A Model for Second Harmonic Generation in a Two-Dimensional Array of Quantum Dots

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We present a model for second harmonic generation in a two-dimensional array of quantum dots. We show that the combined effect of the electromagnetic local field of the array and the intrinsic electronic resonances due to the spatial confinement in the quantum dot produce giant enhancements of the second order non-linear susceptibilities, hence giving a very large efficiency in the second harmonic generation.

**Key words:** quantum dots, second harmonic generation

1. Introduction

Present nanofabrication techniques have opened a new door for research in artificial materials, where novel features arise from the quantum discretization of the dynamical response due to the smallness of the devices. Research in almost all physical aspects, like condensed matter properties, optical responses, opto-electronic characteristics, etc. are currently being pursued by many researchers.\(^1\) In this article we investigate a model for second harmonic generation (SHG) in a two-dimensional (2D) array of quantum dots (QD), where a QD is an structure that confines electrons to a nanoscopic region.\(^2,3\)

We will consider the case for which the QD's are separated from each other in such a way that there is no overlap of the electronic wavefunctions, furthermore, the electronic confinement assumed will be of infinitely rigid walls. Therefore the QD's are isolated from each other as far as the electronic behavior is concerned. The coupling among the QD's will be through the electric local field, i.e. the electric field at a given QD is composed by the external electric field plus the electric field generated by all others QD's.

Two very important features combine to produce giant enhancements of the SHG output in these systems. The first is the fact that the spatial electronic confinement imposed by the QD produces energy separations that resonate in the presence of an external perturbation. The other, is the fact that the geometry of the array can be chosen in such a way that the self-consistent electromagnetic response of all the QD induce resonances as well, through the local field. The origin of these two facts rely on the design of the QD and array itself, thus we show that within present technologies one should be able to observe this giant enhancement of the SHG output.

2. Theory

The model-system that we use for the QD is as follows. First, the electronic density of the 2D electron gas and the size of the QD are such that the number of free electrons per QD is small. Then we assume that the response of the QD is well described by considering only one electron per QD and by neglecting the Coulomb interaction. Thus, we are left with a particle in a box model, for which we assume that the dot itself is like a pillbox of radius \(R_0\) and height \(h_0\) with infinitely rigid walls, with an electron of charge \(e<0\) and effective mass \(m^*\), confined to a potential

\[
U_0(\vec{r}) = \begin{cases} 
\infty & r \geq R_0 \\
\infty & z \geq \pm h_0/2 \\
0 & \text{otherwise}
\end{cases}
\]

where we have chosen \(x\) to be along the axis of the dot and \(x, y\) to coincide with the cross section. The solution to the unperturbed Hamiltonian \(H_0 = p^2/2m^* + U_0\) can be obtained straightforwardly. Also, we consider that the typical QD height, \(h_0(<R_0)\), is such that the energies associated with its confinement will not be experimentally accessible for the range of energies of interest. Therefore, we can consider that the electron would be in the ground state associated with \(z\). Hence, the dynamic response would arise only from the \(xy\) or circular confinement of the QD.

For SHG is necessary to calculate the response of this electron up to second order in the perturbing fields. The polarization hereby attained by the electron is written as

\[
p_i = \alpha_0(\omega)E_0^{(c)} + \chi_{iik}(\omega)E_0^{(c)}E_0^{(c)}
\]

\[
+ \chi_{iikl}(\omega)E_l^{(c)}(P_iE_i)_0^{(c)},
\]

where \(E_0^{(c)}\) is the local electric field at the site of the entity; i.e. the electric field produced by all sources except the entity itself, and \((P_iE_i)_0^{(c)}\) is its gradient. We see that \(\alpha_0(\omega)\) gives the linear susceptibility and \(\chi\) are the non-linear second order susceptibilities. Since we are implicitly assuming a QD where inversion symmetry is satisfied (see Eq. (1)), then it follows that

\[
\chi_{iik} = 0 \quad \text{(inversion symmetry)},
\]

and thus if a SHG signal is to be expected, we need to have gradients of the local field, and a \(\chi_{iikl}\) different from zero.

Using standard quantum time dependent perturbation theory up to second order (i.e., through Kubo formulas\(^4\)), is a simple matter to calculate \(\alpha(\omega)\) and \(\chi\) of Eq. (2), where the perturbing Hamiltonian is given by
with the first term representing a dipolar coupling and the second a coupling to a field with a constant gradient. The results are

$$a_i = a(\omega) \delta_{ji},$$

$$x^{(0)}_{ij} = x^{(i)}_{ij} \delta_{ji} \delta_{k_0} \delta_{l_0} + (x^{(2)}_{ij} \delta_{j} \delta_{l_0} + \delta_{j} \delta_{k_0}) + x^{(4)}_{ij} \delta_{l_0} \delta_{k_0} (1 - \delta_{j} \delta_{l_0} \delta_{k_0}),$$

for $i, j = x$ or $y$. Also, it turns out that the quadrupolar induced moment will radiate an electric field which strength is of the same order of magnitude as the field radiated by the 2nd order dipole polarization. Thus, we obtain the following expression

$$Q_{ij}(2\omega) = x^{(0)}_{ij}(\omega)E^{(0)}_{e}E^{(0)}_{e},$$

where

$$x^{(0)}_{ij} = x^{(i)}_{ij} \delta_{ji} \delta_{k_0} \delta_{l_0} + (x^{(2)}_{ij} \delta_{j} \delta_{l_0} + \delta_{j} \delta_{k_0}) + x^{(4)}_{ij} \delta_{l_0} \delta_{k_0} (1 - \delta_{j} \delta_{l_0} \delta_{k_0}).$$

The functions $a(\omega), x^{(0)}_{ij}(\omega)$ and $x^{(0)}_{ij}(\omega)$ are given elsewhere.\(^{a}\) We mention that the super-script $d$($Q$) is a reminder of a non-linear dipolar (quadrupolar) response.

With the linear and non-linear response of an isolated QD to the local field, we now can make the simplification that the dipole hereby obtained is actually a point dipole localized at the position of the QD. Then, the 2D array of QD can be viewed, as an array of dipoles, called dipolium,\(^{a}\) where the typical separation is $\sim a$. Furthermore we will use the following restriction on the various distances relevant in the problem,

$$h_0 < 2R_0 < a < \epsilon \ell \omega,$$

in this way we are looking for the fields at distances greater than $R_0$ and $h_0$ (in order to ignore higher multipole contributions) and much less than $\epsilon \ell \omega$ (in order to ignore retardation effects, i.e. magnetic fields).

Once this constraint is imposed, we proceed to solve the electrostatic problem. With $R = |\vec{x} - \vec{x}'|$ define $M_{ij} = \partial_\theta \delta_{ji}(1/R)$ and $M_{ij} = -\partial_\theta M_{ij}$. Then, the corresponding electric field and field gradient are

$$E_i(\vec{x}, \vec{x}) = M_{ij} <p_j> + \frac{1}{2} M_{ij} <Q_k>, \ldots,$$

$$\partial_\theta E_i(\vec{x}, \vec{x}) = -M_{ij} <p_k> + \ldots,$$

We solve first for the linear polarization. The dipolium considered in this article will consists of several sub-lattices of dipoles, that due to the long-wavelength approximation given by Eq. (9), the dipole moment of each dipole that belongs to a given sub-lattice will be the same (recall that the array is 2D). Thus the next sums can be carried out as follows:

$$\sum_{\vec{p}} M(\vec{p} - \vec{p}_i) = \sum_{\vec{p}} M(\vec{p}_i - \vec{p}_i) = \mathcal{R}(l, l'),$$

where $\vec{p}_i$ is the position vector of the $l$th sub-lattice. Notice that $l = l'$ is allowed, however the self-interaction of any site is not taken into account on the sum of Eq. (12).

With these, the equation for $\vec{p}_i$ is

$$\rho^{(1)}(\omega, l) = a(\omega)[E_l(\omega) + \sum_{l'} \mathcal{R}_{l, l'}(l, l') \rho^{(1)}(l')],$$

$$= a(\omega)E^{(0)}_{e}(\omega, l) \ldots,$$

with $E_l(\omega)$ the presumed constant applied field, and $E^{(0)}_{e}(\omega, l)$ the local field.

Moving to second order, the self-consistent equation for the quadratic dipole moments is given by:

$$\rho^{(2)}(\omega, l) = a(\omega)[E_l(\omega) + \sum_{l'} \mathcal{R}_{l, l'}(l, l') \rho^{(1)}(l')],$$

$$= a(\omega)E^{(0)}_{e}(\omega, l) \ldots,$$

where from Eq. (10) $E^{(0)}_{e}(\omega, l) = (1/2)M_{ij}Q_{lj}$ is the electric field produced by the non-linear quadrupolar moment of Eq. (7), which in turn induces a non-linear dipolar moment via $\alpha(2\omega)$, and $\rho^{(0)}(\omega, l)$ is given by the third term of Eq. (2) which acts as a non-linear driving moment.

Now is clear that one has to solve Eq. (13) first, for $\rho^{(1)}(\omega, l)$ and use this to solve Eq. (14) self-consistently for the total non-linear dipole moment of each sub-lattice, i.e. $\vec{p}^{(1)}(2\omega, l)$. The formal solution (in the long-wavelength limit) for the total polarization of the array, is simply given by

$$P^{(1)}(2\omega) = \sum_{l} \rho^{(1)}(\omega, l) = \chi^{(0)}_{\Omega}(2\omega)E_l(\omega)E_l(\omega),$$

with

$$\chi^{(0)}_{\Omega}(2\omega) = \sum_{l} \chi_{\Omega k}(2\omega, l),$$

and $\chi_{\Omega k}(2\omega, l)$ can be obtained from above equations.\(^{a}\) We clearly notice that the self-consistency of the dipolium model gives the appropriate macroscopic polarization for a 2D system, i.e. proportional to $\vec{E} \times \vec{E}$ where no bulk term is present. We emphasize that the origin of the non-linearity comes from the microscopic term proportional to $\vec{E} \times \vec{E}$ (see Eq. (2)). Equation (15) imposes a restriction on the 2D array itself towards the generation of second harmonic; if the 2D array has inversion symmetry in the plane, no SHG should be observed, since $\chi^{(0)}_{\Omega k} = 0$. Therefore, staying within 2D arrays, we should design the array with no planar inversion symmetry.

3. Results

With Eq. (15) is a simple matter to calculate the conversion efficiency for SHG, defined through\(^{a}\)

$$\gamma = \frac{2\pi^2 \omega^2}{c^2 A} \sec^2(\theta) \hat{e} \hat{e} \hat{e} \chi^{(2)}(\hat{e} \hat{e} \hat{e} \hat{e}),$$

where $c$ is the speed of light, $\theta$ is the angle of incidence and $A$ the area of the incident beam (assuming a plane wave). Also, $\hat{e}$ and $\hat{e} \hat{e}$ are related to the ingoing polarization of the fundamental beam, and the outgoing polarization in the second harmonic, and to the Fresnel factors for transmission and reflection.\(^{a}\) We only investigate the SHG from the array, and neglect the signal coming from the substrate, which in any case will be small as compared with the array’s SHG enhancements.

In order to break the inversion symmetry within the planar array, we choose as an example the array sketched
in Fig. 1, where we have chosen the $x$ axis as the one that presents the asymmetry (notice the three sub-lattices $I_1$ with triangles, $I_2$ with dots and $I_3$ with crosses). Now, in order to keep to a minimum the number of possible variables upon which SHG could depend for this system, we fix the intrinsic characteristics of the isolated QD and also sub-lattices $I_2$ and $I_3$, leaving as the only variables the ingoing and outgoing polarizations for the fundamental and SH beams, and more importantly, for the effects of the local field, we vary only the $x$ position of the $I_1$ sub-lattice. With the above assumptions and partial results, Eq. (16) gives only the following components different from zero:

$$\chi_{xx}^{\text{eff}}, \chi_{xx}^{\text{eff}}, \text{and } \chi_{xx}^{\text{eff}} \neq \chi_{xx}^{\text{eff}},$$

which are also functions of $\omega$ and $a$.

We choose as example a QD made of GaAs with the following parameters: $R_0=150$ Å, $h_0=50$ Å electronic density of the 2D underlying electron gas of $n=10^{11}$ cm$^{-2}$, effective electron mass of $m^* = 0.068 m_0$, and the lattice constant $b=2000$ Å. With these numbers the energy of the xy ground state is $\omega_0= h k_{2D}^2/2m^* R_0^2 = 14$ meV or 114 cm$^{-1}$, which falls in the infrared region. Finally we introduce an $ad$ hoc damping parameter $1/\tau$ through $\omega^2+\omega^3+i\omega/\tau$, with $\tau$ satisfying $\omega_0\tau=20$.

In order to find the resonance of SHG, one has to sample over the frequency $\omega$ and $x$ position of the $I_1$ sub-lattice. Figure 1 shows the SHG efficiency $\Re$ for $p\rightarrow P$ polarizations, with $\theta=60^\circ$, and azimuthal angle (with respect to $x$) $\phi=0^\circ$, values that maximize the signal. The highest peak has a value of $1.9\times 10^{-14}$ cm$^2$/W, whereas the smaller ones peak at $0.3\times 10^{-14}$ cm$^2$/W. We mention that $\Re$ is symmetric with respect $a/b=-0.5$, and that $\Re(a/b=0.5)=0$. From Fig. 1 is clear that the resonances happen in both frequency and $x$ position of the $I_1$ sub-lattice.

To get a qualitative understanding of this nature we proceed as follows.\textsuperscript{13} The solution to Eq. (14) and thus to Eq. (16), is given by $\chi^{\text{eff}} \sim (1/\text{det}(T_{2\omega})) \times \gamma$, where $\text{det}(T_{2\omega})$ is the determinant of the matrix implied by Eq. (14) and $\gamma$ is proportional to the source term also implied by Eq. (14). By inspection is easy to see that $\text{det}(T_{2\omega})$ has an strong dependence with $a/b$ through $\Re$, thus giving the local field contribution, whereas $\gamma$ depends more on the resonant structure of $\chi^{(4)}_{\text{coh}}$, thus giving the spatial confinement contribution. In Fig. (2) we have plotted $\Im(\Re)$, $\Im(1/\text{det}(T_{2\omega}))$, and the log $\Re$ vs. $\omega/\omega_0$, for the highest peak of Fig. (1). From this figure we can conclude that the overall structure of the SHG efficiency $\Re$ comes from either $\Re$ or $1/\text{det}(T_{2\omega})$, thus giving the double nature of the SHG enhancements.

Finally, we define $\eta$ as an amplifications factor through

$$\eta = \frac{\chi^{\text{coh}}_0}{\chi(2\omega)},$$

where $\chi^{\text{coh}}(2\omega)=\chi_0(2\omega)$, and $\chi_0 = \frac{1}{32\pi^3} \frac{1}{\omega^2 b^3} \frac{n_0 a^3}{\hbar_0, e \omega_{\text{e}}=12.9}$, and we use the largest $\chi(2\omega)_{ijk}$ component. Now, $\chi^{\text{coh}}(\text{GaAs})=4.3\times 10^{-17}$ esu, and $\chi_0=1.12\times 10^{-7}$ esu for this example. Then, for the two peaks in Fig. 1, we get $\eta \approx 20000$ and 8000, where $\chi_{xx}^{\text{coh}}$ dominates. Thus we get giant enhancements with respect to a bulk sample. This kind of enhancements have been experimentally found in quantum wells,\textsuperscript{14} which are yet another example of nanoscopic systems.

It is interesting to notice that the resonant behavior of the local field gives rather sharp resonances in $a/b$, and that the sharpness in $\omega/\omega_0$ is given by $\omega_0\tau=20$ (see Fig. 1). Both of these features would impose a challenge in the quality and precision when building this kind of nanoscopic structures, but we believe that present technologies are within the range of these requirements.

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References

1) We list several conference proceedings, where both experimental and theoretical examples and many further references can be found: Interfaces, Quantum Wells and Superlattices, eds. C.R. Leavens and R. Taylor (Plenum, New York, 1988); Nano-


