Local-field study of the optical second-harmonic generation in a symmetric quantum-well structure

Ansheng Liu and Ole Keller
Institute of Physics, University of Aalborg, DK-9220 Aalborg Øst, Denmark
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A nonlocal analysis of the infrared second-harmonic generation associated with intersubband transitions in a symmetric semiconductor quantum-well structure is presented. Taking as a starting point a fundamental self-consistent integral equation for the local field, the *p*-polarized first-harmonic field inside the quantum well is studied. By using the infinite-barrier wave functions and taking into account only the two lowest subbands, analytical expressions are obtained for the local field. The result of the local-field calculation at the first-harmonic frequency, in turn, is used to calculate the *p*-polarized second-harmonic local field. The conversion efficiency of the second-harmonic generation from the quantum well is thus determined. Numerical calculations of the frequency and angular spectra of the second-harmonic intensities are presented for a symmetric GaAs/$Al_xGa_{1-x}As$ quantum well. The numerical results show that strong second-harmonic generation occurs in the vicinity of the resonance frequencies for both the first- and second-harmonic local field inside the quantum well. The influence of the dynamic local-field interaction of the electrons on the optical second-harmonic generation is investigated. It is demonstrated that the dynamic screening can lead to a notable upward shift of the locations of the resonant peaks in the frequency spectra of the second-harmonic conversion coefficient.

I. INTRODUCTION

Recently there has been an increasing interest in both the linear[1–4] and nonlinear[5–12] optical properties of semiconductor quantum-well (QW) structures. This is mainly because of their possible application in areas such as long-wavelength infrared detection, integrated optics, and optical communications. One of the nonlinear optical phenomena, optical second-harmonic generation (SHG) in asymmetric QW structures, has been extensively investigated both theoretically[6,7,11] and experimentally.[5,8,9] In the previous theoretical treatment the frequency-dependent second-harmonic (SH) susceptibilities of built-in asymmetric QW structures, such as graded band-gap QW's, asymmetric coupled QW's,[6] and symmetric QW's with an applied electric field,[7] were calculated on the basis of the electric-dipole interaction Hamiltonian (local approximation). Large second-order nonlinearities associated with the intersubband transitions in the QW were predicted, and it was found that these depend essentially on the geometry of the well.[6] The influence of the applied electric field on the SH susceptibility of the QW was also studied. It was predicted that the second-order nonlinear effects can be enhanced significantly by applying an electric field across a symmetric QW structure.[7] Experimentally, extremely large second-order nonlinear susceptibilities were observed in asymmetric GaAs/$Al_xGa_{1-x}As$ quantum wells in the infrared region near 10.6 μm.[5,8] In the same frequency range, saturation of the SHG in GaAs/$Al_xGa_{1-x}As$ asymmetric QW's has been also reported by the authors of Ref. 9. In a recent paper by Lue, Lo, and Tzeng[12] the optical SHG from unbiased single $Al_xGa_{1-x}As$ QW's with symmetric structures was measured at 1.06 μm. It was found that the contribution of SH waves from the electron gas and dipole sheet in the near-infrared region is negligibly small in comparison to the second-order nonlinear effects of the bulk GaAs.[12]

It is well known that the second-order nonlinear effects are absent in the electric dipole approximation in materials exhibiting inversion symmetry. For a symmetric QW structure, due to the definite parity of the wave functions of the bound states, the nonlinear polarizability for SHG also vanishes in the local limit.[5] Thus, in order to analyze the optical SHG from QW's with symmetric structures, one has to make use of a nonlocal approach. In the present paper we present a theoretical study of the optical SHG in a single symmetric QW based on a microscopic local-field calculation. The basic framework of our theory is the newly established electromagnetic scattering-theory formalism for mesoscopic media which incorporates both the electronic and electromagnetic nonlocalities in a systematic way (for a review of this formalism the reader is referred to Ref. 13). Recently, this formalism also has been used to study the linear optical diamagnetic[14–16] and paramagnetic[17] responses, the photon-drag effect,[18] and the optical SHG (Ref. 19) of metallic quantum wells.

Our paper is organized as follows. In Sec. II, we present a nonlocal treatment of the optical SHG in a single symmetric GaAs/$Al_xGa_{1-x}As$ QW. From the basic integral equation, the local field inside the QW at the fundamental frequency is derived. By using infinite-barrier wave functions, and by taking into account in the analysis only the two lowest levels within the conduction band, analytical expressions are obtained for the local field in the case where this is *p* polarized. The results for the local field at the first-harmonic (FH) frequency are used to calculate the local field inside the QW at the SH frequency. The conversion efficiency of the SHG from the QW in turn is obtained. In Sec. III, we present detailed numerical calculations of the conversion efficiency of the SHG for different parameters such as the fundamental frequency, the angle of incidence, and the impurity-doping concentration. The numerical calculations demonstrate that there exist two pronounced resonance peaks in the frequency spectra of the SH conversion coefficient. These originate from the local-field resonances at the FH and SH frequencies. Finally, by varying the donor concentration of the QW system, the effects of the dynamic screening of the electrons on the resonance frequency of the SH reflection spectrum are investigated.

II. THEORETICAL TREATMENT

A. Local field at the fundamental frequency

Let us consider the situation where a monochromatic plane wave is incident on a symmetric quantum-well structure consisting of a GaAs layer embedded in an $Al_xGa_{1-x}As$ matrix, and let us assume that the QW system exhibits infinitesimal translational invariance parallel to the plane of the $GaAs/Al_xGa_{1-x}As$ boundaries. In a Cartesian *xyz*-coordinate system, the surfaces of the GaAs slab are placed parallel to the *xy* plane, and the boundaries of the slab are located at $z=0$ and $z=d$, as illustrated in Fig. 1(a).

Due to the assumed translational invariance of our system parallel to the well (*xy* plane) all vector and tensor-field components (F) appearing in the analysis have the generic form

$$F(\mathbf{r},t)=F(z)e^{i(\mathbf{q}_{\parallel}\cdot\mathbf{r}-\omega t)} , \tag{1}$$

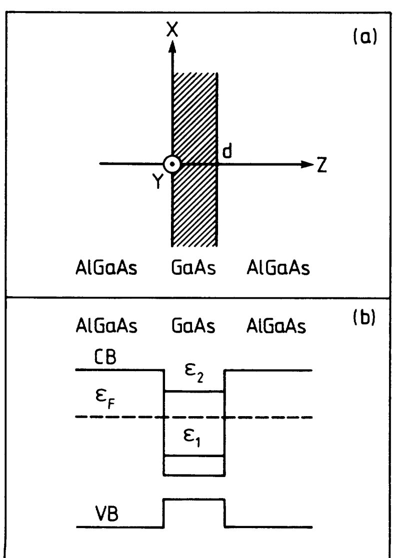

FIG. 1. (a) The Cartesian *xyz* coordinate used in describing the $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ quantum-well structure having a GaAs slab thickness of d. (b) Schematic illustration of a quantum well with two bound states (energies ϵ_1 and ϵ_2) within the conduction band (CB). The Fermi energy (ϵ_F) of the system and the valence band (VB) are also indicated.

where ω and $\mathbf{q}_{\parallel}=(q_{\parallel},0,0)$ denote the angular frequency and the wave-vector projection along the quantum well of the incident electromagnetic wave. Space points are denoted by $\mathbf{r}$ and t is the time. Thus, within the framework of the parametric approximation, the z-dependent local electric field $\mathbf{E}^{(\omega)}(z)$ at the fundamental frequency (ω) satisfies the following wave equation:

$$\overleftrightarrow{\mathcal{L}}^{(\omega)}\cdot\mathbf{E}^{(\omega)}(z)=-i\mu_0\omega\int_{\mathrm{QW}}\overleftrightarrow{\sigma}(z,z')\cdot\mathbf{E}^{(\omega)}(z')dz' . \tag{2}$$

The tensorial operator $\overleftrightarrow{\mathcal{L}}^{(\omega)}$ entering Eq. (2) is given on dyadic form by

$$\overleftrightarrow{\mathcal{L}}^{(\omega)}=\overleftrightarrow{U}\left[\epsilon^B(\omega)\frac{\omega^2}{c^2}-q_{\parallel}^2+\frac{\partial^2}{\partial z^2}\right]-\left[i\mathbf{q}_{\parallel}+\mathbf{e}_z\frac{\partial}{\partial z}\right]\left[i\mathbf{q}_{\parallel}+\mathbf{e}_z\frac{\partial}{\partial z}\right] , \tag{3}$$

where $\overleftrightarrow{U}$ and $\mathbf{e}_z$ are the unit tensor and the unit vector in the z direction, respectively, $\epsilon^B(\omega)$ is the relative dielectric constant of the assumed isotropic and local background medium without the contribution of the conduction electrons in the QW, and c is the speed of light in vacuum. The tensor $\overleftrightarrow{\sigma}(z,z')$ appearing on the right-hand side of Eq. (2) is the appropriate linear nonlocal conductivity response function of the quantum well. By introducing a tensorial Green's function via

$$\overleftrightarrow{\mathcal{L}}^{(\omega)}\cdot\overleftrightarrow{G}^{(\omega)}(z,z')=\overleftrightarrow{U}\delta(z'-z) , \tag{4}$$

where δ is the Dirac δ function, we find that the local electric field $\mathbf{E}^{(\omega)}(z)$ has to be determined from the following self-consistent inhomogeneous integral equation[13]

$$\mathbf{E}^{(\omega)}(z)=\mathbf{E}_0(z)-i\mu_0\omega\times\int\int_{\mathrm{QW}}\overleftrightarrow{G}^{(\omega)}(z,z')\cdot\overleftrightarrow{\sigma}(z',z'')\cdot\mathbf{E}^{(\omega)}(z'')dz''dz' . \tag{5}$$

The incident field (pump field) appearing in the equation above, and satisfying the homogeneous part of Eq. (2), is given by

$$\mathbf{E}_0(z)=\mathbf{E}_0e^{iq_{\perp}z} , \tag{6}$$

where

$$q_{\perp}=\left[\left[\frac{\omega}{c}\right]^2\epsilon^B(\omega)-q_{\parallel}^2\right]^{1/2} \tag{7}$$

is the wave-vector projection perpendicular to the surface. The dyadic Green's function defined in Eq. (4) is given by the following well-known expression:[20]

$$\overleftrightarrow{G}^{(\omega)}(z,z')=\frac{e^{iq_{\perp}|z-z'|}}{2iq_{\perp}}[\mathbf{e}_y\mathbf{e}_y+\Theta(z-z')\mathbf{e}_i\mathbf{e}_i+\Theta(z'-z)\mathbf{e}_r\mathbf{e}_r]+[\epsilon^B(\omega)]^{-1}\left[\frac{c}{\omega}\right]^2\mathbf{e}_z\mathbf{e}_z\delta(z'-z) , \tag{8}$$

where $\mathbf{e}_y=(0,1,0)$, $\mathbf{e}_i=(c/\omega)(q_{\perp},0,-q_{\parallel})/\sqrt{\epsilon^B}$, and

$\mathbf{e}_r=(c/\omega)(-q_\perp,0,-q_\parallel)/\sqrt{\epsilon^B}$ are the relevant unit vectors and Θ is the Heaviside unit step function.

In the present work we restrict ourselves to a study of the nonlinear optical processes associated with the intersubband transitions in the quantum well. In order to focus our attention on the conceptual physical aspects we incorporate in the analysis only the two lowest bound states within the conduction band. Of these one is located above and the other below the Fermi level [see Fig. 1(b)]. Thus, within the framework of the random-phase approximation and in the long-wavelength ($q_\parallel \to 0$) and low-temperature ($T\to 0$) limits, the nonlocal linear conductivity tensor $\overleftrightarrow{\sigma}(z,z')$ takes a diagonalized form, and the diagonal elements are given by[21]

$$\sigma_{xx}(z,z')=\sigma_{yy}(z,z')=\frac{ie^2}{\pi\hbar^2\omega}\frac{(\epsilon_2-\epsilon_1)(\epsilon_F-\epsilon_1)^2}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2}\phi(z)\phi(z') , \tag{9}$$

$$\sigma_{zz}(z,z')=\frac{ie^2}{2\pi m^*\omega}\frac{(\epsilon_2-\epsilon_1)(\epsilon_F-\epsilon_1)}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2}\Phi(z)\Phi(z') , \tag{10}$$

with

$$\phi(u)=\Psi_1(u)\Psi_2(u) , \quad u=z,z' \tag{11}$$

$$\Phi(u)=\Psi_1(u)\frac{d\Psi_2(u)}{du}-\Psi_2(u)\frac{d\Psi_1(u)}{du} , \quad u=z,z' . \tag{12}$$

In the above equations $\Psi_n(z)$ $(n=1,2)$ denotes the (real) wave functions of the two energy eigenstates, ϵ_n $(n=1,2)$ are the corresponding eigenenergies, ϵ_F is the Fermi energy of the system, τ is the relaxation time of the electrons associated with the intersubband transition between state ϵ_1 and state ϵ_2, and finally e and m^* are the charge and effective mass of the electrons, respectively. To keep the calculation of the local field inside the quantum well at an analytical level, we shall make use of infinite-barrier wave functions and eigenenergies, viz.,

$$\Psi_n(z)=\left[\frac{2}{d}\right]^{1/2}\sin\left[\frac{n\pi z}{d}\right] , \quad n=1,2 \tag{13}$$

with

$$\epsilon_n=\frac{n^2\hbar^2\pi^2}{2m^*d^2} , \quad n=1,2 \tag{14}$$

where d denotes the width of the quantum well.

Due to the fact that the z and z' dependence of the linear nonlocal conductivity tensor takes a separable form, it is realized that exact solutions for the local electric field $\mathbf{E}^{(\omega)}(z)$ can be obtained.[17] In the following we limit ourselves to the case where the fundamental field is p polarized, i.e., $\mathbf{E}^{(\omega)}=(E_x^{(\omega)},0,E_z^{(\omega)})$. We do so because the local-field effects to be studied are by far most significant for p-polarized incident fields, these being accompanied by appreciable charge oscillations across the well. By inserting into Eq. (5) the explicit expressions for the conductivity tensor given in Eqs. (9) and (10), one obtains the following results for the FH local field *inside* the QW ($0<z<d$):

$$E_x^{(\omega)}(z)=E_{0x}(z)+F_{xx}(z)N_x+F_{xz}(z)N_z , \tag{15}$$

$$E_z^{(\omega)}(z)=E_{0z}(z)+F_{zx}(z)N_x+F_{zz}(z)N_z , \tag{16}$$

where

$$F_{xx}(z)=a^{(\omega)}\int_{\mathrm{QW}}G_{xx}^{(\omega)}(z,z')\phi(z')dz' , \tag{17}$$

$$F_{xz}(z)=b^{(\omega)}\int_{\mathrm{QW}}G_{xz}^{(\omega)}(z,z')\Phi(z')dz' , \tag{18}$$

$$F_{zx}(z)=a^{(\omega)}\int_{\mathrm{QW}}G_{zx}^{(\omega)}(z,z')\phi(z')dz' , \tag{19}$$

$$F_{zz}(z)=b^{(\omega)}\int_{\mathrm{QW}}G_{zz}^{(\omega)}(z,z')\Phi(z')dz' , \tag{20}$$

with

$$a^{(\omega)}=\frac{\mu_0e^2}{\pi\hbar^2}\frac{(\epsilon_2-\epsilon_1)(\epsilon_F-\epsilon_1)^2}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2} , \tag{21}$$

and

$$b^{(\omega)}=\frac{\mu_0e^2}{2\pi m^*}\frac{(\epsilon_2-\epsilon_1)(\epsilon_F-\epsilon_1)}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2} . \tag{22}$$

By inserting into Eqs. (17)–(20) the explicit expression for the Green's function in Eq. (8), one obtains, after some tedious calculations,

$$F_{xx}(z)=\frac{a^{(\omega)}c^2q_\perp^2d}{2\epsilon^B\omega^2}\left[\frac{2\cos\left[\frac{3\pi z}{d}\right]}{9\pi^2-(q_\perp d)^2}-\frac{2\cos\left[\frac{\pi z}{d}\right]}{\pi^2-(q_\perp d)^2}+\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right](e^{iq_\perp(d-z)}-e^{iq_\perp z})\right] , \tag{23}$$

$$F_{xz}(z)=\frac{3\pi^2b^{(\omega)}c^2q_\parallel}{2i\epsilon^Bd\omega^2}\left[\frac{2\cos\left[\frac{3\pi z}{d}\right]}{9\pi^2-(q_\perp d)^2}-\frac{2\cos\left[\frac{\pi z}{d}\right]}{\pi^2-(q_\perp d)^2}+\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right](e^{iq_\perp(d-z)}-e^{iq_\perp z})\right] , \tag{24}$$

$$F_{zx}(z)=\frac{a^{(\omega)}c^2q_\parallel q_\perp d}{2\epsilon^B\omega^2}\left\{\frac{2\pi}{iq_\perp d}\left[\frac{3\sin\left[\frac{3\pi z}{d}\right]}{9\pi^2-(q_\perp d)^2}-\frac{\sin\left[\frac{\pi z}{d}\right]}{\pi^2-(q_\perp d)^2}\right]+\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right](e^{iq_\perp(d-z)}+e^{iq_\perp z})\right\} , \tag{25}$$

$$F_{zz}(z)=\frac{\pi b^{(\omega)}c^2}{\epsilon^B d^2\omega^2}\left[\sin\left[\frac{3\pi z}{d}\right]-3\sin\left[\frac{\pi z}{d}\right]\right]+\frac{3\pi^2 b^{(\omega)}c^2q_\parallel^2}{i\epsilon^B q_\perp d\omega^2}\left\{\frac{2q_\perp d}{3i\pi}\left[\frac{\sin\left[\frac{3\pi z}{d}\right]}{9\pi^2-(q_\perp d)^2}-\frac{3\sin\left[\frac{\pi z}{d}\right]}{\pi^2-(q_\perp d)^2}\right]\right.$$
$$\left.+\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right](e^{iq_\perp(d-z)}+e^{iq_\perp z})\right\}. \tag{26}$$

To obtain the final expressions for the local field inside the QW one has to calculate the two so-far undetermined constants

$$N_x=\int_{\mathrm{QW}}\phi(z'')E_x^{(\omega)}(z'')dz'' \tag{27}$$

and

$$N_z=\int_{\mathrm{QW}}\Phi(z'')E_z^{(\omega)}(z'')dz'' . \tag{28}$$

By multiplying both sides of Eqs. (15) and (16) by $\phi(z)$ and $\Phi(z)$, respectively, and integrating the two resulting equations over z across the QW, one obtains, after a tedious but straightforward calculation

$$N_x=E_0\frac{S_x(1-a_{22}^{(\omega)})+S_z a_{12}^{(\omega)}}{(1-a_{11}^{(\omega)})(1-a_{22}^{(\omega)})-a_{12}^{(\omega)}a_{21}^{(\omega)}} , \tag{29}$$

$$N_z=E_0\frac{S_x a_{21}^{(\omega)}+S_z(1-a_{11}^{(\omega)})}{(1-a_{11}^{(\omega)})(1-a_{22}^{(\omega)})-a_{12}^{(\omega)}a_{21}^{(\omega)}} , \tag{30}$$

where

$$a_{11}^{(\omega)}=-\frac{a^{(\omega)}c^2q_\perp^2 d}{2\epsilon^B\omega^2}\left[\left[\frac{1}{9\pi^2-(q_\perp d)^2}+\frac{1}{\pi^2-(q_\perp d)^2}\right]+2iq_\perp d(1+e^{iq_\perp d})\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right]^2\right] , \tag{31}$$

$$a_{12}^{(\omega)}=-a_{21}^{(\omega)}\frac{b^{(\omega)}}{a^{(\omega)}}=-\frac{3\pi^2 b^{(\omega)}c^2 q_\parallel}{2i\epsilon^B d\omega^2}\left[\left[\frac{1}{9\pi^2-(q_\perp d)^2}+\frac{1}{\pi^2-(q_\perp d)^2}\right]\right.$$
$$\left.+2iq_\perp d(1+e^{iq_\perp d})\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right]^2\right] , \tag{32}$$

$$a_{22}^{(\omega)}=\frac{5\pi^2 b^{(\omega)}c^2}{\epsilon^B d^3\omega^2}+\frac{3\pi^3 b^{(\omega)}c^2q_\parallel^2}{i\epsilon^B q_\perp d^2\omega^2}\left[\frac{q_\perp d}{3i\pi}\left[\frac{1}{9\pi^2-(q_\perp d)^2}+\frac{9}{\pi^2-(q_\perp d)^2}\right]+6\pi(1+e^{iq_\perp d})\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right]^2\right] , \tag{33}$$

$$S_x=iq_\perp d\cos\theta(1+e^{iq_\perp d})\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right] , \tag{34}$$

$$S_z=-\frac{3\pi^2}{d}\sin\theta(1+e^{iq_\perp d})\left[\frac{1}{9\pi^2-(q_\perp d)^2}-\frac{1}{\pi^2-(q_\perp d)^2}\right] , \tag{35}$$

E_0 and θ being the amplitude of the incident field and the angle of incidence, respectively. From Eqs. (29) and (30), one notes that the local field inside the QW is proportional to the amplitude of the incident field, as it must be. In addition, Eqs. (29) and (30) show that the local field inside the QW is resonantly enhanced when the common denominator of N_x and N_z approaches zero.

B. Local field at the second-harmonic frequency

The wave equation describing the local electric field $\mathbf{E}^{(\Omega)}(z)$ at the SH frequency (Ω) is

$$\overleftrightarrow{\mathcal{L}}^{(\Omega)}\cdot\mathbf{E}^{(\Omega)}(z)=-i\mu_0\Omega\mathbf{J}^{\mathrm{NL}}(z)$$
$$-i\mu_0\Omega\int_{\mathrm{QW}}\overleftrightarrow{\sigma}(z,z')\cdot\mathbf{E}^{(\Omega)}(z')dz' , \tag{36}$$

where the dyadic operator $\overleftrightarrow{\mathcal{L}}^{(\Omega)}$ is obtained by replacing ω and $\mathbf{q}_\parallel$ in Eq. (3) with $\Omega=2\omega$ and $\mathbf{Q}_\parallel=2\mathbf{q}_\parallel$, respectively. The forced nonlinear current density $\mathbf{J}^{\rm NL}$ is related to the FH local electric field by the following constitutive equation:

$$\mathbf{J}^{\rm NL}(z)=\int\int_{\rm QW}\overleftrightarrow{\Sigma}(z,z',z''):\mathbf{E}^{(\omega)}(z'')\mathbf{E}^{(\omega)}(z')dz''dz' \ , \tag{37}$$

where $\overleftrightarrow{\Sigma}$ is the second-harmonic nonlocal conductivity response function of the quantum well. Since in this work we are mainly interested in the p-polarized SH wave, in writing Eq. (37) we have neglected the contribution to the second-harmonic current density from the bulk nonlinear susceptibility. This is justified because the III-V compound semiconductor has only a nonzero local second-harmonic susceptibility $\chi^{(2)}_{14}$, and a p-polarized incident light thus generates only a s-polarized SH wave.[12] Within the same approximations as those adopted to obtain the linear conductivity tensor given by Eqs. (9) and (10), the nonvanishing elements of the nonlocal nonlinear conductivity tensor are given by[22]

$$\Sigma_{xxz}(z,z',z'')=\Sigma_{yyz}(z,z',z'')=\frac{ie^3}{2\pi m^*\omega^2}\frac{(\epsilon_F-\epsilon_1)(\omega+i/\tau)}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2}\times\phi(z)\delta(z'-z)\Phi(z'') \ , \tag{38}$$

$$\Sigma_{zxx}(z,z',z'')=\Sigma_{zyy}(z,z',z'')=\frac{e^3}{4\pi i m^*\omega^2}\frac{(\epsilon_F-\epsilon_1)(2\omega+i/\tau)}{[\hbar(2\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2}\times\Phi(z)\phi(z')\delta(z''-z') \ , \tag{39}$$

$$\Sigma_{zzz}(z,z',z'')=\Sigma_{xxz}(z,z',z'')+\Sigma_{zxx}(z,z',z'') \ . \tag{40}$$

By inserting Eqs. (38)–(40) into Eq. (37), one immediately finds

$$J_x^{\rm NL}(z)=\frac{ie^3N_z}{2\pi m^*\omega^2}\frac{(\epsilon_F-\epsilon_1)(\omega+i/\tau)}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2}\phi(z)E_x^{(\omega)}(z) \tag{41}$$

and

$$J_z^{\rm NL}(z)=\frac{e^3\Phi(z)}{4\pi i m^*\omega^2}\frac{(\epsilon_F-\epsilon_1)(2\omega+i/\tau)}{[\hbar(2\omega+i/\tau)]^2-(\epsilon_2-\epsilon_2)^2}\int_{\rm QW}\phi(z')\{[E_x^{(\omega)}(z')]^2+[E_z^{(\omega)}(z')]^2\}dz' + \frac{ie^3N_z}{2\pi m^*\omega^2}\frac{(\epsilon_F-\epsilon_1)(\omega+i/\tau)}{[\hbar(\omega+i/\tau)]^2-(\epsilon_2-\epsilon_1)^2}\phi(z)E_z^{(\omega)}(z) \ . \tag{42}$$

By use of the electromagnetic scattering-theory formalism it readily follows, cf. the treatment of the local-field problem at the fundamental frequency in the preceding subsection, that the local field inside the QW at the SH frequency is governed by

$$\mathbf{E}^{(\Omega)}(z)=\mathbf{E}_B(z)-i\mu_0\Omega\int\int_{\rm QW}\overleftrightarrow{G}^{(\Omega)}(z,z')\cdot\overleftrightarrow{\sigma}(z',z'')\cdot\mathbf{E}^{(\Omega)}(z'')\times dz''dz' \ , \tag{43}$$

where

$$\mathbf{E}_B(z)=-i\mu_0\Omega\int_{\rm QW}\overleftrightarrow{G}^{(\Omega)}(z,z')\cdot\mathbf{J}^{\rm NL}(z')dz' \tag{44}$$

is the so-called background field, originating in the driven part of the nonlinear current density of the QW, and $\overleftrightarrow{G}^{(\Omega)}$ is the Green's function describing the propagation properties of the electromagnetic field at the SH frequency.

By taking advantage of the fact that the FH and SH local fields inside the QW satisfy the same type of integral equation [see Eqs. (5) and (43)], it is realized that the SH local field readily can be obtained by replacing ω and $q_\parallel$ in Eq. (7) and Eqs. (23)–(26) with Ω and $Q_\parallel$ and by substituting N_x and N_z in Eqs. (15) and (16) with the following two new numbers:

$$N_x'=\frac{S_x'(1-a_{22}^{(\Omega)})+S_z'a_{12}^{(\Omega)}}{(1-a_{11}^{(\Omega)})(1-a_{22}^{(\Omega)})-a_{12}^{(\Omega)}a_{21}^{(\Omega)}} \ , \tag{45}$$

and

$$N_z'=\frac{S_x'a_{21}^{(\Omega)}+S_z'(1-a_{11}^{(\Omega)})}{(1-a_{11}^{(\Omega)})(1-a_{22}^{(\Omega)})-a_{12}^{(\Omega)}a_{21}^{(\Omega)}} \ . \tag{46}$$

In the equations above $a_{11}^{(\Omega)}$, $a_{12}^{(\Omega)}$, $a_{21}^{(\Omega)}$, and $a_{22}^{(\Omega)}$ can be obtained simply by doubling ω and $q_\parallel$ in Eqs. (31)–(33). S_x' and S_z' are related to the background field via

$$S_x'=\int_{\rm QW}\phi(z)E_{Bx}(z)dz \tag{47}$$

and

$$S_z'=\int_{\rm QW}\Phi(z)E_{Bz}(z)dz \ . \tag{48}$$

C. Second-harmonic field outside the quantum well

Once the local field inside the QW has been obtained, one can easily obtain the field distribution outside the QW. In the following we shall calculate the SH field generated in the half space $z<0$. To this end we recall Eq. (43). By inserting the Green's function into Eqs. (43) and (44), and by letting the observation point z be located outside the QW in the half space $z<0$, one finds that the SH field takes the following form:

$$\mathbf{E}^{(<)}(z)=\left(1,0,\frac{Q_\parallel}{Q_\perp}\right)E_x^{(<)}(0)e^{-iQ_\perp z} \ , \tag{49}$$

where

$$Q_\perp = \left[\left[\frac{\Omega}{c} \right]^2 \epsilon^B(\Omega) - Q_\parallel^2 \right]^{1/2} \tag{50}$$

is the z component of the wave vector of the propagating SH wave outside the QW. The x component of the amplitude of the SH field is given by

$$E_x^{(<)}(0) = -\frac{1}{2\epsilon_0 \epsilon^B \Omega} \int_{\mathrm{QW}} e^{iQ_\perp z'} [Q_\perp J_x^{\mathrm{NL}}(z') + Q_\parallel J_z^{\mathrm{NL}}(z')] dz' + \frac{c^2(1+e^{iQ_\perp d})}{2\epsilon^B \Omega^2 d} \times [a^{(\Omega)}(Q_\perp d)^2 N_x' - i3\pi^2 b^{(\Omega)} Q_\parallel N_z'] \left[\frac{1}{9\pi^2 - (Q_\perp d)^2} - \frac{1}{\pi^2 - (Q_\perp d)^2} \right], \tag{51}$$

where ϵ_0 is the vacuum permittivity.

The intensity of the optical SH wave is calculated by the time-averaged Poynting vector $\mathbf{S}^{(\Omega)} = \mathrm{Re}[\mathbf{E}^{(\Omega)}(\mathbf{r}) \times \mathbf{H}^{(\Omega)}(\mathbf{r})^*]/2$, using the relation $\mathbf{H}^{(\Omega)}(\mathbf{r}) = (1/i\mu_0\Omega)\nabla \times \mathbf{E}^{(\Omega)}(\mathbf{r})$. With the help of Eq. (49), one finds that the SH intensity $I^{(\Omega)}$ is given by

$$I^{(\Omega)} = \tfrac{1}{2}\epsilon_0 c [\epsilon^B(\Omega)]^{1/2} |E^{(<)}(0)|^2 = \frac{\epsilon_0 [\epsilon^B(\Omega)]^{3/2} \Omega^2}{2cQ_\perp^2} |E_x^{(<)}(0)|^2 , \tag{52}$$

so that the conversion efficiency (the SH energy reflection coefficient) of the optical second-harmonic generation can be written as

$$\eta = \frac{I^{(\Omega)}}{[I^{(\omega)}]^2} = \frac{2}{\epsilon_0 c} \left[\frac{\Omega}{cQ_\perp} \right]^2 \frac{[\epsilon^B(\Omega)]^{3/2}}{\epsilon^B(\omega)} \frac{|E_x^{(<)}(0)|^2}{|E_0|^4} . \tag{53}$$

III. NUMERICAL CALCULATIONS AND DISCUSSION

In this section we present various numerical calculations of the conversion efficiency of the optical second-harmonic generation from a single symmetric GaAs/$\mathrm{Al}_x\mathrm{Ga}_{1-x}$As quantum well. The parameters used in the calculation for this structure are $d = 100$ Å, $m^* = 0.0665 m_e$,[23] m_e being the mass of the free electrons. The dielectric constant of the background medium is taken to be

$$\epsilon^B(\omega) = \epsilon_\infty \frac{\omega^2 - \omega_{\mathrm{LO}}^2}{\omega^2 - \omega_{\mathrm{TO}}^2} , \tag{54}$$

with $\epsilon_\infty = 11.1$, $\hbar\omega_{\mathrm{LO}} = 36.7$ meV, and $\hbar\omega_{\mathrm{TO}} = 33.6$ meV.[24] For simplicity, we have neglected the small collision frequency of the optical phonons in Eq. (54). For the relaxation time of the electrons associated with the intersubband transition between the two lowest bound states, we have chosen $\hbar/\tau = 5.0$ meV. The Fermi energy of the QW system is determined as is usually done[17] from the charge neutrality condition. Assuming that, by use of the modulation doping technique, the barrier layers of the QW structure are uniformly Si doped with a doping concentration of N_D, and that the donors in the QW are fully ionized, we find that the Fermi energy is determined by

$$\epsilon_F = \epsilon_1 + \frac{\pi\hbar^2}{m^*} N_D D , \tag{55}$$

where D denotes the total thickness of the doped layers. Note that in obtaining Eq. (55), we have assumed that only the lowest level ϵ_1 is occupied. In the calculations we have taken $D = 100$ Å.

In Fig. 2 is shown the SH energy reflection coefficient η as a function of the normalized fundamental frequency, ω/ω_{21}, for three different angles of incidence, i.e., $\theta = 20°$, 40°, and 60°. The characteristic frequency ω_{21} is related to the energy separation between the two subbands by $\omega_{21} = (\epsilon_2 - \epsilon_1)/\hbar = 2.58 \times 10^{14}\ \mathrm{s}^{-1}$. A donor concentration of $N_D = 2.0 \times 10^{18}\ \mathrm{cm}^{-3}$ was used. It appears from Fig. 2 that the SH energy reflection spectrum exhibits two pronounced resonance peaks in the frequency range of $0.4\omega_{21} \sim 1.4\omega_{21}$. One is located somewhat above the characteristic frequency ω_{21}. The other lies a little above the half of the frequency ω_{21}. The two resonance peaks stem from the local-field resonances inside the QW. The one having the highest frequency originates in the FH local-field resonance, and the other one is due to the SH local-field resonance. Note that the FH local-field resonance does not appear at the exact frequency of ω_{21}, whereas it occurs at a frequency somewhat higher than ω_{21}. This is because of the dynamic local-field interaction of the electrons in the QW.[17,21] An exhaustive and heuristic discussion of the general role of local-field effects in mesoscopic systems, as well as a detailed

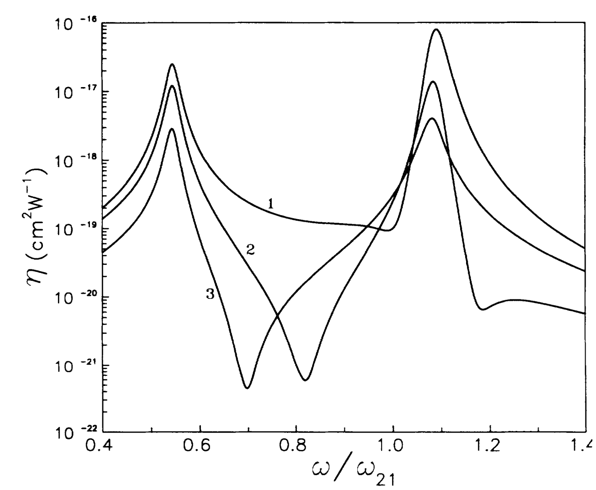

FIG. 2. The SH energy reflection coefficient, η, as a function of the normalized fundamental frequency, ω/ω_{21}, for three angles of incidence, viz., 60° (1), 40° (2), and 20° (3).

analysis of the inherent physics, is given in Ref. 13. In the present context the origin of the local field resonances can be explained in simple terms as follows. Written in vectorial form Eqs. (15) and (16) become $\mathbf{E}^{(\omega)}=\mathbf{E}_0^{(\omega)}+\overleftrightarrow{F}\cdot\mathbf{N}$, where $\overleftrightarrow{F}$ is a two times two matrix (with elements F_{xx}, F_{xz}, F_{zx}, and F_{zz}) and $\mathbf{N}=(N_x,N_z)$. Subjected to the optical field the wave function (and hence the current density) of the two-level quantum well oscillates between the wave functions Ψ_n $(n=1,2)$ of the lower and upper eigenstates. The oscillations are driven not only by the external field ($\mathbf{E}_0^{(\omega)}$), but also by the field ($\overleftrightarrow{F}\cdot\mathbf{N}$) created by the current flow accompanying the change in the wave functions. In mesoscopic systems this field ($\overleftrightarrow{F}\cdot\mathbf{N}$) can be comparable to the external field and is, hence, not negligible.[13] In dynamic local-field calculations the correction of the microscopic current density flows stemming from the radiative reaction of the field generated by these currents on the current themselves are thus considered [cf. the radiative reaction of the field accompanying a moving electron (or an oscillating electric dipole) on the motion of the particle]. A local-field resonance is expected when the current flows are self-sustaining. To obtain a self-sustaining flow, the local field $\mathbf{E}^{(\omega)}$ must be finite in the absence of the external field $\mathbf{E}_0^{(\omega)}$. This requires that $\mathbf{N}$ approaches infinity, and hence that the common denominator of Eqs. (29) and (30) is essentially zero (not completely because irreversible dampings hidden in τ^{-1} are present). Taking as a function of ω, the local-field resonance will not be located at ω_{21} in general (ω_{21} enters the denominator of $\mathbf{N}$ but does not give rise to a pronounced minimum in this denominator). The SH local-field resonance possesses the same character as that of the FH local-field resonance. One also notices from Fig. 2 that the SH energy reflection spectrum does not exhibit any resonant behavior at the two frequencies $\omega=\omega_{21}$ and $\frac{1}{2}\omega_{21}$, although the second-harmonic conductivity response function is resonantly enhanced at these frequencies [see Eqs. (38) and (39)]. The lack of this kind of simple resonance is due to the fact that the conversion efficiency of the SHG is mainly determined by the local field inside the QW. By a comparison in Fig. 2 of the SH energy reflection spectra for different angles of incidence, one sees that although the resonant peak locations are almost independent of the angle of incidence, the magnitude of the conversion efficiency depends strongly on this angle. In addition, it appears from Fig. 2 that there exists a notable minimum in the SH energy reflection spectra for $\theta=20°$ and $40°$ and that the minimum position is displaced when changing the angle of incidence. This is so because for a given angle of incidence, at a certain frequency this angle equals the Brewster angle of the QW structure.

To investigate the influence of the dynamic interaction of the electrons on the optical SHG in the QW structure, we have calculated the SH energy reflection spectra for different doping concentrations. In Fig. 3, we present the conversion efficiency η as a function of the normalized first-harmonic frequency, ω/ω_{21}, for five different values of the donor concentrations, namely, $N_D=0.5\times10^{18}$, 1.0×10^{18}, 1.5×10^{18}, 2.0×10^{18}, and 2.5×10^{18} cm^{-3}. An angle of incidence of 60° was taken in all cases. It appears from Fig. 3 that the resonance frequencies in the SH energy reflection spectra decrease when the doping concentration is decreased. This is to be expected since the current density oscillation of the quantum well decreases in magnitude when less carriers are available to contribute. For a smaller current flow the field stemming from this current in turn becomes less and, hence, smaller relative to the external field. Altogether, the local-field effect decreases in importance when the doping is reduced and displacements of the peaks towards ω_{21} and $\omega_{21}/2$ emerge. The magnitude of the conversion efficiency of the SHG also increases with increasing donor density.

In passing, we should stress that the local-field resonance frequency is strongly affected not only by the dynamic screening, but also by the electrostatic screening effects of the electrons[25] when the electron density in the QW is sufficiently high. Hence, the direct Coulomb in-

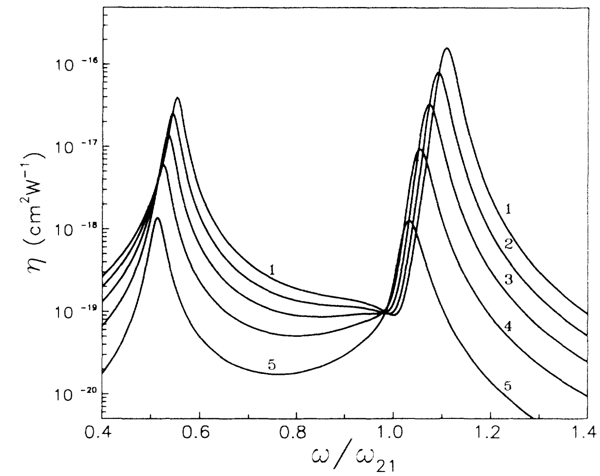

FIG. 3. The SH energy reflection coefficient η as a function of the normalized fundamental frequency, ω/ω_{21}, for five values of donor concentrations (in 10^{18} cm^{-3}), namely, 2.5 (1), 2.0 (2), 1.5 (3), 1.0 (4), and 0.5 (5).

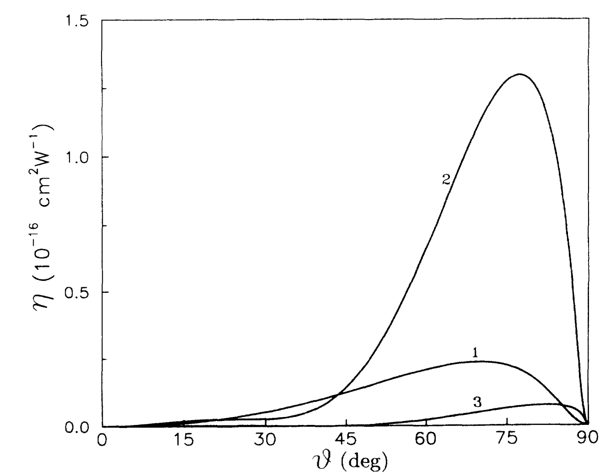

FIG. 4. The SH energy reflection coefficient η as a function of the angle of incidence, θ, for the following three fundamental frequencies: $0.55\omega_{21}$ (1), $1.10\omega_{21}$ (2), and $1.17\omega_{21}$ (3).

teraction and the exchange interaction of the electrons[25] modify the wave functions and the corresponding eigenenergies of the electronic system in the absence of the optical field. This modification leads to an additional shift of the local-field resonance frequency. The electrostatic screening effects are neglected in the present work. However, in order to make a quantitative comparison between theory and experiment, it is in general necessary to take into account both electrostatic and dynamic screening effects in the QW system. We shall deal with this problem in a forthcoming paper.

In Fig. 4 is shown the angular spectra of the SH energy reflection coefficient for three different frequencies, i.e., $\omega/\omega_{21}=0.55$, 1.10, and 1.17. A donor concentration of $N_D=2.0\times10^{18}$ cm^{-3} was used. One sees from Fig. 4 that the angular spectra of the SH energy reflection coefficient show their usual behavior, i.e., the SH reflection coefficient is very small at near-normal incidence and at the large angles of incidence near 90°. In between these limits an often pronounced maximum of the SH energy reflection coefficient occurs. It also appears from Fig. 4 that a change of the fundamental frequency results in a significant effect on the peak height and some effect on the peak location. The maximum is especially pronounced when the frequency of light lies in the vicinity of the local-field resonance. Although the frequency position of the local-field resonance in general depends on the angle of incidence, this dependence usually is weak (cf. the results of Fig. 2, and the discussions in Refs. 13, 18, and 19).

Before closing the present paper, we would like to stress that the upward frequency shifts of the peak locations of the SH energy reflection spectra treated in this work originate solely in the local field mechanism. It is known, however, that, e.g., band-filling effects[26–29] can lead to blueshifts of resonance peaks. In the band-filling mechanism absorption of light leads to a change in the occupation probabilities of electrons and holes in the conduction and valence bands. This change blocks a number of electronic transitions in the quantum well and thus effectively gives rise to an increase in the position of the electronic resonance. Band filling is known to play a role for third-order nonlinearities (Kerr effect, etc.) near interband transitions. In the present calculation the band-filling effect was not incorporated, and to our knowledge such an effect has never been taken into account in the analysis of the optical second-harmonic generation associated with intersubband transitions in quantum wells. It is also known that local heating of a material by the laser beam itself can cause a change in the refractive index, and hence a shift in the position of the various resonance lines.[4,29] Although thermal-index changes might play a role experimentally in some circumstances, these have not been considered in this paper.

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