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Degenerate four waves mixing in multilayer nanoshell

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ABSTRACT

We will present a detailed investigation of intersubband transitions process in core-multi shells quantum dots. The confined wave functions and eigenenergies of electrons in quantum dots have been calculated under the effective-mass approximation by solving a three-dimensional Schrodinger equation. Excellent dependence is found between size effects, time relaxation and degenerate four wave mixing (DFWM). We found that the enhancement of the thickness layers lead to strongly enhancement in peak value of DFWM.

Keywords: *Nanoparticle; Optical susceptibility; Nonlinear effects; Multilayer; Four wave mixing.*

INTRODUCTION

Nano-size semiconductor materials have obtained considerable interest during the past decade [1, 2]. These materials are widely used in various applied technological fields such as optoelectronic and photonic devices or even for advanced biotechnology due to their size dependant physical and optical properties [3-5]. Recently several of type nanocrystallites such as ZnSe, CdS, ZnS, and CdSe are reported in the literature [6-8]. In addition, it has been reported that when the surface of nanocrystals is passivated by ZnS, a core shell type of quantum dot is formed. The third-order nonlinear polarization leads in general to the interaction of four optical waves with frequencies ω_1; ω_2; ω_3;ω_4 and include such phenomena as four-wave mixing (FWM) and parametric amplification. These processes can be used to generate waves at new frequencies. In the degenerate case $\omega_1 = \omega_2 = \omega$ This process can be analytically described for the case when the pulse durations are long enough so that effects of linear dispersion are negligible, the pump pulse is much Optical (FWM) has many applications such as phase conjugation [7], real-time holographic imaging [8], and nonlinear frequency conversion [9]. With electromagnetically induced transparency (EIT) [10], FWM has even been demonstrated at low light level [11].

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More recently, four-wave spontaneous parametric interactions in cold atomic systems have been used to generate narrowband time-energy entangled photon pairs [12–13].Earlier work encompassed phase conjugation with degenerate FWM in two-level systems. Du *et al.* [13,14] showed that in a two-level system there is a destructive interference of the third-order nonlinear susceptibility between two FWM processes. Four waves mixing is a promising technique for wavelength conversion in communication systems and are typically realized in semiconductor optical amplifiers (SOAs) that require external pumping sources [15]. Quantum dots (QDs)have some fundamental advantages over quantum wells for nonlinear optics applications considering the theoretical enhancement by the quantum confinement in more dimensions [16], [17], ultrafast carrier recovery [18] and wide gain spectrum [19]. In this rkwo we present a theoretical computation about third-order nonlinear optical susceptibility $\chi^{(3)}$in the *cdse/zns/cdse/zns* with core-multi shells structure. A schematic representation of studied structure is shown in Figure 1.

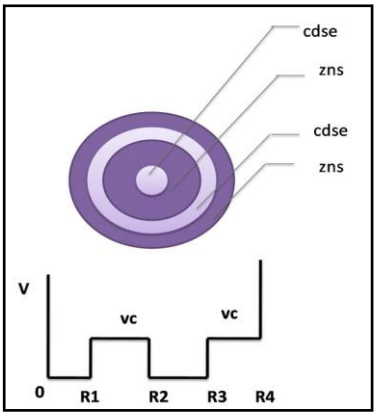

Fig. 1. two-dimensional model and potential diagram of cdse/zns/cdse/zns core-multi shells QD

EXPERIMENTAL

Semiconductor quantum dots represent unique class of quasi-zero dimensional material systems which reveal large optical nonlinearity and hence are potential candidates for optoelectronic and photonic devices. The nature of the nonlinear optical response to an incident light-field can be profoundly influenced by the dimensionality of the material. Nonlinear optical effects are generally strongest in geometries in which the optical intensity is high in the largest possible volume. A one-band effective mass model is used to model the core shell quantum dot (*CSQD),* which is assumed to be perfectly spherical. The potential experienced by the charge carriers is assumed to be infinite outside the dot, giving a definition of the potential as

$$\begin{cases} 0 & 0< r < R_1 \\ V_c & R_1< r < R_2 \\ 0 & R_2< r < R_3 \\ V_c & R_3< r < R_4 \end{cases} \quad (1)$$

Where $V_c$is constant. Similarly, the effective mass of the carrier is defined as

$$\begin{cases} m_1 & 0< r < R_1 \\ m_2 & R_1< r < R_2 \\ m_1 & R_2< r < R_3 \\ m_2 & R_3< r < R_4 \end{cases} \quad (2)$$

Where m_1 and m_2 are also constants. The time-independent Schrodinger equation

$$\{-\frac{\hbar^2}{2m_i^* r^2}[\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right)+\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{\sin\theta^2}\frac{\partial^2}{\partial\varphi^2}]+V_i(r)\}\psi_{nlm}(r,\theta,\varphi)=E\psi nlm(r,\theta,\varphi) \quad (3)$$

Defines the stationary states of a system consisting of a particle with energy E and wavefunction $\psi(r,\theta,\varphi)$. This equation is used as the starting point in the derivation of the required wavefunctions relation. For a central potential such as that given in equation (1) the wavefunction $\psi(r,\theta,\varphi)$ separates into the product of a radial function $R_{nl}(r)$ and a spherical harmonic $Y_{l,m}(\theta,\varphi)$ with orbital and magnetic quantum numbers l and m [13.]. Depending on whether the energy E is greater than or less than the potential V_c, the differential equation which defines $R_{nl}(r)$ is then either the ordinary spherical Bessel or spherical Hankel function.

$$R_{nl}(r) = \begin{cases} A_0 J_l(k_{nl,1}r)+B_0 n_l(k_{nl,1}r) & 0 < r \le R_1 \\ A_1 J_l(k_{nl,2}r)+B_1 n_l(k_{nl,2}r) & R_1 < r \le R_2 \\ A_2 J_l(k_{nl,1}r)+B_2 n_l(k_{nl,1}r) & R_2 < r \le R_3 \\ A_3 J_l(k_{nl,2}r)+B_3 n_l(k_{nl,2}r) & R_3 < r \le R_4 \end{cases} \quad E>V_c \quad (4)$$

Where A_0, B_0, A_1, B_1, A_2, B_2,A_3, and B_3 are normalized constants, and

$$k_{nl,1} = \frac{\left(\sqrt{2m_1 E}\right)}{\hbar} \quad (5)$$

$$k_{nl,2} = \frac{\sqrt{2m_2(E - v_c)}}{\hbar} \quad (6)$$

For other position, namely when $E < V_c$ radial part to some extent different from $R_{nl}(r)$ for$E>Vc$

$$R_{nl}(r) = \begin{cases} A0Jl(knl,1r)+B0nl(knl,1r) & 0 < r \le R1 \\ A1hl(+)(knl,2r)+B1\ hl(-)\ (knl,2r) & R1 < r \le R2 \\ A2Jl(knl,1r)+B2nl(knl,1r) & R2 < r \le R3 \\ A3hl(+)\ (knl,2r)+B3\ hl(-)\ (knl,2r) & R3 < r \le R4 \end{cases} \quad E<V_c \quad (7)$$

Where

$$k_{nl,1} = \frac{\left(\sqrt{2m_1 E}\right)}{\hbar} \quad (8)$$

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With the purpose of determinate wave function, $R_{nl}(r)$ should qualify the consequent boundary, convergence and normalization conditions

$$R_{nl,i}(r)=R_{nl,i+1}(r))_{r=ri} \quad (10)$$

$$\left(\frac{1}{m_1}\frac{dR_{nl,i}}{dr} = \frac{1}{m_2}\frac{dR_{nl,i+1}}{dr}\right)\Big|_{r=Ri} \quad (11)$$

$$\int_0^{R_1} R^*_{nl,0}(r)\, r^2\, R_{nl,0}(r)\, dr + \int_{R_1}^{R_2} R^*_{nl,1}(r)\, r^2\, R_{nl,1}(r)\, dr + \int_{R_2}^{R_3} R^*_{nl,2}(r)\, r^2\, R_{nl,2}(r)\, dr = 1 \quad (12)$$

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Since to get *E* we used presented procedure in the Y Fang ET, al work [20]. They determinated target function *T(E)* which related to *M(E).M(E)* is a matrix that depend upon energy (E). If we express *T (E) =det M (E),* energy can be obtained by solving the equation *T (E) =0* [20]. In case B_0 appeared in $R_{nl}(r)$, Neumann function will singular at origin so, we get $B_0=0$. After that, we should determinate uncertain coefficients (A_0,A_1, B_1, A_2, B_2,A_3 and B_3) these coefficients are obtained by solving equations (10)-(12). Next, we have exact wave functions and the third –order nonlinear optical susceptibility $\chi^{(3)}$ can define exactly in the *core-multilayer* structure. Actually, $\chi^{(3)}$ is optical mixing between two incident light beams with frequencies $\omega_1$and ω_2 that denoted as (13)

$$\chi^3(-2\omega_1+\omega_2,\omega_1,\omega_1,-\omega_2) = \frac{-2iN\mu^4}{[i\hbar(\omega_0-2\omega_1+\omega_2)+\hbar\gamma_\perp]\,[i\hbar(\omega_2-\omega_1)+\hbar\gamma_\parallel]} \times \left[\frac{1}{i\hbar(\omega_0-\omega_1)+\hbar\gamma_\perp}+\frac{1}{i\hbar(\omega_2-\omega_0)+\hbar\gamma_\perp}\right] \quad (13)$$

Where μ indicates the dipole transition matrix element, *N* is the number density of carriers and ω_0 is the transition frequency that related to difference energy levels. $\gamma_\parallel$ ($\gamma_\perp$) is the longitudinal (transverse) relaxation time, since in this structure there is spherical symmetric we can get$\gamma_\perp=\gamma_\parallel=\gamma$ and $\gamma=\tau^{-1}$, τ is the relaxation time.The transition frequency ω_0 and dipole transition matrix element μ read

$$\mu = \langle \Phi_i | e\, r | \Phi_j \rangle \quad (14)$$

$$\omega_0 = \frac{E_j - E_i}{\hbar} \quad (15)$$

RESULTS AND DISCUSSION

We presented equation(13) that can reach us to main aim of this study. We select situation which ($\omega_1=\omega_2$) because we want to consider degenerate four wave mixing. So as to simplify calculation the polarization is assumed along the radius direction. The wave functions are defined completely in former section, so we can achieve the optical susceptibilities $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$. $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$.is a complex term so real and imaginary parts are defined separately. Actually $Re\chi^{(3)}(-\omega,\omega,\omega,-\omega)$, is $\chi_{QEOE}(\omega)$ and $Im\chi^{(3)}(-\omega,\omega,\omega,-\omega)$, is $\chi_{EA}(\omega)$ that relate on direct current (DC) Kerr effect and the electro–absorption process respectively. The used parameters in our calculation are taken as fallow, $m^*_{cdse}=0.13m_0$, $m^*_{zns}=0.28m_0$, (m_0 is the mass of rest electron), $V_c=0.9$ev, $N=5\times10^{24}$ m^{-3} and we assume τ=300fs [20,21]. In order to study quantum size effect three different cases are considered in this paper. In the Figure 2 | $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$|, $\chi_{QEOE}(\omega)$, $\chi_{EA}(\omega)$, for various thickness of first shell namely (R_2-R_1) while other thicknesses were constant are shown. Obviously all of them are depend upon thickness of layers, so the bigger thickness, the larger |$\chi^{(3)}(-\omega,\omega,\omega,-\omega)$|,$\chi_{QEOE}(\omega)$ and $\chi_{EA}(\omega)$, because the bigger size the smaller gap between energy levels, so an saw movement to larger wavelengths. As is shown the $\chi_{QEOE}(\omega)$ in the resonance wavelength change its sign from positive to negative, while $\chi_{EA}(\omega)$ for all wavelengths remain negative. Both of them have one peak because there is one photon absorption. Similar result are obtained for various (R_3-R_2) and (R_4-R_3)(Figures 3 and 4). Dependence of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$, on value of τ, is obvious from equation (13). In order to investigation this dependence, different value for τ are assumed, and after plot these cases are seen which larger τ leads to sharper and bigger peak At the same time it's not means movement place of peak (Figure 5).

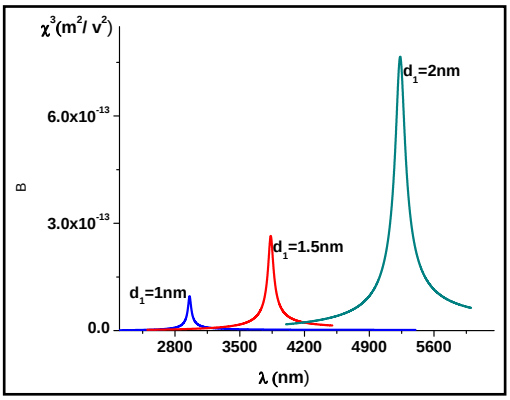

Fig. 2 (a). The modulus of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ versus the wavelength with different d1 and fixed d2, d3 = 1 nm, when E<Vc.

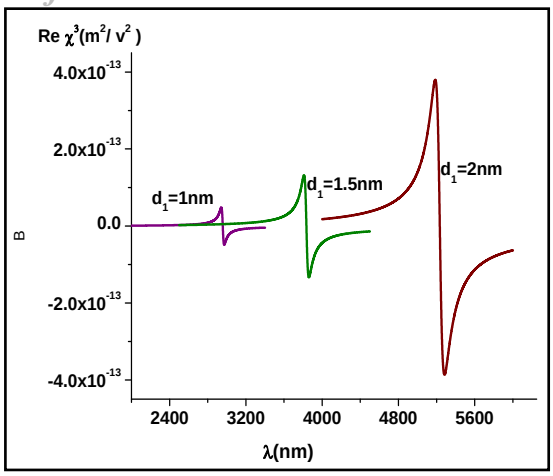

Fig. 2 (b). The Real part of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ versus the wavelength with different d1 and fixed d2, d3 = 1 nm, when E<Vc.

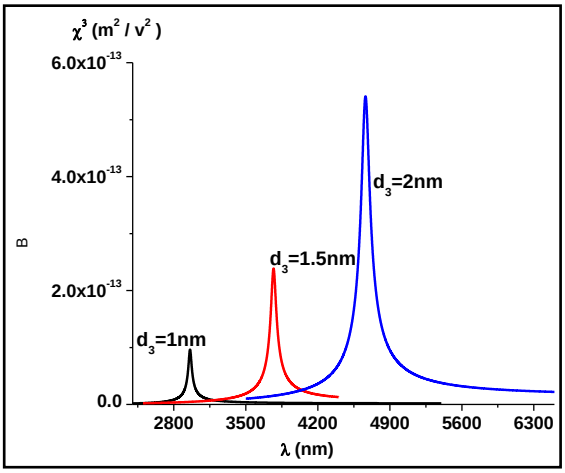

Fig. 4. The modulus of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ versus the wavelength with different d3 and fixed d1, d2= 1 nm, when E<Vc.

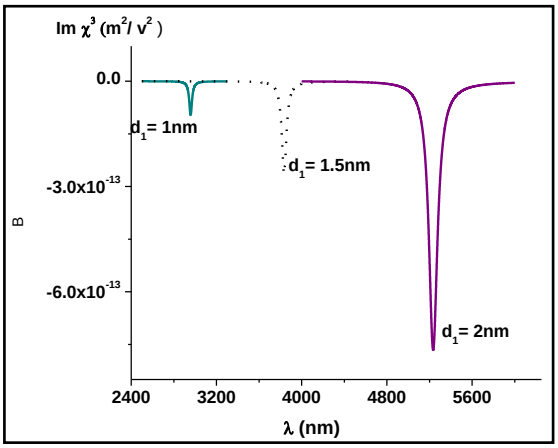

Fig. 2 (c). The imaginary part of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ versus the wavelength with different d1 and fixed d2, d3 = 1 nm, when E<Vc.

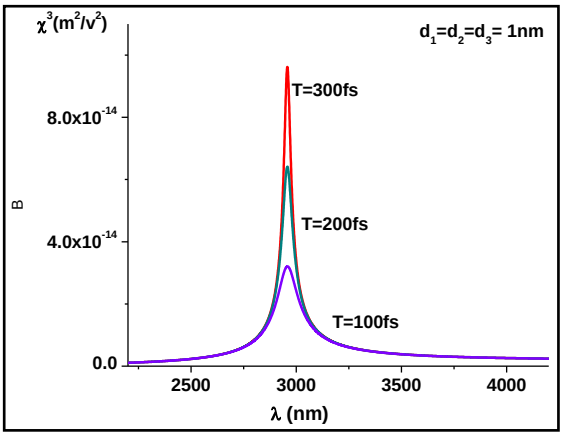

Fig. 5. Peak value of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ as a function of relaxation time τ, in the case that R1 =1 nm, R2 =2 nm,R3=3nm R4=4nm and E<Vc.

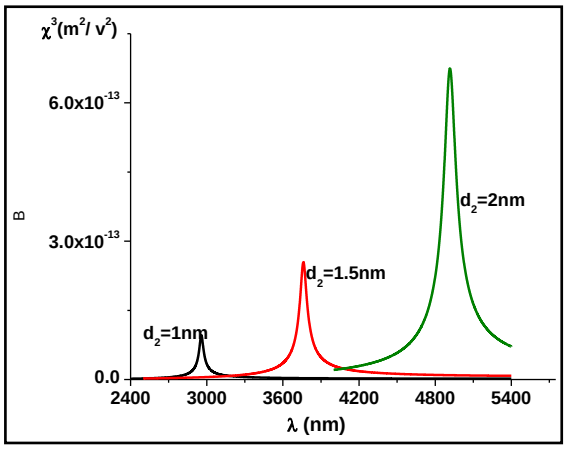

Fig. 3. The modulus of $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ versus the wavelength with different d2 and fixed d1, d3 = 1 nm, when E<Vc.

CONCLUSIONS

Our aim in this theoretical investigation was calculation degenerate four wave mixing effect in the *core-multi shell* structure. Using approximation effective mass and two energy levels model. As we shown, DFWM depends upon thickness of layers and time relaxation. Among various thicknesses, DFWM strongly depends on enhancement of first thickness for $E<V_c$ because of in this situation structure is the biggest. Result of this work can be utilized in fabrication electro optic and photonic devices.

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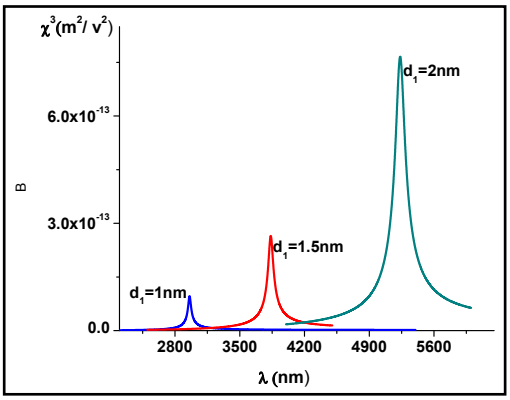

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