magnetic field. More recently, Pereyra et al. [21] investigated magnetic field and quantum confinement asymmetric effects on excitons, again for the case of parabolic confinement. These studies have shown a strong competition between the quantum dot size, Coulomb interaction and magnetic confinement.

In semiconductor quantum dots, the electron–phonon interaction is usually much less effective for relaxation processes of electrons than in the bulk. The polaronic process has become a main research subject in the physics of low-dimensional systems [22–26]. A remarkable exception is when the energies of the longitudinal optical (LO) phonons match the level separation, in this case the electron–phonon interaction can be significantly strong. More recently, many papers have been devoted to this process and to its influence on various physical properties of polar semiconductor quantum wells and quantum dots [23–34]. Some recent optical measurements [27–29,35–39] of the photoluminescence spectra realized on different quantum dots and quantum-well semiconductor structures, also reveal the LO phonon’s effect on the PL line widths. When a spacing between the electron levels is tuned to match the energy of LO phonons by applying magnetic fields, the Rabi splitting of the coherent states has been observed in the absorption spectra.

Since II–VI materials have high ionicity, it is natural to expect the existence of electron and hole polaron states [40]. The comparison of polaron effects in different nanostructures have shown that the maximum polaron effect is in the quantum dot [41].

In this paper we study the exciton–LO phonon interaction of the Fröhlich type in a disk-shaped CdSe semiconducting quantum dot under an applied magnetic field. We theoretically examine the situation where the polarons are observed by the application of a magnetic field. In the strong coupling regime, we calculate the exciton eigen states and eigen energies, then we determine for different disk radius \( R \) and different values of the ratio \( h/R \), where \( h \) denotes the disk-cylinder height, the typical quantum disk size where the strong coupling regime can occur. In this non-adiabatic case a new quasi-particle called exciton–polaron is formed due to the strong interaction between carriers (electron, hole, or exciton) and phonons [42–44]. We focus our analysis on the study of the anti-crossing between the first excitonic state and the ground state plus one LO phonon, such states are directly coupled by the Fröhlich Hamiltonian. We calculate in a two level model the
exciton–polaron modes for different disk radii, and examine also the effect of disk size and an external magnetic field on the exciton–phonon interaction energy and the Rabi splitting between the polaron modes. This calculation leads us to evaluate the effect of such a type of interaction on the integrated photoluminescence (PL) intensity, which represents one of the important parameters characterizing the optical properties of semiconductor structures.

In the effective-mass approximation, the Hamiltonian describing an exciton interacting with LO phonons is given by: \( H = H_{\text{ex}} + H_{\text{LO}} + H_{\text{ex-LO}} \), where \( H_{\text{ex}} \) is the Hamiltonian of an exciton in a parabolic quantum dot under an applied uniform magnetic field, normal to the \( x-y \) plane \( \mathbf{B} = (0, 0, B) \). With the symmetric gauge for the magnetic field, the potential vector \( \mathbf{A} = (B/2)(-y, x, 0) \), the Hamiltonian of the exciton in the vertical quantum dot then reads

\[
H_{\text{ex}} = \frac{p_{e||}^2}{2m_{e||}^*} + \frac{p_{e\perp}^2}{2m_{e\perp}^*} + \frac{p_{h||}^2}{2m_{h||}^*} + \frac{p_{h\perp}^2}{2m_{h\perp}^*} + \frac{1}{2} m_e^* \omega_e^2 r_{e||}^2 + \frac{1}{2} m_h^* \omega_h^2 r_{h||}^2 + \frac{1}{2} m_e^* \omega_e^2 z_e^2 + \frac{1}{2} m_h^* \omega_h^2 z_h^2 + \frac{\omega_e}{2} l_{ez}^2 - \frac{\omega_h}{2} l_{hz}^2 + \frac{1}{2} \left( \frac{\omega_e^2 - \omega_h^2}{\epsilon_\infty(\epsilon_\infty - \epsilon_0)} \right) r_{e||}^2 r_{h||}^2, \tag{1}
\]

where \( m_{e||}(m_{e\perp}) \) and \( m_{h||}(m_{h\perp}) \) are the effective electron and heavy-hole masses for in-plane \((z)\) motions [45]. We focus our attention to the heavy hole (hh) exciton neglecting the coupling between heavy hole and light hole states on account of the strain effects and large energy separation between the zone center states (sub-levels) in small dots;

\[
\omega_{e(h)} = \sqrt{\omega_{e(h)0}^2 + \frac{\omega_{ce(h)}^2}{4}}
\]

is the electron (hole) frequency; \( \omega_{e(h)} = \frac{eB}{cm_{e(h)}} \) and \( \omega_{ce(h)} = \frac{eB}{cm_{h||}} \) are the cyclotron frequencies for the electron and the hole respectively; and \( \omega_{ez}, \omega_{hz}, l_{ez} \) and \( l_{hz} \) are respectively the frequencies and orbital angular momenta along the \( z \)-direction of the electron and the hole.

The optical phonon Hamiltonian is given by \( H_{\text{LO}} = \hbar \omega_{\text{LO}} \sum_q a_q^\dagger a_q \), where each eigenstate \(|\{n_q\}_q\rangle\) corresponds to the energy \( \hbar \omega_{\text{LO}} \), with \( a_q^\dagger \) and \( (a_q) \) being the phonon creation and annihilation operators, and \( q \) is the LO phonon of the wave vector. The interaction Hamiltonian is written as

\[
H_{\text{ex-LO}} = -\frac{\hbar A_f}{q \sqrt{\pi \hbar V N}} \sum_q \left[ a_q^\dagger (e^{iqr_e} - e^{iqr_h}) - a_q (e^{-iqr_e} - e^{-iqr_h}) \right],
\]

where the factor

\[
A_f = \sqrt{\frac{\hbar \omega_{\text{LO}}}{2 \left( \epsilon_\infty - \epsilon_0 \right)}},
\]

it can be expressed as a function of the Fröhlich constant (dimensionless), \( \alpha \) which is related to the strength of the macroscopic polarization field induced by the ion displacements within an elementary cell (CdSe). \( \epsilon_0(\epsilon_\infty) \) is the static (high-frequency) dielectric constant, and \( V \) is the quantum disk volume.
We have evaluated the interaction $H_{\text{ex–LO}}$ form factors $W_{nm'}(q)$ (the indexes $n$ and $n'$ replace both $n_e, m_e$ and $n_h, m_h$ quantum numbers) taking the function of the non-interacting electron and hole system as the first approximation for the exciton wave function $\Phi_{nm}(r_e, r_h) = \psi^{(e)}_{n_e, m_e}(r_e) \psi^{(h)}_{n_h, m_h}(r_h)$, with

$$
\psi^{(i)}_{nm}(r) = \psi^{(i)}_{nm}(r_1, \varphi) \phi^{(i)}(z) = N_{nm}^{(i)} e^{(-i q \varphi)} I_n \left( \frac{r^2}{2 R_{(i)}^2} \right) e^{-i \frac{q^2 x^2}{2 \pi^2}}, \quad \text{(where } i = e, h)$$

where

$$
N_{nm}^{(i)} = \sqrt{\frac{n_1!}{2 \pi R_{(i)}^2 2^{|m|} R_{(i)}^{2|m|} (n + |m|)!}}.
$$

and $R_{e(h)}$ is the effective disk radius under the applied magnetic field effect for the electron (hole), it is written as

$$
R_{e(h)} = \sqrt{\frac{h}{m_{e(h)} \sqrt{\omega_{e(h)}/2}^2 + (\omega_{e(h)}/2)^2}}.
$$

Since $\omega_0 \ll \omega_c^2$, we consider only the first level for the quantization in the $z$-axis direction.

To calculate the exciton eigen states and eigen energies, we have to calculate the Coulomb matrix elements. Introducing $V(q)$, the Fourier transform of the Coulomb interaction in the real space $V(r_2 - r_1)$, the Coulomb matrix elements can be written as: $V(n_e m_e, n_h m_h, n'_e m'_e, n'_h m'_h) = \int d^3 q V(q) I_1(q) I_2(q)$ where $I_1(q) = \int d^3 r_1 \psi_{n_e m_e}^* \psi_{n'_e m'_e} e^{-i q \cdot r_1}$ and $I_2(q) = \int d^3 r_2 \psi_{n_h m_h}^* \psi_{n'_h m'_h} e^{i q \cdot r_2}$. Finally we find that they can be expressed in the following form:

$$
V(n_e m_e, n_h m_h, n'_e m'_e, n'_h m'_h) = (2\pi)^2 N_{n_e, m_e} N_{n_h, m_h} N_{n'_e, m'_e} N_{n'_h, m'_h},
$$

$$
\times \left( \frac{-1}{i! j! k! l!} \right) \frac{1}{2 R_{(e)}^2} \frac{1}{2 R_{(h)}^2} 2^{\mu + \nu - \rho - \sigma - 2} R_{(e)}^{\mu + \nu} R_{(h)}^{\rho + \sigma} \Gamma \left( \frac{\mu - \nu}{2} \right) \Gamma \left( \frac{\rho - \sigma}{2} \right) \int_0^{\infty} dq_2 \int_0^{\infty} dq V(q) q^{\mu + \nu + 1} L_{\mu - \nu - 1}^1 \left( \frac{R_{(e)}^2 q_2^2}{2} \right) \frac{L_{\rho - \sigma - 1}^1}{q_2^2}.
$$

$$
\times \left( \frac{R_{(e)}^2 q_2^2}{2} \right) \exp \left[ -\frac{q_2^2}{2} \left( R_{(e)}^2 + R_{(h)}^2 \right) \right] \exp \left[ -\frac{q_2^2 h^2}{2} \right].
$$

(2)
Fig. 1. Variation of the energy difference between the first excitonic state and the ground state as a function of the disk radius for four values of the magnetic field $B = 0 \, \text{T}, 5 \, \text{T}, 10 \, \text{T}, 20 \, \text{T}$. The dashed lines correspond to the one and two optical phonons energies.

where $\mu \equiv |m_e| + |m'_e| + 2(i + j + 1)$, $\nu \equiv |m_e - m'_e|$, $\rho \equiv |m_h| + |m'_h| + 2(k + p + 1)$, and $\sigma \equiv |m_h - m'_h|$; (the indexes $m_e, m'_e$ and $m_h, m'_h$ denotes respectively the electron and hole quantum numbers).

The strong coupling regime is obtained when the energy distance between the electronic levels in a quantum dot approximately matches the LO phonon energy. In this non-adiabatic case a new quasi-particle, the polaron is formed due to the strong interaction between carriers (electron, hole, or exciton) and the phonon bath. The effect of the non-adiabaticity was shown to be among one of the factors that enhances the electron–phonon interaction in quantum dots [42].

Fig. 1 displays the energy difference between the first excitonic excited state and the ground state versus the quantum disk radius, for a fixed ratio $h/R = 0.2$, and for various magnetic field values. We note that this excitonic energy difference, for quantum disk radius ranging from 27 to 35 Å, is located between the energy of one LO phonon and two LO phonons. This means that for this quantum disk radius domain, the strong coupling regime can be obtained. The effect of the magnetic field does not change significantly this radius domain. We can interpret this result by the fact that the magnetic field effect on the charge carriers is more pronounced for large quantum dots. Hence the typical radius for such a CdSe quantum disk, where the exciton LO phonon strong coupling regime occurs is around 35 Å. Given the confinement effect of the magnetic field on the charge carriers in the quantum dot, the energy difference between the first excitonic excited state and the ground state, in a given quantum dot with a fixed (even relatively large) radius, can be tuned by increasing the magnetic field to match the LO phonon energy and realize the strong coupling regime.
The polaron modes are either following equation:

\[
\gamma
\]

interaction within the strong coupling regime; therefore the quantum dot quantum with the radius essentially exceeding the height. We notice that the intensity of the interaction and since a quantum disk is a cylindrical quantum dot (3) to investigate the quantum disk size and especially the height on the intensity of the interaction increases considerably as the quantum disk increases. We find also that the interaction is more pronounced for quantum disks of small height. In our work we study the exciton–phonon interaction within the strong coupling regime; therefore the quantum disk radius we are interested in will not exceed 40 Å.

Then the considered exciton–phonon interaction element has the form:

\[
W = \sqrt{\sum_q |V_{S,h}(q)|^2}.
\]

We perform our calculations using the following material parameters suitable to the CdSe QD [46]: \( m_e^* = 0.11 m_0 \), \( \epsilon_\infty = 6.23 \), \( \epsilon_0 = 9.56 \), \( \alpha = 0.34 \), the Luttinger parameters \( \gamma_1 = 2.04 \), \( \gamma_2 = 0.58 \) and \( \hbar \omega_{LO} = 26.57 \) meV.

In Fig. 2 we plot the interaction term as a function of the disk radius for different \( \hbar/R \) ratios (1/5), (1/8), (1/10), and (1/15) to investigate the quantum disk size and especially the height on the intensity of the interaction. We notice that the intensity of the interaction increases considerably as the quantum disk increases. We find also that the interaction is more pronounced for quantum disks of small height. In our work we study the exciton–phonon interaction within the strong coupling regime; therefore the quantum disk radius we are interested in will not exceed 40 Å.

The polaronic excitation resulting from the coupling of the first excitonic and the ground state plus one LO phonon can be described by a two level model that takes into account these interacting states. Within this approximation the polaron modes are the solution of the following equation:

\[
\det \begin{pmatrix}
E_S + \hbar \omega - \epsilon & W \\
W^* & E_{P-} - \epsilon
\end{pmatrix} = 0.
\]

The first states of the exciton–polaron spectrum are analytically calculated and given in Table 1 with \( d = 1 \) (the space degeneracy of \(|S_0\rangle\)). The states \(|S, l_q\rangle\) and \(|S, 0_q\rangle\) are identical to the uncoupled states while the \(|\pm\rangle\) represent coupled phonon/exciton states. \( d \) is the space degeneracy of the \( P \) states (\( d = 2 \) if \( B = 0 \), and \( d = 1 \) if \( B \neq 0 \)).

In Fig. 3 we plot the energies of the first exciton state and the ground state plus one LO phonon that corresponds respectively to the \(|P_-, 0_q\rangle\) and \(|S, l_q\rangle\) as a function of the disk radius. We notice that around \( R = 35 \) Å the anti-crossing between the polaron branches corresponds to a large Rabi splitting \( R_S = 21.5 \) meV, and for \( R < 35 \) Å, or \( R > 35 \) Å the polaron modes are either \(|P_-, 0_q\rangle\) like or \(|S, l_q\rangle\). The large Rabi splitting is expected given the intensity of the exciton–phonon in the CdSe quantum disk. It would increase...
significant when reducing the height of the disk since the interaction increases as the ratio $h/R$ decreases.

One of the most recent interests in the area of quantum disk physics has been to investigate the role of carrier charge (electron and hole) interaction not only on the electronic properties, but also on optical ones. The investigation of emission spectra of such structures at different temperatures leads to an understanding of the exciton–phonon coupling.

In relation to the optical experience on these quantum disk structures, integrated photoluminescence intensity is one of the physical parameters able to draw a good picture of their optical properties. The dependence of integrated PL intensity gives by the Arrhenius model [35,47]:

$$\frac{I(T)}{I(0)} = \frac{1}{1 + C_1 \exp \left(-\frac{E_1}{kT}\right) + C_2 \exp \left(-\frac{E_2}{kT}\right)},$$

(5)
Fig. 3. Calculated $R$ dependences of exciton–polaron energy differences $\varepsilon_{|+\rangle} - \varepsilon_{|S_0\rangle}$ and $\varepsilon_{|−\rangle} - \varepsilon_{|S_0\rangle}$ (dotted lines) for a CdSe quantum disk with a fixed ratio $(h/R = 0.2)$ and a fixed magnetic field value $B = 10$ T.

where $I(T)$ and $I(0)$ are the integrated PL intensities emissions at temperatures $T$ and $0$ K, respectively, $E_1$ and $E_2$ are activation energies, and $C_1$ and $C_2$ are constants characterizing the ratio of non-radiative recombination rates.

\[
E_1 = E_e + E_h - E_{\text{ex−polaron}},
\]

\[
E_2 = E_e + E_h + E_{e-LO} + E_{h-LO},
\]

where $E_e$ and $E_h$ are respectively, the electron and hole confinement energies, and $E_{e-LO}$ and $E_{h-LO}$ are the coupling energies between electron and hole with LO phonons, respectively, and $E_{\text{ex−polaron}}$ is the exciton–polaron energy.

Eq. (5), giving the analytical expression of the integrated PL intensity, shows the dependence of this coefficient not only upon the temperature fluctuation, but also upon the activation energies $E_1$ [Eq. (6)] and $E_2$ [Eq. (7)], and the constants $C_1$ and $C_2$, characterizing the ratio of the non-radiative recombination rates. Let us recall that the interaction of the exciton with optical phonons affects only the activation energies $E_1$ and $E_2$, and thus the PL coefficient. Hence it is convenient to know the effect of this type of interaction on the integrated PL intensity curve in terms of temperature. In order to achieve this aim and for the sake of simplicity to obtain the basic information about this process, we take the constants $C_1$ and $C_2$ as equal units.

Figs. 4 and 5 display the behavior of the integrated photoluminescence intensity versus the temperature inverse variation with LO phonon modes, of hh excitons in CdSe quantum disks. In Fig. 4(a) and (b), respectively, the integrated PL intensity is depicted for CdSe quantum disks of height $h = 0.2a_{\text{ex}}^*$ and values of radius $R = 1a_{\text{ex}}^*$, $1.5a_{\text{ex}}^*$, and $2a_{\text{ex}}^*$ (the results are presented in units of the effective Bohr radius $a_{\text{ex}}^*$, which is equal to 38.5 Å). We note that in both cases the magnetic field intensity is assumed to be fixed at the value 1 T.
Fig. 4. Arrhenius plot of the integrated PL intensity using Eq. (3) in the case of CdSe as a function of $T^{-1}$ for an external magnetic field value $B = 1$ T. (a) $h = 0.2a_{ex}^*$ for three values of the dot radius $R = 1a_{ex}^*, 1.5a_{ex}^*, 2a_{ex}^*$ and (b) $R = 1a_{ex}^*$ for three values of the disk height $h = 0, 0.1a_{ex}^*, 0.15a_{ex}^*, 0.2a_{ex}^*$. 

A first analysis of these curves leads to the conclusion that at relatively high temperature and for hh excitons, the LO phonons have a noticeable effect on the PL intensity. Moreover it is convenient to note that the height $h$ of the quantum disk has a great influence on the PL spectra versus the temperature variation. We observe that at high temperature the intensity ratio $I(T)/I(0)$ decreases more rapidly with the increasing of the height [Fig. 4(b)] than of the disk radius $R$ [Fig. 4(a)].
In Fig. 5 we examine the effect of an external applied magnetic field on the integrated photoluminescence behavior versus the temperature inverse, and that for a given quantum disk size namely $R = 1a_{\text{ex}}^*$, and $h = 0.2a_{\text{ex}}^*$. We justify this choice by the fact that for such a disk radius the polaron effect is at its maximum, since it corresponds to the observed anti-crossing between the first exciton state and the ground state plus one LO phonon at resonance. We notice that the magnetic field influences significantly the curves. For a fixed temperature the effect of the magnetic field is to increase the $I(T)/I(0)$ ratio that reaches unity more rapidly. Hence, the calculation of the integrated PL intensity versus the temperature variation, and including the effect of an external magnetic field, shows the noticeable effect of the LO phonon mode on the PL spectra. These phonon modes are very important, and must be taken into account in all electronic or optical investigations.

References
