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Pressure and temperature effects on the third-order nonlinear optical properties in GaAs quantum dots

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ABSTRACT

This work is used in the density matrix formalism and the effective mass approximation to study the third harmonic generation coefficient in a GaAs disc-shaped quantum dot with parabolic confinement potential. It is discussed the strong and weak confinement regime. The results show that the third harmonic generation coefficient is strongly dependent on the excitonic pair localization. The study is extended to consider effects such as hydrostatic pressure and temperature to show that it is possible to induce a blue-shift and/or red-shift on the resonant peaks of the third harmonic generation coefficient.

1. Introduction

Nonlinear optical properties in semiconducting quantum wells, wires and dots have been studied in the recent years both from the theoretical and experimental points of view (see, for instance, Karabulut et al. [1], and references therein). This is motivated mainly by the appearance of giant optical nonlinearities associated with intersubband transitions, which are much attractive for applications in optical communications and integrated optics. It is a well known fact that the symmetry breaking in a quantum well allows for significantly high values of the second order susceptibilities. There are reports about the second order nonlinear effects in step-like quantum wells, coupled quantum wells as well as in quantum wells under the effect of applied electric fields.

Theoretical calculations regarding the effect of exciton and impurity states on the nonlinear optical rectification in quantum dots were presented by Baskoutas and collaborators [2,3] and by Yu et al. [4]. The inclusion of external perturbations such as electric and magnetic fields has been put forward in reports on the calculation of the nonlinear optical rectification in zero- and one-dimensional heterostructures [5–7]. The generation of second and third harmonics in low-dimensional systems is theoretically studied and taking into account the effects of hydrostatic pressure, electron–hole correlation, and applied electric field [8–10]. Several works presenting the calculation of intersubband optical absorption have dealt with V-groove quantum wires, (CDS/ZnSe)/BeTe quantum wells, quantum rings and spherical quantum dots with parabolic potential confinement [11,12]. It is also possible to mention a previous work dealing with the study of nonlinear optical absorption in inverse parabolic quantum wells [13]. The literature also contains several publications dealing with higher harmonics generation in GaAs-based quantum dots with different geometries and confining potential profiles, with or without the inclusion of external electric and magnetic fields, as well as incorporating hydrostatic pressure or excitonic effects [14–21]. However, the literature concerning third harmonics generation in nanostructures is very scarce (see Ref. [22] and references therein). All the nonlinear properties mentioned so far have also been studied in tunable–symmetry systems, as is the case of the Pöschl–Teller quantum wells [23–26].

The exciton-related nonlinear optical rectification modulated by the application of hydrostatic pressure and external DC electric fields was considered by Duque et al. [27]. It is found that the resonant peak exhibits a blueshift as a result of the increase of the degree of carrier confinement. The strengthening of the applied field/hydrostatic pressure is related with a blue/red shift in the energy position of the resonant peak of the nonlinear optical rectification. The present work is an extension of previous studies by Duque et al. [27], and Wang and Guo [28], and has to do with the theoretical study of the combined effects of the electron–hole correlation, hydrostatic pressure, and temperature on third

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harmonic generation in a disc-shaped GaAs quantum dot. The work is organized as follows: In Section 2 the theoretical framework is introduced. Section 3 is devoted to the presentation and discussion of the obtained results, and, finally, Section 4 contains the corresponding conclusions.

2. Theoretical framework

The system under study is that of an exciton confined in a two-dimensional quantum dot with the form of a disc, subjected to a parabolic confining potential. In terms of the relative, $\vec{r} = \vec{r}_e - \vec{r}_h$, and center of mass coordinates, $\vec{R} = (m_e^* \vec{r}_e + m_h^* \vec{r}_h)/M$, the Hamiltonian of the exciton in the parabolic band and effective mass approximations is given by,

$$H = \left[\frac{P^2}{2M} + \frac{1}{2} M\omega_0^2 R^2\right]_{cm} + \left[\frac{p^2}{2\mu} + \frac{1}{2}\mu\omega_0^2 r^2 - \frac{e^2}{\varepsilon r}\right]_{rel}, \tag{1}$$

where ω_0 is the intensity of the parabolic confinement, $\mu = m_e^* m_h^*/M$ is the reduced mass of the electron–hole pair, $M = m_e^* + m_h^*$, and ε is the static dielectric constant of the quantum dot material. The indices cm and rel label, respectively, the center of mass and relative coordinates.

It is possible to define two different scales of length and energy — (L, E) — for the problem under consideration in the form

$$(R_0, \hbar\omega_0) = \left(\sqrt{\frac{\hbar}{\mu\omega_0}}, \frac{\hbar^2}{\mu R_0^2}\right) \tag{2}$$

and

$$(a_B^*, \mathfrak{R}^*) = \left(\frac{\varepsilon\hbar^2}{\mu e^2}, \frac{e^2}{\varepsilon a_B^*}\right). \tag{3}$$

According to such scales, two regimes of confinement can be defined: (i) the strong confinement regime (SC), where $R_0 \ll a_B^*$ (equivalent to neglect the Coulombic interaction associated with the relative coordinate); (ii) the weak confinement (WC) regime, where $R_0 \gg a_B^*$ (which is equivalent to neglect the parabolic term associated with the relative coordinate).

The effects of the hydrostatic pressure and temperature are included via the dependence upon these quantities of the electron and hole (the light one in this case) effective masses and the dielectric constant of the quantum dot material. For the electron effective mass it can be written [29],

$$\frac{m_e^*(P,T)}{m_0} = \left[1 + \frac{15\,020}{E_g(P,T)} + \frac{7510}{E_g(P,T)+341}\right]^{-1}, \tag{4}$$

where

$$E_g(P,T) = \left[1519 + 10.7P - \frac{0.5405T^2}{T+204}\right] \text{meV}. \tag{5}$$

Here, $E_g(P,T)$ is the pressure- and temperature-dependent energy gap of the GaAs. m_0 is the free electron mass. On the other hand, the light hole effective mass is given by Refs. [30,31]

$$\frac{m_h^*(P)}{m_0} = (0.09 - 0.20 \times 10^{-3}P - 3.55 \times 10^{-5}T). \tag{6}$$

The static dielectric constant, ε is taken as [32]

$$\varepsilon(P,T) = \varepsilon_a e^{\alpha_1 T + \alpha_2 P}, \tag{7}$$

where $\varepsilon_a = 12.6498$, $\alpha_1 = 9.4 \times 10^{-5}$ K^{-1} and $\alpha_2 = -1.67 \times 10^{-3}$ kbar^{-1}, for $T < 200$ K, whereas in the range $T \geq 200$ K we have $\alpha_1 = 20.4 \times 10^{-5}$ K^{-1} and $\alpha_2 = -1.73 \times 10^{-3}$ kbar^{-1}. When $T \geq 200$ K, the value of ε_a is obtained by the continuity of $\varepsilon(P,T)$ at $T = 200$ K, for any value of the hydrostatic pressure.

Table 1
Matrix elements for SC and WC.

nn'	$\mu_{nn'}$ (SC)	$\mu_{nn'}$ (WC)
01	$0.43R_0$	$0.16a_B^*$
12	$0.61R_0$	$1.12a_B^*$
23	$0.74R_0$	$2.74a_B^*$
30	$0.06R_0$	$0.04a_B^*$

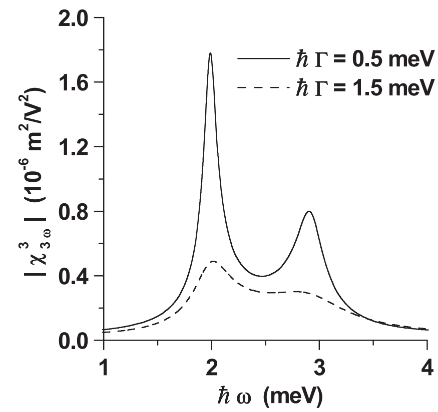

Fig. 1. THG coefficient for an exciton confined in a $Ga_{0.7}Al_{0.3}As$ parabolic quantum dot as a function of the incident photon energy ($\hbar\omega$). The figure corresponds to the weak confinement regime, for two different values of the relaxation constant ($\hbar\Gamma$).

Once the eigenstates of the Hamiltonian (1), and their corresponding eigenvalues, are found; the coefficient of third-harmonic generation (THG) can be calculated in the case of two-photon resonance. The expression to evaluate in this case is obtained within the formalism of the density-matrix when an electromagnetic field — of frequency ω — with polarization vector normal to the quantum dot surface is considered. Then, the expression for the THG is [10]

$$\chi_{3\omega}^{(3)} = -\frac{\mu_{01}\mu_{12}\mu_{23}\mu_{30}\rho_s}{\varepsilon_0 \prod_{k=1}^{3}(k\hbar\omega - E_{k0} + i\hbar\Gamma_{k0})}, \tag{8}$$

where $\hbar\omega$ is the incident photon energy, ρ_s ($= 5 \times 10^{24}$ m^{-3}) is the density of excitons in the quantum dot, ε_0 represents the vacuum permittivity, and $E_{k0} = E_{(0,0)}^{rel} - E_{(k,0)}^{rel}$ is the energy interval between two exciton states. In addition, $\mu_{nn'} = |\langle \varphi_{(n,0)}|er|\varphi_{(n',0)}\rangle|$ $(n, n' = 0,1,2,3,\ldots)$ are the off-diagonal dipole moment matrix elements between exciton states, and Γ_{k0} are the corresponding relaxation rates. In Table 1, these matrix elements are presented in both limits of strong and weak confinement.

3. Results and discussion

In what follows, some results for the THG coefficient — $\chi_{3\omega}^{(3)}$ — are presented for $Ga_{1-x}Al_xAs$ parabolic quantum dots in both the SC and WC regimes. In the calculation, the following values have been taken: $\hbar\Gamma_{10} = \hbar\Gamma$ meV, $\hbar\Gamma_{20} = \hbar\Gamma/2$ meV and $\hbar\Gamma_{30} = \hbar\Gamma/3$ meV [28].

The exciton-related THG in the WC regime is shown in Fig. 1 as a function of the incident photon energy in the situation of a $Ga_{0.7}Al_{0.3}As$ quantum dot with parabolic confinement. Results are presented for two different values of the relaxation constant. As it can be seen, there are two well defined peaks in each curve, associated with the resonant energies of the incident photon. Significant variations are exhibited by such peaks as long as the