Comment to “Effect of an electric field and nonlinear optical rectification of confined excitons in quantum dots” [W. Xie, Phys. Status Solidi B 246, 2257 (2009)]

Jefferson Flórez, Ángela Camacho*, and Hanz Y. Ramírez

Physics Department, Universidad de los Andes, Cra. 1 No. 18A-12, Bogotá, Colombia

Received 12 April 2012, revised 31 May 2012, accepted 27 July 2012
Published online 19 September 2012

Keywords Coulomb effects, excitons, nonlinear optics, quantum dots

*Corresponding author: e-mail acamacho@uniandes.edu.co, Phone +57-1-3324500, Fax +57-1-3324516

In this comment, we review the work by Xie [Phys. Status Solidi B 246, 2257 (2009)] on electric field effects and nonlinear optical properties of excitons confined in spherical quantum dots. We find an inconsistency in the calculation of the dipole moments, which lead to vanishing nonlinear optical results. For the same set of parameters studied by Xie, we obtain new and reliable optical rectification coefficients via a finite element approach.

1 Introduction

In a previous published work [1], Xie takes advantage of the well known solution for a particle in an isotropic three dimensional harmonic potential to study the optical nonlinearities of an exciton in a quantum dot (QD). While the chosen problem is pertinent and its conclusions potentially useful, the showed results regarding the nonlinear optical coefficients are found spurious after a detailed examination.

Xie’s proposal is interesting for two specific reasons. On one hand, it considers a bias field to break the symmetry of the confinement potential in order to get a nonlinear optical response. On the other hand, the author considers the electrostatic Coulomb attraction between the electron–hole pair. Such effect has been neglected so far by other authors in similar systems [2–4].

Nevertheless, we found two issues in Section 3 of Xie’s paper that we consider must be commented. The first one is a lack of precision when the dipole approximation is introduced to describe the interaction of an electromagnetic wave with an exciton confined in a QD. The second one is a serious inconsistency on the dipole moments, which would produce vanishing optical rectification (OR) coefficients.

This comment is organized as follows. In Section 2, we present the correct expressions within the dipole approximation for dealing with the interaction between light and charge carriers. In Section 3, we explain in detail why the OR susceptibilities should be zero according to Xie’s procedure. The results for the OR coefficients obtained by finite element (FE) methodology are shown and analyzed in Section 4. Finally, conclusions are given in Section 5.

2 Dipole approximation

In Section 3 of Xie’s article, the interaction of an electromagnetic wave with an exciton confined in a QD is introduced through the dipole approximation. This approximation is based on the fact that the exciting electromagnetic wave is in the far infrared range, whereas the characteristic QD sizes are on the nanometer scale.

However, the dipole approximation is not well described by Eq. (25) because the polarization of the electromagnetic field is not considered [5]. The corresponding polarization vector describes for each instant of time the direction of the oscillating electric field, which must be collinear to the dipole moment in order to maximize the interaction. Thus, assuming that the incident wave is polarized along the z-axis (in which the asymmetry is introduced by the bias field), the correct Hamiltonian is

\[ H_{\text{e-h}} = eE_0(z_e - z_h) \exp(i\omega t), \]  

or in terms of relative particle coordinates,

\[ H_{\text{e-h}} = eE_0z \exp(i\omega t). \]
Using this Hamiltonian, the calculation of nonlinear optical coefficients may be carried out using the density matrix approach and the perturbation expansion method [6].

### 3 Optical rectification susceptibilities

The OR coefficients are calculated by Xie in Section 3 using the expression (27), where the term \( \delta_{10} \) is defined as

\[
\delta_{10} = |e\langle \Psi_0^0 | z | \Psi_1^0 \rangle| - |e\langle \Psi_0^0 | z | \Psi_0^0 \rangle|. \tag{3}
\]

If we put aside the electron charge, \( \delta_{01} \) is the mean relative particle displacement during the transition in the studied two-level system.

Nevertheless, in the previous section, Xie introduces the Coulomb interaction using the perturbation method for energies up to first order, while for wave functions he keeps unperturbed eigenstates (see Eqs. (12) through (15) of his work). As a consequence, wave functions of the relative particle do not take into account the Coulomb interaction between the electron and hole. Thus, the eigenfunctions of the ground state and the triply degenerate first excited state are those of the three-dimensional harmonic oscillator. When we use these eigenfunctions, specifically Eqs. (12) and (13), to calculate the integrals \( \langle \Psi_0^0 | z | \Psi_0^0 \rangle \) and \( \langle \Psi_0^0 | z | \Psi_1^0 \rangle \), respectively, we find that both results are equal to \(-\beta\), where \( \beta \) is a field dependent length constant defined by Xie in Section 2 of his paper. Therefore, \( \delta_{01} \) is systematically equal to zero as well as the OR susceptibilities regardless the value of \( F \), making not possible that the results shown in Figs. 4 and 5 of Xie’s work correspond to those unperturbed wave functions.

One way to calculate the dipole matrix elements including the Coulomb interaction is to find higher order correction terms for the relative particle wave functions. However, for the sake of full inclusion of correlations, we have chosen a FE approach to obtain such eigenfunctions, and then be able to calculate the nonlinear OR susceptibilities.

### 4 Results

To find the exciton eigenfunctions, we take advantage of the variable separation in terms of relative and center-of-mass coordinates. Then, we solve the corresponding Ben Daniel–Duke equation for the relative Hamiltonian

\[
H_{\text{rel}} = \frac{p_z^2}{2\mu} + \frac{1}{2} \mu \omega_0^2 x^2 + \frac{p_y^2}{2\mu} + \frac{1}{2} \mu \omega_0^2 y^2 + \frac{p_x^2}{2\mu} + \frac{1}{2} \mu \omega_0^2 (z + \beta)^2 - \frac{1}{2} \varepsilon |F\beta - \frac{e^2}{\varepsilon r}|, \tag{4}
\]

where \( \mu \) is the electron–hole reduced mass, \( \omega_0 \) is the strength of the confinement, \( F \) the bias field, \( \beta = |eF/(\mu \omega_0^2)| \), and \( \varepsilon \) the dielectric constant of the material where the electron and the hole are confined.

Using a FE methodology, the QD geometry is discretized and the corresponding algebraic eigenvalue problem solved. To provide high accurate results, computations were carried out by implementing a mesh refinement procedure for the complete geometry [7]. This mechanism ensured that the difference between the results obtained on two subsequent mesh refinement is negligible.

In Table 1, we study the effect of the Coulomb interaction on the relative particle ground state energy for different confinement strengths (Column 1). As a zero approximation (Column 2), we ignore the Coulomb interaction, resulting in a pure three-dimensional harmonic oscillator under a bias field, whose energies are in Eq. (8) of Xie’s article. We obtain more accurate energies (Column 3) introducing the Coulomb interaction via the FE approach (Column 4). Comparing the results of the Column 2 with those of the Column 3, we notice that the perturbation method introduces an important energy correction due to the Coulomb interaction, so that such interaction should not be neglected in these kind of systems. Moreover, the energies in Column 4 include additional correlation effects associated to the full consideration of Coulomb interaction. Although this is a slight correction, it evidences higher accuracy of the FE approach as compared to the perturbation method.

Furthermore, the eigenfunctions obtained in this way include state intermixing due to the Coulomb interaction, which let us calculate the non-zero OR coefficients.

For the same set of parameters as in Ref. [1], we obtain the OR susceptibilities using the expression (27) of Xie’s work and the eigenfunctions and eigenvalues from the FE calculations. In Figs. 1 and 2, we present the corrected versions of Figs. 4 and 5 of Xie’s article, respectively. As a first remark, the amplitude of the peaks are one order of magnitude smaller than those in the revised work. Also, we find dissimilar tendencies and amplitude relations between the OR susceptibilities peaks in both figures.

In Fig. 1, we observe that the larger the confinement energy, the smaller the peak amplitude. This result has its origin in the dipole matrix elements. Both, \( \delta_{10} \) and \( M_{01} \) (with \( M_{01} = e\langle \Psi_0^0 | z | \Psi_1^0 \rangle \)), decrease when \( \hbar \omega_0 \) increases due to a stronger localization. Thus, for more confined carriers the results of the integrals and their differences (in the case of \( \delta_{10} \)) are smaller. Since the OR susceptibility is directly proportional to the product \( \delta_{10} M_{01}^2 \), the net result is a reduction of the peak amplitudes as a function of the confinement energy.

<table>
<thead>
<tr>
<th>( \hbar \omega_0 )</th>
<th>non-interacting electron–hole</th>
<th>1st order perturbation</th>
<th>full Coulomb interaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>35.32</td>
<td>22.51</td>
<td>21.71</td>
</tr>
<tr>
<td>100</td>
<td>140.08</td>
<td>113.97</td>
<td>112.91</td>
</tr>
<tr>
<td>150</td>
<td>220.59</td>
<td>187.14</td>
<td>186.13</td>
</tr>
<tr>
<td>200</td>
<td>297.52</td>
<td>258.46</td>
<td>257.47</td>
</tr>
</tbody>
</table>

All the values are in meV units; \( F = 100 \, \text{kV cm}^{-1} \).
We also observe in Fig. 1 that the resonance condition is satisfied for photon energies around the corresponding confinement energy, yet when this energy increases, the peak positions are farther from the respective $\hbar \omega_0$ values.

The resonance condition is set by $\Delta E$, and without Coulomb interaction, it would be $\hbar \omega_0$. However, when such interaction comes to play, each eigenvalue changes differently according to its wave function distribution, leading to $\Delta E$ values shifted from $\hbar \omega_0$. In addition, this shift is proportional to the Coulomb interaction strength and consequently to the confining energy.

Figure 2 shows that, under a fixed confinement energy, there is an electric field magnitude for which the OR susceptibility is maximum. Within the terms that affect the susceptibility amplitudes, only $\delta_{10}$ registers a maximum for $F = 150 \text{kV cm}^{-1}$, while $M_{01}$ exhibits a monotonous tendency with $F$. We attribute this feature to an interplay between the Coulomb interaction and the electric field: on one side the Coulomb interaction is responsible for state intermixing, while the bias field produces the asymmetry of the system. Since both of them are required for non vanishing $\delta_{10}$, but at the same time the field weakens the Coulomb interaction by separating the electron-hole pair, there is an optimization point where such a quantity is maximized.

In the same way, the interplay between the Coulomb interaction and the electric field also determines the peak position in Fig. 2, where we obtain smaller peak blue-shifts as $F$ grows; i.e. as the relative particle is farther from the origin under the action of $F$, the energy displacement related to the Coulomb interaction is reduced, approaching the peak position to $\hbar \omega_0 = 100 \text{ meV}$.

5 Conclusion  
We have reviewed the nonlinear optical results presented by Xie. First of all, we made an amendment to the dipole approximation in which the polarization of the electromagnetic wave must be included. Secondly, we solved the eigenvalue problem of the relative particle including completely the Coulomb interaction between the electron–hole pair, and conclude that such interaction plays a remarkable role in the OR susceptibilities, affecting both their amplitudes and energy positions. We also found that the competition between the Coulomb interaction and the electric field allows tuning of the optical nonlinearities of such systems.

Finally, we can ensure that the wave functions used by Xie in obtaining the dipole moments cannot correspond to those of Eqs. (12) and (13) of his work, since they would lead to vanishing nonlinear response. Furthermore, given the differences with our results which include Coulomb effects exactly, we found unclear and unreliable the way used in that work to take into account the Coulomb effects on the exciton optical nonlinearities.

References