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Optical rectification and third harmonic generation of spherical quantum dots: Controlling via external factors

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ABSTRACT

In this paper simultaneous effects of pressure, temperature, external electric field and laser radiation on the optical rectification and third harmonic generation of a spherical quantum dot with parabolic confinement and dressed impurity are studied. By means of matrix diagonalization technique, energy eigenvalues and functions are evaluated and used to find the optical rectification coefficient and third harmonic generation of the system via density operator method. It is shown that these nonlinear optical quantities strongly depend on pressure, temperature, electric field, confinement frequency and dressing laser intensity. Obvious effects of these external factors propose new facilities with different effects to control nonlinear optical properties of such systems.

1. Introduction

Semiconductor quantum dots have unique optical properties that turn them into attractive candidates for many optoelectronic applications, such as a new type of lasers [1–3] and single photon source for use as qubits which are building blocks for quantum computers [4–6].

Additionally, a doped hydrogenic impurity in a quantum dot influences both the electronic mobility and its optical properties because of the Coulomb interaction between the electron and the impurity ion [7–10]. The three-dimensional confinement, depending on the size of the dot, causes the electron to move near the impurity ion which is the enhancing effect on the binding energy due to the increasing interaction strength. Moreover a laser radiation can modify the impurity effects which in turn affect on electrical and optical properties of nanostructures [11–13].

In recent years, great attention has been paid to the nonlinear optical properties of low-dimensional semiconductor structures especially quantum dots (QD) in both theoretical and applied physics [14–16]. Enhanced linear and nonlinear optical properties due to the large transition dipole matrix elements and their tunability via external factors and dimensions are the potentials of QDs for a wide range of device applications [17–23]. Among the nonlinear optical properties, more and more attention had been paid to the optical absorbtion coefficient (AC) and refractive index (RI) [24–29], second-order nonlinear optical properties [30,31], such as optical rectification (OR) [32], second-harmonic generation (SHG) [33,34], electro-optic effect (EOE) [35] and less to the third-harmonic generation (THG) [36].

In the current work the matrix diagonalization technique with more accuracy and flexibility is used to find the ORC and THG of a parabolic QD with doped hydrogenic impurity under the combined action of external electric field, pressure, temperature and laser radiation. To our knowledge this study has not been done so far especially for THG. Our results show the considerable effects of these external agents on the ORC and THG of the system.

2. Model

In the effective mass approximation, Hamiltonian for a hydrogenic on-center shallow donor impurity confined in a spherical quantum dot, under the influence of a laser radiation with frequency ω_d, is given by [37]

$$H = \frac{1}{2m_e^*(P,T)}\left[\vec{P}_e + \frac{e}{c}\vec{A}(t)\right]^2 + V_c(\vec{r}) - \frac{e^2}{\varepsilon(P,T)|\vec{r}|} \tag{1}$$

where $\vec{P}_e$ is the momentum operator, $\vec{A}(t) = A_0(\cos\omega_d t\hat{i} + \sin\omega_d t\hat{j})$ is the vector potential of the laser field, P is the hydrostatic pressure in units of kbar, T is temperature in units of Kelvin, $V_c(\vec{r})$ is the confinement potential, $\varepsilon(P,T)$, $m_e^*(P,T)$ are the pressure and temperature dependent dielectric constant and effective mass, respectively, and $\vec{r}$ is the position vector of the electron from the center of

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dot. In the high frequency limit, the laser dressed eigenstates are the solutions of the time independent Schrödinger equation [37,38]

$$\left[-\frac{\hbar^2}{2m_e^*(P,T)}\vec{\nabla}^2 + V_d(\vec{r},\vec{\alpha}_0(P,T)) + V_c(\vec{r})\right]\phi(\vec{r}) = E\phi(\vec{r}), \tag{2}$$

where $V_d(\vec{r},\vec{\alpha}_0(P,T))$ is the laser dressed potential which depends on the laser frequency, laser intensity, I_d, pressure and temperature through $\vec{\alpha}_0(P,T)$

$$\vec{\alpha}(P,T) = \left(\frac{I_d^{1/2}}{\omega_d^2}\right)\left(\frac{e}{m_e^*(P,T)}\right)\left(\frac{8\pi}{c}\right)^{1/2} \tag{3}$$

and in the case of on-center hydrogenic impurity becomes

$$V_d(\vec{r},\vec{\alpha}_0(P,T)) = -\frac{e^2}{\varepsilon(P,T)(r^2+\alpha_0^2(P,T))^{1/2}}. \tag{4}$$

In the current work we have considered the pressure- and temperature-dependent parabolic confinement potential in the form of

$$V_c(\vec{r}) = \frac{1}{2}m_e^*(P,T)|\vec{r}|^2\omega_0^2, \tag{5}$$

where ω_0 is the confinement frequency. In all equations variation of the dielectric constant, $\varepsilon(P,T)$ and effective mass $m_e^*(P,T)$ with temperature and pressure can be found in [39,40].

To find the energy eigenvalues and functions of the system we have constructed the matrix representation of the total Hamiltonian of Eq.(2) by the following eigenfunctions of the 3-dimensional harmonic oscillator as the basis set,

$$\phi_i(\vec{r}) = \frac{1}{r^{3/2}}\mathrm{WhittakerM}\left(\frac{E_i}{2\hbar\omega_0}, \frac{1}{2}l+\frac{1}{4}, \frac{m_e^*(P,T)\omega_0^2r^2}{\hbar}\right). \tag{6}$$

Direct diagonalization gives the energies and related wave functions of the system simultaneously.

3. Optical rectification and third harmonic generation

In order to calculate the second-order ORC of the host material due to the intersubband transitions, we assume the interaction of a polarized monochromatic electromagnetic field with an ensemble of quantum dots. The electric field vector of this optical wave is

$$\mathbf{E}(t) = \tilde{\mathbf{E}}e^{i\omega t} + \tilde{\mathbf{E}}^*e^{-i\omega t}. \tag{7}$$

Due to the time dependent interaction, the time evolution of the matrix elements of one-electron density operator $\hat{\rho}$ is given by the *von-Neumann* equation [41]

$$\frac{\partial\rho_{ij}}{\partial t} = \frac{1}{i\hbar}[\hat{H} - \hat{M}\cdot\mathbf{E}(t), \hat{\rho}]_{ij} - \Gamma_{ij}(\hat{\rho} - \hat{\rho}^{(0)})_{ij}, \tag{8}$$

here $\hat{H}$ is the Hamiltonian of the system in the absence of electromagnetic field E(t), $\hat{M}$ is the dipole moment operator, and $\Gamma_{ij} = 1/T_{ij}$ is the phenomenological relaxation rate, caused by the electron–phonon, electron–electron and other collision processes. Also, $\hat{\rho}^{(0)}$ is the unperturbed density operator. Eq. (8) can be solved using the standard iterative method

$$\hat{\rho}(t) = \sum_{n=0}^{\infty}\hat{\rho}^{(n)}, \tag{9}$$

with

$$\frac{\partial\rho_{ij}^{(n+1)}}{\partial t} = \frac{1}{i\hbar}[H_0, \rho^{(n+1)}]_{ij} - i\hbar\Gamma_{ij}\rho_{ij}^{(n+1)} - \frac{1}{i\hbar}[M, \rho^{(n)}]_{ij}E(t). \tag{10}$$

In addition, the electronic polarization of the system due to the electric field, up to the third order in $\tilde{E}$, can be written as

$$P(t) = (\varepsilon_0\chi_\omega^{(1)}\tilde{E}e^{i\omega t} + \varepsilon_0\chi_0^{(2)}\tilde{E}^2 + \varepsilon_0\chi_{2\omega}^{(2)}\tilde{E}^2e^{2i\omega t} + \varepsilon_0\chi_\omega^{(3)}\tilde{E}^2\tilde{E}e^{i\omega t} + \varepsilon_0\chi_{3\omega}^{(3)}\tilde{E}^3e^{3i\omega t} + c.\,c.\,). \tag{11}$$

where ε_0 is the permittivity of the free space, $\chi_\omega^{(1)}$, $\chi_0^{(2)}$, $\chi_{2\omega}^{(2)}$, $\chi_\omega^{(3)}$ and $\chi_{3\omega}^{(3)}$ are the linear, optical rectification, second harmonic generation, third-order and third harmonic generation susceptibilities, respectively. Hereafter the attention will be confined on the two level system, so from Eqs. (10) and (11) the following expression for the second-order ORC is deduced [32,42]

$$\chi_0^{(2)} = \frac{4\sigma_v}{\varepsilon_0}M_{21}^2\delta_{12}\frac{E_{21}^2(1+\Gamma_2/\Gamma_1) + (\hbar^2\omega^2+\hbar^2\Gamma_2^2)(\Gamma_2/\Gamma_1 - 1)}{[(E_{21}-\hbar\omega)^2+\hbar^2\Gamma_2^2][(E_{21}+\hbar\omega)^2+\hbar^2\Gamma_2^2]}, \tag{12}$$

where, $\delta_{12} = M_{22} - M_{11}$. In addition the analytical expression of the THG within a four level system is obtained as follows [36]

$$\chi_{3\omega}^{(3)} = \frac{\sigma_v}{\epsilon_0} \times \frac{M_{12}M_{23}M_{34}M_{41}}{(\hbar\omega - E_{21} - i\hbar\Gamma_{21})(2\hbar\omega - E_{31} - i\hbar\Gamma_{31})(3\hbar\omega - E_{41} - i\hbar\Gamma_{41})}. \tag{13}$$

In the above equations σ_v is the carrier density, $M_{ij} = |\langle\psi_i|er\cos(\theta)|\psi_j\rangle|(i,j = 1,2,3,4)$ are the matrix elements of the dipole moment, $\psi_i(\psi_j)$ are the eigenfunctions and $E_{ij} = E_i - E_j$ is the energy difference between these states and $\Gamma_{21} = 2\Gamma_{31} = 3\Gamma_{41} = \Gamma_0$ where $\Gamma_j = 1/T_j$ are damping terms associated with the life time of the electrons due to intersubband scattering.

4. Results and discussions

In this section we do our calculations for a typical GaAs QD where its effective mass, m^*, and dielectric constant, ϵ, depend on pressure and temperature. In addition optical nonlinearities originate from an intense laser radiation whose intensity, I, is much larger than the dressing laser intensity, I_d, which dresses the coulomb potential of impurity.

Since ORC is proportional to the geometrical factor (GF), $M_{12}^2\delta_{12}$, directly thus this quantity is calculated and plotted in this section for better explaining the results. In Fig. 1 GF is plotted as a function of electric field strength for different values of laser parameter, α. It is seen that GF increases with increasing F and α. With increasing α effects of Coulomb dressed potential decreases, subband energy difference reduces, wave function overlap increases and in effect $M_{12}^2\delta_{12}$ grows. In Fig. 2 ORC, $\chi_{2\omega}^{(2)}$, is plotted as a function of incident photon energy for different values of α. Since increasing α reduces E_{21} thus resonant peak of ORC moves toward lower energies and its value grows due to the increment of GF. It is not worthy to mention that laser parameter can be tuned via I_d, externally.

To study the effects of dimension on the ORC of the system in the presence of pressure and temperature in Figs. 3 and 4 GF and $\chi_{2\omega}^{(2)}$ are plotted with different confinement frequency, ω_0. Because ω_0 is related to the effective length via the relation $L = \sqrt{\hbar/\omega_0 m^*}$ thus this quantity is a measure for the dimensions of the system. From Fig. 3 it is seen that GF grows as ω_0 decreases (L increases). This confirm the fact that, an increase in the dot size decreases E_{21},