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Electronic structure and nonlinear optical rectification in a quantum dot: effects of impurities and external electric field

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Abstract

The electronic structure of a spherical quantum dot with parabolic confinement that contains a hydrogenic impurity and is subjected to a DC electric field is studied. In our calculations we vary the position of the impurity and the electric field strength. The calculated electronic structure is further used for determining the nonlinear optical rectification coefficient of the quantum dot structure. We show that both the position of the impurity and the strength of the electric field influence the nonlinear optical rectification process.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In the past 15 years the optical properties of semiconductor nanostructures have attracted considerable attention [1, 2]. In this area significant interest has been given to semiconductor quantum wells and quantum dots that are characterized by an asymmetric confining potential [3–22]. The asymmetry of the confining potential can be produced by either advanced material growing technology, such as molecular-beam epitaxy and metallic–organic chemical vapor deposition, or by the application of a static electric field to a nanostructure with symmetric confining potential.

Among the nonlinear optical processes in semiconductor nanostructures, interest has been paid to second-order nonlinear optical properties, such as nonlinear optical rectification [7, 15, 18, 19, 21] and second-harmonic generation [6, 7, 11, 17]. This happens as the second-order nonlinear processes have magnitudes that are usually stronger than those of high-order ones, if the quantum system demonstrates significant asymmetry.

In parallel, impurities such as confined donors or acceptors in quantum dots have been extensively investigated [23–37]. Several numerical methods, both perturbative and exact numerical, have been developed in order to systematically investigate the physical properties of impurities embedded in a quantum dot. In most theoretical investigations the confinement

potential of quantum dots is assumed to possess square (either finite or infinite) or parabolic shape. The parabolic confinement is more appropriate when the quantum dots are fabricated by etching processing on a quantum well, by ion implantation or by application of electrostatic gates.

In the present work we study three-dimensional semiconductor quantum dots with parabolic confinement containing donor impurities. The system can also interact with a DC external electric field. We use the potential morphing method for the calculation of the electronic structure of the system [37–39]. In this work, we concentrate on the calculation of the ground and first excited energy states of the quantum dot system. The effects of the external electric field are found to be more significant for larger dot radii. Moreover, the influence of the external electric field increases as the position of the hydrogenic impurity becomes more off-centre.

Then, by a density matrix approach and a perturbation expansion method, within a two-level system approach, the nonlinear optical rectification coefficient of the system is obtained. For nonzero electric field the system becomes effectively asymmetric, and thus with finite nonlinear optical rectification coefficient. A systematic behaviour is reported, revealing that the increase of the electric field amplitude enhances the maximum value of the nonlinear optical rectification coefficient and shifts this maximum to smaller photon energies. This effect is more pronounced for larger quantum dot sizes. The observed behaviour is attributed to the increase of the asymmetry of the system.

2. Numerical results

The Hamiltonian of the system in three dimensions is given by the expression

$$H = \frac{p^2}{2m^*} + \frac{1}{2}m^*\omega_0^2r^2 - \frac{e^2}{\varepsilon|\vec{r} - \vec{r}_i|} + eE \cdot (z - z_i), \quad (1)$$

where E is the external applied electric field amplitude (taken along the z -axis), z is the position of the electron along the z -axis, z_i is the position of the donor impurity along the z -axis, and $\vec{r}_i = \hat{z}z_i$. Also, m^* is the effective electron mass, ε is the relative dielectric constant, and ω_0 is the characteristic angular frequency of the harmonic oscillator.

We are interested in obtaining the energies and the wavefunctions of the ground and the first excited state of the Hamiltonian (1), as these are needed for the calculation of the optical rectification coefficient. We will use the potential morphing method for this calculation [37–39]. The three-dimensional stationary Schrödinger equation for a particle of mass m in a potential $v_R(\vec{r}) + v_S(\vec{r})$ has the form

$$\left\{ -\frac{\hbar^2}{2m}\nabla^2 + v_R(\vec{r}) + v_S(\vec{r}) \right\} \Phi_{R+S}(\vec{r}) = E_{R+S} \Phi_{R+S}(\vec{r}), \quad (2)$$

where $v_R(\vec{r})$ is a (reference) potential with known eigenfunctions $\Phi_R(\vec{r})$ and eigenvalues E_R , and $v_S(\vec{r})$ is an arbitrary shape interaction potential. The essential point is that the transition from potential $v_R(\vec{r})$ to potential $v_R(\vec{r}) + v_S(\vec{r})$ can be performed by means of the time-dependent Schrödinger equation as follows: using the potentials $v_R(\vec{r})$ and $v_S(\vec{r})$ we formulate a time-dependent Schrödinger equation

$$i\hbar \frac{\partial \Phi(\vec{r}, t)}{\partial t} = \left\{ -\frac{\hbar^2}{2m}\nabla^2 + v_R(\vec{r}) + v_t(\vec{r}) \right\} \Phi(\vec{r}, t), \quad (3)$$

with [38]

$$v_t(\vec{r}) = \sigma(t)v_S(\vec{r}), \quad (4)$$

where $\sigma(t)$ has the following property:

$$\sigma(t) = \begin{cases} 0, & t \leq t_a \\ 1, & t \geq t_b. \end{cases} \quad (5)$$

For $t_a \leq t \leq t_b$ the function $\sigma(t)$ may have any shape, but it should increase monotonically.

We solve equation (3) iteratively. After several iteration steps with respect to time t and for $t > t_b$, the energy eigenvalue E_{R+S} for the potential $v_R(\vec{r}) + v_S(\vec{r})$ is given by

$$E_{R+S} = \int d^3r \Phi_{R+S}^*(\vec{r}) \left\{ -\frac{\hbar^2}{2m} \nabla^2 + v_R(\vec{r}) + v_S(\vec{r}) \right\} \Phi_{R+S}(\vec{r}), \quad (6)$$

where $\Phi_{R+S}(\vec{r})$ is the wavefunction of the system.

In our numerical calculations we take as a reference system ($v_R(\vec{r})$) the usual harmonic oscillator in three-dimensions with eigenfunctions

$$\Psi_{nlm}(r, \theta, \phi) = r^l e^{-r^2/2R^2} {}_1F_1(-n, l + 3/2; r^2/R^2) Y_{lm}(\theta, \phi), \quad (7)$$

with $R = \sqrt{\hbar/m^* \omega_0}$ being the characteristic radius of the quantum dot. In our study we assume a CdS quantum dot and take $m^* = 0.18m_0$ (m_0 is the mass of a free electron) and $\varepsilon = 5.23$. The Bohr radius of this system is 3 nm. Moreover, in our numerical calculations the $v_S(\vec{r})$ interaction potential corresponds to the last two terms of the Hamiltonian of equation (1).

The results of the energies of the ground and the first excited state (the state that has been obtained by the 010 reference state, and it is denoted as the 010 excited state later on) of the system as a function of the external electric field amplitude E are shown in figure 1 for several values of the radius of the quantum dot and for the hydrogenic impurity taken both on-centre and off-centre. We choose the quantum dot radius to be $R = 2$ nm, just below the Bohr radius, $R = 3$ nm, at the Bohr radius, and $R = 5$ nm, above the Bohr radius. It is found that for the case of zero external field the difference between the first excited state and the ground state is linearly proportional to the inverse of the square of the quantum dot radius. We also find that for a specific quantum dot radius and position of the impurity the energy difference is weakly influenced by the external electric field, but one may note that the increase of the electric field amplitude leads to a decrease of the energy difference of the first excited state and the ground state of the system. The latter is more pronounced in figure 1(f).

We would like to mention that the reference system of the three-dimensional harmonic oscillator has three-fold degeneracy in the first excited state. This also holds in the case of an on-centre impurity with zero electric field. As it is shown in figure 2, the electric field will gradually lift this degeneracy and one obtains a nondegenerate first excited state from state 010 and two degenerate states from states 011 and 01-1. We stress here that from these states only the state that we denote with 010 contributes to the nonlinear optical rectification coefficient, as the corresponding matrix elements of the ground state with the other two states have zero values.

The nonlinear optical rectification coefficient of an asymmetric quantum dot can be obtained by a density matrix approach and a perturbation expansion method, and it can be written, within a two-level system approach, as [7, 15, 18, 19, 21]

$$\chi_0^{(2)}(\omega) = 4 \frac{e^3 \sigma_s}{\varepsilon_0 \hbar^2} \mu_{01}^2 \delta_{01} \frac{\omega_{01}^2 \left(1 + \frac{T_1}{T_2}\right) + \left(\omega^2 + \frac{1}{T_2^2}\right) \left(\frac{T_1}{T_2} - 1\right)}{\left[(\omega_{01} - \omega)^2 + \frac{1}{T_2^2}\right] \left[(\omega_{01} + \omega)^2 + \frac{1}{T_2^2}\right]}, \quad (8)$$

where

$$\mu_{01} = \langle \Phi_0 | z | \Phi_1 \rangle \quad (9)$$

$$\delta_{01} = \langle \Phi_1 | z | \Phi_1 \rangle - \langle \Phi_0 | z | \Phi_0 \rangle, \quad (10)$$

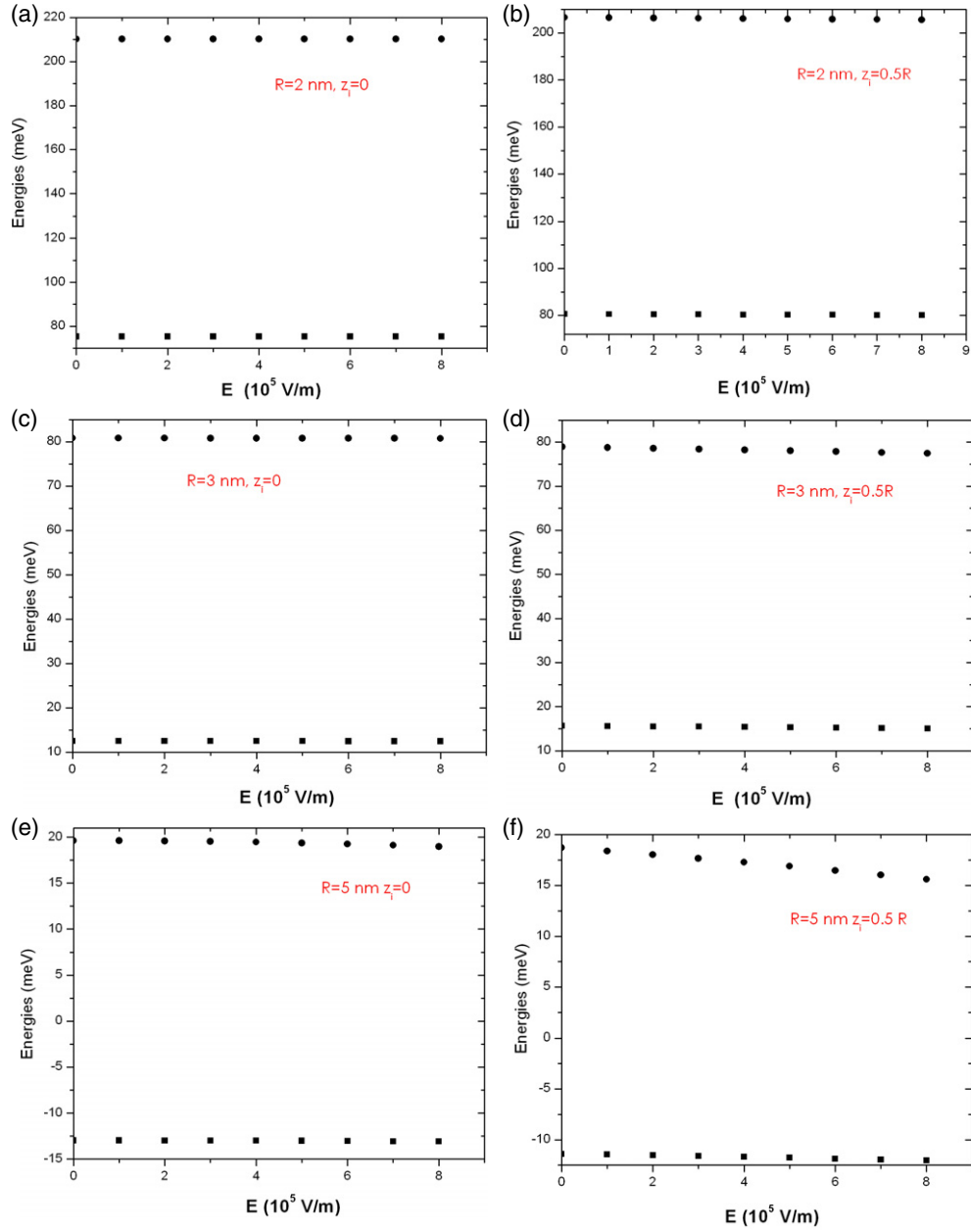


Figure 1. Plot of the energies of the ground (squares) and the first excited state (010) (circles) of the system as a function of the external electric field amplitude E . In (a) $R = 2$ nm, $z_i = 0$, (b) $R = 2$ nm, $z_i = 0.5R$, (c) $R = 3$ nm, $z_i = 0$, (d) $R = 3$ nm, $z_i = 0.5R$, (e) $R = 5$ nm, $z_i = 0$ and (f) $R = 5$ nm, $z_i = 0.5R$.

with Φ_0 , Φ_1 being the wavefunctions of the electron in the ground state and in the first excited state, respectively. Also, ω_{01} is the transition frequency that corresponds to the energy difference of the first excited state and the ground state, σ_s is the density of electrons in the

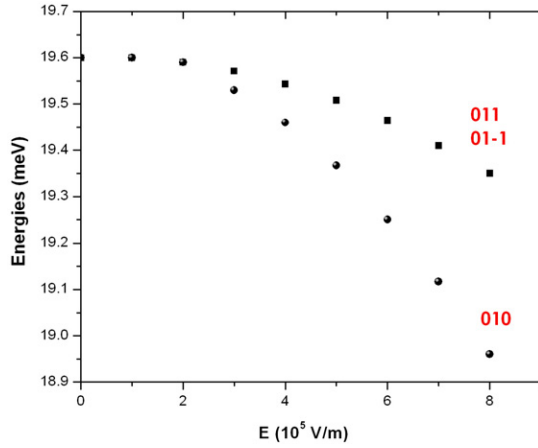


Figure 2. Plot of the energies of the excited states (010) (circles), (0 1 1), (01 - 1) (squares) of the system as a function of the external electric field amplitude E for the case that $R = 5$ nm, $z_i = 0$.

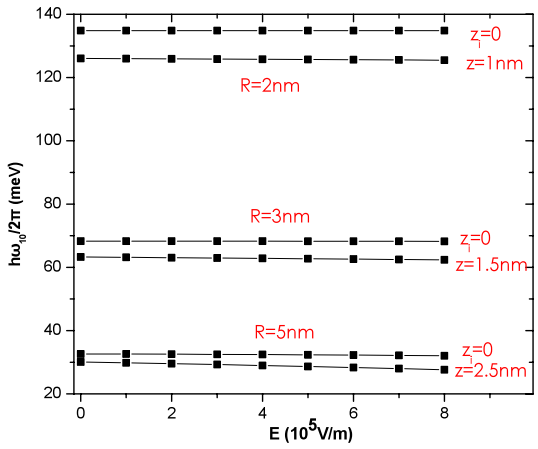


Figure 3. Plot of the energy difference of the first excited state (010) and the ground state of the system as a function of the external electric field amplitude E for several values of the quantum dot radius and the position of the impurity.

quantum dot, ϵ_0 is the vacuum permittivity, T_1 is the longitudinal relaxation time and T_2 is the transverse relaxation time.

For $\omega \approx \omega_{01}$, there is a peak value of $\chi_0^{(2)}$ ($\equiv \chi_{0,\max}^{(2)}$), estimated by the expression

$$\chi_{0,\max}^{(2)} = \frac{2e^3 T_1 T_2 \sigma_s}{\epsilon_0 \hbar^2} \mu_{01}^2 \delta_{01}. \quad (11)$$

As we have noted above, when we calculate the factor $\mu_{01}^2 \delta_{01}$ separately for the ground state and the states ((010), (011), (01 - 1)), we find that this factor obtains a nonzero value only for the state (010). In our calculations the relaxation times are set to $T_1 = 1$ ps, $T_2 = 0.2$ ps [40] and the electron density is taken as $\sigma_s = 5 \times 10^{24} \text{ m}^{-3}$.

As we have noted above, the position of the maximum of the nonlinear optical rectification coefficient is found at $\omega \approx \omega_{01}$. In figure 3 we plot the values of $\hbar\omega_{01}$ as a function of the electric field amplitude for various values of the quantum dot radius and the position of the impurity. It can be seen that for given values of the quantum dot radius and the position of the impurity, ω_{01} depends weakly on the electric field amplitude. From all the cases the most significant influence of the electric field is found for larger quantum dot radius. In the latter case, we can also note that the increase of the electric field amplitude leads to a decrease of the energy difference of the first excited state and the ground state of the system. In addition,

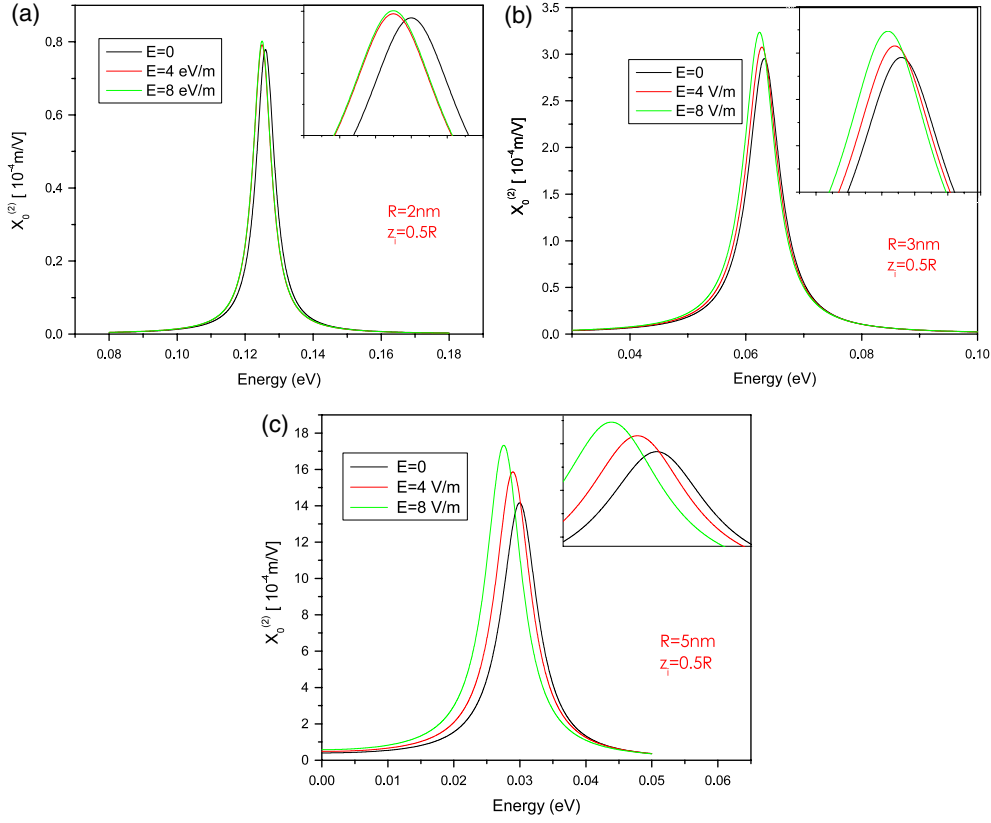


Figure 4. Plots of the nonlinear optical rectification coefficient $\chi_0^{(2)}(\omega)$. In all figures $z_i = 0.5R$, $E = 0$ (black curve), $E = 4 \text{ V m}^{-1}$ (red curve), $E = 8 \text{ V m}^{-1}$ (green curve). In (a) $R = 2 \text{ nm}$, (b) $R = 3 \text{ nm}$ and (c) $R = 5 \text{ nm}$. The insets focus on the maxima of $\chi_0^{(2)}(\omega)$.

the increase of the position of the impurity shifts ω_{01} to smaller values for a given quantum dot radius. The larger influence with the position of the impurity is found for smaller values of the quantum dot radius.

Typical nonlinear optical rectification spectra are shown in figures 4 and 5. We find that the increase of the electric field amplitude enhances the maximum value of the nonlinear optical rectification coefficient and shifts this maximum to smaller photon energies. These facts are more pronounced in the case of figure 4(c), where $R = 5 \text{ nm}$. The enhancement of the nonlinear optical rectification coefficient can be attributed to the increase of the asymmetry of the system and thus to the enhancement of the factor $\mu_{01}^2 \delta_{01}$. The shift of the nonlinear optical rectification coefficient maximum is attributed to the decrease of the energy difference with the increase of the electric field amplitude, that we noted in figures 1 and 3. In addition, the larger values of the nonlinear optical rectification coefficient are obtained for larger quantum dot radius, i.e. in the case that $R = 5 \text{ nm}$. Finally, as we can see from figure 5, for a given value of the electric field amplitude and the quantum dot radius the increase of the position of the impurity leads to a strong increase of the maximum value of the nonlinear optical rectification coefficient. The reason for this is that the system becomes more asymmetric with the increase of the position of the impurity. This increase also shifts the maximum of the spectrum to smaller frequencies, as was also noted in figure 3.

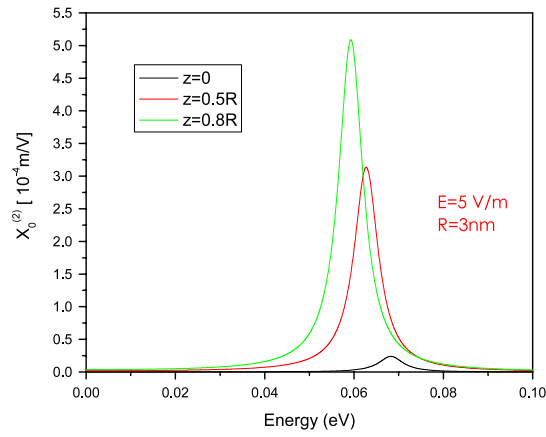


Figure 5. Plot of the nonlinear optical rectification coefficient $\chi_0^{(2)}(\omega)$ for three values of the position of the impurity, $z_i = 0$ (black curve), $z_i = 0.5R$ (red curve) and $z_i = 0.8R$ (green curve). In addition, $E = 5 \text{ V m}^{-1}$ and $R = 3 \text{ nm}$.

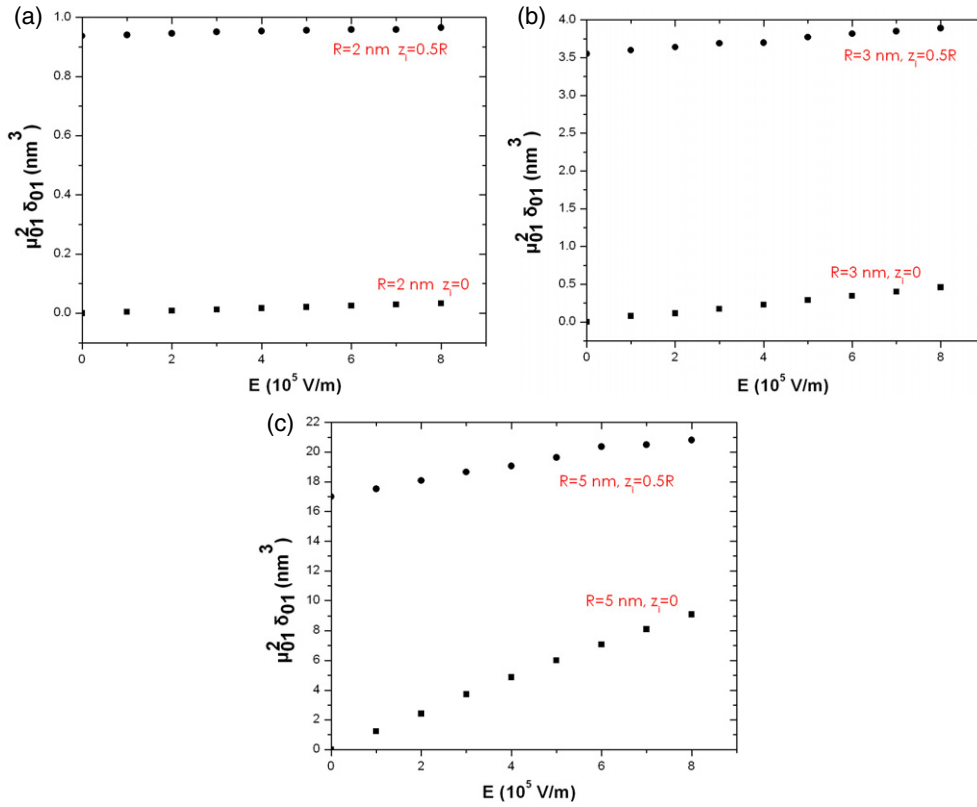


Figure 6. Plots of $\mu_{01}^2 \delta_{01}$ as a function of the external electric field amplitude E . With squares we denote the case that $z_i = 0$ and with circles the case that $z_i = 0.5R$. In (a) $R = 2 \text{ nm}$, (b) $R = 3 \text{ nm}$ and (c) $R = 5 \text{ nm}$.

The above findings regarding the maximum value of the nonlinear optical rectification coefficient are also shown in figure 6, where we depict the values of $\mu_{01}^2 \delta_{01}$ as a function of

the applied electric field amplitude for all the cases we studied. In addition, for an on-centre impurity and no applied electric field we see that, for all radii we have studied, the factor $\mu_{01}^2 \delta_{01}$ equals zero. This is expected, as in this case the system is symmetric, so there is no nonlinear optical rectification process. We also find from figure 6 that for the same electric field amplitude and the same quantum dot radius the case of off-centre impurities gives larger maximum values of the nonlinear optical rectification coefficient than the case of on-centre impurities, as was also found from figure 5.

3. Summary

In this work we have studied numerically the electronic structure of a spherical quantum dot with parabolic confinement that contains a donor impurity and interacts with a DC electric field. Both the impurity and the electric field lead to an asymmetry in the otherwise symmetric confining potential. In our calculations we have varied the position of the impurity and have taken an electric field of varying strength along the z -direction. The calculated electronic structure is then used for the determination of the nonlinear optical rectification coefficient of the quantum dot structure. We show that both the position of the impurity and the strength of the electric field influence the electronic structure of the system and the nonlinear optical rectification process.

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