Effects of the Coulomb interaction on intersubband doubling and difference-frequency mixing in asymmetric double quantum wells

Effects of the Coulomb interaction on intersubband doubling and difference-frequency mixing in asymmetric double quantum wells

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Abstract. We study theoretically the influence of the direct and indirect Coulomb interaction on second-harmonic generation (SHG) and difference-frequency generation (DFG) connected with intersubband transitions in asymmetric double quantum wells. Our approach is based on the time-dependent local density approximation. The exact analytical expressions have been derived for SHG (in a two-subband system) and DFG (in a three-subband system) surface susceptibilities. Unlike in our previous papers, we take into account the nonresonant terms and nonlinear dependence of the exchange–correlation potential upon the electron density. Numerical calculations have been performed for GaAl/AlGaAs asymmetric coupled double-quantum-well structures with small energy separation (about 10 meV) between the ground and excited subband when the effect of the Coulomb interaction is most pronounced and the nonresonant terms become essential. The results obtained show that Coulomb interaction not only shifts the peak positions in SHG and DFG spectra, but also modifies their height. The height modification can be particularly strong (even more than one order of magnitude) in the case of the SHG. The nonlinearity of the exchange–correlation potential is found to lead to a novel feature that cannot be ignored in a correct description of SHG and DFG spectra. The electron-density dependence of the many-body effect is analysed. A critical comparison of the results obtained with those reported in the literature is also given.

1. Introduction

Second-order nonlinear intersubband optical properties of asymmetric quantum wells have been investigated experimentally and theoretically by many groups [1–10]. However, in most of the theoretical works, the authors neglect the influence of Coulomb interaction on the nonlinear optical response despite the fact that in the case of linear intersubband absorption the above influence can play a very important role. Ando *et al* [11] have shown, employing the time-dependent local density approximation (TDLDA), that when ground (1) and excited (2) subbands are parabolic, the electron–electron interaction leads to a shift of the intersubband absorption energy from the corresponding intersubband energy separation E_{21}. This shift is determined by:

(i) the depolarization effect (DE) (which arises from the direct Coulomb interaction and causes an increase of the absorption energy [12]) and

(ii) the exciton-like effect (EE) (which results from the exchange–correlation interaction and leads to a reduction in the absorption energy [11]).

The above-mentioned shift can be particularly strong (comparable with E_{21}) in structures with $E_{21} \leqslant 10$ meV and large electron concentration ($N_S \geqslant 10^{11}$ cm^{-2}). A coupled asymmetric double quantum well (CADQW) [3,4] is a typical example of such a structure.

Recently Heyman *et al* [3] (see also reference [4]) have shown experimentally that in CADQWs not only the intersubband absorption peak position but also the low-energy peak position in the SHG spectrum can be very strongly affected by the Coulomb interaction. For interpretation of experimental results, the authors used an analytical expression for $\chi^{(2)}(2\omega)$ derived in the two-subband limit employing the TDLDA. Unfortunately, it is not clear how reliable this expression is because the authors did not present details of the calculations. Moreover, the expression for $\chi^{(2)}(2\omega)$ reported in the above-mentioned references contradicts a well known rule stating that the poles of the susceptibility should lie in the lower half of the complex-frequency plane [13]. The SHG in two-level structures was also discussed theoretically (in the framework of the TDLDA) by Gusmão and Mahan [5]. However, the authors restricted consideration to numerical calculations concentrating just on the influence of the Coulomb interaction on the low-energy peak position in SHG spectra.

The strong influence of the many-body interaction on the doubly resonant difference-frequency-generation (DFG) spectra in CADQWs was observed experimentally by Sirtori *et al* [6]. A simplified description of the above effect was given in our previous paper [9], where the calculations were performed using the time-dependent Hartree approximation and including only resonant terms.

In this paper we present a detailed derivation of the exact analytical expressions for

(i) the second-order susceptibility, $\chi^{(2)}(2\omega)$, connected with the SHG in two-subband asymmetric double QWs and

(ii) the second-order susceptibility, $\chi^{(2)}(\omega_3 = \omega_1 - \omega_2)$, connected with the DFG in doubly resonant three-subband asymmetric double QWs.

Our approach is based on the TDLDA and the density matrix formulation (in the nonretarded electric dipole approximation) employed in our previous papers [8–10]. The formulae obtained in this work include the nonresonant terms which are shown to be essential for the correct description for the structures with small intersubband energy interaction and cannot be ignored as in reference [9]. Both the direct and indirect contribution to the Coulomb interaction are included. Considering the effects induced by the indirect Coulomb interaction, we take into account the nonlinear dependence of the exchange–correlation potential upon the electron concentration. It is found that this nonlinearity affects the peak values of the spectra but does not influence the peak position.

The paper is organized as follows. In section 2 we derive analytical expressions for the SHG and DFG susceptibilities. The numerical results and detailed discussion are given in section 3. Section 4 contains conclusions.

2. Theory

We start from the one-band effective mass Hamiltonian

$$\hat{H} = p^2/2m + V_{\text{EFF}}(z) \tag{1}$$

where m is the effective mass of an electron. In the LDA approach the electron is assumed to move in an effective potential

$$V_{\text{EFF}}(z) = \mathcal{V}_{\text{CONF}}(z) + \mathcal{V}_{\text{H}}(z) + \mathcal{V}_{\text{XC}}(z)$$

where $\mathcal{V}_{\text{CONF}}(z)$ determines the shape of the confining quantum well potential, $\mathcal{V}_{\text{H}}(z)$ is the electrostatic Hartree potential, and $\mathcal{V}_{\text{XC}}(z)$ denotes the exchange–correlation potential energy. We adopt a standard parametrized form (originally due to Gunnarson and Lundqvist [14]) for this potential:

$$\mathcal{V}_{\text{XC}}(z) = -\left[1 + 0.7734\,X(z)\ln\left(1 + \frac{1}{X(z)}\right)\frac{2\,\text{Ryd}^*}{\pi\alpha r_s(z)}\right]$$

where

$$r_s(z) = [4\pi a_*^3 n(z)/3]^{-1/3}$$

$$X(z) = r_s(z)/21$$

$$\alpha = (4/9\pi)^{1/3}.$$

Also,

$$a_* = 4\pi\epsilon_0\epsilon\hbar^2/me^2$$

is the effective Bohr radius,

$$\text{Ryd}^* = me^4/2(4\pi\epsilon_0\epsilon\hbar)^2$$

is the effective Rydberg, ϵ is the averaged dielectric constant, and $n(z)$ is the electron density.

The eigenfunction of Hamiltonian (1) can be written as $|i, \mathbf{k}_\parallel\rangle = \exp(\mathrm{i}\mathbf{k}_\parallel \mathbf{r}_\parallel)\varphi_i(z)$ where $\mathbf{k}_\parallel$ and $\mathbf{r}_\parallel$ are the wave vector and the position vector in the x–y plane, respectively. $\varphi_i(z)$ is a solution of the one-dimensional Schrödinger equation

$$[p_z^2/2m + V_{\text{EFF}}(z)]\varphi_i(z) = E_i\varphi_i(z)$$

where E_i is the minimum energy of the ith subband. In this paper we neglect, for simplicity, the position and energy dependence of the effective mass. Thus, $\varphi_i(z)$ and the intersubband energy, $E_{ij} = E_i - E_j$, can be treated as independent of $\mathbf{k}_\parallel$.

The equation of motion for the matrix elements of the density matrix ρ (in the representation of $|i, \mathbf{k}_\parallel\rangle$ ($i = 1, 2, 3$)) is given by

$$\frac{\partial\rho_{ij}}{\partial t} = \frac{1}{\mathrm{i}\hbar}[\hat{H} + \Delta V, \rho]_{ij} - \frac{\Delta\rho_{ij}}{\tau_{ij}} \tag{2}$$

where $\Delta V \equiv \Delta V(z, t)$ is the effective perturbing Hamiltonian, τ_{ii}^{-1} is the relaxation rate from the ith subband, $\tau_{ij}^{-1} = \tau_{ji}^{-1}$ is the off-diagonal elastic dephasing rate connected with $i \rightarrow j$ transitions, $\Delta\rho = \rho - \rho^{(0)}$, and $\rho^{(0)}$ is the unperturbed density matrix. The diagonal element $\rho_{jj}^{(0)} \equiv \rho_{jk_\parallel jk_\parallel}^{(0)}$ is equal to the thermal equilibrium occupation probability, $F_j(k_\parallel)$, of the corresponding state. The equilibrium surface density of the electrons in the jth subband is given by

$$N_j = 2\sum_{\mathbf{k}_\parallel} F_j(k_\parallel).$$

Note that in the approximation used here, $\rho_{ik_\parallel jk_\parallel'} = \delta_{\mathbf{k}_\parallel, \mathbf{k}_\parallel'}\rho_{ik_\parallel jk_\parallel}$.

The electric field, $E(t)$, of the pumped radiation modifies the density distribution of electrons. (Because we are interested in the SHG and DFG connected with intersubband optical transitions, we take into account only the z-component of the electric field of the radiation.)

The change of the electron distribution $\Delta n(z, t) = n(z) - n_0(z)$ ($n_0(z)$ is the unperturbed electron distribution) can be expressed through the density matrix as

$$\Delta n(z, t) = 2\,\text{Tr}[\Delta\rho\,\delta(z - z')]. \tag{3}$$

The modification of the carrier distribution leads to the modification of the effective perturbing potential, $\Delta V(z, t)$, which in the electrostatic limit can be written in the form

$$\Delta V(z, t) = \Delta V_{\text{EXT}}(z, t) + \Delta V_{\text{H}}(z, t) + \Delta V_{\text{XC}}(z, t) \tag{4}$$

where $\Delta V_{\text{H}}(z, t)$ takes into account the direct Coulomb interaction whereas $\Delta V_{\text{XC}}(z, t)$ describes the exchange–correlation interaction [11].

As in most previous papers, we assume that the external perturbation, $\Delta V_{\text{EXT}}(z, t) = eE(t)z$, is small. In this limit, a self-consistent solution of equations (2)–(4) can be obtained perturbatively by expanding $\Delta\rho$, ΔV, and Δn (generally denoted as ΔX) in powers of the external electric field E as

$$\Delta X(z, t) = \sum_{n>0} X^{(n)}(z, t). \tag{5}$$

Substituting equation (5) into equation (2) and using the usual iterative method we get

$$\frac{\partial \rho_{ij}^{(n)}}{\partial t} = \frac{1}{\text{i}\hbar}[\hat{H}, \rho^{(n)}]_{ij} + \frac{1}{\text{i}\hbar}\sum_{k>0}^{n}[V^{(k)}, \rho^{(n-k)}]_{ij} - \frac{\rho_{ij}^{(n)}}{\tau_{ij}}. \tag{6}$$

The surface electronic polarization, $P_s(t)$, can also be a series expansion like equation (5). The nth-order term, $P_s^{(n)}(t)$, is connected with $\rho^{(n)}$ by the relation [1]

$$P_s^{(n)}(t) = \frac{-e}{\epsilon_0}\,\text{Tr}[\rho^{(n)}(z, t)z]. \tag{7}$$

2.1. Second-harmonic susceptibility

First we consider the SHG in ACDQW structures whose energy spectrum can be approximated by a two-subband model, i.e., we assume that $E_{21} \ll E_{l2}$ ($l > 2$). The electric field of pumped radiation is taken in the form $E(t) = \tilde{E}\exp(-\text{i}\omega t) + \text{c.c.}$. This implies that

$$\Delta V_{\text{EXT}}(z, t) = V^{(\text{ext})}(z, \omega)\exp(-\text{i}\omega t)$$

where $V^{(\text{ext})}(z, \omega) = e\tilde{E}z$.

When for simplicity we neglect the rectification effect, then

$$X^{(n)}(z, t) = X^{(n)}(z, \Omega_n)\exp(-\text{i}\Omega_n t)$$

where $\Omega_n = n\omega$ ($n = 1, 2$). (The validity of this approximation is discussed at the end of section 3.1.) Thus, a second-order correction to the surface electronic polarization can be written as

$$P_s^{(2)}(t) = \epsilon_0\chi^{(2)}(2\omega)\tilde{E}^2\exp(-\text{i}\,2\omega t) + \text{c.c.} \tag{8}$$

where $\chi^{(2)}(2\omega)$ is the SHG surface susceptibility.

From equations (8) and (7) we find that

$$\chi^{(2)}(2\omega) = \frac{-e}{\epsilon_0\tilde{E}^2}\sum_{i,j}\bar{\rho}_{ij}^{(2)}(2\omega)z_{ji} = \frac{-e}{\epsilon_0\tilde{E}^2}[\bar{\rho}^{(2)}(2\omega)z_{21} + \bar{\rho}_{11}^{(2)}(2\omega)z_{11} + \bar{\rho}_{22}^{(2)}(2\omega)z_{22}] \tag{9}$$

where

$$z_{ij} = z_{ji} = \int_{-\infty}^{\infty}\varphi_i(z)z\varphi_j(z)\,\text{d}z$$

$$\bar{\rho}_{ij}^{(n)}(\Omega_n) = 2\sum_{k_\parallel}\rho_{ij}^{(n)}(\Omega_n)$$

$$\bar{\rho}^{(n)}(\Omega_n) = \bar{\rho}_{12}^{(n)}(\Omega_n) + \bar{\rho}_{21}^{(n)}(\Omega_n).$$

Employing equations (5) and (6) we get

$$\bar{\rho}_{ij}^{(n)}(\Omega_n) = \frac{1}{\hbar\Omega_n + i\Gamma_{ij} - E_{ij}} \sum_{k>0}^{n} [V^{(k)}(z, \Omega_k), \bar{\rho}^{(n-k)}(z, \Omega_{n-k})]_{ij} \tag{10}$$

with $\Gamma_{ij} = \hbar\tau_{ij}^{-1}$.

In the quasi-static approximation, the direct Coulomb contribution to the nth-order effective potential, $V^{(n)}(z, \Omega_n)$, can be written in terms of $n^{(n)}(z, \Omega_n)$:

$$V_{\text{H}}^{(n)}(z, \Omega_n) = -\frac{e^2}{\epsilon_0\epsilon_\infty} \int_{-\infty}^{z} \mathrm{d}z' \int_{-\infty}^{z'} \mathrm{d}z'' \, n^{(n)}(z'', \Omega_n). \tag{11}$$

Expanding $\mathcal{V}_{\text{XC}}[n(z, t)]$ in powers of $\Delta n(z, t)$ up to second order, we find that the contribution to $V^{(n)}(z, \Omega_n)$ $(n = 1, 2)$ connected with exchange–correlation potential is given by [5]

$$V_{\text{XC}}^{(n)}(z, \Omega_n) = V_{\text{XC}}'(z)n^{(n)}(z, \Omega_n) + \frac{1}{2}V_{\text{XC}}''(z)[n^{(1)}(z, \omega)]^2\delta_{n2} \tag{12}$$

where $V_{\text{XC}}'(z)$, $(V_{\text{XC}}''(z))$ is the first (second) functional derivative of $\mathcal{V}_{\text{XC}}(z)$ with respect to the equilibrium density distribution of the electrons. It must be emphasized that the second term in the above equation arises due to a nonlinear dependence of the exchange–correlation potential upon the sheet electron density. Since this term does not appear when we consider the linear response, we call it an 'additional driving term' (ADT). Equation (12) shows that it is the nonlinearity (or ADT) by means of which the first-harmonic of electron-density modification, $n^{(1)}(z, \omega)$, influences the second harmonic of the effective perturbing potential, $V^{(2)}(z, 2\omega)$. Note that in this paper we work in a standard static approximation, i.e. we neglect the frequency dependence of the exchange–correlation potential (for details, see reference [15]).

Using equations (3), (11), and (12) we find the following expression for the matrix elements of $V^{(n)}(z, \Omega_n)$:

$$V_{ij}^{(n)}(\Omega_n) = V_{ij}^{(\text{ext})}(\omega)\delta_{1n} + \sum_{k,l} \gamma(i, j; k, l)\bar{\rho}_{kl}^{(n)}(\Omega_n) + Z(i, j; \omega)\delta_{2n} \tag{13}$$

where

$$\gamma(i, j; k, l) = \alpha(i, j; k, l) + \beta(i, j; k, l) \tag{14}$$

with

$$\alpha(i, j; k, l) = \frac{e^2}{\epsilon_0\epsilon_\infty} L(i, j; k, l)$$

and

$$L(i, j; k, l) = \int_{-\infty}^{\infty} \mathrm{d}z \left[\int_{-\infty}^{z} \mathrm{d}z' \, \varphi_i(z')\varphi_j(z')\right]\left[\int_{-\infty}^{z} \mathrm{d}z' \, \varphi_k(z')\varphi_l(z')\right] \tag{15}$$

$$\beta(i, j; k, l) = \int_{-\infty}^{\infty} \mathrm{d}z \, \varphi_i(z)\varphi_j(z)\varphi_k(z)\varphi_l(z)V_{\text{XC}}'(z) \tag{16}$$

$$Z(i, j; \omega) = \frac{1}{2}\int_{-\infty}^{\infty} \mathrm{d}z \, \varphi_i(z)\varphi_j(z)V_{\text{XC}}''(z)\left[\sum_{k,l} \bar{\rho}_{kl}^{(1)}(\omega)\varphi_k(z)\varphi_l(z)\right]^2. \tag{17}$$

One can check that

$$Z(1, 2; \omega) = Z(2, 1; \omega) = \left[\bar{\rho}^{(1)}(\omega)\right]^2 \bar{Z}(1, 2) \tag{18}$$

where

$$\bar{Z}(1, 2) = \bar{Z}(2, 1) = \frac{1}{2}\int_{-\infty}^{\infty} \mathrm{d}z \, [\varphi_1(z)\varphi_2(z)]^3 \, V_{\text{XC}}''(z). \tag{19}$$

Equations (10) and (13) form a set of algebraic equations for $\bar{\rho}_{ij}^{(n)}(\Omega_n)$. Their application for $n = 1$ yields

$$\bar{\rho}_{ij}^{(1)}(\omega) = \frac{N_{ij}V_{ij}^{(1)}(\omega)}{E_{ij} - \hbar\omega - \mathrm{i}\Gamma} = \frac{N_{ij}}{E_{ij} - \hbar\omega - \mathrm{i}\Gamma}\left[V_{ij}^{(\mathrm{ext})}(\omega) + \gamma(1,2;1,2)\bar{\rho}^{(1)}(\omega)\right] \tag{20}$$

where $N_{ij} = N_i - N_j$ and $\Gamma = \Gamma_{12}$.

The expression for $\bar{\rho}^{(1)}(\omega)$, resulting from the above equation, can be written as

$$\bar{\rho}^{(1)}(\omega) = -\frac{2N_{12}\,V_{12}^{(\mathrm{ext})}(\omega)E_{21}}{\tilde{E}_{21}^2 - (\hbar\omega + \mathrm{i}\Gamma)^2} \tag{21}$$

where

$$\tilde{E}_{21} = E_{21}[1 + 2\gamma(1,2;1,2)N_{12}/E_{21}]^{1/2}$$

is the intersubband energy shifted by the DE and the EE [11].

Employing equations (13)–(21), we find after some manipulations that

$$V_{ij}^{(1)}(\omega) = V_{ij}^{(\mathrm{ext})}(\omega) + \gamma(1,2;i,j)\bar{\rho}^{(1)}(\omega) = V_{ij}^{(\mathrm{ext})}(\omega)C_{ij}(\omega) \tag{22}$$

with

$$C_{ij}(\Omega_n) = \frac{E_{21}^2 - (\hbar\Omega_n + \mathrm{i}\Gamma)^2}{\tilde{E}_{21}^2 - (\hbar\Omega_n + \mathrm{i}\Gamma)^2} \qquad i \neq j \tag{23}$$

and

$$C_{ii}(\Omega_n) = \frac{E_{ii}'^2 - (\hbar\Omega_n + \mathrm{i}\Gamma)^2}{\tilde{E}_{21}^2 - (\hbar\Omega_n + \mathrm{i}\Gamma)^2} \tag{24}$$

where

$$E_{ii}'^2 = E_{21}^2\left\{1 + 2\left[\gamma(1,2;1,2) - \gamma(1,2;i,i)\frac{z_{12}}{z_{ii}}\right]\frac{N_{12}}{E_{21}}\right\}.$$

(We assume here that $z_{ii} \neq 0$.)

Taking $n = 2$ and assuming for simplicity that $\Gamma_{11} = \Gamma_{22} = \bar{\Gamma}$, we get from equations (10) and (13) the following relations:

$$\bar{\rho}_{11}^{(2)}(2\omega) = \frac{V_{12}^{(1)}(\omega)[\bar{\rho}_{21}^{(1)}(\omega) - \bar{\rho}_{12}^{(1)}(\omega)]}{2\hbar\omega + \mathrm{i}\bar{\Gamma}} = -\frac{2[V_{12}^{(\mathrm{ext})}(\omega)]^2 N_{12}}{2\hbar\omega + \mathrm{i}\bar{\Gamma}}\frac{(\hbar\omega + \mathrm{i}\bar{\Gamma})[E_{21}^2 - (\hbar\omega + \mathrm{i}\Gamma)^2]}{[\tilde{E}_{21}^2 - (\hbar\omega + \mathrm{i}\Gamma)^2]^2} \tag{25}$$

$$\bar{\rho}_{22}^{(2)}(2\omega) = -\bar{\rho}_{11}^{(2)}(2\omega) \tag{26}$$

and

$$\bar{\rho}^{(2)}(2\omega) = \frac{2E_{21}\gamma_{21}\bar{\rho}_{11}^{(2)}(2\omega)N_{12}}{\tilde{E}_{21}^2 - (2\hbar\omega + \mathrm{i}\Gamma)^2} - \frac{2Z(1,2;\omega)N_{12}E_{21}}{\tilde{E}_{21}^2 - (2\hbar\omega + \mathrm{i}\Gamma)^2}$$
$$- [V_{22}^{(1)}(\omega) - V_{11}^{(1)}(\omega)]\frac{(E_{21} + 2\hbar\omega + \mathrm{i}\Gamma)\bar{\rho}_{21}^{(1)}(\omega) + (E_{21} - 2\hbar\omega - \mathrm{i}\Gamma)\bar{\rho}_{12}^{(1)}(\omega)}{\tilde{E}_{21}^2 - (2\hbar\omega + \mathrm{i}\Gamma)^2} \tag{27}$$

where $\gamma_{21} = \gamma(1,2;2,2) - \gamma(1,2;1,1)$.

Substitution of equations (25), (26), and (27) into equation (9) gives the final expression for the second-harmonic susceptibility in the TDLDA as a sum of three terms which represent different aspects of the electron–electron interaction effect:

$$\chi^{(2)}(2\omega) = \chi_{(A)}^{(2)}(2\omega) + \chi_{(B)}^{(2)}(2\omega) + \chi_{(C)}^{(2)}(2\omega) \tag{28}$$

where

$$\chi^{(2)}_{(A)}(2\omega) = \frac{-2e^3 N_{12} z_{12}^2 (z_{22} - z_{11})[E_{21}^2 - (\hbar\omega + i\Gamma)^2]}{\epsilon_0[\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2}$$
$$\times \left[\frac{E_{21}^2 + (2\hbar\omega + i\Gamma)(\hbar\omega + i\Gamma)}{\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2} \frac{\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2}{E_{21}^2 - (\hbar\omega + i\Gamma)^2} + \frac{\hbar\omega + i\Gamma}{2\hbar\omega + i\bar{\Gamma}} \right] \qquad (29)$$

$$\chi^{(2)}_{(B)}(2\omega) = \frac{e^3 N_{12}^2 z_{12}^3 \gamma_{21} 2 E_{21}[E_{21}^2 - (\hbar\omega + i\Gamma)^2]}{\epsilon_0[\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2[\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2]}$$
$$\times \left[\frac{E_{21}^2 + (2\hbar\omega + i\Gamma)(\hbar\omega + i\Gamma)}{E_{21}^2 - (\hbar\omega + i\Gamma)^2} + \frac{\hbar\omega + i\Gamma}{2\hbar\omega + i\bar{\Gamma}} \right] \qquad (30)$$

and

$$\chi^{(2)}_{(C)}(2\omega) = \frac{8e^3 N_{12}^3 z_{12}^3 \bar{Z}_{12} E_{21}^3}{\epsilon_0[\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2[\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2]}. \qquad (31)$$

The first term in equation (28), $\chi^{(2)}_{(A)}(2\omega)$, plays dominant role only in the range of small electron concentration. In the one-electron limit ($\Delta V(z, t) = \Delta V_{\text{EXT}}(z, t)$) it reduces to the expression derived by Tsang *et al* [7]:

$$\chi^{(2)}_{(0)}(2\omega) = \frac{-2e^3 N_{12} z_{12}^2 (z_{22} - z_{11})}{\epsilon_0[E_{21}^2 - (\hbar\omega + i\Gamma)^2]} \left[\frac{E_{21}^2 + (2\hbar\omega + i\Gamma)(\hbar\omega + i\Gamma)}{E_{21}^2 - (2\hbar\omega + i\Gamma)^2} + \frac{\hbar\omega + i\Gamma}{2\hbar\omega + i\bar{\Gamma}} \right]. \qquad (32)$$

The last two terms in equation (28) arise due to a many-body interaction. They become important at higher electron concentrations since $\chi^{(2)}_{(B)}(2\omega) \propto N_{12}^2$ and $\chi^{(2)}_{(C)}(2\omega) \propto N_{12}^3$.

The second term, $\chi^{(2)}_{(B)}(2\omega)$, results from the coupling between the intersubband and intrasubband transitions induced by $V_{\text{H}}^{(1)}(z, \omega)$ and $V_{\text{XC}}^{(1)}(z, \omega)$. The strength of this coupling is controlled by factor γ_{21} (see equation (22)).

The third term, $\chi^{(2)}_{(C)}(2\omega)$, has its origin directly in the nonlinearity of the exchange–correlation potential. (More exactly, it appears due to the presence of the ADT in the expression for $V_{\text{XC}}^{(2)}(z, 2\omega)$). Equation (31) shows that inclusion of the ADT affects only the peak value of the SH susceptibility and does not affect the resonance photon energies at all. It is a new feature of the exchange–correlation interaction. The physical reason for this is that the resonance condition $\tilde{E}_{21} = \hbar\omega$ ($\tilde{E}_{21} = 2\hbar\omega$) is affected only by $n^{(1)}(z, \omega)$ through $V^{(1)}(z, \omega)$ ($n^{(2)}(z, 2\omega)$ through $V^{(2)}(z, 2\omega)$). The role of the ADT (i.e. the nonlinearity) is to take into account the influence of $n^{(1)}(z, \omega)$ on $V^{(2)}(z, 2\omega)$. Thus, by definition, the nonlinearity cannot affect the resonance conditions. The above facts lead us to the conclusion that, in contrast with that of the linear response, the influence of the exchange–correlation interaction on the SH susceptibility cannot be considered as a simple correction to the DE.

The commutation of any function $f = f(z, t)$ with z results in the following relation:

$$(f_{22} - f_{11})/f_{12} = (z_{22} - z_{11})/z_{12}$$

where $f_{ij} = \langle i|f|j \rangle$. Employing this relation for the function $\gamma(1, 2; i, j)$, the expression for $\chi^{(2)}(2\omega)$ can be transformed to the following form:

$$\chi^{(2)}(2\omega) = \frac{-2e^3 N_{12} z_{12}^2 (z_{22} - z_{11})}{\epsilon_0} \frac{[E_{21}^2 - (\hbar\omega + i\Gamma)^2]}{[\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2][\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2}$$
$$\times E_{21}^2 \left[1 + \frac{\hbar\omega + i\Gamma}{2\hbar\omega + i\bar{\Gamma}} + \frac{(2\hbar\omega + i\Gamma)(\hbar\omega + i\Gamma)}{E_{21}^2} \frac{i(\bar{\Gamma} - \Gamma)}{2\hbar\omega + i\bar{\Gamma}} \right] + \chi^{(2)}_{(C)}(2\omega). \qquad (33)$$

By using the facts that $\Gamma \ll \hbar\omega$ and $(\hbar\omega + i\Gamma)/(2\hbar\omega + i\bar{\Gamma})$ is practically equal to $1/2$ for the near-resonance regime, the above equation can be simplified to

$$\chi^{(2)}(2\omega) = \frac{-e^3 N_{12} z_{12}^2 (z_{22} - z_{11})}{\epsilon_0} \frac{3E_{21}^2 [E_{21}^2 - (\hbar\omega + i\Gamma)^2]}{[\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2][\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2} + \chi_{(C)}^{(2)}(2\omega).$$

(34)

Now we compare our expression for $\chi^{(2)}(2\omega)$ with that reported by Heyman *et al* [3] given by

$$\chi^{(2)}(2\omega) = \frac{-e^3 N_{12} z_{21}^2 (z_{22} - z_{11})}{\varepsilon_0} \frac{3(E_{21} + i\Gamma)^2 [(E_{21} + i\Gamma)^2 - (\hbar\omega)^2]}{[(\tilde{E}_{21} + i\Gamma)^2 - (2\hbar\omega)^2][(\tilde{E}_{21} + i\Gamma)^2 - (\hbar\omega)^2]^2}.$$

(35)

The difference between equation (35) and our formula, equation (34), is readily seen. As was mentioned in section 1, equation (35) has a drawback: it contradicts the well known rule stating that the poles of the susceptibility should lie in the lower half of the complex-frequency plane [13]. This feature is responsible for incorrect sign of $\mathrm{Im}[\chi^{(2)}(2\omega)]$ predicted by equation (35). It is important to note that, as a result, equation (35) does not reduce in the one-electron limit to that derived by Tsang *et al*. Nevertheless, in this limit the difference between the numerical values of $|\chi^{(2)}(2\omega)|$ predicted by equation (35) and the expressions given by Tsang *et al* is negligibly small for CADQW. Also, for the parameters used in our paper, the numerical difference of $|\chi^{(2)}(2\omega)|$ predicted by equation (35) and our simplified equation (34) with $\chi_{(C)}^{(2)}(2\omega) = 0$ can be ignored.

2.2. Difference-frequency-generation susceptibility

Now we derive the expression for the doubly resonant DFG susceptibility connected with the intersubband transitions in three-subband asymmetric double-QW structures. The electric field of the incident radiation is taken in the form

$$E(t) = \tilde{E}(\omega_1) \exp(-i\omega_1 t) + \tilde{E}(\omega_2) \exp(-i\omega_2 t) + \text{c.c.}$$

where ω_1 and ω_2 ($<\omega_1$) are angular frequencies of pumped waves. To be close to experiment, we assume that the structure is similar to that studied by Sirtori *et al* [6], i.e., $E_{21} \ll E_{32} \approx \hbar\omega_t$ ($t = 1, 2$). For simplicity, we also assume that only the ground subband is occupied ($N_i = \delta_{1i} N_S$). The surface second-order electronic polarization oscillating with the difference frequency $\omega_3 = \omega_1 - \omega_2$ takes the form

$$P_s^{(2)}(t) = \epsilon_0 \chi^{(2)}(\omega_3) \tilde{E}(\omega_1) \tilde{E}^*(\omega_2) \exp(-i\omega_3 t) + \text{c.c.}$$

(36)

where $\chi^{(2)}(\omega_3)$ is the second-order susceptibility associated with the DFG.

From equations (36) and (7) we find that

$$\chi^{(2)}(\omega_3) = \frac{-e}{\epsilon_0 \tilde{E}(\omega_1) \tilde{E}^*(\omega_2)} \mathrm{Tr}[\rho^{(2)}(z, \omega_3) z] = \frac{-e z_{12}}{\epsilon_0 \tilde{E}(\omega_1) \tilde{E}^*(\omega_2)} \bar{\rho}^{(2)}(\omega_3)$$

(37)

where $\bar{\rho}^{(2)}(\omega_3) = \bar{\rho}_{12}^{(2)}(\omega_3) + \bar{\rho}_{21}^{(2)}(\omega_3)$.

Starting from equation (6) and performing manipulations similar to that used in the case of SHG (see also reference [9]), we get

$$\rho_{ij}^{(1)}(\omega_t) = \frac{V_{ij}^{(1)}(\omega_t)(\rho_{jj}^{(0)} - \rho_{ii}^{(0)})}{E_{ij} - \hbar\omega_t - i\Gamma_{ij}}$$

(38)

for $t = 1, 2$, and

$$\rho_{ij}^{(2)}(\omega_3) = \frac{[(V^{(1)}(z, \omega_2))^*, \rho^{(1)}(z, \omega_1)]_{ij} + V_{ij}^{(2)}(\omega_3)(\rho_{jj}^{(0)} - \rho_{ii}^{(0)})}{E_{ij} - \hbar\omega_3 - i\Gamma_{ij}}.$$

(39)

The direct and exchange–correlation contributions to $V^{(1)}(z, \omega_t)$ and $V^{(2)}(z, \omega_3)$ can be written in the form

$$V_H^{(1)}(z, \omega_t) = -\frac{e^2}{\epsilon_0 \epsilon_\infty} \int_{-\infty}^{z} dz' \int_{-\infty}^{z'} dz'' \, n^{(1)}(z'', \omega_t) \tag{40}$$

$$V_{XC}^{(1)}(z, \omega_t) = V_{XC}'(z) n^{(1)}(z, \omega_t) \tag{41}$$

and

$$V_H^{(2)}(z, \omega_3) = -\frac{e^2}{\epsilon_0 \epsilon_\infty} \int_{-\infty}^{z} dz' \int_{-\infty}^{z'} dz'' \, n^{(2)}(z'', \omega_3) \tag{42}$$

$$V_{XC}^{(2)}(z, \omega_3) = V_{XC}'(z) n^{(2)}(z, \omega_3) + V_{XC}''(z) n^{(1)}(z, \omega_1)[n^{(1)}(z, \omega_2)]^*. \tag{43}$$

The second term in equation (43) results from the nonlinear dependence of the exchange–correlation potential upon the sheet electron density. Thus, similarly to the case for the SHG, we call this term the ADT. The ADT (or nonlinearity) takes into account the influence of the first harmonics of the electron-density modification, $n^{(1)}(z, \omega_1)$ and $n^{(1)}(z, \omega_2)$, on the second harmonic of the effective perturbing potential, $V^{(2)}(z, \omega_3)$.

The matrix elements of $V^{(1)}(z, \omega_t)$ and $V^{(2)}(z, \omega_3)$ obey the following relations:

$$V_{ij}^{(1)}(\omega_t) = V_{ij}^{(ext)}(\omega_t) + \sum_{k,l} \gamma(i, j; k, l) \bar{\rho}_{kl}^{(1)}(\omega_t) \tag{44}$$

$$V_{ij}^{(2)}(\omega_3) = \sum_{k,l} \gamma(i, j; k, l) \bar{\rho}_{kl}^{(2)}(\omega_3) + Z_d(i, j) \tag{45}$$

where $\gamma(i, j; k, l)$ is given by equation (14), while

$$Z_d(j, i) = \int_{-\infty}^{\infty} dz \, \varphi_i(z) \varphi_j(z) V_{XC}''(z)$$

$$\times \left[\sum_{k,l} \varphi_k(z) \varphi_l(z) \bar{\rho}_{kl}^{(1)}(\omega_1) \right] \left[\sum_{k,l} \varphi_k(z) \varphi_l(z) [\bar{\rho}_{kl}^{(1)}(\omega_2)]^* \right]. \tag{46}$$

Calculating matrix elements of $\rho^{(1)}$, we employ the fact that only the resonant term can be considered when $\hbar\omega_1$ is close to E_{31} and the Coulomb correction to the resonance energy corresponding to the $1 \to 3$ transitions is small. This term has the following form:

$$\bar{\rho}_{31}^{(1)}(\omega_t) = -\frac{V_{31}^{(1)}(\omega_t) N_S}{E_{31} - \hbar\omega_t - i\Gamma_{31}}. \tag{47}$$

The second-order terms resulting from equation (39) are given by

$$\bar{\rho}_{21}^{(2)}(\omega_3) = -\frac{[V_{23}^{(1)}(\omega_2)]^* \bar{\rho}_{31}^{(1)}(\omega_1) + V_{21}^{(2)}(\omega_3) N_S}{E_{21} - \hbar\omega_3 - i\Gamma_{21}} \tag{48}$$

and

$$\bar{\rho}_{12}^{(2)}(\omega_3) = -\frac{V_{12}^{(2)}(\omega_3) N_S}{E_{21} + \hbar\omega_3 + i\Gamma_{21}}. \tag{49}$$

Note that in reference [9], only resonant term $[\bar{\rho}_{21}^{(2)}(\omega_3)]$ was taken into account. Omission of the off-resonant term $\bar{\rho}_{12}^{(2)}(\omega_3)$ has a good justification only when $\tilde{E}_{21} - E_{21} \ll E_{21}$.

From equations (44) and (45), we get the following expressions for the matrix elements appearing in equations (47)–(49):

$$V_{31}^{(1)}(\omega_t) = V_{31}^{(ext)}(\omega_t) + \gamma(3, 1; 3, 1) \bar{\rho}_{31}^{(1)}(\omega_t) \tag{50}$$

$$V_{32}^{(1)}(\omega_2) = V_{23}^{(1)}(\omega_2) = V_{32}^{(\text{ext})}(\omega_2) = e\tilde{E}(\omega_2)z_{23} \tag{51}$$

and

$$V_{12}^{(2)}(\omega_3) = V_{21}^{(2)}(\omega_3) = \gamma(1, 2; 2, 1)\bar{\rho}^{(2)}(\omega_3) + \bar{Z}_d\bar{\rho}_{31}^{(1)}(\omega_1)[\bar{\rho}_{31}^{(1)}(\omega_2)]^* \tag{52}$$

with

$$\bar{Z}_d = \int_{-\infty}^{\infty} \mathrm{d}z\, \varphi_1^3(z)\varphi_2(z)\varphi_3^2(z)V_{\text{xc}}''(z). \tag{53}$$

After some algebra, we obtain the expression for $\bar{\rho}^{(2)}(\omega_3)$ as

$$\bar{\rho}^{(2)}(\omega_3) = \frac{e^2\tilde{E}(\omega_1)\tilde{E}^*(\omega_2)z_{12}z_{31}N_S}{(\tilde{E}_{21} - \hbar\omega_3 - \mathrm{i}\Gamma_{21})(\tilde{E}_{31} - \hbar\omega_1 - \mathrm{i}\Gamma_{31})}D(\omega_2, \omega_3) \tag{54}$$

with

$$D(\omega_2, \omega_3) = \frac{E_{21} + \hbar\omega_3 + \mathrm{i}\Gamma_{21}}{\tilde{E}_{21} + \hbar\omega_3 + \mathrm{i}\Gamma_{21}} - \frac{2\bar{Z}_d N_S^2 E_{21}z_{31}/z_{23}}{(\tilde{E}_{21} + \hbar\omega_3 + \mathrm{i}\Gamma_{21})(\tilde{E}_{21} + \hbar\omega_2 + \mathrm{i}\Gamma_{21})} \tag{55}$$

where

$$\tilde{E}_{21} = E_{21}[1 + 2\gamma(1, 2; 1, 2)N_S/E_{21}]^{1/2}$$

$$\tilde{E}_{31} = E_{31} + \gamma(1, 3; 1, 3)N_S.$$

The final expression for $\chi^{(2)}(\omega_3)$ takes the form

$$\chi^{(2)}(\omega_3) = \frac{-e^3 N_S}{\epsilon_0}\frac{z_{12}z_{23}z_{31}}{(\tilde{E}_{21} - \hbar\omega_3 - \mathrm{i}\Gamma_{21})(\tilde{E}_{31} - \hbar\omega_1 - \mathrm{i}\Gamma_{31})}D(\omega_2, \omega_3). \tag{56}$$

The second term in the expression for $D(\omega_2, \omega_3)$ comes into being solely due to the ADT. Naturally, for the same reason as for the SHG, the ADT does not affect the resonance condition for $\chi^{(2)}(\omega_3)$. We have checked numerically that, in contrast with the case for SHG, this term has a negligible influence on the spectral shape of $\chi^{(2)}(\omega_3)$. Employing this fact, we can make the following approximation:

$$D(\omega_2, \omega_3) \cong D(\omega_3) = \frac{E_{21} + \hbar\omega_3 + \mathrm{i}\Gamma_{21}}{\tilde{E}_{21} + \hbar\omega_3 + \mathrm{i}\Gamma_{21}}. \tag{57}$$

If the depolarization correction is small $((\tilde{E}_{21} - E_{21}) \ll E_{21})$, the above function can be replaced by unity. In this limit,

$$\chi^{(2)}(\omega_3) = \frac{-e^3 N_S}{\epsilon_0}\frac{z_{12}z_{23}z_{31}}{(\tilde{E}_{21} - \hbar\omega_3 - \mathrm{i}\Gamma_{21})(\tilde{E}_{31} - \hbar\omega_1 - \mathrm{i}\Gamma_{31})}. \tag{58}$$

Since $(\tilde{E}_{21} - E_{21}) \ll E_{21}$, the intersubband resonance energy appearing in the above equation can be approximated by

$$\tilde{E}_{21} = E_{21}[1 + \gamma(1, 2; 1, 2)N_S/E_{21}].$$

Note that in the Hartree approximation, i.e. when only direct Coulomb interaction is considered, the above expression for $\chi^{(2)}(\omega_3)$ reduces to that derived in our previous paper [9].

Making in equation (58) the substitution $\tilde{E}_{i1} \rightarrow E_{i1}$, we get a well known one-electron formula for the doubly resonant difference-mixing-frequency susceptibility [6].

3. Numerical results and discussion

Numerical calculations reported in this paper have been performed for modulation-doped CADQWs (similar to that studied in reference [3]), with 85 Å and 75 Å GaAs wells separated by a 25 Å $Al_{0.3}As_{0.7}Ga$ barrier (see the inset in figure 1). We have used the following parameters: the conduction band offset $\delta E_c = 233$ meV; the electron effective masses $m = 0.066\, m_0$ (the same for the wells and barriers). The dephasing parameters were taken to be $\Gamma_{1i} = 0.42$ meV ($i = 1, 2, 3$) unless otherwise stated and $\bar{\Gamma} = 0.1$ meV. For simplicity, we restrict consideration to the case where only the ground subband is occupied.
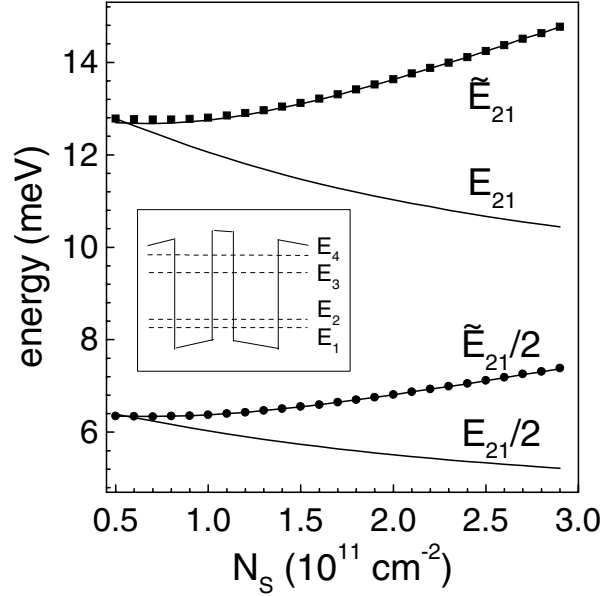

Figure 1. Dependences of $\tilde{E}_{21}$ and E_{21} on N_S. For comparison, we present also the N_S-dependence of $\hbar\omega_{res}^H$ (squares) and $\hbar\omega_{res}^L$ (circles). The inset shows the energy band diagram of the CADQW structure with $N_S = 2.5 \times 10^{11}$ cm^{-2}. Shown are the positions of the calculated energy levels: $E_1 = 35$ meV, $E_2 = 47$ meV, $E_3 = 144$ meV, $E_4 = 187$ meV.

3.1. SHG spectra

Figure 1 presents the electron-density dependence of E_{21} and $\tilde{E}_{21}$ in the above-mentioned structure. From this figure we find that the intersubband resonance energy $\tilde{E}_{21}$ increases with N_S due to many-body effects despite the fact that the bare intersubband gap E_{21} decreases with increasing N_S. At large electron concentration, the difference between $\tilde{E}_{21}$ and E_{21} becomes substantial, and therefore we can expect a very strong influence of the Coulomb interaction on the spectral shape of the SHG susceptibility. This suggestion is supported by figure 2 which shows the spectral shape of $|\chi^{(2)}(2\omega)|^2$ for the structure with $N_S = 2.5 \times 10^{11}$ cm^{-2}. The lower (L) and higher (H) resonance photon energies are denoted by $\hbar\omega_{res}^L$ and $\hbar\omega_{res}^H$, respectively. It is important to note that $\hbar\omega_{res}^L$ ($\hbar\omega_{res}^H$) practically coincides with $\tilde{E}_{21}/2$ ($\tilde{E}_{21}$) (see figure 1).

Inspection of figure 2 shows that, in contrast with the case for linear intersubband absorption, not only the position but also the height of the L- and H-peaks in the SHG spectrum
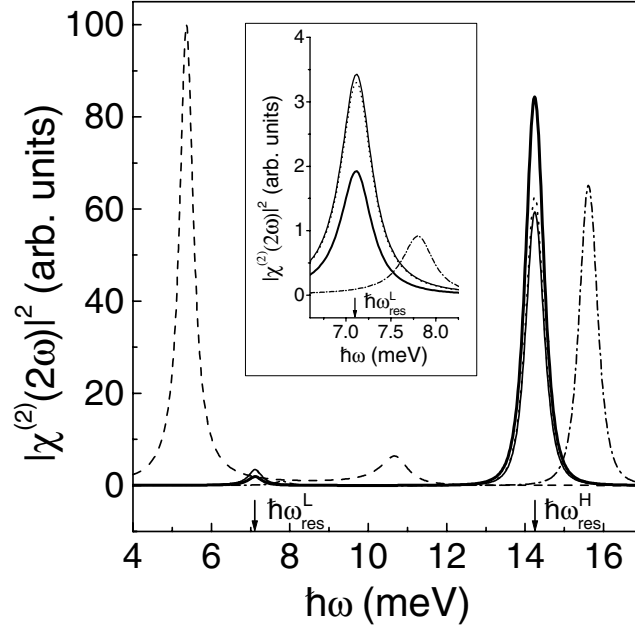
Figure 2. Spectral dependences of $|\chi^{(2)}(2\omega)|^2$ in: the TDLDA (thick solid line), the TDLDA but without the ADT (thin solid line), the Hartree approximation (dash–dot line), the one-electron approximation (dashed line), the simplified equation (34) without the ADT (dotted line). The inset shows the detailed behaviour of $|\chi^{(2)}(2\omega)|^2$ for ω near ω_{res}^L.

(i.e. $|\chi^{(2)}(2\omega)|^2_{max} = |\chi^{(2)}(2\omega_{res}^{L,H})|^2$) is strongly modified by the Coulomb interaction. Our numerical results reveal that the modification of the peaks is essentially different. Many-body interaction dramatically reduces the height of the L-peak and enhances the height of the H-peak. (Note that the Coulomb interaction practically does not affect the width of the peaks.) The numerical calculations indicate that the above-mentioned modification is particularly strong in the case of the L-peak and high electron concentration (see figures 3 and 4). For example, at $N_S \approx 3 \times 10^{11}$ cm^{-2} the height of the L-peak is reduced by more than two orders of magnitude while the H-peak is enhanced by up to about one order of magnitude with respect to the one-electron result. It should be stressed that the peak height modification is determined mainly by the direct Coulomb interaction.

It is important to note that, due to the Coulomb interaction, the height of the L-peak is not a monotonic function of N_S, as it is in the one-electron approach (see figure 3). There is some optimum electron concentration (N_S^*) at which the SHG is maximal. (In the structure considered, $N_S^* \approx 1.1 \times 10^{11}$ cm^{-2}.)

Inspection of numerical results leads to an interesting conclusion that the modification of the H-peak is much more sensitive to the value of the line broadening parameter Γ than the modification of the L-peak (see figure 4). This difference is revealed best in the N_S-dependence of the factor

$$R_{SHG} = |\chi^{(2)}_{max}(2\omega)|^2/|\chi^{(2)}_{(0),max}(2\omega)|^2$$

which determines the strength of the peak modification induced by the Coulomb interaction. The insets in figure 4 present the N_S-dependence of this factor for H-peaks (R_{SHG}^H) and L-peaks (R_{SHG}^L) for different values of Γ (taken as a constant dependent upon N_S). (The N_S-dependence of Γ was recently reported by Williams *et al* [16].) One can see that R_{SHG}^L, being less than
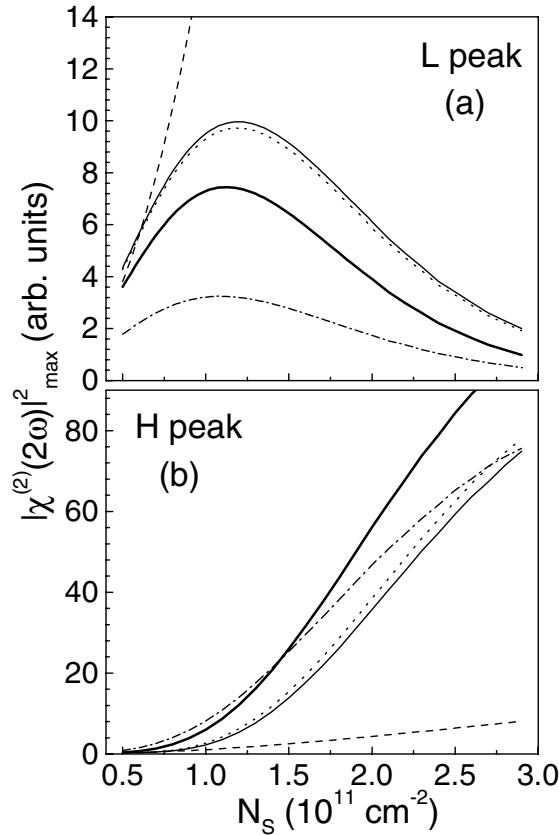
Figure 3. The height of the *L*-peak (a) and that of the *H*-peak (b) versus N_S. The meaning of the lines is the same as in figure 2.

1, decreases nearly exponentially with increasing electron concentration and is practically independent of Γ. The behaviour of R_{SHG}^H, which is bigger than 1, is quite different. It increases with increasing N_S (when N_S is not too large) and decreases very slowly with further increase of Γ. Unfortunately, the experimental verification of this feature for the *H*-peak would be a major challenge, since $\hbar\omega_{res}^H$ practically coincides with the intersubband resonant energy $\tilde{E}_{21}$. (Since $\bar{\Gamma} \ll \tilde{E}_{21}$, the dependence of $\chi^{(2)}(2\omega)$ on $\bar{\Gamma}$ is negligibly small. The term $(\hbar\omega + i\Gamma)/(2\hbar\omega + i\bar{\Gamma})$ in equations (29) and (30) can be replaced by $\frac{1}{2}$. Our numerical calculations verify that this approximation works very well.)

The numerical results presented in figure 3 illustrate also our previous statement that inclusion of the ADT affects only the height of the peaks. The modification of the peak height induced by the ADT is substantial and cannot be ignored when we study the influence of the indirect interaction on SHG spectra.

It is pertinent to note that the direct and indirect Coulomb interactions are shown to be always competing in their effect on the position and height of the *L*- and *H*-peaks.

Finally, we should make some remarks about the validity of our approach. We suppose that the range of power of the incident radiation considered lets us restrict to a second-order correction in equation (8) and neglect the rectification (see reference [17]). Indeed our results on the shift of the *L*-peak coincide with the experimental results of Sherwin *et al* [4], while
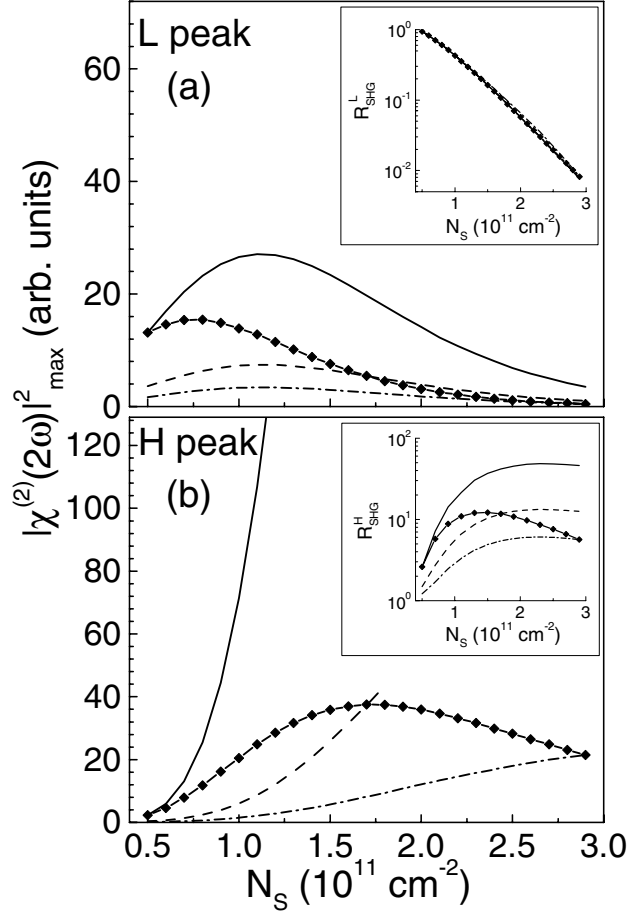
Figure 4. Dependences of $|\chi^{(2)}(2\omega)|^2_{\text{max}}$ and the factor R_{SHG} (insets) on N_S for the L-peak (a) and H-peak (b) calculated for different values of the broadening parameter Γ: 0.22 meV (solid line), 0.42 meV (dashed line), 0.62 meV (dash–dot line), and Γ increasing linearly from 0.22 meV (at $N_S = 0.5 \times 10^{11}$ cm^{-2}) to 0.62 meV (at $N_S = 3.0 \times 10^{11}$ cm^{-2}) (squares).

the L-peak-value reduction is in qualitative agreement with reference [4] (direct comparison is impossible, because in reference [4] N_S was changed by an applied voltage which changed also other parameters of the quantum structure). However, as mentioned in reference [4], since the excitation is resonant and saturation is expected to occur, the H-peak of the second harmonic will be difficult to detect. Nevertheless, this problem may be solved by working at higher temperature [4].

3.2. DFG spectra

The spectral shapes of $|\chi^{(2)}(\omega_3)|^2$ for the CADQW with $N_S = 2.5 \times 10^{11}$ cm^{-2} and $\hbar\omega_1 = \tilde{E}_{31}$ are presented in figure 5. We find that due to the Coulomb interaction, the peak position is shifted (from E_{21} to $\tilde{E}_{21}$) and the peak height is reduced. (Note that approximate equation (58) does not predict the peak reduction.) The reduction is controlled by the factor

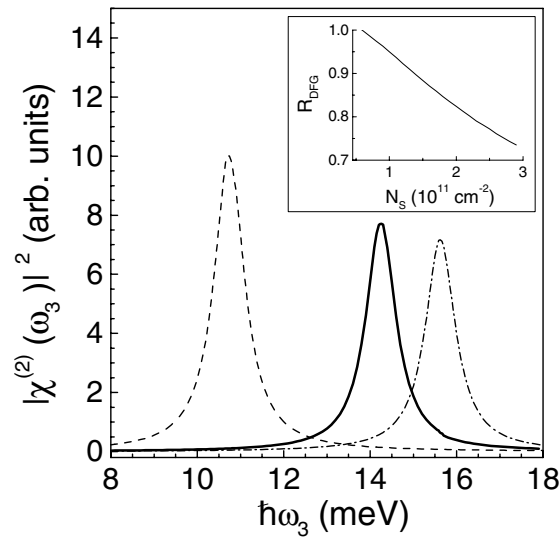$$R_{\text{DFG}} = |D(\omega_3 = \tilde{E}_{21}/\hbar)|^2.$$

Figure 5. Spectral dependences of $|\chi^{(2)}(\omega_3)|^2$ in: the TDLDA without the ADT (thick solid line) (inclusion of the ADT has practically no effect), the Hartree approximation (dash–dot line), the one-electron approximation (dashed line). The inset shows the N_S-dependence of the factor R_{DFG}.

The inset in figure 5 presents the N_S-dependence of R_{DFG}. We see that as long as $(\tilde{E}_{21} - E_{21})/E_{21} < 1$, the above-mentioned peak reduction is rather small and can be neglected in the first approximation. Experimental results of reference [6] confirm qualitatively our results on the position shift and value reduction of the peak (quantitative comparison is impossible because of the uncertainty of the measurement accuracy pointed out in reference [6]).

4. Conclusions

In this paper we have considered, employing the TDLDA, the influence of the direct and indirect Coulomb interaction on the singly resonant SHG and the doubly resonant DFG in asymmetric double-quantum-well structures. We have found that (in contrast with our previous results [9]) the nonresonant terms must be taken into account when describing the Coulomb interaction effect in CADQWs with small subband separation. The many-body interaction is shown to affect the position as well as the height of the peaks in both spectra. The peak height modification is found to be particularly strong (even two orders of magnitude) in the case of the SHG. The low- (high-) energy peak is suppressed (enhanced) by the many-body effect with growing sheet electron density. A novel feature of the influence of the indirect Coulomb interaction connected with nonlinearity of the exchange–correlation potential is found: the nonlinearity induces a considerable change of the peak value and does not affect the peak position. Inclusion of this effect is essential for the correct description of the above-mentioned peak height modification in SHG spectra.

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