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On the Second-Harmonic Generation in Two-Level Quantum-Well Systems

Generacja drugiej harmonicznej w dwupoziomowych studniach kwantowych

1. INTRODUCTION

The second-order nonlinear optical properties of asymmetric quantum wells (AQWs) (connected with the intersubband transitions) have been investigated experimentally and theoretically by several groups [1–3]. In most of the papers the authors neglect the dynamical screening. The effect of this screening on the linear response is well known. In the systems with parallel subbands it leads to a depolarization shift between the intersubband spacing and the intersubband infrared absorption resonance [3, 4].

Recently Heyman et al. [3] have shown experimentally that in AQWs with two subbands the second harmonic generation (SHG) spectrum is also affected by the depolarization effect (DE) i.e. resonance in $\chi^{(2)}(2\omega)$ occurs when $2\hbar\omega$ coincides with the depolarization shifted intersubband energy, and not the bar intersubband spacing. The authors also presented the analytical expression for $\chi^{(2)}(2\omega)$. Unfortunately, the details of the calculations were not given. In this paper we present the detailed derivation of $\chi^{(2)}(2\omega)$ for a two-subband system and show that the expression reported in Ref. [3] is not fully correct.

Our analysis is based on the density matrix formulation in the electric dipole approximation developed in our previous paper [6]

2. DENSITY-MATRIX FORMALISM

We start from the one band effective mass Hamiltonian given by $\hat{H} = \mathbf{p}^2/2m + V_{\rm SCF}(z)$ where $V_{\rm SCF}(z)$ is the self-consistent quantum well potential and m is the effective mass of the electron. The eigenfunction of this Hamiltonian can be written as $| i, \mathbf{k}_{\|}\rangle = \exp(i\mathbf{k}_{\|}\mathbf{r}_{\|})\varphi_i(z)$ where $\mathbf{k}_{\|}$ and $\mathbf{r}_{\|}$ are the wave vector and the position vector in the x-y plane. $\varphi_i(z)$ is the solution of the one dimensional Schrödinger equation $[\hat{p}_z^2/2m + V_{\rm SCF}(z)]\varphi_i = E_i\varphi_i$ where E_i is the minimum energy of the subband.

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

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

where $V \equiv V(z,t)$ is the effective perturbing Hamiltonian, τ_{ii}^{-1} is the relaxation rate from the ith subband, $\tau_{12}^{-1} = \tau_{21}^{-1} (\equiv \tau^{-1})$ is the off-diagonal elastic dephasing rate and $\Delta\rho = \rho - \rho^{(0)}$ where $\rho^{(0)}$ is the unperturbed density matrix. The diagonal element $\rho_{jj}^{(0)} \equiv \rho_{j\mathbf{k}_{\|}j\mathbf{k}_{\|}}^{(0)}$ equals the thermal equilibrium occupation probability of the corresponding state. The equilibrium surface density of the electrons in the jth subband is given by $N_j = 2\sum_{\mathbf{k}_{\|}} \rho_{jj}^{(0)}$.

The electric field $E(t) = \tilde{E}\exp(-i\omega t) + {\rm c.c.}$ of the pumped radiation modifies the density distribution of electrons. The change of the distribution $\Delta n(z,t)$ can be expressed through the density matrix as

$$\Delta n(z,t) = 2\sum_{\mathbf{k}_{\|}} {\rm Tr}\,[\Delta\rho\delta(z - z')]\,. \tag{2}$$

Modification of the carrier distribution leads to the modification of the effective perturbing potential $V(z,t)$. In the electrostatic limit this potential can be written in the form [5, 6]

$$V(z,t) = eE(t)z - \frac{e^2}{\epsilon_0\epsilon}\int_{-\infty}^{z} dz' \int_{-\infty}^{z'} dz''\Delta n(z'',t)\,. \tag{3}$$

where ϵ is the average dielectric constant neglecting any difference between the dielectric properties of the well and the barrier.

As in most of the previous papers we assume that the external perturbation $V^{\text{ext}}(z,t) = eE(t)z$ is small. Then the self-consistent solution of Eqs. (1–3) can be obtained perturbatively by expanding $\Delta\rho$, V and Δn in powers of $\tilde{E}$ as

$$\Delta\rho = \sum_{n>0} \rho^{(n)}\,, \tag{4}$$

$$V = \sum_{n>0} V^{(n)}\,, \tag{5}$$

$$\Delta n = \sum_{n>0} n^{(n)}\,. \tag{6}$$

Substituting Eqs. (4–6) into Eq. (1) and using the usual iterative method we get

$$\frac{\partial \rho_{ij}^{(n)}}{\partial t} = \frac{1}{i\hbar}[\hat{H}, \rho^{(n)}]_{ij} + \frac{1}{i\hbar}\sum_{k=1}^{n}[V^{(k)}, \rho^{(n-k)})]_{ij} - \frac{\rho_{ij}^{(n)}}{\tau_{ij}}\,. \tag{7}$$

The surface electronic polarization of the AQW can also be a series expansion as Eqs. (4–6). We shall limit ourselves to the first two orders, i.e.,

$$\begin{aligned} P_s(t) &= \epsilon_0\chi^{(1)}(\omega)\tilde{E}\exp(-i\omega t) + \\ &+ \epsilon_0\chi^{(2)}(2\omega)\tilde{E}^2\exp(-i2\omega t) + \text{c.c.} + \epsilon_0\chi^{(2)}(0)|\tilde{E}|^2\,, \end{aligned} \tag{8}$$

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

If we neglect, for simplicity, the effects connected with rectification then Eqs. (4–6) can be rewritten in the forms

$$\Delta\rho(t) = \sum_{n=1,2} \rho^{(n)}(\omega_n)\exp(-i\omega_n t) + \text{c.c.}\,, \tag{9}$$

$$V(z,t) = \sum_{n=1,2} V^{(n)}(z,\omega_n)\exp(-i\omega_n t) + \text{c.c.}\,, \tag{10}$$

$$\Delta n(z,t) = \sum_{n=1,2} n^{(n)}(z,\omega_n)\exp(-i\omega_n t) + \text{c.c.}\,, \tag{11}$$

where $\omega_n = n\omega$.

The nth order surface electronic polarization $[P_s^{(n)}(\omega_n)]$ is given by [1]

$$P_s^{(n)}(\omega_n) = \frac{-e}{\epsilon_0} 2 \sum_{\mathbf{k}_\parallel} \mathrm{Tr}\,[\rho^{(n)}(\omega_n) z] \,, \tag{12}$$

From Eqs. (12) and (9) we find that the second-order surface electronic susceptibility can be written as

$$\begin{aligned} \chi^{(2)}(2\omega) &= \frac{-e}{\epsilon_0 \tilde{E}^2} 2 \sum_{\mathbf{k}_\parallel, i, j} \rho_{ij}^{(2)}(2\omega) z_{ji} = \\ &= \frac{-e}{\epsilon_0 \tilde{E}^2} [\tilde{\rho}^{(2)}(2\omega) z_{21} + \bar{\rho}_{11}^{(2)}(2\omega) z_{11} + \bar{\rho}_{22}^{(2)}(2\omega) z_{22}] \,, \end{aligned} \tag{13}$$

where $z_{ij} = z_{ji} = \int_{-\infty}^{\infty} \varphi_i(z) z \varphi_j(z) dz$, $\bar{\rho}_{ij}^{(n)}(\omega_n) = 2\sum_{\mathbf{k}_\parallel} \rho_{ij}^{(n)}(\omega_n)$, and $\tilde{\rho}^{(n)}(\omega_n) = \bar{\rho}_{12}^{(n)}(\omega_n) + \bar{\rho}_{21}^{(n)}(\omega_n)$.

Employing Eqs. (7–11) we find after some manipulations that

$$\bar{\rho}_{ij}^{(n)}(\omega_n) = \frac{1}{\hbar\omega_n - E_{ij} + i\Gamma_{ij}} \sum_{k=1}^{n} [V^{(k)}(\omega_{k,}), \bar{\rho}^{(n-k)}(z, \omega_{n-k})]_{ij} \,, \tag{14}$$

with

$$V_{ij}^{(n)}(\omega_n) = V_{ij}^{\mathrm{ext}}(\omega)\delta_{1n} + \frac{e^2}{\epsilon_0 \epsilon_\infty} \sum_{k,l} L(i,j;k,l) \bar{\rho}_{kl}^{(n)}(\omega_n) \,, \tag{15}$$

where

$$L(i,j;k,l) = \int_{-\infty}^{\infty} dz \left[\int_{-\infty}^{z} dz' \varphi_i(z') \varphi_j(z') \right] \left[\int_{-\infty}^{z} dz' \varphi_k(z') \varphi_l(z') \right] \tag{16}$$

is the Coulomb matrix element, $E_{ij} = E_i - E_j$ and $\Gamma_{ij} = \hbar\tau_{ij}^{-1}$.

One can see that Eqs. (14) and (15) form a set of algebraic equations for $\bar{\rho}_{ij}^{(n)}(\omega_n)$. In the next sections we will solve them and calculate $\chi^{(2)}(2\omega)$.

3. SECOND-HARMONIC SUSCEPTIBILITY

The application of Eq. (14) to $n = 1$ yields

$$\tilde{\rho}_{ij}^{(1)}(\omega) = \frac{N_{ij}V_{ij}^{(1)}(\omega)}{E_{ij} - \hbar\omega - i\Gamma} = \frac{N_{ij}}{E_{ij} - \hbar\omega - i\Gamma}\left[V_{ij}^{\text{ext}}(\omega) + \alpha\frac{E_{21}}{2N_{12}}\tilde{\rho}^{(1)}(\omega)\right], \tag{17}$$

where $\alpha = \frac{2e^2N_{12}}{\epsilon_0\epsilon_\infty E_{21}}L(1,2;1,2)$, $N_{ij} = N_i - N_j$ and $\Gamma = \hbar\tau^{-1}$.

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

$$\tilde{\rho}^{(1)}(\omega) = -\frac{N_{12}V_{12}^{\text{ext}}(\omega)2E_{21}}{\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2}, \tag{18}$$

where $\tilde{E}_{21} = E_{21}(1+\alpha)^{1/2}$ is the depolarization shifted intersubband energy.

Since $\tilde{\rho}_{11}^{(1)}(\omega) = \tilde{\rho}_{22}^{(1)}(\omega) = 0$, Eq. (15) can be rewritten (for $n = 1$) in the form

$$\begin{aligned} V_{ij}^{(1)}(\omega) &= V_{ij}^{\text{ext}}(\omega) + \frac{e^2}{\epsilon_0\epsilon_\infty}L(i,j;1,2)\tilde{\rho}^{(1)}(\omega) \\ &= V_{ij}^{\text{ext}}(\omega)\frac{E_{ij}'^2 - (\hbar\omega + i\Gamma)^2}{\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2}, \end{aligned} \tag{19}$$

where $E_{ij}'^2 = E_{21}^2[1+\delta_{ij}(\alpha - \alpha_i)]$ and $\alpha_i = \frac{2e^2N_{12}}{\epsilon_0\epsilon_\infty E_{21}}\frac{z_{12}}{z_{ii}}L(1,2;i,i)$. (We assume that $z_{ii} \neq 0$.)

Taking $n = 2$ and assuming that $\Gamma_{11} = \Gamma_{22} = \hat{\Gamma}$ we get from Eqs. (14) and (17) the following relations

$$\begin{aligned} \tilde{\rho}_{11}^{(2)}(2\omega) &= \frac{V_{12}^{(1)}(\omega)[\tilde{\rho}_{21}^{(1)}(\omega) - \tilde{\rho}_{12}^{(1)}(\omega)]}{2\hbar\omega + i\bar{\Gamma}} = \\ &= -\frac{V_{12}^{\text{ext}}(\omega)^2N_{12}}{2\hbar\omega + i\bar{\Gamma}}\frac{2(\hbar\omega + i\bar{\Gamma})[E_{21}^2 - (\hbar\omega + i\Gamma)^2]}{[\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2}, \end{aligned} \tag{20}$$

$$\tilde{\rho}_{22}^{(2)}(2\omega) = -\tilde{\rho}_{11}^{(2)}(2\omega), \tag{21}$$

and for $i \neq j$

$$\tilde{\rho}_{ij}^{(2)}(2\omega) = \frac{[V_{ii}^{(1)}(\omega) - V_{jj}^{(1)}(\omega)]\tilde{\rho}_{ij}^{(1)}(\omega) - \alpha_{12}\frac{E_{21}}{2}\tilde{\rho}_{11}^{(2)}(2\omega) - \alpha\frac{E_{21}}{2}\tilde{\rho}^{(2)}(2\omega)}{-E_{ij} + 2\hbar\omega + i\Gamma}. \tag{22}$$

where $\alpha_{12} = \alpha_1 - \alpha_2$.

The last equation leads to the following expression for $\tilde{\rho}^{(2)}(\omega)$

$$\tilde{\rho}^{(2)}(2\omega) = -\frac{E_{21}^2 \alpha_{12} \bar{\rho}_{11}^{(2)}(2\omega)}{\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2} - [V_{22}^{(1)}(\omega) - V_{11}^{(1)}(\omega)] \quad (23)$$
$$\times \frac{[(E_{21} + 2\hbar\omega + i\Gamma)\rho_{21}^{(1)}(\omega) + (E_{21} - 2\hbar\omega - i\Gamma)\rho_{12}^{(1)}(\omega)]}{\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2} .$$

Substituting Eq. (23) into Eq. (13) we get the final expression for the second-harmonic susceptibility

$$\chi^{(2)}(2\omega) = \frac{-e^3 2 N_{12} z_{12}^2}{\epsilon_0} \frac{E_{21}^2 - (\hbar\omega + i\Gamma)^2}{[\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2]^2}$$
$$\times \left\{ (z_{22} - z_{11}) \left[\frac{(2\hbar\omega + i\bar{\Gamma})(\hbar\omega + i\Gamma) + E_{21}^2}{\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2} + \frac{\hbar\omega + i\Gamma}{2\hbar\omega + i\bar{\Gamma}} \right] - \right.$$
$$- z_{12}\alpha_{12} \frac{E_{21}^2}{\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2}$$
$$\left. \left[\frac{(2\hbar\omega + i\bar{\Gamma})(\hbar\omega + i\Gamma) + E_{21}^2}{E_{21}^2 - (\hbar\omega + i\Gamma)^2} \frac{\tilde{E}_{21}^2 - (\hbar\omega + i\Gamma)^2}{\tilde{E}_{21}^2 - (2\hbar\omega + i\Gamma)^2} + \frac{\hbar\omega + i\Gamma}{2\hbar\omega + i\bar{\Gamma}} \right] \right\} . \quad (24)$$

(In obtaining this equation we have used relations (17) and (18))

In limit $\alpha, \alpha_i \to 0$ (i.e., when the DE is neglected) Eq. (24) reduces to that derived by Tsang et al. [7].

For comparison, the expression reported by Heyman et al. [3] is given by

$$\chi^{(2)}(2\omega) = \frac{-e^3 N_{12} z_{21}^2 (z_{22} - z_{11})}{\varepsilon_0}$$
$$\frac{3(E_{21} + i\Gamma)^2[(E_{21} + i\Gamma)^2 - (\hbar\omega)^2]}{[(\tilde{E}_{21} + i\Gamma)^2 - (2\hbar\omega)^2][(\tilde{E}_{21} + i\Gamma)^2 - (\hbar\omega)^2]^2} . \quad (25)$$

Eqs. (24) and (25) have a rather different form. However, the numerical values (for $\left|\chi^{(2)}(2\omega)\right|$) resulting from them do not differ substantially if we take into account the fact that usually $\Gamma_{ij} \ll \tilde{E}_{21}$ and neglect, in our expression, the term containing α_{12}. We would like to note that the correction to the SHG connected with omitting this term can be substantial only when $2\hbar\omega \approx \tilde{E}_{21}$.

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STRESZCZENIE

W niniejszym artykule analizowano od strony teoretycznej wpływ efektu depolaryzacyjnego na kształt widmowy $\chi^{(2)}(2\omega)$ w dwupoziomowych studniach kwantowych. Otrzymane wyniki porównano z danymi literaturowymi.