Efficient control of coulomb enhanced second harmonic generation from excitonic transitions in quantum dot ensembles†

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In this work, the second harmonic generation from excitonic transitions in semiconductor quantum dots is computationally studied. By integrating a density matrix treatment with a partial configuration interaction approach, we obtain the second order susceptibility as a function of externally applied electric and magnetic fields for highly confined neutral and charged excitons. Our results show an enhancement in the nonlinear response with respect to analogous optical processes based on intraband transitions, and predict their efficient tunability by taking advantage of the interplay between Coulomb effects and field-driven wave function manipulation.

I. Introduction

The nonlinear optical properties of low dimensional systems have become an interesting field of research because these nanostructures promise higher performance than other nonlinear materials such as polar crystals and organic polymers.1,2 When this is combined with some peculiar features of quantum dots, for instance the possibility of engineering the carrier wave function by means of externally applied fields, the potential applications of their improved and controllable nonlinear optical characteristics can be diverse and remarkable.

Specifically, second harmonic generation (SHG) has been found to be useful in a wide range of applications including entanglement generation,3,4 determination of microscopic orientations,5–7 bioimaging,8,9 and surface characterization.10,11 However, the absence of dipole moments for transitions between states of the same parity inhibits SHG in centrosymmetric structures.12–14 Then, here we propose to take advantage of the trade-off between symmetry breaking and state intermixing generated by external fields and Coulomb interaction, respectively, to overcome such a limitation. This represents a possibility for induction and manipulation of second order nonlinearities in low-polar or non-polar systems.

Many studies have previously considered SHG in zero dimensional systems, generally finding better optical nonlinearities than those observed in structures of higher dimensionality.13,15 In particular Brunhes et al., in ref. 16, calculated and measured SHG susceptibilities for intraband transitions finding two main features: good agreement of the experimental results with the predicted values obtained through a density matrix treatment of the nonlinear optical response, and surprisingly large values for these nonlinearities.

Following up, some studies on this matter have consistently found nonlinear optical intensities of several orders of magnitude greater than those in bulk materials depending on geometrical and strain effects, although these studies dismissed the Coulomb interaction in strongly confined nanostructures under the argument of their much lower typical energy scale.17–23 Further on, several studies have called the attention on the relevance of electrostatic effects on the nonlinear response of low dimensional systems,24–28 thus stressing the importance of including the Coulomb impact on the modeling of those kinds of optical processes.

In this work we study the SHG from exciton transitions in quantum dot ensembles, and show how manipulation of the Coulomb interaction through applied fields and/or carrier injection, both of them being external and post fabrication control mechanisms,29–31 allows for efficient modulation of this second order susceptibility in the strongly confined regime. Such a result represents a significant contribution towards all-tunable nonlinear materials.

This paper is organized as follows. Section II presents the models used in describing the nonlinear optical response and
the confined excitons in quantum dots. The application of a time independent electric field, inclusion of the Coulomb interaction, and their competing interplay are shown and discussed in Sections III and IV. Section V reports the effects of applying a magnetic field, and finally the influence of additional charge in an exciton complex is addressed in Section VI. Section VII provides a summary and conclusion.

II. Theoretical model

In our approach to study nonlinearities of quantum dot ensembles, two main components are to be integrated. On the one hand, the model to establish the strength of the nonlinear optical response to the stimulating light in a collection of nanostructures with fully discretized energy levels. On the other hand, the particular description at the single dot level of the transition energies and dipole moments that are involved in the modeled optical process.

A. Optical model

The light–matter interaction is studied here using the density matrix formalism, which allows in a very convenient way to introduce decoherence effects in the calculation of the susceptibility.\textsuperscript{32,33}

We will assume quantum dot size homogeneity along the ensemble, which despite of being an idealization of the system is a reasonable approximation considering the continuous improvements in the growth techniques and the significant simplification that it allows in the calculations.

The equation of motion for $\hat{\rho}$ is

$$\frac{\partial \hat{\rho}}{\partial t} = \frac{i}{\hbar} \left[ \hat{H}_0 + \hat{H}', \hat{\rho} \right] - i \gamma \left( \hat{\rho}_g - \hat{\rho}_g^{(0)} \right),$$

where $\hat{\rho}^{(0)}$ is the density matrix operator for the non-interacting system at thermal equilibrium, and $\gamma_i = T_i^{-1}$ ($i = j$) and $\gamma_{ij} = T_{ij}^{-1}$ ($i \neq j$) are decoherence rates which absorb size inhomogeneities along the sample as well as relaxation and dephasing processes.\textsuperscript{16,34} $\hat{H}_0$ is taken to be the time independent part of the Hamiltonian, so that $|i\rangle$ and $E_i$ are the corresponding eigenstates and eigenvalues satisfying $\hat{H}_0 |i\rangle = E_i |i\rangle$, while $\hat{H}'$ is the time dependent part of the Hamiltonian which describes light–matter coupling.

Given that the wavelength of the stimulating radiation is around three orders of magnitude larger than the average quantum dot constituting the artificial gas, the dipole approximation is used,\textsuperscript{15,36} and $\hat{H}'$ becomes

$$\hat{H}' = - e \hat{z} \hat{E}(t)$$

where $e > 0$ is the electron charge and $\hat{E}(t) = \hat{E}(\omega)e^{i\omega t} + \text{c.c.}$ is the electric field of the incident electromagnetic wave, assumed here to be polarized along the $z$-axis.

Eqn (1) is solved iteratively using the expansion

$$\hat{\rho} = \hat{\rho}^{(0)} + \hat{\rho}^{(1)} + \hat{\rho}^{(2)} + \ldots$$

This expansion inserted into eqn (1) allows us to calculate the induced second order polarization (and then $\chi^{(2)}$) as the ensemble average:

$$\langle \hat{P}^{(2)} \rangle = \text{Tr}(\hat{P}^{(2)} \hat{\rho}).$$

where $\hat{P} = n_0 e \hat{\varepsilon}$ with $n_0$ being the three-dimensional quantum dot density.

Keeping only the double-frequency contribution to the second order polarization, which gives rise to the well-known SHG process, and defining $\chi^{(2)}_{2\omega}(\omega) = \langle \hat{P}^{(2)}_{2\omega} / i e_0 \hat{\varepsilon}^2(\omega) \rangle$ as the second order susceptibility for the SHG process, for a three-level system one obtains

$$\chi^{(2)}_{2\omega}(\omega) = \frac{n_0 e^4 \hat{P}^{(0)}_{11}}{e_0 \hbar^2} \frac{z_{12} z_{23} z_{31}}{(2\omega - \omega_{31} - i\Gamma_{13})(\omega - \omega_{21} - i\Gamma_{12})}. $$

Thus, the magnitude of this nonlinear response can be calculated in terms of the dipole moments $z_{ij} \equiv \langle \langle i | z | j \rangle \rangle$ ($i, j = 1, 2, 3$), and of the transition energies $\hbar \omega_{31}$ and $\hbar \omega_{31}$.

B. Quantum dot model

The constituting unit of the ensemble (single quantum dot) is modeled by an axially symmetric 3D harmonic confinement potential for each carrier. In contrast to other studies, we allow different confining frequencies for the electron and hole, which are much more appropriate considering the differences in the offsets and effective masses between the valence and conduction bands.\textsuperscript{37,38}

Under this confinement, the single particle (electron or hole) Hamiltonian reads

$$\hat{H}^{\text{e/h}} = - \frac{\nabla^2}{2m_e^{\text{e/h}}} + \frac{m_e^{\text{e/h}}}{2}(\omega_e^{\text{e/h}})^2 \left( x_e^{2} + y_e^{2} + \frac{2}{\gamma_e^{2}} \right),$$

where $m_e^{\text{e/h}}$ is the electron/hole effective mass, and $\omega_e^{\text{e/h}}(\omega_e^{\text{e/h}})$ the electron/hole frequency associated with the harmonic in-plane (vertical) confinement.

The eigenstates of the Hamiltonians in eqn (6) are the familiar Fock–Darwin states $\Phi_{l}^{\text{e/h}}$, where the electron/hole label $\text{e/h}$ is an index composed of the three quantum numbers $n_e^{\text{e/h}}, m_e^{\text{e/h}},$ and $q^{\text{e/h}}$. These first two numbers correspond to in-plane excitations, while the third one refers to the $z$ direction.

Focusing on the single particle ground states $n_e^{\text{e/h}} = m_e^{\text{e/h}} = q^{\text{e/h}} = 0$, the corresponding wave functions have characteristic extents directly related to the confining frequencies according to

$$l_x^{\text{e/h}} = \sqrt{\frac{\hbar}{2m_e^{\text{e/h}}\omega_e^{\text{e/h}}}}, \quad l_z^{\text{e/h}} = \sqrt{\frac{\hbar}{2m_e^{\text{e/h}}\omega_z^{\text{e/h}}}}.$$

In real quantum dots, the electron parameters are usually larger than those of the hole. Although the main features of the nonlinear response are not substantially affected by it, in this work, we consider $e$–$h$ asymmetry indices of $l_x/l_z = 0.5$ and $l_x/l_z = 0.75$.\textsuperscript{39}
Because of such an asymmetry, it is convenient to define the hybridized e–h wave function parameters,

\[ t_{e}^{ph} \equiv \sqrt{\frac{(F_{z})^{2} + (F_{r})^{2}}{2}} \quad t_{h}^{ph} \equiv \sqrt{\frac{(F_{z})^{2} + (F_{r})^{2}}{2}}, \tag{8} \]

based on which the quantum dot aspect ratio can be defined as \( a \equiv t_{h}^{ph}/t_{e}^{ph}. \)

Regarding energies, the single particle eigenvalues are given by

\[ E_{n_{e}n_{h}, n_{e}^{ph}n_{h}^{ph}}^{e/h} = \hbar \omega_{c}^{e/h} \left(n^{e/h} + m^{e/h} + 1\right) + \hbar \omega_{s}^{e/h} \left(q^{e/h} + \frac{1}{2}\right). \tag{9} \]

It is important to note that within this model, existence of electron–hole pairs in the ensemble is assumed, which can be achieved by means of optical pumping on the sample. Regarding dynamic control of the nonlinear response, \(40,41\) this actually represents an advantage of such an interband scheme compared to intraband systems where the presence of confined carriers in the quantum dot ensemble is achieved by conventional doping. Furthermore, if compared to systems where population is controlled by gate voltages, \(42\) this optically activated occupation of the dots seems to be still more efficient and deterministic, because using voltages to depopulate may require very strong fields to deplete deeply bound states (far below the band offset).

### III. Electric field effects

Although self-assembled dots generally exhibit some asymmetry along the growth direction, in this study vertically centrosymmetric structures are considered. This means to evaluate the Coulomb and field effects in the worst scenario for nonlinearities, because as described by eqn (3), non-zero transition dipole moments are required in the polarization direction to observe SHG. Thus, since the ignored asymmetries would contribute to the transition dipole moments, the SHG susceptibilities calculated in what follows would be even larger, and the enhancement in their magnitudes with respect to those of intraband transitions should be more substantial.

In those vertically symmetric quantum dots, occurrence of non-vanishing dipole moments requires a symmetry breaking which in our model is achieved and controlled by applying a bias field. \(43,44\)

The corresponding term in the Hamiltonian reads

\[ \hat{H}^{bf} = |e|Fz_{e} - z_{h}), \tag{10} \]

where \( F \) is the magnitude of a time independent bias applied in the \( z \) direction.

When added to the non-interacting e–h part of the Hamiltonian in eqn (6), the bias term shifts the origins of the harmonic confinements in the growth direction, by quantities with units of length linearly depending on \( F \) equal to

\[ \beta_{e/h} \equiv \pm eF \frac{1}{m_{e/h} c^{2}(\omega_{c}^{e/h})^{2}}. \tag{11} \]

and introduces an energy offset given by

\[ E_{bf} = \frac{(eF)^{2}}{2} \frac{1}{m_{e} c^{2}(\omega_{c}^{e})^{2}} - \frac{1}{m_{h} c^{2}(\omega_{c}^{h})^{2}}. \tag{12} \]

The distance \( \beta = |\beta_{e} - \beta_{h}| \) represents the bias-driven separation between the electron and the hole, which simultaneously accounts for the reduction in the Coulomb interaction and the increase in the dipole moments.

### IV. Coulomb effects

Spin influence is neglected in this work, given that the g-factor of heterostructures made of wide bandgap materials is known to be small, \(45-48\) and also because the magnitude of the e–h exchange energy is normally much smaller than the exciton direct and correlation Coulomb interactions, whose effects are the main scope of this work. \(39,49,50\) Thus, the complete time independent Hamiltonian reads

\[ \hat{H}^{T} = \hat{H}^{e} + \hat{H}^{h} + \hat{H}^{bf} + \hat{H}^{eh}, \tag{13} \]

in which, along with the single particle and bias terms, the e–h Coulomb interaction is included. Such a term explicitly reads

\[ \hat{H}^{eh} = - \sum_{n_{i,j,k,m}} V_{n_{i,j,k,m}}^{eh} a_{i,j,k}^{h \dagger} c_{k,m}^{e}. \tag{14} \]

where \( h^{\dagger} \) and \( c^{\dagger} \) (\( h \) and \( c \)) are the electron and hole creation (annihilation) operators, respectively. The corresponding matrix elements are given by

\[ V_{n_{i,j,k,m}}^{eh} = \int \frac{d^{3}r_{e} d^{3}r_{h}}{4\pi e_{0}^{2} |r_{e} - r_{h}|} \phi_{n}^{h \dagger}(r_{h}) \phi_{m}^{e}(r_{e}). \tag{15} \]

With the purpose of characterizing the magnitude of the Coulomb interaction and its effects, we carry out calculations for obtaining eigenenergies at three levels of approximation: non-interacting e–h, first order perturbation, and configuration interaction (CI). In the last case, \( S, P, \) and \( D \)-like single particle orbitals are considered.

To find the eigenenergies within each of the chosen approaches, we either suppress all interaction terms, retain only diagonal Coulomb terms, or include both diagonal and off-diagonal terms, respectively.

Therefore in the CI calculation, by diagonalizing the Hamiltonian of eqn (13) written on the basis defined by the direct product of \( \hat{H}^{e} \) and \( \hat{H}^{h} \) eigenstates, the eigenenergies and envelope wave functions of the confined exciton are acquired.

To obtain the bias dependent e–h Coulomb matrix elements, we add a label \( i \) or \( f \) to electron and hole quantum numbers depending on their association with the initial or final transition state, and extend the result by Chen et al. in ref. 48 so that holes and bias effects can be incorporated. Thus, for the considered dots with aspect ratio \( a \leq 1 \) we use
where \( u \equiv m^e_i + m^h_i + n^e_i + n^h_i - (p_1 + p_2 + p_4 + p_5) \) and \( v \equiv q^e_i + q^h_i + q^e_i + q^h_i - 2p_1 - 2p_5 \). \( \Gamma(x_i) \) and \( 2F_1(x_1, x_2, x_3, x_4) \) represent the Euler Gamma and hypergeometric \( 2F_1 \) functions, respectively. \( \delta_{q^e_i + q^h_i + q^e_i + q^h_i} \) preserves the vertical parity, while \( \delta_{e_i} \) ensures the conservation of the \( z \) component of angular momentum, with \( L_z \equiv (n^e_i - n^h_i) + (m^h_i - n^h_i) \) and \( L_x \equiv (n^e_i - m^h_i) + (m^h_i - n^h_i) \).

Fig. 1(a) illustrates the studied system, in which a neutral exciton \((X^0)\) is confined in an axially symmetric quantum dot under the action of the electric field \( \vec{F} \).

In order to evaluate Coulomb effects, the relative energy differences \( \Delta E_{\text{UP,CI}} \equiv \frac{E_{\text{CI}} - E_{\text{UP}}}{E_{\text{CI}}} \) and \( \Delta E_{\text{PT,CI}} \equiv \frac{E_{\text{CI}} - E_{\text{PT}}}{E_{\text{CI}}} \) are defined for the \( i \)th eigenstate, where \( E_{\text{UP}}, E_{\text{PT}}, \) and \( E_{\text{CI}} \) are the corresponding eigenenergies obtained by the non-interacting system, first order perturbation, and CI approach, respectively. Thus, taking the CI calculation as reference, \( \Delta E_{\text{UP,CI}} \) measures the composed influence of direct and indirect Coulomb terms, while \( \Delta E_{\text{PT,CI}} \) accounts only for the indirect ones (correlations).

In what follows, some specific material parameters have to be chosen in order to carry out numerical simulations that elucidate the required Coulomb effects. In this case, InGaAs/GaAs dots are considered and close dependence between such particular parameters and the obtained magnitudes must be taken into account. Nevertheless, the assumptions behind this approach allow us to extrapolate the observed trends to a wide range of III–V or even II–VI direct-gap semiconductor dot samples, since the studied underlying physics is in principle common to all of them. Moreover, it could be expected that the more polar the considered material (e.g. CdTe dots), the stronger the enhancement effect.

Fig. 1(b) shows those relative energy differences as functions of the applied bias field. The expected reduction of the Coulomb interaction for stronger fields is verified and the tiny influence of correlations on eigenenergies is evidenced. It is also consistent with the known fact that electrostatic effects are more noticeable on the ground energy than on excited levels.

The norm of the second harmonic generation susceptibility (SHGS), \( \chi^{(2)}_{C,2} \), is plotted in Fig. 2(a) as a function of the stimulating photon energy and the bias field. In Fig. 2(b), the SHGS is shown as a function of the stimulating photon energy for some chosen bias values.

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**Fig. 1** (a) Schematics of the artificial atom where a neutral exciton is confined. (b) Direct Coulomb and correlation effects on the three lowest eigenenergies.
For the sake of verification of the order of magnitude obtained for the maximum, those computed SHGS may be collated with the ones shown in Fig. 8 of ref. 16, being aware that resonant conditions and vertical confinement asymmetry are dissimilar in both studies. In doing that, our results are found at the same order of magnitude as the experimental ones reported there, and one order of magnitude larger than their theoretical prediction. This leads us to think that the discrepancy between experimental and theoretical results in that work might be attributed to the disregarding of the Coulomb interaction, which must be somehow present given that the calculation parameters used there reflect a number of carriers 1.5 times the number of dots, thus making the average occupation larger than 1. If Coulomb effects would be considered, external stationary fields would also play an interesting role in that kind of intraband system.

V. Magnetic field effects

Pursuing additional control on the nonlinear response, we added a time independent magnetic field to the system. To do that, it is necessary to change the canonical momentum operator \(-\hat{\nabla}^2/2m_e\hbar^2\) by the kinematical one \(-\hat{\nabla}^2/2m_e\hbar^2 \pm |e|A(r_e/\hbar)\), where \(A(r_e/\hbar) = B/2(-y_e/\hbar, x_e/\hbar, 0)\) is the magnetic potential taken in the Coulomb gauge. This results in modifications of the electron and hole harmonic frequencies according to

\[
\omega_{e\hbar}^{c/\hbar} = \sqrt{\left(\omega_e^{c/\hbar}\right)^2 + \left(\omega_h^{c/\hbar}/2\right)^2},
\]

Fig. 2 (a) Norm of the SHG susceptibility as a function of the stimulating frequency and the bias field. (b) Profile of the SHG susceptibility as a function of the stimulating frequency for some specific values of \(F\) [four of the five chosen fields are highlighted in (a)].

As can be noted in eqn (5), the SHGS has two distinctive peaks in \(\hbar\omega = E_2 - E_1\) and \(2\hbar\omega = E_3 - E_1\), with the latter being larger than the former within this model because \(2(E_2 - E_1) > (E_3 - E_1)\). Without Coulomb effects, the harmonic exciton confinement implies \(2(E_2 - E_1) = (E_3 - E_1)\), which would produce the so-called double resonance condition (DRC). Strong bias fields lead the system to recover the DRC by weakening the Coulomb interaction between the electron and the hole.

Taking into account that because of the well-defined parity of the single carrier wave functions, Coulomb related state intermixing is required to obtain the non-vanishing \(z_{31}\) dipole moment within our model, it is interesting to observe the interplay between the bias-related enhanced asymmetry and the reduced Coulomb interaction, so that the system reaches a SHGS maximum at some electric field value. Such an optimization is clearly evidenced in Fig. 2(b).

The red-shift with increasing bias can be understood in terms of the reduction in the push-down Coulomb effect experienced by the ground state.
However, the growing trend is not monotonic as can be observed for a field of 8 T. Under that field, the calculated SHGS is even smaller than that when no magnetic field was applied. This evidences that larger Coulomb matrix elements are not the only necessary thing for increasing the nonlinear response.

Fig. 4 provides some enlightening information for understanding the modulated behavior. We use $V_{eh} = \langle 0,0,0,0,0 | H_{e-h}^2 | 0,0,0,0,0 \rangle$ (the main direct Coulomb matrix element) to get an estimation of the interaction magnitude. In Fig. 4(a) the competition between vertical asymmetry and electrostatic interaction is elucidated, and the increase of the Coulomb matrix elements with the magnetic field is manifested. Fig. 4(b) shows the product of the involved dipole moments as a function of the bias field for different magnetic field values. The inset presents some particular non-interacting e–h eigenenergies and the corresponding off-diagonal matrix elements.

It is worth mentioning that Fig. 4(a) and (b) evidence strong correlation between the magnitude of SHGS and the effectiveness of the Coulomb interaction in causing state intermixing. Indeed, calibration calculations carried out as part of this work showed that if the e–h electrostatic interaction is artificially turned off, the dipole moment $z_{31}$ vanishes even for very high bias fields.\cite{44, 57, 58}
VI. Charged exciton

In the last part of this work, charge effects on the SHGS for an exciton complex are studied. We calculate the SHGS for a negatively charged exciton (X⁻) as a function of the bias field with and without the magnetic field.

The modified systems are depicted in Fig. 5(a), where the additional considered electron is shown. The negatively charged exciton was chosen because in this configuration the Coulomb effects are expected to be potentiated because of the more extended spatial distribution of the electron wave function as compared to that of the hole.59,60

The Coulomb interaction matrix elements for the three particle system ⟨|H_Coul|⟩ are obtained according to

\[
\langle n^2, m^2, q^2 | n^1, m^1, q^1 | H_{\text{Coul}} | n^1, m^1, q^1, n^2, m^2, q^2 \rangle = \delta_{q^1, q^2} \delta_{m^1, m^2} \delta_{n^1, n^2} \delta_{e_{q^1, q^2}} \delta_{e_{m^1, m^2}} \delta_{e_{n^1, n^2}}
\]

where the labels e1 and e2 have been introduced referring to either of the two electrons. The expression of the matrix element for the e–e interaction is analogous to that for the e–h one, but differs in that the former does not depend on the bias field, and conservation of the z component of angular momentum is now given by δLₚ/Lₚ with Lₚ = (m₁² + m₂²) − (n₁² + n₂²) and Lₚ = (m¹² + m²²) − (n¹² + n²²).

In Fig. 5(b), the intensity of the direct and correlation Coulomb effects can be observed for the negatively charged exciton as functions of the bias field. Although the direct Coulomb effects are stronger in the X⁰ case,61 correlations which are responsible for state intermixing are noticeably larger in the X⁻ case. The corresponding values for B = 0 are also shown as dashed lines, and allow us to observe how the magnetically driven wave function shrinking increases electrostatic interaction for lower bias fields but leads to faster reduction of the Coulomb influence for higher bias values.
The $X^-$ SHGS as a function of the stimulating photon energy is shown in Fig. 6(a) for different bias values and is displayed in such a way that the results can be directly compared with the corresponding $X^0$ case.

The SHGS improvement in around one order of magnitude is clear, and Fig. 6(b) evidences that the gain is rooted in the larger product of dipole moments for the charged exciton. This product is in turn favored by the Coulomb driven hybridization of states. Defining the ratio $E_2 - E_1$, which accounts for breaking of the DRC for each of the $X^0$ and $X^-$ cases, the strength of the Coulomb related intermixing can be visualized (i.e. the farther this ratio is from 1, the stronger the mix of states), as done in the inset of Fig. 6(b). As presumed, stronger Coulomb effects in the charged exciton configuration enhance the nonlinear response of the system.

VII. Summary and conclusions

In this work, manipulation of the Coulomb effects on the second harmonic generation from quantum dot ensembles was studied. Significant amplification and efficient control of the corresponding second order susceptibility under application of electric and magnetic external fields are predicted. Besides neutral excitons, further improvement of the nonlinear response was obtained for negatively charged excitons as compared to neutral excitons.

The magnitude of Coulomb matrix elements and their consequent state intermixing were addressed as the underlaying mechanisms for efficient on-demand modulation and important gain in the maximum of the second harmonic resonance. Thus, interband setups are found to be superior to pure intraband schemes (in which no significant role is played by electrostatic interaction), for second harmonic generation and its associated applications.

Beyond pointing out the relevance of electrostatic interaction on the nonlinear optical properties of quantum dot ensembles, it was demonstrated how all-external wave function control in zero dimensional systems leads to tunable nonlinearities via Coulomb correlations, in spite of their characteristic scale being several orders of magnitude smaller than energy spacing due to quantum confinement.

Although these results were obtained for ensembles formed by identical dots, the inherent physics is anticipated to stand under moderate size inhomogeneities. Thus, the main predicted features should be susceptible of observation in real high quality artificial gases. This might represent a significant contribution toward nanostructured tunable nonlinear materials.

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References


