

Temperature Dependence of the Confined Exciton States in CdTe/Cd_{1-x}Zn_xTe Cylindrical Quantum Dots

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In the framework of the effective-mass approximation and using a variational approach, we present a systematic study of the excitonic states in CdTe/Cd_{1-x}Zn_xTe cylindrical quantum dot as a function of the dot size, temperature and composition. The photoluminescence line intensity temperature dependence is also investigated. Our results show that the stability of the exciton is more influenced by the dot size and temperature factors. Furthermore, the photoluminescence intensity depends strongly on the temperature, dot size and composition.

1. Introduction

In recent years, a great attention has been devoted to the study and engineering of high-quality devices of very low dimensions, such as quasi-two-dimensional quantum wells (QW), quasi-one-dimensional quantum well wires (QWW) and quasi-zero-dimensional quantum dots [1–14]. Correlated electron-hole pairs form excitons in semiconductor heterostructures. It is well known that spatial confinement in semiconductor nanostructures increases the overlap of the electron and hole wave functions and then causes an enhancement of the electron-hole interaction. Therefore an enhancement of the exciton binding energies is expected as the dimensionality of these structures is reduced. The binding energy increases when going from bulk to quantum well systems, and then to quantum

well wire and quantum dots systems. In view of rapid development in crystal growth and nanofabrication technology it becomes increasingly necessary to address the concern of the exciton losing its enhanced effects in the ultrasmall quantum structures, due to the increased penetration of the exciton wave function into the barrier materials in the direction of the decreasing spatial confinement. There has been much interest in the study of electronic and optical properties in quantum dots where the charge carriers are confined in all three dimensions. The properties of confined exciton in those systems have been the subject of many theoretical works [15–20]. It was shown in Refs. [21–22] that the excitons in low-dimensional semiconductor structures play much more important role compared to excitons in bulk semiconductors. Dramatic impact can be expected on the optical properties of such structures even at room temperature. Let us mention that the authors of Refs. [15–16] have used variational approach and effective-mass approximation to investigate properties of excitons in cylindrical quantum dots and quantum discs structures.

Temperature effect, on the binding energy of a hydrogenic donor impurity [23–24] and photoluminescence line intensity, is one of the interesting physical parameters which are essential to understand the experimental observation of the semiconductor optical spectra [25–28]. Some recent optical measurements [29–35] of the photoluminescence spectra realized on different quantum dot and quantum well structures reveal also the dependence of the luminescence and photoluminescence line widths on the dot sizes and temperatures. In this work we report a variational calculation in the effective mass approximation of the exciton binding energy in CdTe cylindrical quantum dot (well material), of radius R and height H embedded in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (barrier material). The quantum confinement induced by the barrier material is described by a finite depth potential well. The effect of the temperature on the light (heavy) hole exciton binding energy for different values of the QD sizes and the potential level is investigated. Using the exciton binding energy dependence on temperature, the effect of QDs sizes and potential level induced by barrier material on the integrated photoluminescence PL intensity is evaluated. Let us notice that this PL intensity is one of the important parameter characterizing the optical properties of semiconductor structures. The paper is organized as follows. After a brief introduction we present in Section 2 the Hamiltonian of the system and calculation method. In Section 3 we describe some numerical results for CdTe/ $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ systems and we summarize by a conclusion in Section 4.

2. Theory

We consider a confined exciton in a cylindrical quantum dot of radius R and height $H = 2d$ (well material) embedded in barrier material semiconductors, in the non-degenerate band approximation. The effective mass Hamiltonian of this exciton at temperature T can be written as follows:

$$H_{ex}(r_e, r_h, T) = -\frac{\hbar^2}{2m_e^*} \nabla_e^2 - \frac{\hbar^2}{2m_h^*} \nabla_h^2 - \frac{e^2}{\varepsilon_0(T)|r_e - r_h|} + V_w^e(r_e, T) + V_w^h(r_h, T), \quad (1)$$

where m_e^* and m_h^* are the effective masses of the electron and hole, respectively, and $r_e = (\rho_e, z_e)$ and $r_h = (\rho_h, z_h)$ are the spatial coordinates of the electron and hole, respectively. $V_w^e(r_e, T)$ ($V_w^h(r_h, T)$) is the corresponding electron (hole) confining potential at temperature T :

$$V_w^i(r_i, T) = \begin{cases} 0, & \text{if } \rho_i \leq R \text{ and } |z_i| \leq d \\ V_i(T) & \text{elsewhere, } (i = e, h) \end{cases}, \quad (2)$$

where $-e^2/\varepsilon_0(T)|r_e - r_h|$ is the Coulomb potential and $\varepsilon_0(T)$ is the static dielectric constant at temperature T .

$V_i(T)$ is obtained from the temperature dependent band-gap discontinuity. Since there is a strain effect, the conduction and hole valence band offsets are a complex function of the temperature and concentration x . In this work the temperature dependence of the energy band gaps of group IV and III-V semiconductors is given by Varshni formula [36] $E_g(T) = E_g(0) - aT^2/(T+d)$, where d is the Debye temperature and a is a fitting parameter. From the reference [37], the valance band offset equals $20\% \Delta E_g$. We can then write the expressions for $V_e(x, T)$ and $V_h(x, T)$ as follows:

$$V_e(x, T) = 0.80 \Delta E_g(x, T), \quad V_h(x, T) = 0.20 \Delta E_g(x, T), \quad (3)$$

where

$$\Delta E_g(x, T) = E_{g \text{ CdTe/Cd}_{1-x}\text{Zn}_x\text{Te}}(x, T) - E_{g \text{ CdTe}}(x, T). \quad (4)$$

To deal with the Hamiltonian of this system, we shall adopt the variational treatment for quasi-zero-dimensional systems developed in reference [15]. The effective Hamiltonian, in the atomic units system ($a_{ex}^* = \varepsilon_0(0) \frac{\hbar^2}{\mu e^2}$ is the excitonic Bohr radius and $R_{ex}^* = \mu e^4 / 2\varepsilon_0^2(0) \hbar^2$ is the Rydberg energy), reads

$$H_{eff} = -\frac{1}{1+\sigma} \left[\frac{\partial^2}{\partial \rho_e^2} + \frac{\partial}{\rho_e \partial \rho_e} + \frac{\rho_{eh}^2 + \rho_e^2 - \rho_h^2}{\rho_e \rho_{eh}} \frac{\partial^2}{\partial \rho_e \partial \rho_{eh}} + \frac{\partial^2}{\partial z_e^2} \right] - \quad (5)$$

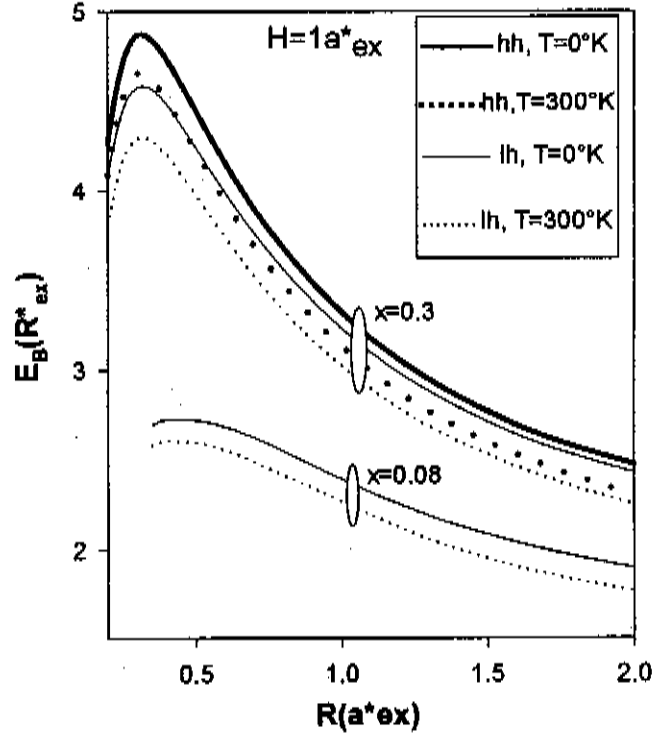


Figure 1. Binding energy of the light (lh) and heavy (hh) hole excitons in a CdTe QD as a function of the cylinder radius R for a fixed height $H = a_{ex}^*$, two values of temperature T and zinc concentration x .

$$\begin{aligned}
 &= -\frac{\sigma}{1+\sigma} \left[\frac{\partial^2}{\partial \rho_h^2} + \frac{\partial}{\rho_h \partial \rho_h} + \frac{\rho_{eh}^2 + \rho_h^2 - \rho_h^2}{\rho_h \rho_{eh}} \frac{\partial^2}{\partial \rho_h \partial \rho_{eh}} + \frac{\partial^2}{\partial z_h^2} \right] - \\
 &\quad - \left[\frac{\partial^2}{\partial \rho_{eh}^2} + \frac{\partial}{\rho_{eh} \partial \rho_{eh}} \right] - \frac{\epsilon_0(0)}{\epsilon_0(T)} \frac{2}{\sqrt{\rho_{eh}^2 + (z_e - z_h)^2}} + \\
 &\quad + V_w^e(\rho_e, z_e, T) + V_w^h(\rho_h, z_h, T),
 \end{aligned}$$

where $\sigma = m_e^*/m_h^*$ and $\epsilon_0(0)$ is the static dielectric constant for QD at temperature $T = 0$ K. In order to calculate the exciton binding energy, we choose the following wave function [15]:

$$\psi_{ex}(\rho_e, \rho_h, z_e, z_h, T) = F_e(\rho_e, z_e, T) F_h(\rho_h, z_h, T) F_{eh}(\rho_{eh}, |z_e - z_h|), \quad (6)$$

with

$$F_{eh}(\rho_{eh}, |z_e - z_h|) = \exp(-\alpha \rho_{eh}) \exp(-\gamma (z_e - z_h)^2), \quad (7)$$

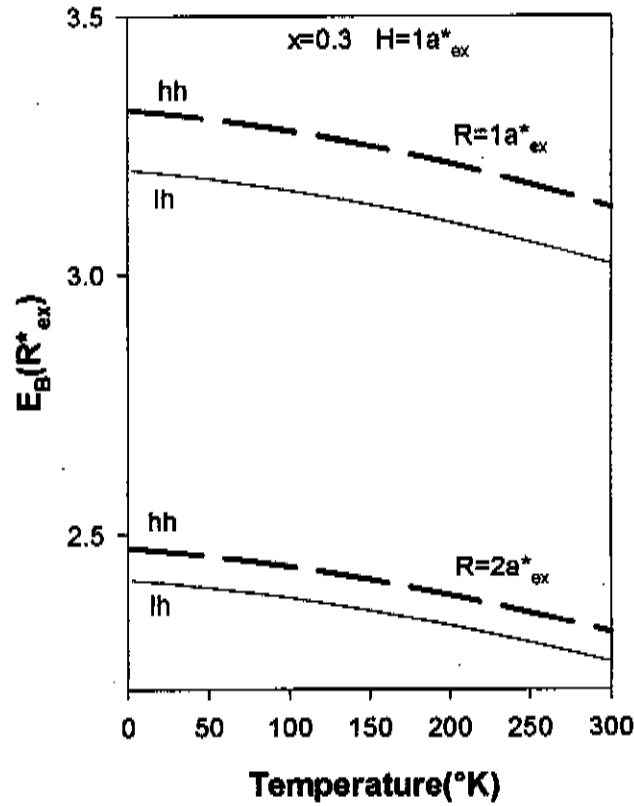


Figure 2. Variation of light (solid line) and heavy (dashed line) hole exciton binding energy as a function of temperature for a fixed height H and zinc concentration x and two values of the dot radius R .

and

$$F_i(\rho_i, z_i, T) = f_i(\rho_i, T)g_i(z_i, T), \quad (i = e, h). \quad (8)$$

Respectively, the corresponding 2D (lateral direction) and 1D (longitudinal direction) effective-mass Schrödinger equations are

$$\left\{ -\frac{\hbar^2}{2m_i^*} \nabla_i^2 + V_w^i(\rho_i, T) \right\} f_i(\rho_i, T) = E_i(\rho_i, T) f_i(\rho_i, T) \quad (i = e, h), \quad (9)$$

$$\left\{ -\frac{\hbar^2}{2m_i^*} \nabla_i^2 + V_w^i(z_i, T) \right\} g_i(z_i, T) = E_i(z_i, T) g_i(z_i, T) \quad (i = e, h), \quad (10)$$

with solutions of the form

$$f_i(\rho_i, T) = \begin{cases} J_0(\theta_i(T) \frac{\rho_i}{R}) & \text{for } \rho_i \leq R, \quad (i = e, h) \\ A_i(T) K_0(\beta_i(T) \rho_i) & \text{for } \rho_i > R \end{cases} \quad (11)$$

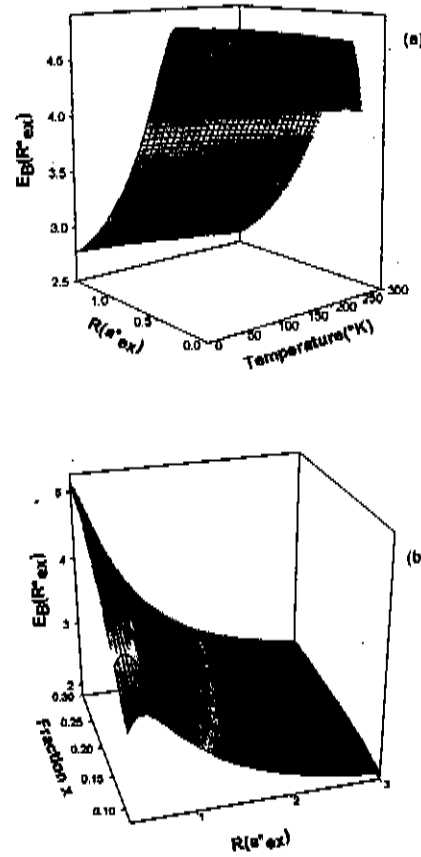


Figure 3. Variation of the heavy hole (hh) exciton binding energy in cylindrical quantum dot of height equal to $H = 1a_{ex}^*$ as a function of: (a): the radius R and temperature T for a fixed Zn concentration $x = 0.3$, (b): the radius R and fraction x of zinc at temperature $T = 300$ K.

$$g_i(\rho_i, T) = \begin{cases} \cos(\pi_i(T)\frac{z_i}{2d}) & \text{for } |z_i| \leq d, \quad (i = e, h) \\ B_i(T) \exp(k_i(T)|z_i|) & \text{for } |z_i| > d \end{cases} \quad (12)$$

Here J_0 and K_0 are the modified Bessel functions of m^{th} order. $\theta_i(T)$, $\beta_i(T)$, $\pi_i(T)$, $k_i(T)$, $A_i(T)$ and $B_i(T)$ are determined from the boundary conditions at $\rho_i = R$ and $|z_i| = d$.

In Eq.(7), α and γ are the variational parameters. The temperature dependent variational binding energy of the exciton system is given by

$$E_B(T) = E_c(T) + E_h(T) - E(T), \quad (13)$$

where $E(T)$ is the temperature dependent variational bound-state energy of the

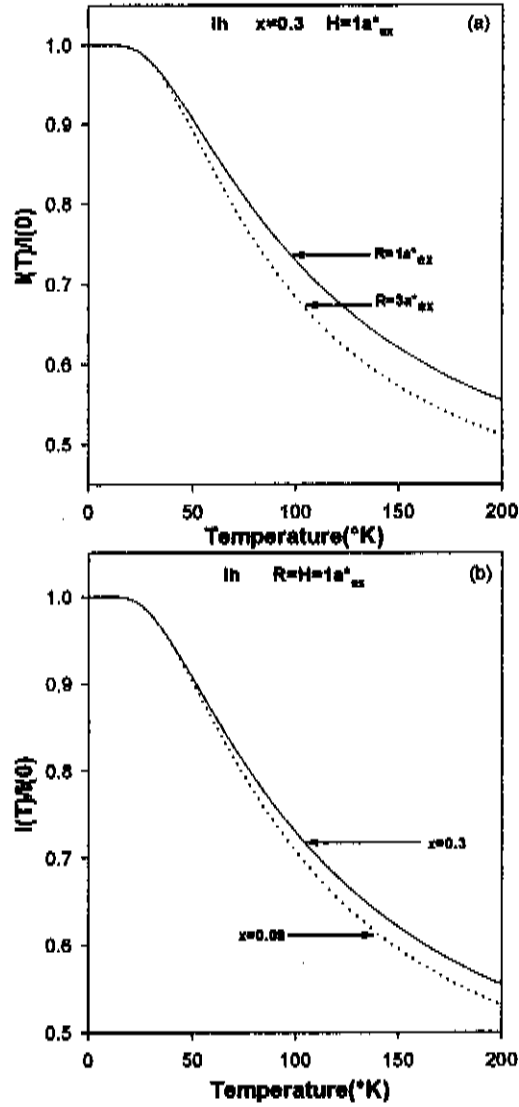


Figure 4. Arrhenius plot of the integrated photoluminescence intensity using Eq.(15) for the light hole exciton, in the case of CdTe/Cd_{1-x}Zn_xTe QD, as function of the temperature T for a fixed dot height $H = 1a^*_{ex}$. (a): $x = 0.3$, for two values of the dot radius $R = 1a^*_{ex}$ and $R = 3a^*_{ex}$ (b): $R = 1a^*_{ex}$, for two values of the zinc concentration $x = 0.08$, $x = 0.3$.

exciton system which can be written as:

$$E(T) = \min_{\alpha, \gamma} \langle \psi_{ex} | H_{ex} | \psi_{ex} \rangle \quad (14)$$

where $E_e(T)$ and $E_h(T)$ are the free electron and hole energies.

In relation to the optical experiment on these quantum dot structures, the integrated photoluminescence intensity is one of the physical parameters which can draw a good picture of their optical properties. The temperature dependence of integrated photoluminescence intensity is given by the Arrhenius model [25,38]:

$$\frac{I(T)}{I(0)} = \frac{1}{1 + C_1 \exp(-E_1(T)/kT) + C_2 \exp(-E_2(T)/kT)}, \quad (15)$$

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

$$E_1(T) = E_e(T) + E_h(T) - \min_{\alpha, \gamma} \frac{\langle \psi_{ex} | H_{eff} | \psi_{ex} \rangle}{\langle \psi_{ex} | \psi_{ex} \rangle}, \quad (16)$$

$$E_2(T) = E_e(T) + E_h(T), \quad (17)$$

$E_e(T)$ and $E_h(T)$ are, respectively, the electron and hole energies at temperature T .

3. Numerical results and discussions

The numerical calculations are performed for a cylindrical quantum dot made of CdTe embedded in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ material. The physical parameters corresponding to the (well) polar crystal CdTe are: $m_{lh}^* = 0.11m_0$, $m_{hh}^* = 0.662m_0$, $m_e^* = 0.098m_0$, (m_e^* , m_{lh}^* and m_{hh}^* are respectively the electron, the light and heavy hole effective masses). The dependence of the static dielectric constant on temperature T is given in Ref. [39] as $\epsilon_0(T) = \epsilon_0(0) + 8.3 \cdot 10^{-4}T + 3.3 \cdot 10^{-6}T^2$ where $\epsilon_0(0) = 9.808$. Our numerical results are presented in units of the effective Rydberg energy $R_{ex}^* = 7.62$ meV and the effective Bohr radius $a_{ex}^* = 98.17$ Å at low temperature.

Figure 1 shows the variation of the light hole exciton binding energy in a QD of a fixed height $H = 1a_{ex}^*$, as a function of the cylinder radius R , for two different values of temperature ($T = 0$ K and 300 K) and for two zinc concentrations ($x = 0.08$ and 0.3). In the same figure we illustrate the variation of heavy hole exciton binding energy for zinc concentration $x = 0.3$ and the same two values of temperature. At fixed temperature, the exciton binding energy increases as the radius decreases (well width is reduced), reaches a peak

at a critical confinement width (which depends on the height of the potential model considered), and then decreases. At small R values, discrete exciton levels are absent in the well and the electron and hole wave functions are distributed outside the cylinder. When increasing the cylinder radius R , the exciton energy levels fall from the continuum spectrum into the well. For a fixed radius R , it is seen from this figure that the exciton binding energy at low temperature $T = 0$ K is greater than that at room temperature $T = 300$ K. Similar result is obtained in the case of a donor impurity in Ref. [23]. We should emphasize that for two different values of temperature ($T = 0$ K and 300 K) and fixed zinc concentrations ($x = 0.08$ and 0.3), the difference between the heavy and light hole exciton binding energies becomes smaller with increasing dot radius. The heavy hole exciton is more stable than the light hole exciton in cases of smaller QD which can be attributed to the heavier effective mass which provides much more confinement in these situations and hence an enhancement of the exciton binding energy. Our result agrees with that obtained previously by Huang and Jain [18]. Figure 2 displays the heavy (light)-hole exciton binding energy versus the temperature for different radius ($R = 1a_{ex}^*$, $R = 2a_{ex}^*$). The concentration of zinc is fixed at $x = 0.3$ and the dot height is $H = 1a_{ex}^*$. This figure shows that the exciton binding energy decreases with increasing temperature. This is due to the fact that with increasing temperature the exciton has a tendency to dissociate into an electron-hole pair. We notice also that the temperature effect on the exciton binding energy is relatively similar for both heavy and light hole excitons. To get a good picture of the effect of zinc concentration, radius and temperature on the exciton binding energy, we have plotted the heavy hole (hh) exciton binding energy as a function of the dot radius and temperature (Fig. 3(a)) and also versus the radius and zinc concentration at room temperature (Fig. 3(b)). The dot height is fixed at $H = 1a_{ex}^*$. The exciton binding energy increases when the temperature and cylinder radius decrease and also when zinc concentration increases in the barrier material. This means that more confinement of the electron and hole wave functions can be reached by adjusting the zinc concentration and dot sizes.

More interests, in the area of nanostructure physics, have been focused on the study of the role of carrier charge (electron or hole) interaction not only on the electronic properties but also on the optical ones. Investigation of the emission spectra in such structures versus temperature allows to find the effect of different parameters on the luminescence and photoluminescence. Equation (15), which gives the analytical expression for the integrated photoluminescence intensity, shows its dependence not only on temperature fluctuation, but also on the activation energies $E_1(T)$ (Eq.16) and $E_2(T)$ (Eq.17), and the constants

C_1 and C_2 characterizing the ratio of non-radiative to radiative recombination rates. In order to achieve this aim and for the sake of simplicity to obtain basic information about this process, we take the constants C_1 and C_2 equal to unit.

Figure 4 (a) displays the behavior of the integrated photoluminescence intensity of light hole exciton in CdTe quantum dot embedded in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ barrier material of zinc fraction x equal to 0.3. The integrated photoluminescence intensity is depicted versus temperature for CdTe quantum dots of height $H = 1a_{ex}^*$ and different radii $R = 1a_{ex}^*$ and $3a_{ex}^*$. It is important to note that the sizes of the QD as well as the temperature affect the photoluminescence spectra. The intensity ratio $I(T)/I(0)$ decreases with increasing radius R for temperatures approximately higher than 50 K.

In Figure 4(b) we show, for a given CdTe quantum dot of sizes $R = 1a_{ex}^*$ and $H = 1a_{ex}^*$, how the depth of the barrier potential induced by the $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ material modifies the intensity of the integrated photoluminescence ratio $I(T)/I(0)$. This figure is depicted for two values of the zinc fraction ($x = 0.08$ and $x = 0.3$) which correspond to different values of the potential barriers levels. We remark that at low temperatures the height of the potential barrier has no effect on the photoluminescence intensity ratio $I(T)/I(0)$ but when temperature becomes greater than a certain critical value (around 60 K), the well potential barrier influences notably the photoluminescence spectra by adjusting zinc concentrations.

4. Conclusion

Using the variational method, we have investigated two effects on the heavy (light)-hole excitonic binding energy for a finite depth potential: the temperature and the dot sizes effects. Our results show that due to the finite confining capacity of the QD there is a critical radius for which the exciton is no longer confined. Indeed, as the confining potential decreases, this critical radius becomes bigger. The exciton binding energy is enhanced by decreasing temperature which allows the exciton to become more stable. Calculation of the integrated photoluminescence intensity versus the temperature variation shows the noticeable effect of the dot size and the zinc concentration on the photoluminescence spectra.

We expect that the present study will be useful for a good understanding of experimental works concerning the existence and stability of the exciton in nanostructures. This stability is dependent on the temperature and the dimensions of the nanostructures.

Acknowledgements

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