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Exciton related nonlinear optical properties of a spherical quantum dot

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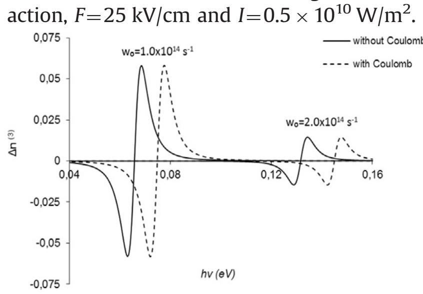
HIGHLIGHTS

- The exciton related nonlinear optical properties of a quantum dot is calculated analytically.
- The roles of confinement, Coulomb interaction and electric field are studied.
- No assumptions are made about the strength of confinement.

GRAPHICAL ABSTRACT

Non-linear refractive index change as a function of photon energy, with and without Coulomb interaction, $F=25$ kV/cm and $I=0.5\times10^{10}$ W/m^2.



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ABSTRACT

The nonlinear optical properties of an exciton in a spherical quantum dot (QD) is studied analytically. The nonlinear optical coefficients are calculated within the density matrix formalism. The electronic problem is solved within the effective mass approximation. The contributions from the competing effects of the confinement, the Coulomb interaction, and the applied electric field are calculated and compared with each other. We have made no assumptions about the strength of the confinement. We concentrate the effect of the Coulomb interaction. Our results may provide an input for optimization of the nonlinear optical coefficients.

1. Introduction

The study of the exciton related nonlinear optical properties of quantum dots is of great importance since it is technologically relevant and it provides challenging physics [1–3].

There are a few important factors that affect the linear and nonlinear optical properties. The first is the confinement of the exciton. The exciton in a QD is not free to move in any directions. This gives way to the quantization of the electronic levels. The second important factor is the Coulomb interaction between the electron and the hole. In the strong confinement limit this interaction is neglected or treated as a perturbation. This is regularly done in many earlier investigations [4–6]. However, this interaction is important and it is this interaction that forms the exciton at the first place. The third factor is the presence of the externally applied electric and magnetic fields. They influence the electronic structure and thus the optical properties. The fourth one is the intensity of the incoming photons, which has a direct effect on the nonlinear contributions. One can also investigate other factors such as the presence of impurities, strain, temperature etc. But in this work, we consider the effects of the confinement, the Coulomb interaction and the applied electric field on the exciton related optical properties of a spherical quantum dot.

There have been a number of investigations of the problem of

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exciton-related nonlinear optical properties of a QD [7–20]. The majority of these works are either numerical or considering only the strong confinement limit. It is well understood that the nonlinear optical coefficients are considerably larger when excitonic effects are taken into account. Some of the previous works make an approximation about the shape of the dot. Some take it as one-dimensional [7,13,17], two-dimensional [8,16], disk-like [10,11,15], or semi-spherical [9]. This may simplify the calculation of the electronic structure of the QD. A number of investigations consider only the strong confinement limit where the Coulomb interaction may be neglected [7,8,10,13]. This obviously simplifies the calculation. There is also a scatter in the nonlinear optical coefficients taken into consideration. Some of the works consider only the refractive index change, and/or the optical rectification coefficient.

The computational method used also varies from the direct numerical matrix diagonalization to a variational calculation. The Coulomb potential is considered as a perturbation by Wenfang Xie's group [21–23]. Their work needs to be extended to more nonlinear optical coefficients and emphasis should be given to the effects of the Coulomb interaction.

It is thus well established, through a combination of fragmented studies that the nonlinear optical properties of QDs depend mainly on the dot size, external fields and the incident photon intensity. We now need an analytical calculation that may show the various competing effects clearly.

In this work, no assumptions are made about the strength of the confinement. An analytical solution is done for the electronic structure of the QD, by including a parabolic potential to describe the confinement, and an external electric field in the Hamiltonian. We have considered small electric fields and treated the Coulomb term as a perturbation [24]. The main aim of the paper is to find out the effect of the Coulomb interaction on the nonlinear optical properties of a QD in both strong and weak confinement limits.

2. Theoretical framework

The Hamiltonian for an exciton in a spherical QD with a parabolic potential, within the effective mass approximation is

$$H = -\frac{\hbar^2}{2m_e}\vec{\nabla}_e^2 + \frac{1}{2}k_e r_e^2 - \frac{\hbar^2}{2m_h}\vec{\nabla}_h^2 + \frac{1}{2}k_h r_h^2 - \frac{\gamma}{\left|\vec{r}_h - \vec{r}_e\right|} + e\vec{F}\cdot\left(\vec{r}_e - \vec{r}_h\right), \tag{1}$$

where, $m_e(m_h)$ and $\vec{r}_e(\vec{r}_h)$ denote the effective mass, and the position vector of the electron (hole), respectively. $k_e(k_h) = (m_h)\omega_o^2$, where ω_o defines the parabolic confining potential. $\gamma = e^2/\epsilon$ is a positive constant, e is the absolute value of the electron charge, ϵ is the dielectric constant and $\vec{F}$ is the external electric field applied in z direction. The details are given in our earlier paper [21]. The eigen values and the corresponding eigen functions are

$$E_n = \left[n + \frac{1}{2}\left(3 - \alpha\beta^2\right)\right]\hbar\omega_o, \tag{2}$$

$$\psi_{n,l,m} = Ne^{-\frac{\rho^2}{2}+im\phi}(\rho^2)^{l/2}L_n^{l+1/2}\left[\rho^2\right]P_l^m[\cos\theta], \tag{3}$$

where,

$$\alpha = \frac{\mu\omega_o}{\hbar},\ \beta = -\frac{qF}{\mu\omega_o^2} \text{ and } n = 2\kappa + l,\ \text{with } \kappa = 0, 1, 2, \ldots,\ l = 0, 1, 2, \ldots,$$
$$\rho^2 = \frac{\mu\omega}{h}(x^2 + y^2 + (z-\beta)^2) \tag{4}$$

and $\mu = m_e m_h/M$ is the electron–hole reduced mass.

We consider an optical radiation of angular frequency ω applied to the system with the polarization along the growth direction. The incident field can be written as

$$F(t) = \sum_j F(\omega_j)\,\mathrm{Exp}\left(-i\omega_j t\right) \tag{5}$$

where the summation is over all frequencies. Using the density matrix formalism, one can write the first-and the third-order susceptibilities as [25]

$$\chi^{(1)}(\omega) = \frac{\rho_s\left|\mu_{10}\right|^2}{E_{10} - \hbar\omega - i\hbar\Gamma_o}, \tag{6}$$

$$\chi^{(3)}(\omega, I) = \frac{2\pi I\rho_s\left|\mu_{10}\right|^4}{n_r c(E_{10} - \hbar\omega - i\hbar\Gamma_o)} \times \left[\frac{4}{(E_{10} - \hbar\omega)^2 + (\hbar\Gamma_o)^2} - \frac{\left|\mu_{11} - \mu_{00}\right|^2}{\left|\mu_{10}\right|^2}\frac{1}{(E_{10} - \hbar\omega - i\hbar\Gamma_o)(E_{10} - i\hbar\Gamma_o)}\right], \tag{7}$$

and the second-order nonlinear optical rectification coefficient is given by [26–28]

$$\chi_0^{(2)} = \frac{4\rho_s\left(\mu'_{01}\right)^2\left|\mu'_{11} - \mu'_{00}\right|\left\{\left(E'_{10}\right)^2\left(1 + \frac{T1}{T2}\right) + \left[(\hbar\omega)^2 + \left(\frac{\hbar}{T2}\right)^2\right]\left(\frac{T1}{T2} - 1\right)\right\}}{\varepsilon_o\left[\left(E'_{10} - \hbar\omega\right)^2 + \left(\frac{\hbar}{T2}\right)^2\right]\left[\left(E'_{10} + \hbar\omega\right)^2 + \left(\frac{\hbar}{T2}\right)^2\right]}, \tag{8}$$

where μ_{ij}, μ'_{ij} are the matrix elements of the unperturbed and the perturbed electric dipole moments. The change in the linear, the third-order nonlinear and the total refractive index (RI) due to the incident field are given by

$$\Delta n^{(1)}(\omega) = \frac{1}{2\pi\varepsilon_o n_r}\mathrm{Re}\left[\chi^{(1)}(\omega)\right], \tag{9}$$

$$\Delta n^{(3)}(\omega, I) = -\frac{1}{2\pi\varepsilon_o n_r}\mathrm{Re}\left[\chi^{(3)}(\omega)\right], \tag{10}$$

$$\Delta n^{(total)}(\omega, I) = \Delta n^{(1)}(\omega) + \Delta n^{(3)}(\omega, I). \tag{11}$$

Here, I is the intensity of the incident field, n_r is the refractive index, c is the speed of light and $\Gamma_0 = 1/T_0$, where T_0 is the relaxation time.

The simplicity of the expressions obtained is solely due to the parabolic confinement assumed and the resulting harmonic oscillator wave functions considered in this work. It is well known that the first-order perturbation treatment within the customary infinite-barrier confinement leads to an infinite series when one evaluates the first-order correction to the unperturbed exciton wave function.

3. Results and discussion

The numerical values are chosen for a typical GaAs/Al_xGa_{1-x}As system with $x = 0.3$. The input parameters are taken as, the electron density $\rho_s = 5.0 \times 10^{22}\ \mathrm{m}^{-3}$, reduced mass $\mu = 0.0549m_o$, $m_e = 0.067m_o$, $m_h = 0.340m_o$, where m_o is the free electron mass. The relaxation time is $T_o = 0.14$ ps, the dielectric constant as $\epsilon = 12.4$.