Influence of the depolarization effect on second-harmonic generation in asymmetric quantum wells

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The effect of the depolarization field on the second-harmonic generation due to resonant intersubband transitions in asymmetric quantum wells is discussed. Calculations are performed in the framework of the perturbative density-matrix formulation with relaxation-time approximation. The results obtained show that in the near-double-resonant system the depolarization effect not only shifts the peak, in the second-harmonic-generation spectrum, but also changes its shape and enhances the maximum value.

I. INTRODUCTION

Recently there has been a strong interest in the second-order nonlinear optical phenomena connected with the intersubband transitions in asymmetric quantum wells (AQW's).[1–13] This interest results both from a fundamental physics point of view as well from the possible practical applications in the area of integrated optics and optical communications.

The calculation of the electromagnetic response (of the low-dimensional electron gas), with the standard mean-field approaches, consists of two steps.[14,15] First, determination of electron states of the system by self-consistent solution of Schrödinger's equation and Poisson's equation. (The exchange-correlation interaction can be incorporated within the density-functional scheme.) Second, the response of the system to the electromagnetic radiation is obtained by employing the random-phase approximation (RPA) or the time-dependent local-density approximation (TDLDA),[15] if exchange-correlation effects are to be included. To obtain an accurate representation of the response we have to include both steps.

In most of the theoretical works devoted to the second-order nonlinear responses, the second step is omitted and the authors concentrate on the optical coupling between the ground occupied subband and several excited subbands. By manipulating the well shape or/and by applying a static electric field, one can enhance matrix elements and make the energy dominator very small, leading to very strong nonlinear responses. Omitting the second step is equivalent (when we work in the RPA) to neglecting the depolarization effect (DE). Some authors[3,11,12] have made an attempt to take into account the DE. In Ref. 11, the DE was included by simply replacing (in the one-electron expression for the second-order susceptibility) the intersubband energy by the depolarization-shifted intersubband energy. The self-consistent treatment (based on the TDLDA) of the second-harmonic generation (SHG) in the two-level AQW system was reported by Heyman *et al.*[12] The three-level system was considered by Fejer, Yoo, and Byer.[3] The authors of Ref. 3 calculated the SHG coefficient using the self-consistent approach similar to that developed by Allen, Tsui, and Vinter.[14] Unfortunately, (i) the calculations have been performed only numerically, and (ii) the coupling between the different intersubband-subband transitions was neglected. A more sophisticated approach to the SHG in quantum wells subject to an applied electric field was reported in Ref. 13. This approach is based on the electromagnetic scattering theory presented in Ref. 16, including the retardation effects. Unfortunately, the authors of Ref. 13 concentrated only on the two subband model. Results obtained in Refs. 3, 12, and 13 indicate that the DE leads mainly to the blue shift of the resonance in the SHG spectrum.

In this paper, our aim is to discuss the modification the SHG spectrum (in the three-level system) induced by the DE, making allowance for the coupling between different intersubband transitions. We show that in the case of the near-double-resonant structures the effects induced by the DE are much more complicated than was suggested in Refs. 3 and 11.

II. THEORY

Our analysis is based on the density-matrix formulation with relaxation-time approximation. Like in the previous papers, we consider the three subband system assuming that only the ground subband is occupied in the absence of the radiation.

For a parabolic conduction band, the energies of the subbands can be expressed as

$$E_i(\mathbf{k}_t)=E_i+\hbar^2k_t^2/2m^*, \quad i=1,2,3 , \tag{1}$$

where $\mathbf{k}_t$ is the wave vector in the x-y plane, $E_i=E_i(\mathbf{k}_t=0)$, and m^* is the effective mass of electrons in the conduction band. (We neglect for simplicity the difference between the effective masses inside the well and in the barrier.)

The corresponding wave functions (normalized to the unit area of the system) can be written as

$$\Psi_i(\mathbf{k}_t,\mathbf{r}_t)=\exp(i\mathbf{k}_t\cdot\mathbf{r}_t)\varphi_i(z) , \tag{2}$$

where $\mathbf{r}_t$ is the position vector in the x-y plane and $\varphi_i(z)$ is the solution of the one-dimensional Schrödinger equation $\mathcal{H}_0\varphi_i=E_i\varphi_i$ with $\mathcal{H}_0=p_z^2/2m^*+V_{\mathrm{SCF}}(z)$. [$V_{\mathrm{SCF}}(z)$ is the self-consistent quantum-well potential.[2,7,14]]

Since in our system the intersubband energy

$E_{ji}=E_j(\mathbf{k}_t)-E_i(\mathbf{k}_t)$ is independent on $\mathbf{k}_t$, the electron gas can be treated as a sum of noninteracting N_s three-level systems, where N_s is the surface density of the carrier in the QW.

The equation for the matrix elements of the density matrix [in the representation of $\varphi_i(z)$] is given by[5,17–19]

$$\frac{\partial\rho_{ij}}{\partial t}=(1/i\hbar)[\mathcal{H}_0+\mathcal{V},\rho]_{ij}-\Delta\rho_{ij}/\tau_{ij} , \tag{3}$$

where $\Delta\rho=\rho-\rho^{(0)}$, $\rho^{(0)}$ is the unperturbed density matrix having only diagonal term $\rho_{ij}^{(0)}=\delta_{i1}\delta_{j1}$. $\mathcal{V}\equiv\mathcal{V}(z,t)$ is the effective perturbing Hamiltonian, τ_{ii}^{-1} is the relaxation rate from the ith subband, and τ_{ij}^{-1} is the off-diagonal elastic dephasing rate. (We assume for simplicity that $T=0$.)

Since the operator $\mathcal{V}(z,t)$ is Hermitian, we have the following relations: $\mathcal{V}_{ij}=\mathcal{V}_{ji}^*$ and $\rho_{ij}=\rho_{ji}^*$.

Under usual experimental conditions, the wavelength of the light is much larger than the effective thickness of the system. Thus, we can work in the dipole approximation taking the electric field of the incident radiation (applied in the z direction) in the form

$$D(t)=\tilde{D}\exp(-i\omega t)+\tilde{D}\exp(i\omega t) . \tag{4}$$

(At this point, we should note that in the case of a weakly asymmetric system the correct description of the SHG must take into account the z dependence of the external radiation.[13,16])

The field $D(t)$ modifies the density distribution of electrons. The change of the distribution $[\Delta n(z,t)]$ can be expressed through the density matrix as

$$\Delta n(z',t)=N_s\mathrm{Tr}[\Delta\rho\delta(z-z')] . \tag{5}$$

Modification in the carrier distribution leads to the modification of the Hartree potential. Thus, the effective perturbing potential, appearing in Eqs. (3), can be written in the form[14,15]

$$\begin{aligned}\mathcal{V}(z,t)&=\mathcal{V}^{\mathrm{ext}}(z,t)+\Delta\mathcal{V}(z,t)\\&=eD(t)z-(e^2/\varepsilon_0\varepsilon_\infty)\int_{-\infty}^{z}dz'\int_{-\infty}^{z'}dz''\Delta n(z'',t) ,\end{aligned} \tag{6}$$

where ε_∞ is the background dielectric constant and $-e$ is the electron charge.

(In this paper, we shall restrict ourselves to the electrostatic limit where the velocity of light can be taken to be infinite, and the electric field can be expressed as gradient of a potential.)

As in most of the previous papers, we assume that the external perturbation $[\mathcal{V}^{\mathrm{ext}}(z,t)]$ is small. Then the self-consistent solution of Eqs. (4)–(6) can be obtained perturbatively by expanding $\Delta\rho$, Δn, and $\Delta\mathcal{V}$ in powers of $\tilde{D}$ as

$$\Delta\rho=\sum_{n>0}\rho^{(n)} , \tag{7a}$$

$$\Delta n=\sum_{n>0}n^{(n)} , \tag{7b}$$

$$\Delta\mathcal{V}=\sum_{n>0}\mathcal{V}^{(n)} . \tag{7c}$$

The surface electronic polarization $[P_s(t)]$ can also be written in the similar form. We shall limit ourselves to the first two orders, i.e.,

$$\begin{aligned}P_s(t)=&\varepsilon_0\chi^{(1)}(\omega)\tilde{D}\exp(-i\omega t)+\varepsilon_0\chi^{(2)}(2\omega)\tilde{D}^2\exp(-2i\omega t)\\&+\mathrm{c.c.}+\varepsilon_0\chi^{(2)}(0)\tilde{D}^2 ,\end{aligned} \tag{8}$$

where $\chi^{(1)}(\omega)$, $\chi^{(2)}(2\omega)$, and $\chi^{(2)}(0)$ are the linear, SHG, and optical rectification coefficients, respectively.

If we neglect, for simplicity, the effects connected with rectification then, in the approximation used here, Eq. (7) reduces to

$$\begin{aligned}\Delta\rho(t)=&\rho^{(1)}(\omega)\exp(-i\omega t)\\&+\rho^{(2)}(2\omega)\exp(-i2\omega t)+\mathrm{c.c.},\end{aligned} \tag{9a}$$

$$\begin{aligned}\Delta n(z,t)=&n^{(1)}(z,\omega)\exp(-i\omega t)\\&+n^{(2)}(z,2\omega)\exp(-i2\omega t)+\mathrm{c.c.},\end{aligned} \tag{9b}$$

$$\begin{aligned}\Delta\mathcal{V}(z,t)=&\mathcal{V}^{(1)}(z,\omega)\exp(-i\omega t)\\&+\mathcal{V}^{(2)}(z,2\omega)\exp(-i2\omega t)+\mathrm{c.c.}\end{aligned} \tag{9c}$$

It is well known that, in the case of the finite systems, the polarizability $[\mathbf{P}(\mathbf{r},t)]$ is related to the induced charge density $[\Delta n(\mathbf{r},t)]$ by

$$\nabla\cdot\mathbf{P}(\mathbf{r},t)=e\Delta n(\mathbf{r},t) . \tag{10}$$

Using the above relation and Eqs. (8) and (9b), we find that the SHG coefficient can be written as

$$\chi^{(2)}(2\omega)=\frac{e}{\tilde{D}^2}\int_{-\infty}^{\infty}dz\int_{-\infty}^{z}dz'n^{(2)}(z',2\omega) . \tag{11}$$

Thus, the knowledge of the induced charge density at frequency 2ω is sufficient to determine the SHG coefficient. (In the absence of the electron-electron interaction, it is more convenient to use the relation $\chi^{(2)}(2\omega)=N_s\mathrm{Tr}[\rho^{(2)}(2\omega)(-e)z]/\varepsilon_0\tilde{D}^2$ for the calculation of the SHG coefficient.[5,17])

Successive orders of the density matrix can be obtain from Eqs. (3) and (7a) using the usual iterative method:

$$\begin{aligned}\frac{\partial\rho_{ij}^{(n)}}{\partial t}=&(1/i\hbar)[\mathcal{H}_0,\rho^{(n)}]_{ij}-\rho_{ij}^{(n)}/\tau_{ij}\\&+\sum_{k>0}(1/i\hbar)[\mathcal{V}^{(k)},\rho^{(n-k)}]_{ij} .\end{aligned} \tag{12}$$

Using Eqs. (5)–(7) and (9), we find that

$$\rho_{ij}^{(1)}(\omega)=\frac{\mathcal{V}_{ij}^{(1)}(\omega)(\rho_{jj}^{(0)}-\rho_{ii}^{(0)})}{\hbar\omega-E_{ij}+i\Gamma_{ij}} , \tag{13}$$

$$\begin{aligned}\rho_{ij}^{(2)}(2\omega)=&\frac{1}{2\hbar\omega-E_{ij}+i\Gamma_{ij}}[\mathcal{V}^{(1)}(z,\omega),\rho^{(1)}(\omega)]_{ij}\\&+\frac{\mathcal{V}_{ij}^{(2)}(2\omega)(\rho_{jj}^{(0)}-\rho_{ii}^{(0)})}{2\hbar\omega-E_{ij}+i\Gamma_{ij}} ,\end{aligned} \tag{14}$$

with

$$\mathcal{V}^{(n)}(z,\omega_n)=\mathcal{V}^{\text{ext}}(z,\omega)\delta_{n1} -(e^2/\varepsilon_0\varepsilon_\infty)\int_{-\infty}^{z}dz'\int_{-\infty}^{z'}dz''n^{(n)}(z'',\omega_n)\ , \tag{15}$$

and

$$n^{(n)}(z,\omega_n)=N_s\sum_i\sum_j\varphi_i(z)\varphi_j(z)\rho_{ij}^{(n)}(\omega_n)\ , \tag{16}$$

where $\Gamma_{ij}=\hbar\tau_{ij}^{-1}$, $\mathcal{V}^{\text{ext}}(z,\omega)=e\tilde{D}z$, $\omega_1=\omega$, and $\omega_2=2\omega$. Using Eqs. (16) and (13) we get

$$n^{(1)}(z,\omega)=-N_s\sum_{k=2,3}\varphi_1(z)\varphi_k(z)\frac{2\mathcal{V}_{k1}^{(1)}(\omega)E_{k1}}{E_{k1}^2-(\hbar\omega+i\Gamma_{k1})^2}\ . \tag{17}$$

Unfortunately, the second-order correction $[n^{(2)}(z,2\omega)]$ resulting from inserting Eq. (14) to Eq. (16) is much more complicated. However, in the case of the near-double-resonant structures ($E_{21}\approx E_{32}\approx\hbar\omega$), we can neglect the off-resonant terms (assuming that the relative line broadening is small $\Gamma_{ij}\ll E_{ij}$). Then

$$n^{(2)}(z,2\omega)=n_1^{(2)}(z,2\omega)+n_2^{(2)}(z,2\omega)\ , \tag{18}$$

where

$$n_1^{(2)}(z,2\omega)=N_s\varphi_1(z)\varphi_3(z) \times\frac{\mathcal{V}_{21}^{(1)}(\omega)\mathcal{V}_{32}^{(1)}(\omega)}{(E_{31}-2\hbar\omega-i\Gamma_{31})(E_{21}-\hbar\omega-i\Gamma_{21})}\ , \tag{19a}$$

and

$$n_2^{(2)}(z,2\omega)=-N_s\varphi_1(z)\varphi_3(z)\frac{\mathcal{V}_{31}^{(2)}(2\omega)}{E_{31}-2\hbar\omega-i\Gamma_{31}}\ . \tag{19b}$$

For comparison, in the two subband model and single resonance condition ($2\hbar\omega\approx E_{21}$) the expressions for $n_1^{(2)}(z,2\omega)$ and $n_2^{(2)}(z,2\omega)$ can be approximated by

$$n_1^{(2)}(z,2\omega)=N_s\varphi_1(z)\varphi_2(z) \times\frac{\mathcal{V}_{21}^{(1)}(\omega)[\mathcal{V}_{22}^{(1)}(\omega)-\mathcal{V}_{11}^{(1)}(\omega)]}{(E_{21}-2\hbar\omega-i\Gamma_{21})(E_{21}-\hbar\omega-i\Gamma_{21})} \tag{20}$$

and

$$n_2^{(2)}(z,2\omega)=-N_s\varphi_1(z)\varphi_2(z)\frac{\mathcal{V}_{21}^{(2)}(2\omega)}{E_{21}-2\hbar\omega-i\Gamma_{21}}\ . \tag{21}$$

From Eqs. (18)–(21), we find that in the double resonant (single-resonant) structure, calculation of the influence of the DE on the SHG spectrum reduces to the calculation of $\mathcal{V}_{21}^{(1)}(\omega)$, $\mathcal{V}_{32}^{(1)}(\omega)$, and $\mathcal{V}_{13}^{(2)}(2\omega)$ [$\mathcal{V}_{11}^{(1)}(\omega)$, $\mathcal{V}_{22}^{(1)}(\omega)$, $\mathcal{V}_{21}^{(1)}(\omega)$, and $\mathcal{V}_{12}^{(2)}(2\omega)$].

The use of Eqs. (13), (15), and (17) gives the following relation, which determines $\mathcal{V}^{(1)}$:

$$\mathcal{V}^{(1)}(z,\omega)=\mathcal{V}^{\text{ext}}(z,\omega)-\frac{2e^2N_s}{\varepsilon_0\varepsilon_\infty}\int_{-\infty}^{z}dz'\int_{-\infty}^{z'}dz''\sum_k\varphi_k(z'')\varphi_1(z'')\frac{E_{k1}\mathcal{V}_{k1}^{(1)}(\omega)}{E_{k1}^2-(\hbar\omega+i\Gamma_{k1})^2}\ . \tag{22}$$

Taking a matrix element (between the states φ_k and φ_1), we get the set of the equations for $\mathcal{V}_{k1}^{(1)}(\omega)$ identical with that discussed in Refs. 14 and 15. When $\hbar\omega$ is close to E_{k1}, then in summation over k only the resonant term ($k=k'$) can be retained. In this diagonal approximation, $\mathcal{V}_{21}^{(1)}(\omega)$ is given by

$$\mathcal{V}_{21}^{(1)}(\omega)=\mathcal{V}_{21}^{\text{ext}}(\omega)\frac{E_{21}^2-(\hbar\omega+i\Gamma_{21})^2}{\tilde{E}_{21}^2-(\hbar\omega+i\Gamma_{21})^2} \approx\mathcal{V}_{21}^{\text{ext}}(\omega)\frac{E_{21}-\hbar\omega-i\Gamma_{21}}{\tilde{E}_{21}-\hbar\omega-i\Gamma_{21}}\ , \tag{23}$$

where

$$\tilde{E}_{k1}=E_{k1}(1+\alpha_{kk})^{1/2}\ , \tag{24}$$

with

$$\alpha_{kk}=\frac{2e^2N_s}{E_{k1}\varepsilon_0\varepsilon_\infty}\int_{-\infty}^{\infty}dz\left[\int_{-\infty}^{z}dz'\varphi_1(z')\varphi_k(z')\right]^2\ . \tag{25}$$

It is the well-known result, indicating that (in the linear approximation) the DE only shifts the resonant energy from E_{k1} to $\tilde{E}_{k1}$. (It is easy to check that diagonal approximation works well only when $\alpha_{kk}\ll 1$.)

Substituting Eq. (23) in Eq. (22) and taking the matrix element [between the states $\varphi_3(z)$ and $\varphi_2(z)$], we obtain the following equation, which determines $\mathcal{V}_{32}^{(1)}(\omega)$:

$$\mathcal{V}_{32}^{(1)}(\omega)=\mathcal{V}_{32}^{\text{ext}}(\omega)-\frac{2e^2N_s}{\varepsilon_0\varepsilon_\infty}\sum_k\frac{L(3,2;k,1)E_{k1}\mathcal{V}_{k1}^{\text{ext}}(\omega)}{\tilde{E}_{k1}^2-(\hbar\omega+i\Gamma_{k1})^2}\ , \tag{26}$$

where

$$L(i,j;k,l)=\int_{-\infty}^{\infty}dz\left[\int_{-\infty}^{z}dz'\varphi_i(z')\varphi_j(z')\right] \times\left[\int_{-\infty}^{z}dz'\varphi_k(z')\varphi_l(z')\right] \tag{27}$$

is the Coulomb matrix element.[14,15,20] Retaining in Eq. (26) only the near resonant term ($k=2$), we find

$$\mathcal{V}_{32}^{(1)}(\omega)=\mathcal{V}_{32}^{\text{ext}}(\omega)\frac{\tilde{E}_{21}^2-(\hbar\omega+i\Gamma_{21})^2}{\tilde{E}_{21}^2-(\hbar\omega+i\Gamma_{21})^2} \approx\mathcal{V}_{32}^{\text{ext}}(\omega)c(\omega)\ , \tag{28}$$

with

$$\bar{E}_{21}=E_{21}(1+\alpha_{22}-\bar{\alpha}_{32})^{1/2} . \tag{29}$$

where

$$\bar{\alpha}_{32}=\frac{2e^2N_s}{E_{21}\varepsilon_0\varepsilon_\infty}\frac{z_{21}}{z_{32}}L(3,2;2,1) \tag{30}$$

and

$$c(\omega)=\frac{\bar{E}_{21}-\hbar\omega-i\Gamma_{21}}{\tilde{E}_{21}-\hbar\omega-i\Gamma_{21}} . \tag{31}$$

[We assume that $(\alpha_{22}-\bar{\alpha}_{32})\ll 1$.] It is interesting to note that function $c(\omega)$ appears as a result of the coupling between $|1\rangle\rightarrow|2\rangle$ and $|2\rangle\rightarrow|3\rangle$ transitions [more exactly due to the influence of the depolarization field connected with $|1\rangle\rightarrow|2\rangle$ on the matrix element $\mathcal{V}_{23}^{(1)}(\omega)$].

Using Eqs. (14)–(19), (23), (25), and (28), we find easily

$$\mathcal{V}^{(2)}(z,2\omega)=(N_se^2/\varepsilon_0\varepsilon_\infty)\times\int_{-\infty}^{z}dz'\int_{-\infty}^{z'}dz''\varphi_1(z'')\varphi_3(z'')\times[-A(\omega)+B(\omega)\mathcal{V}_{31}^{(2)}(2\omega)] , \tag{32}$$

with

$$A(\omega)=\frac{\mathcal{V}_{21}^{(1)}(\omega)\mathcal{V}_{32}^{(1)}(\omega)}{(E_{31}-2\hbar\omega-i\Gamma_{31})(E_{21}-\hbar\omega-i\Gamma_{21})}$$

$$=\frac{\mathcal{V}_{21}^{\text{ext}}(\omega)\mathcal{V}_{32}^{\text{ext}}(\omega)}{(E_{31}-2\hbar\omega-i\Gamma_{31})(\tilde{E}_{21}-\hbar\omega-i\Gamma_{21})}c(\omega) , \tag{33}$$

where $B(\omega)=1/(E_{31}-2\hbar\omega-\Gamma_{31})$.

Taking the matrix element of Eq. (32), we get

$$\mathcal{V}_{31}^{(2)}(2\omega)=\alpha_{33}A(\omega)(E_{31}/2)/(1+\alpha_{33}B(\omega)E_{31}/2) . \tag{34}$$

Having the necessary matrix elements [Eqs. (23), (26), and (34)], we can calculate $n^{(2)}(z,2\omega)$ defined by Eqs. (18) and (19). After some manipulations, we get

$$n^{(2)}(z,2\omega)=N_s\varphi_1(z)\varphi_3(z)A(\omega)\left[1-\frac{\alpha_{33}B(\omega)(E_{31}/2)}{[1+\alpha_{33}B(\omega)E_{31}/2]}\right]$$
$$\approx N_s\varphi_1(z)\varphi_3(z)A(\omega)(E_{31}-2\hbar\omega-i\Gamma_{31})/(\tilde{E}_{31}-2\hbar\omega-i\Gamma_{31}) . \tag{35}$$

Deriving the above equation, we have used the relation $\tilde{E}_{31}=E_{31}(1+\alpha_{33})^{1/2}\approx E_{31}(1+\alpha_{33}/2)$ assuming that $\alpha_{33}\ll 1$.

Substituting Eq. (35) in Eq. (11) and performing the integration, we find

$$\chi^{(2)}(2\omega)=\frac{-e^3N_s\varepsilon_0^{-1}z_{23}z_{21}z_{31}}{(\tilde{E}_{31}-2\hbar\omega-i\Gamma_{31})(\tilde{E}_{21}-\hbar\omega-i\Gamma_{21})}c(\omega) . \tag{36}$$

It can be easily checked that when we neglect the DE, i.e., when we replace $\tilde{E}_{ij}$ and $\bar{E}_{ij}$ by E_{ij}, then Eq. (36) reduces to the one-electron expression used in the earlier papers.

The second-harmonic-generation spectrum is proportional to

$$|\chi^{(2)}(2\omega)|^2=e^6N_s^2\mathcal{P}\varepsilon_0^{-2}\frac{1}{(\tilde{E}_{31}-2\hbar\omega)^2+\Gamma_{31}^2}\times\frac{1}{(\tilde{E}_{21}-\hbar\omega)^2+\Gamma_{21}^2}C(\omega) , \tag{37}$$

where $C(\omega)=|c(\omega)|^2=[(\bar{E}_{21}-\hbar\omega)^2+\Gamma_{21}^2]/[(\tilde{E}_{21}-\hbar\omega)^2+\Gamma_{21}^2]$ and $\mathcal{P}=(z_{21}z_{31}z_{32})^2$.

The application of Eq. (22) for the two-level system shows that when $2\hbar\omega\approx E_{21}$, the influence of the DE on the $\mathcal{V}^{(1)}$ can be neglected in the first approximation, i.e., $\mathcal{V}_{ij}^{(1)}=\mathcal{V}_{ij}^{\text{ext}}$. In the similar way, like in the case of three-level system, we can show [using Eqs. (14)–(19)] that

$$\mathcal{V}_{21}^{(2)}(2\omega)=\alpha_{22}\bar{A}(\omega)(E_{21}/2)/(1+\alpha_{22}\bar{B}(\omega)E_{21}/2) , \tag{38}$$

where

$$\bar{A}(\omega)=\frac{\mathcal{V}_{21}^{\text{ext}}(\omega)[\mathcal{V}_{22}^{\text{ext}}(\omega)-\mathcal{V}_{11}^{\text{ext}}(\omega)]}{(E_{21}-2\hbar\omega-i\Gamma_{21})(E_{21}-\hbar\omega-i\Gamma_{21})} \tag{39}$$

and

$$\bar{B}(\omega)=\frac{1}{E_{21}-2\hbar\omega-i\Gamma_{21}} . \tag{40}$$

From Eqs. (38)–(40), (21), and (11), we find the following expression for the SHG spectrum in the two subband system:

$$|\chi^{(2)}(2\omega)|^2=e^6N_s^2\varepsilon_0^{-2}z_{21}^4(z_{22}-z_{11})^2\times\frac{1}{(\tilde{E}_{21}-2\hbar\omega)^2+\Gamma_{21}^2}\frac{1}{(E_{21}-\hbar\omega)^2+\Gamma_{21}^2} . \tag{41}$$

The above equation shows that in the single-resonant structure, the DE only shifts the peak position in the SHG spectrum from E_{21} to $\tilde{E}_{21}$. This conclusion is consistent with that reported in Refs. 12 and 13.

III. NUMERICAL RESULTS AND DISCUSSION

In this section, we calculate numerically the SHG spectrum [defined by Eq. (37)] for the near-double-resonant

GaAs asymmetric-step-quantum-well (ASQW) structure shown in inset of Fig. 1(a). (We neglect for simplicity the band-bending effects. They will be briefly discussed at the end of this section.) This asymmetrical structure is experimentally realized by varying the Al composition in $Al_xGa_{1-x}As$ layers during growth. The parameters are as follows: $m^*=0.066m_0$, $\varepsilon_\infty=11.1$, and $N_s=10^{12}\text{cm}^{-2}$. We also use the usually quoted value for $\Gamma_{ij}\equiv\Gamma$ of 5 meV. The total thickness (L) of the ASQW and the thickness of the narrow well (L_N) are assumed to be 125 and 50 Å, respectively. The barrier height (V_b) is 350 meV.

As an illustration, the values of E_{21}, $\bar{E}_{21}$, $\tilde{E}_{21}$, E_{31}, and $\tilde{E}_{31}$ are calculated as a function of the height of the step (V). For comparison, we also calculated the above energies in the presence of the external electrostatic potential eFz (neglecting the carrier screening effect[21]) for the arbitrary value of the potential step, e.g., 110 meV. The results are shown in Fig. 1. From Fig. 1 (see also Refs. 21, 22 and 23), we find that the level separations in ASQW's can be tuned in wide range by changing the height of the step or/and by an external bias field. (The similar dependence of the level separation on the external bias field was reported for asymmetric double-quantum-well systems.[11]) We have also calculated the dependence of the product of the dipolar matrix elements $\mathcal{P}$ on V. Within the potential range 90–130 meV, this dependence is negligible small [$\mathcal{P}(V=90\text{ meV})\approx\mathcal{P}(V=130\text{ meV})\approx2.7\times10^7$ Å^6]. (It is interesting to note that the above value of $\mathcal{P}$ is very close to the value resulting from the optimization procedure[5] for the photon energy $\hbar\omega\approx100$ meV.)

First, we discuss the case of noninteracting electrons. When the electron concentrating is small ($N_s\leq10^{11}$ cm^{-2}), the DE can be neglected and the double-resonance condition has the form $E_{21}=E_{31}/2=\hbar\omega$ [see Eq. (36) or (37)]. Figure 1 shows that in our structure this condition is fulfilled at $V=V_{\text{res}}=115.6$ meV. The peak in the SHG spectrum appears then for $\hbar\omega=\hbar\omega_{\text{res}}=102$ meV. From Fig. 2, we find that the full

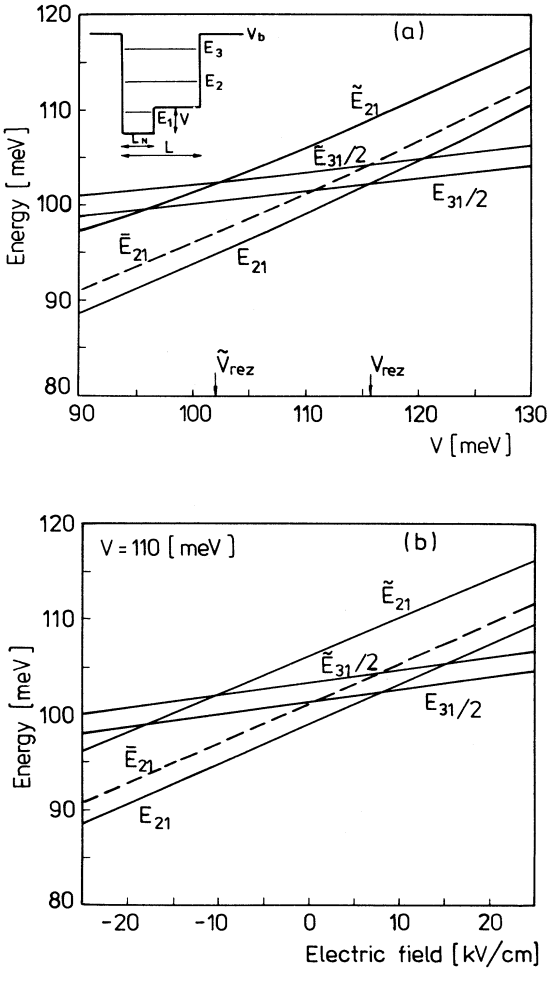

FIG. 1. The energies E_{21}, $E_{31}/2$, $\bar{E}_{21}$, $\tilde{E}_{21}$, and $\tilde{E}_{31}/2$ for a GaAs step QW (shown in inset) with $L=125$ Å, $L_N=50$ Å, and $V_b=350$ meV as a function of (a) the height of the step V and (b) the strength of the electrostatic field F at $V=110$ meV.

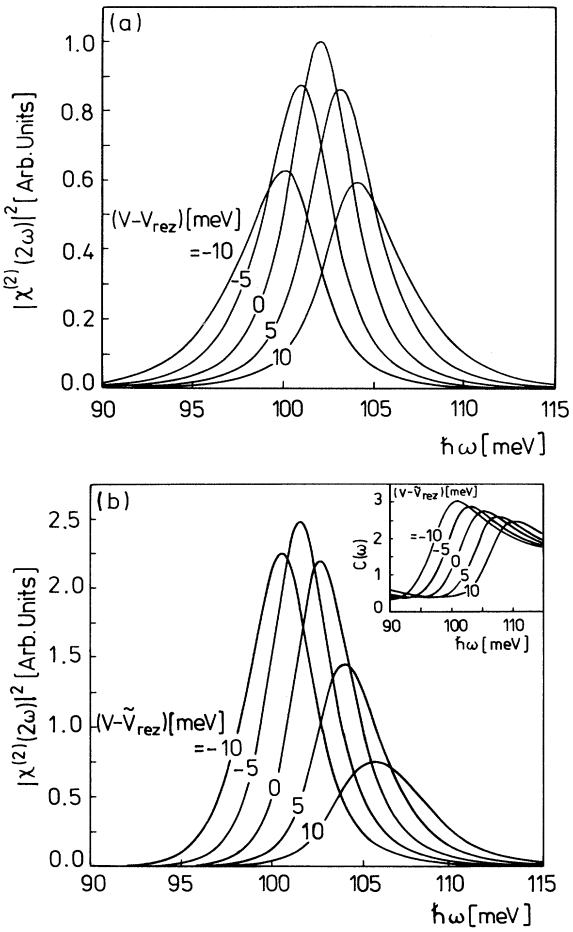

FIG. 2. The dependence of $|\chi^{(2)}(2\omega)|^2$ on the photon energy $\hbar\omega$ for GaAs step QW with $L=125$ Å, $L_N=50$ Å, $V_B=350$ meV, $N_s=10^{12}$ cm^{-2}, and various height of the potential step: (a) the results obtained neglecting the DE and (b) the results obtained taking into account the DE. The unit of $|\chi^{(2)}(2\omega)|^2$ is normalized to the maximum of $|\chi^{(2)}(2\omega)|^2$ calculated in the one-electron approximation at $V=V_{\text{res}}$. The inset shows the dependence of $C(\omega)$ on the photon energy.

width at half maximum (FWHM) is 4.2 meV, i.e., nearly two and half times smaller than the intersubband absorption linewidth ($=2\Gamma$). The deviation of the height of the step from $V_{\rm res}$ leads to the increase (decrease) of the FWHM (the peak value). The peak is shifted from $\hbar\omega_{\rm res}$ to $\hbar\omega^*$, which (for $\Gamma_{21}\simeq\Gamma_{31}$) is very close to $E_{31}/2$.

The situation is more complicated in the systems with high carrier concentration where the DE must be taken into account. When we neglect the coupling between $|1\rangle\rightarrow|2\rangle$ and $|2\rangle\rightarrow|3\rangle$ transitions (the diagonal approximation), the DE can be included by replacing, in the one-electron expression for the SHG coefficient, E_{21} (E_{31}) by $\tilde{E}_{21}$ ($\tilde{E}_{31}$). The double-resonance condition then takes the form $\tilde{E}_{21}=\tilde{E}_{31}/2=\hbar\omega$. From Fig. 2, we find that (in the system with $N_s=10^{12}$ cm^{-2}) the above condition is fulfilled for $V=\tilde{V}_{\rm res}=102.2$ meV and $\hbar\omega=\hbar\tilde{\omega}_{\rm res}=102.3$ meV (if we neglect the band-bending effect). The deviation of V from $\tilde{V}_{\rm res}$ modifies the spectrum in the similar way as in the one-electron case. The coupling between the intersubband transitions (which is controlled by a dimensionless parameter $\bar{\alpha}_{32}$) changes this picture substantially. Calculations show that in the system considered here, $\bar{\alpha}_{32}$ is comparable with α_{22}. Consequently, the function $C(\omega)$, appearing in Eq. (37), rather strongly depends on photon energy when the depolarization shift $\Delta_{21}(=\tilde{E}_{21}-E_{21})$ is comparable with the linewidth (see Fig. 2). In the presence of the coupling, the SHG spectrum has the maximum peak value $|\chi^{(2)}_{\rm max}(2\omega)|^2$ at $V=\tilde{\tilde{V}}_{\rm res}=97.2$ meV for $\hbar\omega=\hbar\tilde{\tilde{\omega}}_{\rm res}=102.2$ meV. It is interesting to note that, in our system, the maximum peak value calculated including the coupling is larger than that obtained in the diagonal (or one-electron) approximation by a factor of 2.5. The FWHM linewidth is then slightly smaller (a few percent) compared to the one-electron case (with $V=V_{\rm res}$). Figure 2(b) shows that a deviation of V from $\tilde{\tilde{V}}_{\rm res}$ leads, like in the one-electron case, to the decrease (increase) of the peak value (linewidth). Consequently, at $V=V_{\rm res}$ ($\equiv\tilde{\tilde{V}}_{\rm res}-18.4$ meV) the peak value (linewidth) is about two times smaller (larger) than that obtained neglecting the DE. The above presented results indicate that in the near-double resonant system, the DE can lead to the substantial enhancement of the maximum peak value.

As it was mentioned in Sec. I, the SHG spectrum is affected not only by the dynamic screening, but also by electrostatic screening effect. In real devices, based on the intersubband transitions, modulation-doped QW's are usually used (in order to lower the ionized impurity-scattering induced line broadening). In such system, the spatial separation between the charged dopand atoms and the free carriers creates the additional potential V_H, which leads to the modification of the energy levels and wave functions of the confined states. This effect was neglected in our calculations. The electrostatic self-consistent potential $V_H(z)$ is the solution of the Poisson equation with the total equilibrium charge distribution determined by the ionized donor doping profile $N_D^+(z)$ and the electron-density distribution $n(z)$.[15,22–24] To estimate the influence of this potential on the SHG spectrum, we assume (following Fishman[20]) that $n(z)$ can be approximated by the electron distribution in the absence of the Coulomb interaction. We have checked numerically that the above approximation works very well due to very fast[2,23] convergence of the self-consistent calculations. We have solved the Schrödinger and Poisson equation, assuming for simplicity that ionized donors are placed adjacent (the spacer thickness $L_S=0$) to the well in the external barriers in regions measuring L_D with constant density $n_D^+=N_S/2L_D$. Obtained in this way, the results show that in the system considered here the band bending affects the SHG spectrum mainly through the modification of the subband separation energies. Consequently, the value of V (and/or F) at which the double-resonance condition is achieved is also modified compared to the case $V_H=0$. Taking, for example, $F=0$ and $L_D=100$ Å, we find that the electrostatic Coulomb interaction shifts $V_{\rm res}$ ($\tilde{V}_{\rm res}$) from 115.6 meV (102.3 meV) to 142.8 meV (132.5 meV) and $\hbar\omega_{\rm res}$ ($\hbar\tilde{\omega}_{\rm res}$) from 102 meV (102.3 meV) to 99.5 meV (96 meV). The parameters $\mathcal{P}$ and $\bar{\alpha}_{23}$ (affecting strongly the peak value of the SHG spectrum) are rather moderately enhanced by the band-bending effect. The results presented above suggest that the main conclusion of the paper, also holds true when the electrostatic Coulomb interaction is included. However, in some situations the influence of the band-bending effect on the SHG spectrum can be more complex. For example, in the case of the modulation-doped multiple-quantum AQW systems with $L_S\geq 50$ Å, we an expect the strong influence of the deep potential minima in the barriers generated by the ionized donors.[24] Unfortunately, the more detailed discussion of this problem is beyond the scope of this paper.

At the end, we note that the approach developed here can be easily extended on the case of the doubly resonant difference frequency mixing or triply resonant third-harmonic generation observed experimentally in Refs. 11 and 10, respectively. These problems will be discussed in future articles.

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[1]M. K. Gurnick and T. A. De Temple, IEEE J. Quantum Electron. **QE-19**, 791 (1983).
[2]Z. Ikonic, V. Milanovic, and D. Tiapkin, IEEE J. Quantum Electron. **QE-25**, 54 (1989).
[3]M. M. Fejer, S. J. B. Yoo, and R. L. Byer, Phys. Rev. Lett. **62**, 1041 (1989).
[4]P. Boucaurd, F. H. Julien, D. D. Yang, J. M. Lourtioz, E. Rosencher, Ph. Bois, and J. Nagle, Appl. Phys. Lett. **57**, 215

(1990).
[5]E. Rosencher and Ph. Bois, Phys. Rev. B **44**, 11 315 (1991).
[6]S. J. B. Yoo, M. M. Fejer, R. L. Byer, and J. S. Harris, Jr., Appl. Phys. Lett. **58**, 1724 (1991).
[7]P. Boucaurd and F. H. Julien, J. Phys. (France) III **1**, 13 (1991).
[8]Z. Chen, M. Li, D. Cui, H. Lu, and G. Yang, Appl. Phys. Lett. **62**, 1502 (1993).
[9]H. C. Chui, E. L. Martinet, G. L. Woods, M. M. Fejer, J. S. Harris, Jr., C. A. Rella, B. I. Richman, and H. A. Schwettman, J. Appl. Phys. **64**, 3365 (1994).
[10]F. Capasso, C. Sirtori, and A. Y. Cho, IEEE J. Quantum Electron. **QE-30**, 1313 (1994), and references therein.
[11]C. Sirtori, F. Capasso, J. Faist, L. N. Pfeffer, and K. W. West, Appl. Phys. Lett. **65**, 445 (1994).
[12]J. N. Heyman, K. Kraig, B. Galdrikian, M. S. Sherwin, K. Campman, P. F. Hopkins, S. Fafard, and A. C. Gossard, Phys. Rev. Lett. **72**, 2183 (1994).
[13]A. Liu and O. Keller, Proc. SPIE **2139**, 342 (1994).
[14]S. J. Allen, Jr., D. C. Tsui, and B. Vinter, Solid State Commun. **20**, 425 (1976).
[15]T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).
[16]A. Liu and O. Keller, Phys. Rev. B **49**, 13 616 (1994).
[17]N. Blombergen, *Nonlinear Optics* (Benjamin, New York, 1965).
[18]A. Yariv, *Quantum Electronics* (Wiley, New York, 1975).
[19]K. Blum, *Density Matrix. Theory and Applications* (Plenum, New York, 1981).
[20]G. Fishman, Phys. Rev. B **27**, 7611 (1983).
[21]F. Y. Huang, J. Li, Lie-Ming Li, and H. Morkoç, Appl. Phys. Lett. **63**, 1669 (1993).
[22]P. F. Yuh and K. L. Wang, J. Appl. Phys. **65**, 4377 (1989).
[23]W. Q. Chen and T. G. Anderson, J. Appl. Phys. **73**, 4484 (1993).
[24]T. Ando and S. Mori, J. Phys. Soc. Jpn. **47**, 1518 (1979).