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# The control of the nonlinear optical response of semiconductor quantum dots

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ABSTRACT

In the present work, we investigate the nonlinear optical properties emerged from excitonic features in an experimentally realized spherical parabolic semiconductor quantum dot (QD). The lowest exciton states together with relevant wave functions are calculated through the expansion method with direct matrix diagonalization method within the effective mass approximation. The effect of the size of QD and confinement potential in exciton state is studied in details. Results show that with increasing the size of the QD the energy of exciton decreases because of decreasing of the effect of coulomb potential. Using the compact density matrix formalism second order nonlinear optical rectification ($\chi^{(2)}$) are obtained. By means of the applied electric and magnetic field we manipulate the exciton states and control the nonlinear optical response in a typical GaAs, InAs, CdSe QDs. Our model system presents a way to control the performance of excitonic optoelectronic devices based on semiconductor nanostructures.


## 1. Introduction

During past decades remarkable progresses in fabrication techniques of semiconductor nanostructures pave the way towards the implementation of relevant optical phenomena [1–18]. Optical properties of semiconductor nanostructures offer a wide range potential application for optoelectronic devices [19–23]. Interestingly, semiconductor QDs (SQDs) have very important role for device applications based on nonlinear optics. In this case, quantized energy levels in each direction prevent carrier–reservoir interaction in very high level. This leads to a lot of low noise photonic devices emerged from QDs [24,25].

It was shown that the exciton effects have remarkable effects on the nonlinear optical absorption coefficient [26]. In the case of SQDs the analysis of exciton states is inevitable due to the effect of confinement of carriers that increases the oscillator strength of electron–hole excitations. The enhancement of the binding energy of excitons results in the stability of excitons even at room temperature. Due to the importance of excitons for fundamental physics and device application, it is necessary to control and manipulate these states. The control of excitonic optical response can be done by the changing size, composition, and the geometry of QDs and also by applying external electric and magnetic fields. Recently, a number of theoretical investigations of excitonic effects in QDs have been published (for example see references [27–29] and references therein). Most of them are related to the studies of nonlinear optical properties in parabolic or one-dimensional QD including the effect of one of applied external fields. In reference [27] authors studied the optical rectification in the presence of electric and magnetic fields without taking the excitonic effect into account. The study of exciton effects in the model spherical parabolic QDs in the presence of both external fields is still rare.

The second order nonlinear optical rectification plays key role due to its simplicity and lowest order nonlinear effect. We focus on the investigationion of the single particle and exciton spectrum under the effect of applied electric and magnetic field. The density matrix approach within two level system and effective mass approximation are used to calculate the optical rectification of the exciton. Our results present engineering the nonlinear optical response of QDs to produce desirable nonlinear optical rectification.

## 2. Theory

We consider an electron and a hole moving in a spherical QD confined in a parabolic confinement potential under electric and

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magnetic fields applied simultaneously in $z$ direction. The Hamiltonian of this system with the effective mass approach reads [21]:

$$\begin{aligned} H &= H_e + H_h - \frac{e^2}{\varepsilon r} \\ H_e &= \frac{1}{2m_e}\left(p_e - \frac{e}{c}A_e\right)^2 + \frac{1}{2}m_e\Omega^2 r_e^2 - eFz_e \\ H_h &= \frac{1}{2m_h}\left(p_h + \frac{e}{c}A_h\right)^2 + \frac{1}{2}m_h\Omega^2 r_h^2 + eFz_h \\ A_e &= \frac{1}{2}B\times(r_e - r_h),\ A_h = -\frac{1}{2}B\times(r_e - r_h) \end{aligned} \quad (1)$$

where $\Omega$ is the frequency of parabolic confinement taken same for electron and hole and $F$ and $B$ are external electric and magnetic fields, respectively and other parameters have usual meaning.

In terms of the relative and center of mass coordinate the Hamiltonian can be rewritten as:

$$\begin{aligned} H &= H_R + H_r + H_c \\ H_R &= \frac{P^2}{2M} + \frac{1}{2}M\Omega^2 R^2 \\ H_r &= \frac{P^2}{2\mu} + i\mu_B g_B B\frac{\partial}{\partial\varphi} + \frac{1}{2}\mu\Omega^2 r^2 + \frac{1}{2}\mu\omega^2\left(x^2+y^2\right) - \frac{e^2}{\varepsilon r} - eFz \\ H_c &= \frac{i\hbar e}{M}Br\times\nabla_R \end{aligned} \quad (2)$$

where

$$\begin{aligned} \mu_B &= \frac{\hbar e}{2gc} \\ g_B &= \left(\frac{1}{m_e} - \frac{1}{m_h}\right) \\ \omega &= \frac{Be}{\mu c} \end{aligned} \quad (3)$$

Indices $r$, $R$ indicates relative and center of mass motions. One can take the center of mass as the reference of energy and neglect $H_c$ that indicates the coupling of relative motion with center of mass motion. Hamiltonian has spherical symmetry which implies that the total orbital angular momentum, $L$, is a conserved quantity, and the eignestates of the exciton in spherical (cylindrical) QDs can be classified according to the total orbital angular momentum.

The total Hamiltonian can be diagonalized in the space by expanding wave function with a fixed quantum numbers ($n$,$l$,$m$) as:

$$\Phi_m = \sum_i c_i\psi_i(\vec{r}) \quad (4)$$

$\psi_i(\vec{r})$ is 3D spherical harmonic oscillator eigenvectors as $\psi_i(\vec{r}) = R_{n_i l_i}(\vec{r})Y_{l_i m_i}(\theta,\varphi)$ with the corresponding eigenvalue $E_i = \left(2n_i + l_i + \frac{3}{2}\right)\hbar\omega_0$. Matrix elements of the relative part of Hamiltonian reads:

$$\begin{aligned} \left\langle \Phi_m \left| H \right| \Phi_m \right\rangle &= \sum_{i,j}\left\{\left[2n_i + l_i + 3/2\right]\hbar\omega_0\delta_{ij} + V_{ij}\right\}, \\ V_{ij} &= -\int d\Omega\int_0^\infty R_{n_i l_i}(r)Y^*_{l_i,m}(\theta,\varphi)\left[\frac{e^2}{r} + efr\cos\theta - i\mu_B g_B B\frac{\partial}{\partial\varphi} + \frac{1}{2}\mu\omega^2(x^2+y^2)\right]\times R_{n_j l_j}(r)Y_{l_j,m}(\theta,\varphi)r^2 dr. \end{aligned} \quad (5)$$

### 2.1. Optical response

The Hamiltonian of light-carrier interaction in a QD can be described in dipole approximation and can be expressed in terms of the relative coordinate and does not depend on the center of mass coordinate. We assume that interband transitions can be neglected, so-that the matrix element of the polarization operator in the second quantization is given by [30]:

$$P_z^+ = M_{cv}\int d^3r\psi_e^+(\vec{r})\psi_h^+(\vec{r}) + h.c. \quad (6)$$

where $M_{cv}$ is the valence-conduction band dipole matrix element and $\psi_e^+$, $\psi_h^+$ are the electron, hole creation operators, respectively. The relevant dipole matrix elements are those between vacuum ($\left|0\right\rangle$), exciton ($\left|1\right\rangle$) states and denoted $M_{12}$. Relevant dipole transition can be written as following well-known relation:

$$M_{12} = \left\langle 1\right| P^+ \left|0\right\rangle = M_{cv}\int d^3r\Phi_m. \quad (7)$$

Now, the oscillator strength of excitonic transition can be calculated (not shown here). By starting with equation of motion for the density matrix of a two level system and applying some algebraic manipulation, one obtain second order nonlinear optical rectification ($\chi^{(2)}$) of exciton as a two level system can be written as [17–19]:

$$\chi_0^{(2)} = \frac{4\sigma_v}{\varepsilon_0}M_{21}^2\delta_{21}\frac{E_{21}^2\left(1+\frac{T_1}{T_2}\right) + \left[(\hbar\omega)^2 + \left(\frac{\hbar}{T_2}\right)^2\right]\left(\frac{T_1}{T_2} - 1\right)}{\left[(E_{21}-\hbar\omega)^2 + \left(\frac{\hbar}{T_2}\right)^2\right]\left[(E_{21}+\hbar\omega)^2 + \left(\frac{\hbar}{T_2}\right)^2\right]}. \quad (8)$$

$E_{21}$ indicates the transition energy, $M_{21}$ denotes the dipole moment element between two levels. $T_1$, $T_2$ are dephasing times that we take their magnitudes as 1 ps and 2 ps, respectively. $\sigma_v$ is electron density in QD that is set to $5\times10^{18}$ cm$^{-3}$ [31].

**Table 1**
Values of physical parameters for three semiconductors: the electron mass $m_e$, the hole mass $m_h$.

| | $m_e$ | $m_h$ |
|---|---|---|
| GaAs | $0.067m_0$ | $0.625m_0$ |
| CdSe | $0.13m_0$ | $0.22m_0$ |
| InAs | $0.023m_0$ | $0.41m_0$ |

## 3. Results and discussion

In this work we consider the freestanding GaAs, CdSe and InAs QDs. The relevant parameters used in the calculation are given in Table 1. The behavior of exciton energy versus the QD radius is depicted in Fig. 1. It is clear that by increasing the QD radius the exciton energy is reduced because of the weakening of confinement effect, also for small radii the effect of effective mass on exciton energy is negligible while for larger QDs exciton energy has higher value for bigger values of effective mass. From Table and arrangement of curves in Fig. 1, it can be seen that the electron effective mass plays main role in calculations to determine exciton energy. Fig. 2 shows the second order nonlinear optical rectification versus incident photon energy for different value of confining potential. It can be seen that by increasing the confining potential the peak of $\chi^{(2)}$ blueshiftted and its magnitude reduced. This behavior returns to the reduction of electron–hole wave functions overlapping and reduction of parameter $M_{12}$ and increasing the single particle level spacing. In Fig. 3 we present the frequency dispersion of $\chi^{(2)}$ around the one photon resonance at $\omega = E_{21}/\hbar$ for different values of