Effects of excitons in nonlinear optical rectification in semiparabolic quantum dots

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We study the effects of excitons in nonlinear optical rectification in one-dimensional semiparabolic quantum dots. We consider the cases that the electron and the hole are confined in semiparabolic potentials (i) with the same oscillator frequency and (ii) with the same width. In the first case we present approximate analytical results in the strong confinement regime and find a closed-form relation between the nonlinear optical rectification coefficient of an exciton and the nonlinear optical rectification coefficient when only one electron exists in the structure. In the second case we use the Hartree-Fock approximation and the potential morphing method and present results for the nonlinear optical rectification coefficient in all confinement regimes.

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Nonlinear optical properties in semiconductor nanostructures have attracted considerable interest due to their relevance in several applications.[1,2] Among the nonlinear optical properties, significant theoretical[3–12] and experimental[13–18] attention has been paid to second-order nonlinear optical properties, such as nonlinear optical rectification (OR) and second harmonic generation. This happens as the second-order nonlinear processes are the simplest and the lowest-order nonlinear effects and their magnitudes are usually stronger than those of high-order ones, if the quantum system demonstrates significant asymmetry. In general, these asymmetries are produced by two methods. One is by using advanced material growing technology, such as molecular beam epitaxy and metallic-organic chemical vapor deposition, to obtain nanostructures with asymmetric confining potential.[3,5,7,9–12,14–18] The other is through the application of a static electric field to a structurally symmetric system in order to get an asymmetric nanostructure.[4,6,8,13]

Recently, Yu *et al.*[19] studied the effects of an exciton (taken as an interacting electron-hole pair) on the nonlinear OR in one-dimensional asymmetric semiparabolic quantum dots that are strongly confined in the x and y directions and are confined by a semiparabolic potential in the z direction. We note that the semiparabolic confining potential has been also used in other studies in semiconductor quantum wells and quantum dots.[10–12,20–22] Yu *et al.* in their study[19] assumed that the electron and the hole are confined in semiparabolic potentials with the same oscillator frequency. Using analytical approximate results they showed that in the strong confinement regime the excitonic effects enhance significantly the nonlinear OR coefficient, in comparison to the value that one obtains if only one electron exists in the structure.[19]

In the present work we also study the effects of excitons in nonlinear OR in one-dimensional semiparabolic quantum dots. We first revisit the problem studied by Yu *et al.*[19] and present approximate analytical results in the strong confinement regime. With the use of these results we find a closed-form relation between the nonlinear OR coefficient in the case that one exciton is confined in the quantum dot structure and the corresponding nonlinear OR coefficient when only one electron exists in the structure. It is found that the OR coefficients are related only by the effective masses of the electron and the hole. These results are also verified by numerical calculations performed with the potential morphing method (PMM).[23–26]

We also consider the case where the electron and the hole are confined in a quantum dot structure with the same width. This problem is quite different from that studied by Yu *et al.*[19] In our study, we take into account the interaction of electron and hole and use the Hartree-Fock approach and the PMM for the calculation of the necessary energy eigenvalues and eigenfunctions.[23–26] We find that the magnitude of the nonlinear OR coefficient of this system in the strong confinement regime is lower than the corresponding coefficient when only one electron exists in the structure. In addition, we show that larger values of the nonlinear OR coefficient are found for larger quantum dot structures.

In the effective mass approximation the Hamiltonian for the electron-hole system in one dimension can be written as[27,28]

$$H = -\frac{\hbar^2}{2m_e^*}\frac{\partial^2}{\partial z_e^2} - \frac{\hbar^2}{2m_h^*}\frac{\partial^2}{\partial z_h^2} + V_0^e(z_e) + V_0^h(z_h) - \frac{e^2}{\epsilon|z_e - z_h|}. \tag{1}$$

Here e refers to the electron and h to the hole. Also, $m_{e(h)}^*$ is the effective electron (hole) mass, ϵ is the relative dielectric constant, and $V_0^{e(h)}$ is the confinement potential for the electron (hole).

In this Brief Report we consider the case that the electron and the hole are confined in a semiparabolic potential of the form $V_0^{e(h)}(z_{e(h)}) = \frac{1}{2}m_{e(h)}^*\omega_{e(h)}^2 z_{e(h)}^2$, if $z_{e(h)} \geqslant 0$ and $V_0^{e(h)}(z_{e(h)}) \to \infty$, if $z_{e(h)} < 0$, where ω_e, ω_h are the oscillator frequencies of the electron and the hole confining potentials, respectively.

First case: $\omega_e = \omega_h = \omega_0$. In this case it is useful to rewrite the Hamiltonian of Eq. (1), in the case that $z_e, z_h \geqslant 0$, in the center-of-mass and relative motion coordinates as

$$H = -\frac{\hbar^2}{2M}\frac{\partial^2}{\partial Z^2} + \frac{1}{2}M\omega_0^2 Z^2 - \frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial z^2} + \frac{1}{2}\mu\omega_0^2 z^2 - \frac{e^2}{\epsilon|z|}, \tag{2}$$

where $M = m_e^* + m_h^*$ is the total mass, $\mu = m_e^* m_h^*/M$ is the reduced mass, $Z = (m_e^* z_e + m_h^* z_h)/M$ is the center-of-mass motion coordinate and $z = z_e - z_h$ is the relative motion coordinate along the z-axis. The form of the Hamiltonian of Eq. (2) allows us to separate the center-of-mass and the relative position motions. Therefore we take $\Psi(z_e, z_h) = \Phi(Z)\phi(z)$ for the total (envelope) wave fuction of the system, and $E = E_Z$

$+E_z$ for the total energy of the system. The center-of-mass motion wave functions and eigenergies are[19] $\Phi(Z)=\Phi_N(Z) =[\sqrt{\pi}2^{2N}(2N+1)!/\alpha]^{-1/2}\exp(-\alpha^2Z^2/2)H_{2N+1}(\alpha Z)$, $E_Z=E_N =\hbar\omega_0(2N+3/2)$, with $N=0,1,2,\ldots$ and $\alpha=\sqrt{M\omega_0/\hbar}$. Here, H_{2N+1} are Hermite polynomials of order $2N+1$. The relative motion wave functions and eigenergies are determined by the equation

$$\left[-\frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial z^2}+\frac{1}{2}\mu\omega_0^2z^2-\frac{e^2}{\epsilon|z|}\right]\phi(z)=E_z\phi(z). \quad (3)$$

The above equation can be solved analytically in the strong confinement regime[27,28] where the Coulomb interaction term is omitted. Then $\phi(z)=\phi_n(z)=[\sqrt{\pi}2^{2n}(2n+1)!/\beta]^{-1/2}\exp(-\beta^2z^2/2)H_{2n+1}(\beta z)$, $E_z=E_n=\hbar\omega_0(2n+3/2)$, with $n=0,1,2,\ldots$ and $\beta=\sqrt{\mu\omega_0/\hbar}$.

The nonlinear OR coefficient of an asymmetric one-dimensional 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[2,7,11,12,19]

$$\chi_0^{(2)}(\omega)=\frac{4e^3\sigma_s\mu_{01}^2\delta_{01}}{\epsilon_0\hbar^2}\times\frac{\omega_{01}^2\left(1+\frac{T_1}{T_2}\right)+\left(\omega^2+\frac{1}{T_2^2}\right)\left(\frac{T_1}{T_2}-1\right)}{\left[(\omega_{01}-\omega)^2+\frac{1}{T_2^2}\right]\left[(\omega_{01}+\omega)^2+\frac{1}{T_2^2}\right]}, \quad (4)$$

where $\mu_{01}=\langle\Psi_0|z_e-z_h|\Psi_1\rangle$, $\delta_{01}=|\langle\Psi_1|z_e-z_h|\Psi_1\rangle-\langle\Psi_0|z_e-z_h|\Psi_0\rangle|$, with Ψ_0, Ψ_1 the total wave functions of the exciton in the ground state and in the first excited state, respectively. Also, ω_{01} is the transition frequency, σ_s is the density of electrons in the 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 $\chi_{0,max}^{(2)}=2e^3T_1T_2\sigma_s\mu_{01}^2\delta_{01}/(\epsilon_0\hbar^2)$. The matrix elements μ_{01}, δ_{01} in this case are given by $\mu_{01}=\langle\phi_0|z|\phi_1\rangle$, $\delta_{01}=|\langle\phi_1|z|\phi_1\rangle-\langle\phi_0|z|\phi_0\rangle|$. Using the above analytical results we obtain $\mu_{01}=\sqrt{2}/(\sqrt{3\pi}\beta)$, $\delta_{01}=1/(\sqrt{\pi}\beta)$, and $\omega_{01}=2\omega_0$. Then, $\chi_{0,max}^{(2)}=4e^3T_1T_2\sigma_s/(3\pi^{3/2}\epsilon_0\hbar^{1/2}\mu^{3/2}\omega_0^{3/2})$.

In the case that there is only one electron in the quantum dot structure then the matrix elements are given by[12,19] $\mu_{01}=\sqrt{2}/(\sqrt{3\pi}\gamma)$, $\delta_{01}=1/(\sqrt{\pi}\gamma)$, with $\gamma=\sqrt{m_e^*\omega_0/\hbar}$. Also, $\omega_{01}=2\omega_0$. Therefore the following relation exists between the nonlinear OR coefficient when there is an exciton in the quantum dot structure ($\chi_0^{(2)}$) and when there is only one electron in the structure ($\bar{\chi}_0^{(2)}$)

$$\chi_0^{(2)}(\omega)=\left(1+\frac{m_e^*}{m_h^*}\right)^{3/2}\bar{\chi}_0^{(2)}(\omega). \quad (5)$$

The above relation is dependent only on the effective masses of the electron and the hole and explains analytically the results of Ref. 19. For our study we assume a GaAs/AlGaAs structure and take[19] $m_e^*=0.067m_0$, $m_h^*=0.09m_0$ (m_0 is the mass of a free electron) and $\epsilon=12.53$. In addition,[19] the relaxation times are set to $T_1=1$ ps, $T_2=0.2$ ps and the electron

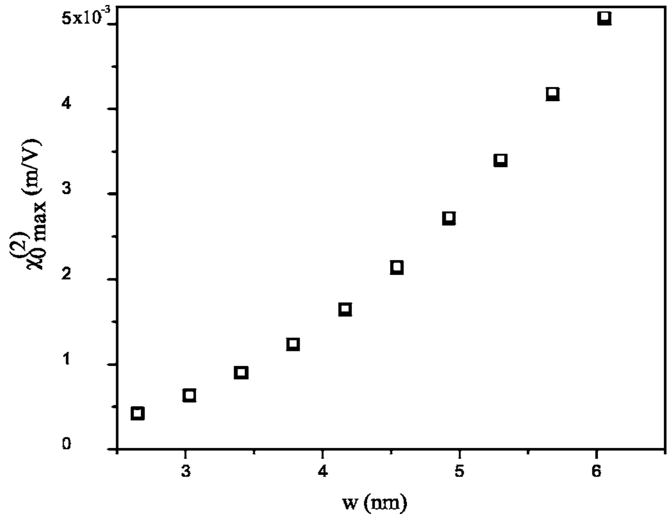

FIG. 1. The peak value $\chi_{0,max}^{(2)}$ as a function of the electronic quantum dot width. Closed squares represent the numerical results and open squares the analytical results.

density is taken $\sigma_s=5\times10^{24}$ m^{-3}. In this case $(1+m_e^*/m_h^*)^{3/2}=2.304$, therefore in the strong confinement regime the nonlinear OR coefficient for an exciton in the structure is enhanced by this value.

The above relation is based on the approximate analytical results that have been obtained after the omission of the Coulomb term in Eq. (3). We will now assess the validity of this approximation by obtaining the values of the nonlinear OR coefficient numerically without making this approximation. We therefore solve Eq. (3) numerically with the PMM and obtain the relevant energies, wave functions, and matrix elements.[23] We then calculate the nonlinear OR coefficient as a function of the "electronic width" of the one-dimensional quantum dot, $w=\gamma^{-1}$. These results are shown in Fig. 1. As it can be seen for the system under study the numerical and the approximate analytical results agree very well.

Second case: $m_e^*\omega_e=m_h^*\omega_h$. In this case we take the parameters of the system such that $m_e^*\omega_e=m_h^*\omega_h$, so that the electron and the hole are confined in a quantum dot with well-defined width.[24–27] In this case the system cannot be separated in relative motion and center-of-mass motion and the relevant wave functions and energies will be determined numerically using the Hartree-Fock approach. We note that the Hartree-Fock approach has been used with success in explaining experimental results in quantum dot and quantum rod structures.[25,26] In our approach we solve the iterative Hartree-Fock equations for the electron and the hole with PMM. For a detailed description of the relevant equations and the numerical methodology see Refs. 25 and 26.

We calculate the nonlinear OR coefficient with the Hartree-Fock method for a GaAs/AlGaAs structure. The rest of the parameters are taken the same as before. As it is clearly shown in Figs. 2(a) and 2(b) for the strong and intermediate confinement regimes $\chi_{0,max}^{(2)}$ increases as the quantum dot width increases. However, as it can be seen from Fig. 2(c) in the weak confinement $\chi_{0,max}^{(2)}$ decreases as the quantum dot width increases. This behavior is also clearly shown in Fig. 3, where the maximum value of the nonlinear OR coef-

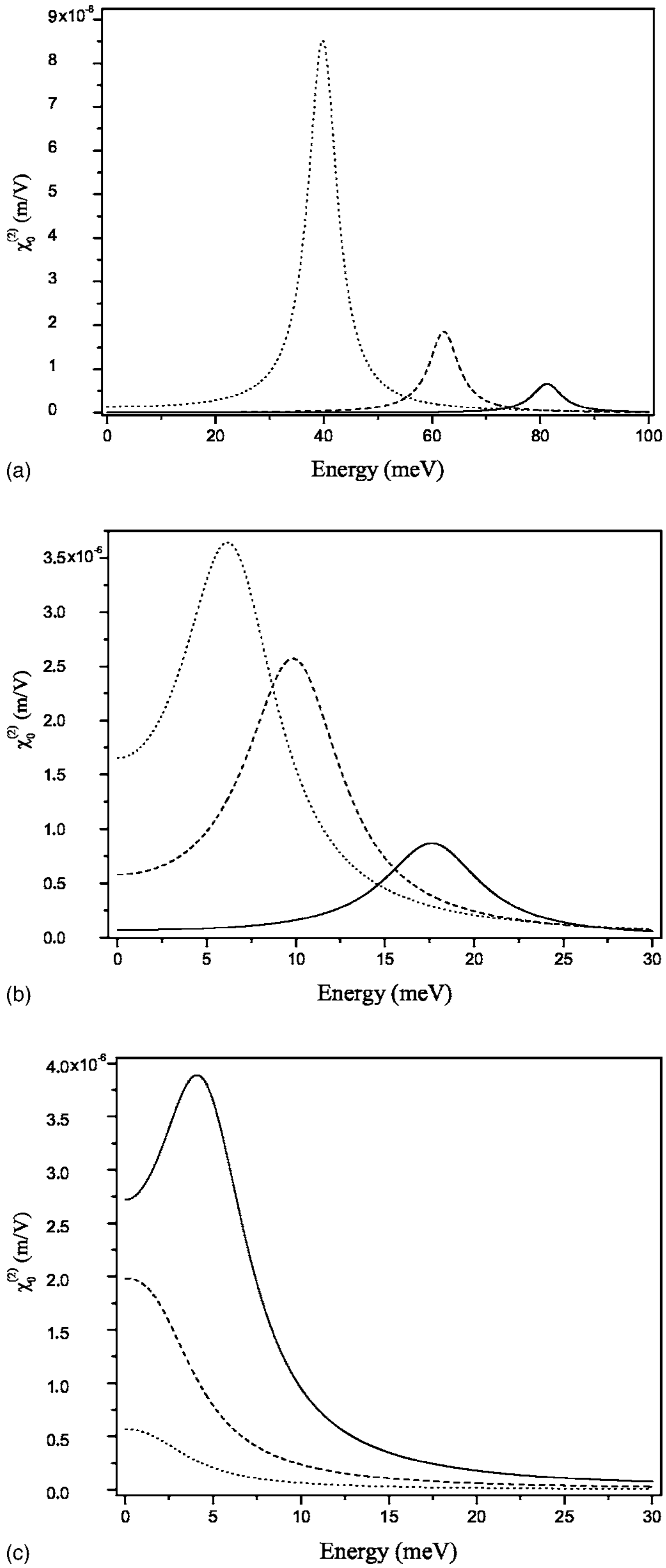

FIG. 2. The nonlinear OR coefficient $\chi_0^{(2)}(\omega)$: (a) in the strong confinement regime, for quantum dot width $w=7$ nm (solid curve), $w=8$ nm (dashed curve), and $w=10$ nm (dotted curve); (b) in the intermediate confinement regime, for quantum dot width $w=15$ nm (solid curve), $w=20$ nm (dashed curve), and $w=25$ nm (dotted curve); and (c) in the weak confinement regime, for quantum dot width $w=30$ nm (solid curve), $w=50$ nm (dashed curve), and $w=60$ nm (dotted curve).

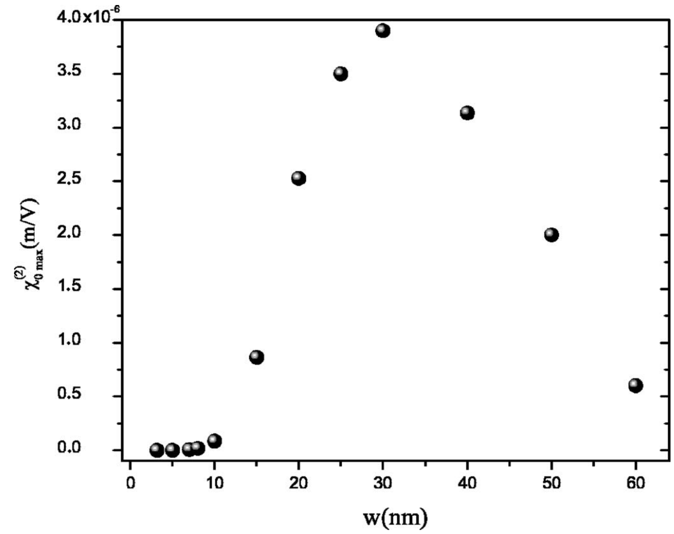

FIG. 3. The peak value $\chi_{0,max}^{(2)}$ as a function of the quantum dot width.

ficient is plotted as a function of the quantum dot width.

Comparing these results with those obtained previously we find that in this case the maximum value of the nonlinear OR coefficient is significantly reduced. In addition, even if we compare these results with those obtained in the case that only one electron exists in the structure we find that the existence of an exciton in the structure decreases the maximum value of the nonlinear OR coefficient. This is in contrast to the results presented above for the case that $\omega_e=\omega_h$. Therefore for this structure excitonic effects decrease the nonlinear OR coefficient in the strong confinement regime. In this case that the electron and the hole have the same width, in the strong confinement regime, their wave functions obtain approximately the same functional form. Due to the use of the Slater wave function for the exciton[29,30] in the Hartree-Fock calculations,[25,26] the relevant matrix elements obtain small values, much smaller than those obtained in the case of a single electron in the quantum dot structure. This explains the decrease of the OR coefficient in the strong confinement regime. As our results further indicate, for the system under study, the larger values of the nonlinear OR coefficient are found for large widths of the quantum dot, around $w\approx 30$ nm.

In summary, in the present work we study excitonic effects in nonlinear OR in one-dimensional semiconductor quantum dots with semiparabolic confinement. We present numerical and analytical results for two cases, the case that the electron and the hole are confined in one-dimensional semiparabolic potentials with the same oscillator frequency, and the case that the electron and the hole are confined in a quantum dot with well-defined width. We intend to extend these results in two and three dimensions in another publication.

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