Influence of the depolarization effect on the nonlinear intersubband absorption spectra of quantum wells

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The effect of the depolarization field on the nonlinear intersubband absorption spectrum of a rectangular quantum well (described by the two-subband model) is discussed. Calculations are performed in the framework of the density-matrix formulation with use of the relaxation-time approximation. Obtained results show that the depolarization effect not only shifts the peak by an amount depending on the intensity of the incident light but also leads to a significant distortion (asymmetry) of the line shape.

Recently there has been a strong interest in the nonlinear optical phenomena connected with the intersubband transitions in semiconductor quantum wells (QW's).[1-9] This interest results both from a fundamental physics point of view as well as from the possible device applications.[8]

The calculation of the electromagnetic response (of the low-dimensional electron gas), with the standard mean-field approaches, consists of two steps.[10,11] First, determination of the final and initial 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[11] 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 nonlinear intersubband absorption, the second step is omitted. It is equivalent (when we work in the RPA) in neglecting the depolarization effect (DE). Some authors[1] try to include the DE, assuming, that like in the case of the linear intersubband absorption, the DE shifts only the resonance frequency by an amount independent on the intensity of the incident radiation. In this paper, we show (employing RPA) that in the case of the nonlinear absorption in typical rectangular QW's, the error induced by this simplified treatment of the DE can be substantial.

Our analysis is based on the density-matrix formulation with relaxation-time approximation. As in previous papers, we consider the two-subband system, assuming that only the ground subband is occupied in the absence of the radiation. For a parabolic conduction band, the energies of these two subbands, E_1 and E_2, can be expressed as

$$E_n(\mathbf{k}_t)=E_n+\hbar^2\mathbf{k}_t^2/2m^*, \quad n=1,2 , \tag{1}$$

where $\mathbf{k}_t$ is the wave vector in the x-y plane, m^* is the effective mass of electrons in the conduction band, and $E_n=E_n(\mathbf{k}_t=0)$.

The corresponding wave functions can be written as

$$\Psi_n(\mathbf{k}_t,\mathbf{r}_t)=\exp(i\mathbf{k}_t\mathbf{r}_t)\chi_n(z) , \tag{2}$$

where $\mathbf{r}_t$ is the position vector in the x-y plane and $\chi_n(z)$ is the envelope wave function in the z direction.

The evolution of the density matrix ρ is given by[12-14]

$$\frac{\partial\rho}{\partial t}=(1/i\hbar)[\mathcal{H}_0+\mathcal{H}'(t),\rho]-R\rho , \tag{3}$$

where $\mathcal{H}_0$ is the Hamiltonian for the unperturbed system, $\mathcal{H}'(t)$ is the effective perturbing Hamiltonian, and R is the relaxation operator.

The equations for the matrix elements of the density matrix [in the representation of (2)] resulting from Eq. (3) can be written in the form[12]

$$\dot\rho_{12}=i(\omega_{21}+i/T_2)\rho_{12}+(i/\hbar)\mathcal{H}'_{12}(t)\Delta\rho , \tag{4a}$$

$$\dot\rho_{21}=-i(\omega_{21}-i/T_2)\rho_{21}-(i/\hbar)\mathcal{H}'_{21}(t)\Delta\rho , \tag{4b}$$

$$(\dot\rho_{11}-\dot\rho_{22})=(2i/\hbar)[\mathcal{H}'_{21}(t)\rho_{12}-\rho_{21}\mathcal{H}'_{12}(t)] -(\Delta\rho-\Delta\rho^{(0)})/T_1 , \tag{4c}$$

where T_2 is the intrasubband relaxation time, T_1 is the intersubband relaxation time, $\mathcal{H}'_{ij}(t)=\langle\chi_i|\mathcal{H}'(t)|\chi_j\rangle$, $(i,j=1,2)$, $\Delta\rho=\rho_{11}-\rho_{22}$, $\Delta_\rho^{(0)}=\rho_{11}^{(1)}-\rho_{22}^{(0)}$, and $\rho^{(0)}$ is the unperturbed density matrix having only diagonal terms. Since the operator $\mathcal{H}'(t)$ is Hermitian, we have the following relations: $\mathcal{H}'_{12}(t)=\mathcal{H}'^*_{21}(t)$ and $\rho_{12}=\rho^*_{21}$.

When external electric field $D(t)$ is applied in the z direction, it modifies the density distribution of electrons, and consequently the Hartree potential. Let $\Delta n(z,t)$ be the change of the distribution due to this field. Then the effective perturbing potential, appearing in Eqs. (3) and (4), can be written in the form[10,11]

$$\mathcal{H}'(t)=eD(t)z-(4\pi e^2/\epsilon)\int_{-\infty}^{z}dz'\int_{-\infty}^{z'}dz''\Delta n(z'',t) , \tag{5}$$

where ϵ is the background dielectric constant.

(In this paper we are restricted 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.)

At this point it is interesting to note that the electrostatic field of the optically generated space-charge shifts the subband energy.[2,8] However, in the case of the typical rectangular QW's this shift is very small and can be neglected in the first approximation.[2,15]

Under usual experimental conditions the wavelength of the light is much larger than the thickness of the system. When the absorption is not strong, which is usually the case, the electric field of the incident radiation can be taken into the form

$$D(t)=\tilde{D}\exp(-i\omega t)+\tilde{D}^{*}\exp(i\omega t)\ . \tag{6}$$

To solve the system of Eqs. (4)–(6) we adopt the rotating-wave approximation[12–14] (RWA) and restrict only to the steady-state responses. In the case considered here the RWA is equivalent to the assumption that the nondiagonal matrix elements of $\rho(t)$ and $\mathcal{H}'(t)$ are in the following forms:

$$\mathcal{H}'_{ij}(t)=\tilde{\mathcal{H}}'_{ij}(\omega)\exp(-i\omega t)+\tilde{\mathcal{H}}'_{ij}(-\omega)\exp(i\omega t)\ , \tag{7}$$

$$\rho_{ij}(t)=\tilde{\rho}_{ij}(\omega)\exp(-i\omega t)+\tilde{\rho}_{ij}(-\omega)\exp(i\omega t)\ , \tag{8}$$

where $\tilde{\rho}_{12}(\pm\omega)$, $\tilde{\rho}_{21}(\pm\omega)$, $\mathcal{H}'_{12}(\pm\omega)$, and $\mathcal{H}'_{21}(\pm\omega)$ are assumed to be time independent. The diagonal terms ρ_{11} and ρ_{22} can also be treated as time independent.

Substituting Eqs. (7) and (8) into the density-matrix equations (4a), (4b), and (4c), and considering only the $\exp(-i\omega t)$ and $\exp(i\omega t)$ terms for ρ_{12} and ρ_{21}, and the time-independent terms ρ_{11} and ρ_{22}, after some manipulations we get

$$\tilde{\rho}_{12}(\omega)=-\Delta\rho\tilde{\mathcal{H}}'_{12}(\omega)[\hbar(\omega_{21}+\omega+i/T_2)]^{-1}\ , \tag{9a}$$

$$\tilde{\rho}_{12}(-\omega)=-\Delta\rho\tilde{\mathcal{H}}'_{12}(-\omega)[\hbar(\omega_{21}-\omega+i/T_2)]^{-1}\ , \tag{9b}$$

$$\tilde{\rho}_{21}(\omega)=-\Delta\rho\tilde{\mathcal{H}}'_{21}(\omega)[\hbar(\omega_{21}-\omega-i/T_2)]^{-1}\ , \tag{9c}$$

$$\tilde{\rho}_{21}(-\omega)=-\Delta\rho\tilde{\mathcal{H}}'_{21}(-\omega)[\hbar(\omega_{21}+\omega-i/T_2)]^{-1}\ , \tag{9d}$$

$$\Delta\rho=\Delta\rho^{(1)}\left[1+\frac{4|\tilde{\mathcal{H}}_{21}(\omega)|^2}{\hbar^2}\frac{T_1}{T_2}\times\frac{2(\omega_{21}^2+\omega^2)}{(\omega_{21}^2-\omega^2)^2+(2\omega T_2^{-1})^2}\right]^{-1}\ , \tag{10}$$

where $\hbar\omega_{21}\equiv E_{21}=E_2-E_1$.

The Fourier component of $\Delta n(t,z)$ with $\exp(-i\omega t)$, time dependence can be written in the form

$$\Delta n(\omega,z)=\partial\sum_{\mathbf{k}_t}[\tilde{\rho}_{12}(\omega)+\tilde{\rho}_{21}(\omega)]\chi_1(z)\chi_2(z)\ . \tag{11}$$

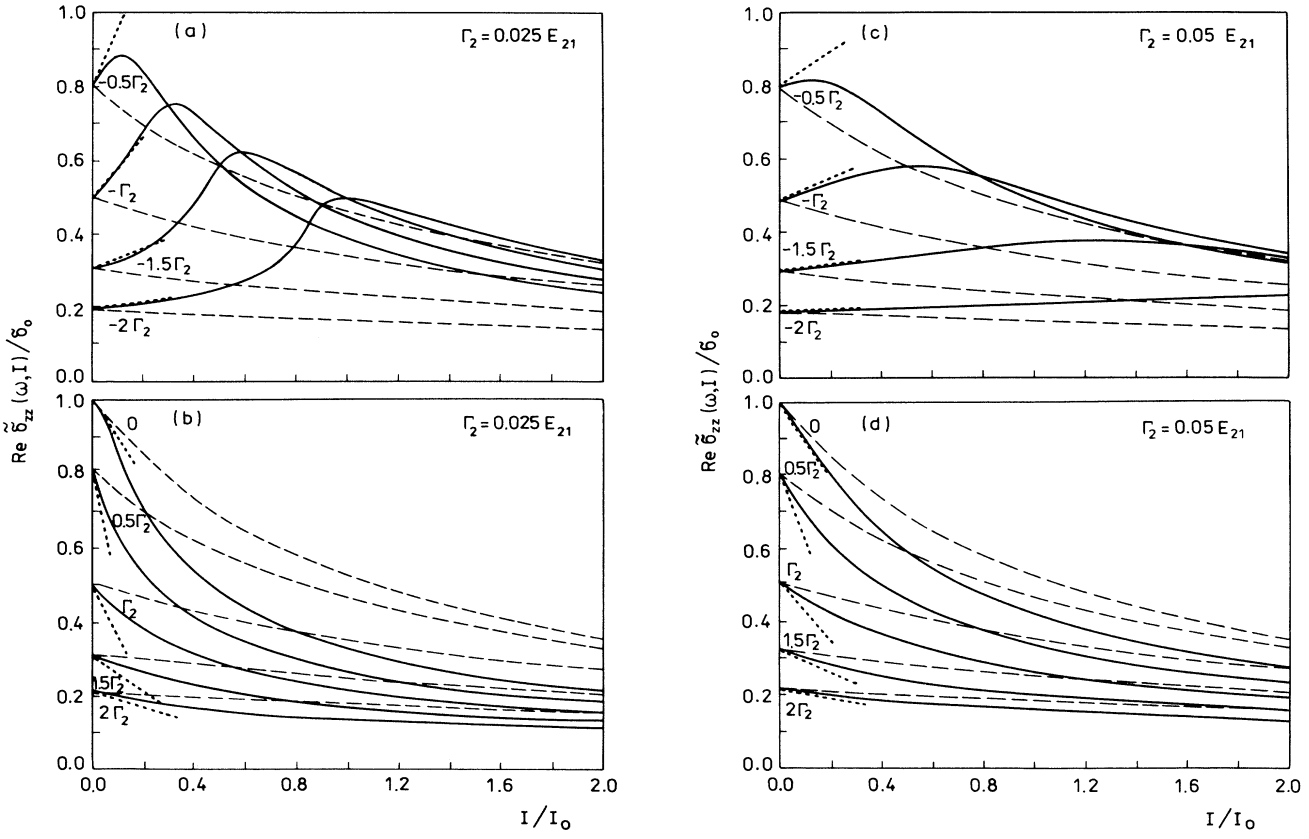

FIG. 1. Dependence of the real part of the nonlinear conductivity $\bar{\sigma}_{zz}(\omega,I)$ on the intensity of the radiation for $\delta=0$, ±0.5, ±1, ±1.5, and $\pm2\Gamma_2$ in GaAs QW with $\alpha_{11}=0.2$ and (a,b) $\Gamma_2=0.025E_{21}$ and (c,d) $\Gamma_2=0.05E_{21}$. The solid lines represent exact results. The dotted lines are obtained with the help of Eq. (20). The dashed lines represent $\alpha_{11}=0$ results but with E_{21} replaced by $\tilde{E}_{21}$.

The Fourier component of the induced current can be calculated from Eq. (11) by the use of the equation of continuity, and is given by

$$j(\omega,z)=-(-e)(-i\omega)\int_{-\infty}^{z}dz'\Delta n(\omega,z)\ . \tag{12}$$

The absorption ($\mathcal{P}$) in a unit area is given by[10,11]

$$\mathcal{P}(\omega,I)=\tfrac{1}{2}\mathrm{Re}\tilde{\sigma}_{zz}(\omega,I)|\tilde{D}|^2\ , \tag{13}$$

where

$$\tilde{\sigma}_{zz}(\omega,I)=(1/\tilde{D})\int_{-\infty}^{\infty}j(\omega,z)$$
$$=\frac{e^2(-i\omega)N_s}{m^*}f_{21}\frac{\mathcal{W}(\omega,I)H_{12}}{E_{21}^2-(\hbar\omega)^2-i2\hbar\omega\Gamma_2} \tag{14}$$

is the nonlinear conductivity and

$$\mathcal{W}(\omega,I)=\left[1+\frac{I}{I_0}\frac{|H_{12}|^2\Gamma_2^2[E_{21}^2+(\hbar\omega)^2]}{[E_{21}^2-(\hbar\omega)^2]^2+(2\hbar\omega\Gamma_2)^2}\right]^{-1}\ . \tag{15}$$

Here N_s is the electron concentration in unit area, $f_{21}=2m^*|\langle\chi_2|z|\chi_1\rangle|^2\omega_{21}/\hbar$, $H_{12}=\tilde{\mathcal{H}}'_{12}(\omega)/e\tilde{D}\langle\chi_1|z|\chi_2\rangle$, $\Gamma_i=\hbar/T_i$, and $I=c\epsilon^{1/2}|\tilde{D}|^2/2\pi$ is the intensity of the incident optical radiation. $I_0=c\epsilon^{1/2}\Gamma_1\Gamma_2/8\pi e^2|\langle\chi_2|z|\chi_1\rangle|^2$ is the saturation intensity.

Taking the matrix element of Eq. (5) we have the following equation, which determines H_{12}:

$$H_{12}\left[1+\frac{\alpha_{11}E_{21}^2}{E_{21}^2-(\hbar\omega)^2-i2\hbar\omega\Gamma_2}\mathcal{W}(\omega,I)\right]=1\ , \tag{16}$$

where

$$\alpha_{11}=\frac{8\pi N_s e^2}{\epsilon E_{21}}\int_{-\infty}^{\infty}dz\left[\int_{-\infty}^{\infty}dz'\chi_2(z')\chi_1(z')\right]^2\ . \tag{17}$$

In typical rectangular GaAs QW with $E_{21}\sim 100$ meV and $N_s\sim 10^{12}$ cm^{-2} (such as those in which the saturation of the intersubband transitions was observed[5]) parameter α_{11} is rather small ~ 0.2.[16,17]

It is easy to check that $\mathcal{W}(\omega,I=0)=1$ and

$$H_{12|_{I=0}}=[E_{21}^2-(\hbar\omega)^2-i2\hbar\omega\Gamma_2]/[\tilde{E}_{21}^2-(\hbar\omega)^2-i2\hbar\omega\Gamma_2]\ , \tag{18}$$

where $\tilde{E}_{21}^2=E_{21}^2(1+\alpha_{11})$.

Substituting Eq. (18) into Eq. (14) we get the well-known result,[10] indicating that (in the two-subband approximation) the DE only shifts the resonant energy from E_{21} to $\tilde{E}_{21}$.

Assume now that α_{11} is vanishingly small (what takes place when $N_s<10^{11}$ cm^{-2}). Then $H_{12}=1$ and the expression for the nonlinear conductivity (14) becomes consistent with that employed in the previous papers[8,15] discussing the third-order nonlinearities in QW's. Moreover, it reduces (near the resonance) to the result obtained by Roan and Chuang,[7] if we put in our formula $\Gamma_1=\Gamma_2$ (= the intrasubband relaxation time). This suggests that the approach used by Roan and Chuang for the description of the electron-scattering effects is not fully correct. It is interesting to note that in QW's with E_{21} much larger than LO-phonon energy $\Gamma_2\gg\Gamma_1$.[9] The above facts are probably the main reason why calculated by Roan and Chuang the saturation intensity is about five times larger than that obtained in experiment.[5]

Consider now the more interesting case when both α_{11} and I are finite. Then, the simple analytical solution of Eq. (16) can be obtained only in the low saturation limit ($I\ll I_0$). When $I\ll I_0$ the expression for $\mathcal{W}(\omega,I)$ can be approximated by

$$\mathcal{W}(\omega,I)=\left[1-\frac{I}{I_0}\frac{|H_{12|_{I=0}}|^2\Gamma_2^2[E_{21}^2+(\hbar\omega)^2]}{[E_{21}^2-(\hbar\omega)^2]^2+(2\hbar\omega\Gamma_2)^2}\right]\ . \tag{19}$$

After some manipulation, we get from Eqs. (19), (16), and (14) the following approximated expression for $\mathrm{Re}\tilde{\sigma}_{zz}(\omega,I)$:

$$\mathrm{Re}\tilde{\sigma}_{zz}(\omega,I)=\sigma_0\frac{\Gamma_2^2}{\delta^2+\Gamma_2^2}\left[1-\frac{I}{I_0}\frac{\Gamma_2^2}{\delta^2+\Gamma_2^2}\times\left[1+\alpha_{11}\frac{\tilde{E}_{21}\delta}{\delta^2+\Gamma_2^2}\right]\right]\ , \tag{20}$$

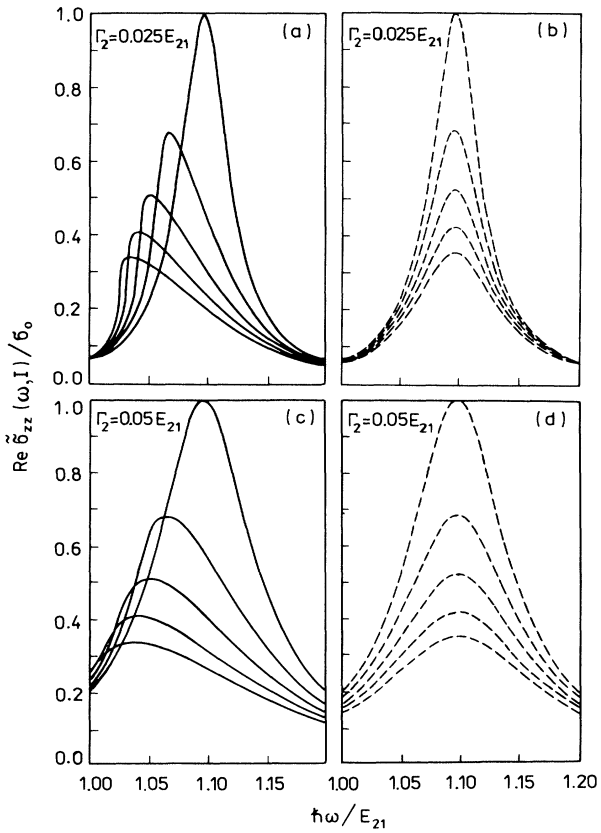

FIG. 2. Dependence of the real part of the nonlinear conductivity $\tilde{\sigma}_{zz}(\omega,I)$ on the photon energy in GaAs QW with $\alpha_{11}=0.2$ and (a,b) $\Gamma_2=0.025E_{21}$ and (c,d) $\Gamma_2=0.05E_{21}$. The intensities of the radiation are 0, 0.5, 1, 1.5, and 2 I_0, from the highest to the lowest peak. The solid lines represent exact results. The dashed lines represent $\alpha_{11}=0$ results, but with E_{21} replaced by $\tilde{E}_{21}$.

where $\sigma_0=N_s e^2 f_{21}\hbar/2m^*\Gamma_2$, $\delta=\hbar\omega-\tilde{E}_{21}$ is detuning. (In obtaining the above equation we have assumed that photon energy $\hbar\omega$ is close to $\tilde{E}_{21}$ and the factor α_{11} is small.) Equation (20) is equivalent to that obtained in our previous paper,[15] with the help of the less-sophisticated method.

At high intensity of the incident light, Eq. (16) can be solved only numerically. We have performed the numerical calculations, taking $\alpha_{11}=0.2$. (This value of the parameter α_{11} is very close to that obtained by Bloss[16] for an 85-Å GaAs QW doped to $N_s=10^{12}$ cm^{-2} with 240-meV barriers of $Al_xGa_{1-x}As$.) The results are presented in Figs. 1 and 2. For comparison we also present results obtained with the help of Eq. (20) as well as results obtained assuming that the DE only shifts the resonant energy from E_{21} to $\tilde{E}_{21}$. From these figures we find that when the intensity of the radiation is comparable with I_0 the shift of the peak induced by the DE decreases with increasing the intensity and the absorption line becomes strongly asymmetric. Consequently, the influence of the resonant screening on the behavior of the nonlinear intersubband absorption (as a function of I) depends not only on α_{11} but also on the sign of δ. When $\delta>0$ then the DE results in the decreasing absorption, while for $\delta<0$ the nonlinear conductivity has a maximum at some value of I. This maximum shifts to higher intensity with increasing δ and is particularly well pronounced in QW's with $I=0$ line broadening ($2\Gamma_2$), much smaller than the depolarization shift ($\sim\alpha_{11}E_{21}/2$).

It is also interesting to note that the range of the intensity for which the approximated formula (20) works well is much larger for $\delta\leq-\Gamma_2$ than for $\delta\geq 0$.

In conclusion, we have shown that in the case of the nonlinear intersubband absorption the simplified treatment of the depolarization effect (assuming only the shift of the resonant energy from E_{21} to $\tilde{E}_{21}$) can introduce substantial errors.

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[1]S. Y. Yuen, Appl. Phys. Lett. **43**, 813 (1983).
[2]D. J. Newson and A. Kurobe, Appl. Phys. Lett. **51**, 1670 (1987).
[3]D. Ahn and S. L. Chuang, J. Appl. Phys. **62**, 3052 (1987).
[4]D. Ahn and S. L. Chuang, IEEE J. Quantum Electron. **QE-23**, 2196 (1987).
[5]F. H. Julien, J.-M. Lourtioz, N. Herschkorn, D. Delacourt, J. P. Pocholle, M. Papuchon, R. Planel, and G. Le Roux, Appl. Phys. Lett. **53**, 116 (1988).
[6]Kelin J. Kuhn, Guita U. Iyengar, and Sinclar Yee, J. Appl. Phys. **70**, 5010 (1991).
[7]E. Roan and S. L. Chuang, J. Appl. Phys. **69**, 3249 (1991).
[8]J. Khurgin and S. Li, Appl. Phys. A **53**, 527 (1991).
[9]D. Walrod, S. Y. Auyang, P. A. Wolff, and M. Sugimoto, Appl. Phys. Lett. **59**, 2932 (1991).
[10]S. J. Allen, Jr., D. C. Tsui, and B. Vinter, Solid State Commun. **20**, 425 (1976).
[11]T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).
[12]N. Blombergen, *Nonlinear Optics* (Benjamin, New York, 1965).
[13]A. Yariv, *Quantum Electronics* (Wiley, New York, 1975).
[14]K. Blum, *Density Matrix Theory and Applications* (Plenum, New York, 1981).
[15]M. Załużny, Appl. Phys. Lett. (to be published).
[16]W. L. Bloss, J. Appl. Phys. **66**, 3639 (1989).
[17]G. Jogai, J. Vac. Sci. Technol. B **9**, 2473 (1991).