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Pressure induced nonlinear optical rectification of a confined exciton in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ strained quantum dot

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ABSTRACT

Pressure dependent exciton binding energy in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ strained quantum dot is investigated with the geometrical confinement within the single band effective mass approximation. The interband transition energy as functions of dot radius and the hydrostatic pressure is brought out. Computations are carried out using variational formulism. The pressure induced oscillator strength and the nonlinear optical rectification coefficient of an exciton in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot are studied using matrix method. Our results show that the exciton binding energy, interband emission energy and the nonlinear optical rectification coefficient are strongly dependent on the geometrical confinement and the applied hydrostatic pressure values.

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Introduction

ZnTe based II-VI semiconductors are of great interests because they have possible quantum efficiency in the visible spectrum region [1]. ZnTe semiconductor is an attractive material for the usage of terahertz wave imaging applications among nonlinear optical materials [2]. It is considered to be one of the best II-VI semiconductor heterostructures having a potential application of green wavelength light emitting diodes [3]. ZnTe doped with Cd ternary compounds can be used for tailoring the properties of any potential applications for device fabrications. CdTe/ZnTe semiconductors are quite interesting due to their lattice matched nature by adjusting ratio of Cd:Zn. The effects of band offsets and the lattice mismatch on strained-layer CdTe/ZnTe superlattices have been studied earlier [4]. Some nonlinear optical properties such as absorption coefficients and changes of refractive index bring out the quantum size effects for any QD semiconductors can be fabricated by some latest technologies such as Molecular beam epitaxy, Metal organic chemical vapour deposition and electron lithorgraphy [6-8]. CdZnTe materials are used to produce photo-detectors, solar cells, laser diodes and light emitting diodes.

Excitons and biexcitons play a vital role in the optoelectronic semiconductor devices and the application of pressure affects the band structure and thereby electronic states and optical properties are altered. The effects of pressure in low dimensional semiconductors are quite different from their counterpart bulk materials. The effects of pressure on excitons and their related properties in a quantum dot have been studied earlier [9]. The pressure induced exciton binding energies, the oscillator strength and some nonlinear optical properties of excitons have been dealt using matrix diagonalization method [10]. The combined effects of pressure and temperature on the optical properties in a disc shaped parabolic quantum dot have been investigated [11]. The pressure coefficients for bulk and low dimensional semiconducting structures have been investigated earlier [12,13]. Kozyrev [14] has studied the infrared spectroscopy of lattice vibrations in a multiperiod ZnTe/CdTe superlattices with CdTe quantum dots recently.

In the present work, the exciton binding energy in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ is computed with the effects of geometrical confinement and the hydrostatic pressure using a single band effective mass approximation. Computations are followed using an effective mass approximation with the variational formulism. The pressure dependent interband emission energy as a function of dot radius is studied. The oscillator strength and the nonlinear optical rectification coefficient are investigated in the presence of hydrostatic

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pressure. In Section 2, the theoretical model used in our calculations of strain effect, the obtained eigen functions and eigen energies of electron (hole) states and the nonlinear rectification coefficient are discussed. The results and discussion are presented in Section 3. A brief summary and results are presented in the last Section.

Theoretical frame work

Consider an exciton in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ strained quantum dot semiconductor heterostructure. The Schrödinger wave equation for the exciton, within the single band effective mass and non-degenerate parabolic band approximation, in the presence of hydrostatic pressure, is given by

$$\hat{H}\Psi(r_e, r_h) = [E - E_g^{\Gamma}(P)]\Psi(r_e, r_h)$$
⁽¹⁾

where the Hamiltonian operator is given by

$$\hat{H} = -\frac{\hbar^{2}}{2m_{e}^{*}(P)}\frac{\partial^{2}}{\partial z_{e}^{2}} + V_{e}(\rho, z_{e}, P) - \frac{\hbar^{2}}{2m_{h\perp}^{*}(P)}\frac{\partial^{2}}{\partial z_{h}^{2}} + V_{h}(\rho, z_{h}, P) \qquad (2)$$

$$-\frac{\hbar^{2}}{2}\left(\frac{1}{2m_{e}^{*}(P)} + \frac{1}{2m_{h\parallel}^{*}(P)}\right)\nabla_{\rho}^{2} + q_{j}Fz - \frac{e^{2}}{\varepsilon(P)|\bar{r}_{e} - \bar{r}_{h}|}$$

where z_e and z_h are the electron and hole coordinates along the growth direction, ρ is the inplane relative coordinate, $V_{e(h)}(z_{e(h)}, P)$ is the pressure dependent barrier height of conduction and valence band and e is the absolute value of electron charge. The subscript, j, refers the electron and hole and the charge q_j is e for an electron and -e for a hole. $|\bar{r}_e - \bar{r}_h|$ denotes the relative distance between the electron and the hole. $\varepsilon(P)$ is the pressure dependent dielectric constant of the material inside the quantum dot.

It is well known that the hydrostatic pressure affects the geometrical dimension of any semiconductor nanostructure. The lattice constants, dot sizes, barrier height, effective masses and the dielectric constants of semiconductor material are altered with the application of pressure. The pressure dependent radius of the quantum dot, R(P), may be obtained from the fractional change in volume given by $\Delta V/V_0 = -3P(S_{11}(P) + S_{12}(P))$, here $S_{11}(P)$ and $S_{12}(P)$ are the pressure dependent compliance constants of inner dot of the material given by

$$S_{11}(P) = \frac{(C_{11}(P) + C_{12}(P))}{[(C_{11}(P) - C_{12}(P))(C_{11}(P) + 2C_{12}(P))]}.$$
(3)

$$S_{12}(P) = \frac{C_{12}(P)}{\left[(C_{11}(P) - C_{12}(P))(C_{11}(P) + 2C_{12}(P))\right]}.$$
(4)

where $C_{11}(P)$ and $C_{12}(P)$ are the pressure dependent elastic constants of $Cd_{0.8}Zn_{0.2}Te$.

The pressure dependent effective masses of electron and heavy hole are given by

$$\frac{m_{e}^{*}(P)}{m_{0}} = \left[1 + E_{p}\left(\frac{2}{E_{g}^{\Gamma}} + \frac{1}{E_{g}^{\Gamma}} + \Delta\right)\right]^{-1}$$
(5)

and

$$\frac{m_{hh\perp,\parallel}^*(P)}{m_0} = \frac{m_{hh\perp,\parallel}^*}{m_0} + \alpha P + \beta P^2 \tag{6}$$

where E_p is the kane energy of the inner material, α , β are the pressure coefficients and Δ is the spin orbit splitting energy. The heavy hole mass in the direction perpendicular and parallel to the growth direction is given by

$$\frac{m_{hh\perp}^*}{m_0} = (\gamma_1 - 2\gamma_2)^{-1}$$
⁽⁷⁾

and

$$\frac{m_{hh\parallel}^{*}}{m_{0}} = (\gamma_{1} + \gamma_{2})^{-1}$$
⁽⁸⁾

where m_0 is the free electron mass and the γ_1 and γ_2 are the Luttinger parameters as given Table.1.

Parameter	CdTe	ZnTe	Zn _x Cd _{1-x} Te
ε	10.4	9.4	10.4-1.0x
<i>a</i> (nm)	0.6481	0.61037	0.6481-0.03773x
$C_{11}(GPa)$	53.8	72.2	53.8+18.397x
C_{12} (GPa)	37.4	40.9	37.4+3.471x
$e_{14}~(C/m^2)$	0.91	0.028	0.91 -0.88x
$E_{g}^{r}\left(eV ight)$	1.51	2.27	1.51+0.45x+0.31x ²
∆(eV)	4.67	4.789	4.67-0.119
γ_1	4.14	3.96	4.14-0.18x
γ_2	1.019	0.86	1.019-0.159x
$a_{c}\left(eV ight)$	-3.96	-5.83	-3.96 -1.87x
$a_v (eV)$	0.55	0.79	0.55+0.24x
Ե (eV)	-1.0	-1.4	-1.0-0.4x

Table 1. Material parameters* used in the calculations (all the other parameters are linearly interpolated)

The strain related pressure dependent band gap of Cd_{0.8}Zn_{0.2}Te quantum dot is given by

$$E_{g,d(b)}(P) = E_{g,d(b)}(P) + (a_{d(b)}^{c} - a_{d(b)}^{v})(2\varepsilon_{xx,d(b)} + \varepsilon_{zz,d(b)})$$
(9)

where the subscripts d,b denote the dot and barrier material and the pressure dependent band gap is given by

$$E_{g,d(b)}(P) = E_{g,d(b)}(0) + \alpha P + \beta P^2$$
⁽¹⁰⁾

where $E_{g,d(b)}(0)$ is the band gap of Cd_{0.8}Zn_{0.2}Te at zero pressure given by the following expression [15]

$$E_{g} = 1.51 + 0.45x + 0.31x^{2} \tag{11}$$

where x is the concentration of Zn alloy content in $Cd_{1-x}Zn_xTe$ semiconducting material and the biaxial lattice mismatched induced strain in the dot is given by

$$\mathcal{E}_{xx,d} = \mathcal{E}_{yy,d} = \mathcal{E}_{\parallel,d}^c = \frac{a_b(P) - a_d(P)}{a_d(P)}$$
(12)

and in the barrier is given by

$$\varepsilon_{xx,b} = \varepsilon_{yy,b} = \varepsilon_{\parallel,b}^{c} = \frac{a_d(P) - a_b(P)}{a_b(P)}$$
(13)

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and

$$\varepsilon_{zz} = \varepsilon_{\perp} = -\frac{2C_{12}}{C_{11}}\varepsilon$$
⁽¹⁴⁾

where C_{ij} are the elastic constants of the materials.

The pressure dependent lattice constant is expressed as [16]

$$a_{d,b} = a_{d,b}(0) \left(1 - \frac{P}{3B} \right)^{.}$$
(15)

where B is the bulk modulus of the material.

The inner and outer materials in the dot have different built-in internal fields caused by the spontaneous and piezo-electric polarizations. The semiconducting heterostructure will have spontaneous polarization (P_{SP}) and the piezo electric polarization (P_{PZ}) due to strain caused by the lattice mismatch between the inner and the outer dot material. In general, the wurzite semiconductors possess both the spontaneous and piezoelectric components. The total polarization, \vec{P}_{tot} , is the addition of spontaneous polarization, \vec{P}_{SP} , and the piezoelectric polarization, \vec{P}_{PE} , in the absence of the external electric field. Thus the built-in internal field is due to the existence of polarization of the quantum dot. The expression for the total internal field is given by [17]

$$F_{ZnCdTe} z_{ZnCdTe} + F_{ZnTe} z_{ZnTe} = 0$$
⁽¹⁶⁾

and using the boundary condition of the continuity of the electric displacement vector at the heterointerfaces of the quantum dot, we get,

$$\varepsilon_{e(ZnTe)}\varepsilon_{0}F_{ZnTe} - \varepsilon_{e(ZnCdTe)}\varepsilon_{0}F_{ZnCdTe} = P_{SP(ZnCdTe)} + P_{PE(ZnCdTe)} - P_{SP(ZnTe)}$$
(17)

where \mathcal{E}_0 is the dielectric constant of the respective material. Using, Eq.(16) and Eq.(17), the built-in electric field can be arrived at

$$F_{ZnTe} = \frac{(P_{SP(ZnCdTe)} + P_{PE(ZnCdTe)} - P_{SP(ZnTe)})z_{ZnCdTe}}{\mathcal{E}_0(\mathcal{E}_{e(ZnTe)}z_{(ZnCdTe)} + \mathcal{E}_{e(ZnCdTe)}z_{(ZnTe)})}$$
(18)

and

$$F_{ZnCdTe} = -\frac{(P_{SP(ZnCdTe)} + P_{PE(ZnCdTe)} - P_{SP(ZnTe)})z_{ZnTe}}{\varepsilon_0(\varepsilon_{e(ZnTe)}z_{(ZnCdTe)} + \varepsilon_{e(ZnCdTe)}z_{(ZnTe)})}$$
(19)

The piezo electric polarization along the c-axis is given by

$$P^{PZ} = e_{31}(\varepsilon_{xx} + \varepsilon_{yy}) + e_{33}\varepsilon_{zz}$$
⁽²⁰⁾

where e_{ij} are the piezo electric constants. Thus the total polarization is given by

$$\vec{P} = \vec{P}^{PZ} + \vec{P}^{SP}.$$
⁽²¹⁾

The confinement potential due to the band offset between Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot structure is given by

$$V(\rho, z) = \begin{cases} V(\rho) & z_0 < z < z_1 \\ V_{II} & otherwise \end{cases}$$
(22)

$$V(\rho) = \begin{cases} 0 & \rho \le R \\ V_I & \rho > R \end{cases}$$
(23)

where R is the dot radius.

In order to compute the binding energy of an exciton in the $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot, the trial wave function is chosen as

$$\Psi = \psi_e(\rho, \phi, z)\psi_h(\rho, \phi, z)\exp(-\delta\rho^2 - \lambda z^2)$$
⁽²⁴⁾

where $\psi_{e(h)}(\rho, \varphi, z)$ is the eigen function of the Hamiltonian according to the Eq.(2) and δ, λ are the variational parameters. Thus the energy of the exciton in the Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot is obtained by minimizing, the Hamiltonian, Eq.(2),

$$E_{\min} = \min_{\delta,\lambda} \frac{\langle \Psi | \hat{H} | \Psi \rangle}{\langle \Psi | \Psi \rangle}$$
(25)

Thus, the pressure dependent exciton binding energy is calculated as

$$E_{exc} = E_{e(h)} - E_{\min}$$
⁽²⁶⁾

where $E_{e(h)}$ is the ground state energy of the electron (hole).

The interband emission energy E_{ph} associated with the exciton is calculated using the following equation

$$E_{ph} = E_e + E_h + E_g^{\Gamma} - E_{exc} \tag{27}$$

where E_e and E_h are the ground state energies of the electron and hole in the Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot respectively. E_a^{Γ} is

the band gap energy in the $Cd_{0.8}Zn_{0.2}Te$ material.

The behaviour of recombination rate of electron-hole pair in the presence of pressure effects is investigated here. The decay time is closely related to the recombination rate [18]. The oscillator strength of the exciton in the $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot is expressed as

$$f(P) = \frac{E_p}{E_{exc}} \left| \int_V \psi_e(r_e) \psi_h(r_h) dr_e dr_h \right|^2$$
(28)

where E_p is the Kane energy of $Cd_{0.8}Zn_{0.2}Te$ and E_{exc} is the exciton binding energy.

The second order nonlinear optical rectification coefficient, using density matrix formulism, is given by the expression as [19,20]

$$\chi_0^2 = \frac{q^3 \sigma_s \mu_{01}^2 \delta_{01}}{\varepsilon_0} \frac{2\Delta E^2}{\left[\left(\Delta E - \hbar\omega\right)^2 + \left(\hbar\Gamma_0\right)^2\right] \left[\left(\Delta E + \hbar\omega\right)^2 + \left(\hbar\Gamma_0\right)^2\right]}$$
(29)

where σ_s is the electron density in the quantum dot, ε_0 is the permittivity in vacuum, $\Gamma = 1/\tau$ is the relaxation rate for states between 1 and 2. The matrix element, $\mu_{01} = \langle \psi_0 | z | \psi_1 \rangle$ is defined as the electric dipole moment of the transition from the ground state (ψ_0) to the first excited state ψ_1 with $\delta_{01} = \langle \psi_1 | z | \psi_1 \rangle - \langle \psi_0 | z | \psi_0 \rangle \cdot \Delta E$ is the absorption energy from ground state to the first excited states. The relaxation rate is taken as 1ps and the electron density is considered to be 1×10^{24} m⁻³.

Results and discussion

The binding energy of a heavy hole exciton as functions of dot radius and the pressure values in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot is computed numerically using variational formulism and thereby the interband optical transition energy as a function of dot radius is calculated. The oscillator strength and the non-linear optical rectification coefficient of a confined exciton in the presence of hydrostatic pressure values are investigated with the geometrical confinement effect using matrix formulism. Application of pressure changes all the structural parameters. The strain effect has been included through the shifts due to strain in the conduction (valence) band edges.

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Fig.1 Exciton binding energy as a function of dot radius of Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot for various pressure values; the insert figure shows the variation of exciton binding energy as a function of pressure for different dot radii

We present the exciton binding energy as a function of dot radius in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot for various pressure values in Fig.1 and the insert figure displays the variation of exciton binding energy as a function of pressure for different dot radii. It is observed that the exciton binding energy is found to be larger for smaller dot radius and smaller for larger dot radius. This is due to the geometrical confinement. The exciton binding energy increases as the quantum dot radius decreases, it reaches a peak value for a critical geometrical confinement and then starts decreasing for all the applied hydrostatic pressure values. The enhancement of exciton binding energy with the decrease of dot radius is due to the localization of electron (hole) wave function in the dot and the enhancement of Coulomb interaction. It is because the finite confinement potential is taken in the problem here. The decrease of exciton binding energy for the smaller dot radius, after the critical confinement, is due to the squeezing of wave functions of electron and hole through the barrier. When the pressure is increased, the exciton becomes more confined and thereby exciton binding energy increases for all the dot radii. It is brought out clearly from the insert figure. The exciton binding energy is enhanced for all the dot radii when the pressure is applied. This is because there are changes in structural parameters when the hydrostatic pressure is applied. And an increase in effective mass and decrease in dielectric constant are obtained when the pressure is applied for any semiconductor. Moreover, it is found that smaller dot radii have more exciton binding energy than the larger dot radii due to the additional confinement of hydrostatic pressure.

Fig.2 displays the interband emission energy as a function of dot radius in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot for various pressure values and the insert figure shows the interband emission energy as a function of pressure for two different dot radii. The interband band emission energy increases as the dot radius is decreased also the interband emission energy is found to be enhanced with the hydrostatic pressure values due to the additional confinement. The confinement energies of electron and hole increase with the pressure values. Thus it is brought out that the effect of pressure has influence on the exciton binding energy and the strong confinement region. Large blue shift takes place mainly with the dependence of pressure of the band gap of $Cd_{0.8}Zn_{0.2}Te$. It is because the changes in effective mass and the dielectric constant when the pressure is applied. Eventually, the pressure and the geometrical confinement alter the exciton binding energy and thereby the interband optical energy is also modified.



Fig.2 Interband emission energy as a function of dot radius of Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot for various pressure values and the insert figure shows the interband emission energy as a function of pressure for two different dot radii

We present the variation of oscillator strength as a function of dot radius in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot for various pressure values in Fig.3 and the insert figure shows the oscillator strength as a function of pressure for different dot radii. It is noticed that the pressure induced oscillator strength of the exciton increases with the decrease in dot radius and the oscillator strength increases with the pressure values. This is because the exciton binding energy increases with the pressure as seen earlier. Also, the enhancement of recombination rate of the exciton and the reduction of transition energy are observed as the dot radius is increased whereas the overlapping of electron-hole wave function increases as the spatial confinement decreases eventually making the decay time to decrease. Also, the electron (hole) wave function is more strongly localized inside the quantum dot in the strong confinement region and thus the recombination rate is decreased [21].



Fig.3 Variation of oscillator strength as a function of dot radius in a Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot for various pressure values and the insert figure shows the oscillator strength as a function of pressure for different dot radii

In Fig.4, we present the variation of second order nonlinear coefficient as a function of incident energy of a confined exciton for various dot radii in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot. It is observed that the resonant peak shifts towards the blue region as the dot radius is decreased in the $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot. It is seen earlier that the exciton binding energy increases with the decrease in dot radius. The magnitude of the resonant peak of nonlinear optical rectification is found to be around 10^{-4} m/V. This strength of nonlinear optical rectification is closely related to the other investigator [22]. This strong value is due to the existence of larger dipole matrix elements in the ZnCdTe quantum dot, however, we notice that the variation of magnitude of nonlinear optical rectification coefficient decreases with the dot radius. The spacing between the energy levels increases due to decrease in dot radius. It is because the exciton binding energy decreases when the dot radius is increased. Optical rectification in a ZnCdTe brings out an increasing efficiency of terahertz beam generation with a proper incorporation of Cd [23]. Thus, the resonant frequencies are important and it should be taken into account in studying the optical properties of exciton in the quantum nanostructures.

Fig.5 displays the variation of second order nonlinear coefficient as a function of incident energy of a confined exciton for various pressure values with the constant dot radius (40 Å) of a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ quantum dot. It is noticed that the resonant rectification constant peak shifts towards the blue region when the pressure is applied. This is because the higher transition energy takes place when the pressure is applied as shown in Eq.(29). The energy interval between two different states increases with the pressure. Further it is noticed that the resonant values are found to be constant when the hydrostatic pressure is applied. It is due to two competing factors of energy interval and the dipole matrix element.



Fig.4. Variation of second order nonlinear coefficient as a function of incident energy of a confined exciton for various dot radii in a Cd0.8Zn0.2Te/ZnTe quantum dot



Fig.5. Variation of second order nonlinear coefficient as a function of incident energy of a confined exciton for various pressure values with the constant dot radius (40 Å) of a Cd_{0.8}Zn_{0.2}Te/ZnTe quantum dot

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In conclusion, the exciton binding energy as functions of dot radius and the hydrostatic pressure in a $Cd_{0.8}Zn_{0.2}Te/ZnTe$ in a quantum dot has been calculated using a single band effective mass approximation with the variational method. The pressure induced interband emission energy with the spatial confinement has been studied. The oscillator strength and the nonlinear optical rectification coefficient have been investigated in the presence of hydrostatic pressure. Our result shows that the nonlinear optical rectification coefficient is strongly dependent on the effect of pressure and the geometrical confinement. We hope that our results would stimulate further research works on ZnTe based II-VI semiconductors for some potential applications emitting green wavelength required for optical devices such as optical fibers and laser diodes.

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