Second-order optical susceptibility of biased quantum wells in the interband regime

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We develop a theory within the effective-mass approximation of the second-order susceptibility of a quantum well subject to an applied electric field. We show that close to the midgap of the structure it is possible within that approximation to reduce the normally derived triple sums over subbands to a simple sum over all the optically allowed transitions. We have demonstrated that in realistic structures, higher-energy transitions play a strong role for quantitative results. This strong influence of the higher-energy transitions on the second-order susceptibility has the consequence that it is not generally possible to define a quantum-well susceptibility independent of the structure in which the well is inserted. Numerical results indicate that the quantum wells should not lead to spectacular enhancements of the second-harmonic-generation susceptibility.

INTRODUCTION

In an ever increasing body of work, the nonlinear properties of intersubband transitions (ISBT's) in quantum wells (QW's) have been explored.[1–6] Strong second-order optical susceptibilities have shown, due to large asymmetry of bound states within a given band, huge optical dipole moments and double resonance effects. Inversion symmetry is usually broken by using asymmetric coupled quantum wells,[5] growing asymmetric composition gradients of Al (Ref. 7), or biasing a symmetric QW electrically.[4] Previous works have concentrated on processes whose energies are limited by the energy difference between two subbands in the conduction band, i.e., mostly dealing with midinfrared radiations. Therefore, the emphasis has recently been put on phenomena that should permit us to achieve frequency conversion in a wider range of frequencies, especially in the near infrared region where optical integration could be really completed. This means considering band-to-band transitions. In his pioneering work, Khurgin suggested that large second-order susceptibility in the near infrared could be obtained in asymmetric QW's by using virtual ISBT's.[3] Among the different sources of asymmetry, electric-field biasing of quantum wells is of particular interest both from a fundamental and application point of view. As far as the fundamental aspects are concerned, it is a way to study the influence of a growing asymmetry. For applications, the possibility to monitor the second-harmonic-generation coefficient with a bias opens the way to new devices, such as, e.g., electrically controlled quasiphase match.[3]

The purposes of this paper are the following. We shall present a compact and very illustrative formulation of the second-order susceptibility near midband gap in asymmetric quantum wells. In particular, we will use a sum rule to get rid of the unnecessary intersubband transitions, which complexify the work of Khurgin.[3] The obligation of having at least two subbands to obtain second-order effects is, for instance, lifted. Moreover, the contribution of levels in the continuum above the barriers, which was neglected in the previous works,[3,8–11] is shown to significantly reduce the nonlinearity relative to the contribution of confined levels. The analysis of our simple expression will give us a way to study the influence of different parameters on second-harmonic-generation coefficients and, thus, optimize the structures.

THEORY

We shall focus on a two-band Kane semiconductor model.[12] We shall not consider here light holes since their calculated contribution happens to be negligible. This stems from the fact that the ratio of the light hole and electron effective masses is very close to the conduction- to valence-band offset ratio.[3,13] Besides, in the frame of the two-band approximation, the transitions between light holes and heavy holes have vanishing optical matrix elements so we shall not consider those in the following. We also neglect excitonic effects. This is justified considering that the excitonic contribution to the nonlinearity is narrowly peaked[11] around excitonic resonances, while for applications some detuning is necessary to avoid linear absorption at the fundamental and second-harmonic frequencies.

The wave functions of the bound states in a QW for such a two-band system are then

$$\begin{aligned} |e_n,\mathbf{k}_\parallel\rangle &= e_n(z)u_c(\mathbf{r})e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\ , \\ |\mathrm{hh}_m,\mathbf{k}_\parallel\rangle &= \mathrm{hh}_m(z)u_v(\mathbf{r})e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\ , \end{aligned} \tag{1}$$

where u_c and u_v are the periodic parts of the Bloch functions in the conduction and valence bands, respectively, at $k=0$, and $\mathbf{k}_\parallel$ and $\mathbf{r}_\parallel$ are the wave vector and the position in the xy plane, respectively.

The electron $e_n(z)$ and heavy hole $\mathrm{hh}_m(z)$ envelope wave functions are solutions of the one-dimensional Schrödinger equations in the growth direction z:

$$\begin{aligned} H_e e_n(z) &= e_n e_n(z)\ , \\ H_{\mathrm{hh}}\mathrm{hh}_m(z) &= \mathrm{hh}_m \mathrm{hh}_m(z)\ , \end{aligned} \tag{2}$$

where e_n (respectively, hh_m) is the transverse energy of the nth conduction subband (respectively, mth valence subband) and the z parts of the Hamiltonian are

$$H_e=\frac{p_z^2}{2m_e}+V_e(z)\ ,$$
$$H_{\mathrm{hh}}=\frac{p_z^2}{2m_{\mathrm{hh}}}+V_h(z)\ . \tag{3}$$

Here $V_e(z)$ [respectively, $V_h(z)$] represents the profile of the conduction-band potential (respectively, valence-band potential). The eigenenergies of the Schrödinger equation in the effective-mass Hamiltonian approximation are given by

$$E(e_n,\mathbf{k}_\parallel)=e_n+\frac{\hbar^2\mathbf{k}_\parallel^2}{2m_e}+E_g\ ,$$
$$E(\mathrm{hh}_m,\mathbf{k}_\parallel)=-\mathrm{hh}_m-\frac{\hbar^2\mathbf{k}_\parallel^2}{2m_{\mathrm{hh}}}\ , \tag{4}$$

where m_e and m_{hh} are the conduction and heavy-hole band effective masses and E_g the band gap of the bulk well material; the second terms represent the kinetic energies of motion parallel to the interfaces. The zero energy has been taken at the top of the heavy-hole valence band.

Let us write

$$\hbar\omega_{ij}(\mathbf{k}_\parallel)=E_i-E_j \tag{5}$$

as the difference between the energies of the ith and jth subband states of whatever bands are concerned. We take the dipolar interaction Hamiltonian as

$$W_D(t)=\frac{q}{2}\mathbf{E}\cdot\mathbf{r}e^{i\omega t}+\mathrm{c.c.} \tag{6}$$

The electronic second-order susceptibility $\chi_{ijk}(2\omega)$ of the material is defined by

$$P_i(2\omega)=\varepsilon_0\sum_{jk}\chi_{ijk}(2\omega)E_jE_k\ , \tag{7}$$

where $P_i(2\omega)$ is the second-harmonic polarization along the i direction, ε_0 is the vacuum permittivity, and E_j and E_k are components of two electric fields at frequency ω along the j and k directions, respectively.

The second-order susceptibility of such a two-band system can be derived by using the density-matrix formalism as[14,15]

$$\chi_{ijk}(2\omega)=-\frac{q^3}{\varepsilon_0 V}\sum_{n,m,l}\frac{r^i_{nm}r^i_{ml}r^k_{ln}}{(2\hbar\omega+\hbar\omega_{mn}-i\hbar\gamma_{mn})(\hbar\omega+\hbar\omega_{ln}-i\hbar\gamma_{ln})}(N_l-N_n)$$
$$-\frac{r^i_{nm}r^j_{ln}r^k_{ml}}{(2\hbar\omega+\hbar\omega_{mn}-i\hbar\gamma_{mn})(\hbar\omega+\hbar\omega_{ml}-i\hbar\gamma_{ml})}(N_m-N_l)+j\leftrightarrow k\ , \tag{8}$$

where r^i_{nm} is the position matrix element for direction i, N_l is the lth subband state occupation, and the different line broadenings γ_{ln} have been included.

Keeping the gauge, and allocating the levels between the two bands, one can compute the second-order susceptibility for the bulk

$$\chi_{ijk}(2\omega)=-\frac{q^3}{\varepsilon_0 V}\sum_{\mathbf{k}}N_v(\mathbf{k})r^i_{vc}[(r^j_{vv}-r^j_{cc})r^k_{vc}+j\leftrightarrow k]$$
$$\times\left[\frac{1}{(2\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)}-\frac{1}{(2\hbar\omega+\hbar\omega_{cv}-i\hbar\gamma)(\hbar\omega+\hbar\omega_{cv}-i\hbar\gamma)}\right]\ , \tag{9}$$

where the index c is for the electrons of the conduction band and the index v for the heavy holes of the valence band. In this approach, the second-order susceptibility stems from the different symmetry of the conduction and valence Bloch states, i.e., the differences between r_{vv} and r_{cc} terms.

The result is different in QW's. If we apply the result of (8) to a quantum well, taking the scalar potential interaction and considering the conduction band empty and the valence band completely filled, we obtain

$$\chi_{ijk}(2\omega)=-\frac{q^3}{\varepsilon_0 V}\sum_{\mathbf{k}_\parallel}N_v(\mathbf{k}_\parallel)\sum_{c,v,m}\left[\frac{r^i_{mc}r^j_{vm}r^k_{cv}}{(2\hbar\omega+\hbar\omega_{cm}-i\hbar\gamma_{cm})(\hbar\omega+\hbar\omega_{cv}-i\hbar\gamma_{cv})}+\frac{r^i_{cm}r^j_{mv}r^k_{vc}}{(2\hbar\omega+\hbar\omega_{mc}-i\hbar\gamma_{mc})(\hbar\omega+\hbar\omega_{vc}-i\hbar\gamma_{vc})}\right]$$
$$-\left[\frac{r^i_{mv}r^j_{cm}r^k_{vc}}{(2\hbar\omega+\hbar\omega_{vm}-i\hbar\gamma_{vm})(\hbar\omega+\hbar\omega_{vc}-i\hbar\gamma_{vc})}+\frac{r^i_{vm}r^j_{mc}r^k_{cv}}{(2\hbar\omega+\hbar\omega_{mv}-i\hbar\gamma_{mv})(\hbar\omega+\hbar\omega_{cv}-i\hbar\gamma_{cv})}\right]+j\leftrightarrow k\ , \tag{10}$$

where $N_v(k_\parallel)=2$ is the number of electrons in each level of the valence band. Note that now the indices c and v vary in the conduction and valence band, respectively, while m varies in both bands.

Assuming that the light propagates in the y direction and using the polarization selection rules, we can easily derive

$$\chi_{xzx}(2\omega)=+\frac{q^3}{\varepsilon_0 V}\sum_{\mathbf{k}_\parallel,v}N_v(\mathbf{k}_\parallel)\sum_{c,c'}\left[\frac{x_{c'v}z_{cc'}x_{vc}}{(2\hbar\omega-\hbar\omega_{c'v}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)}\right]-\sum_{c,v'}\left[\frac{x_{cv'}z_{v'v}x_{vc}}{(2\hbar\omega-\hbar\omega_{cv'}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)}\right], \tag{11}$$

where we have neglected the nonresonant terms [that is the $(\omega+\omega_{cv})$ and $(2\omega+\omega_{cv})$ terms] and have considered the same linewidth γ for the different transitions. The $\chi^{(2)}$ tensor has also χ_{zxx}-type components, but we do not consider them because they are not resonant at midgap.[3]

We now explicitly consider the midgap frequency region. For $\hbar\omega\approx E_g$, we can perform a first-order development in power of $\omega_{cc'}$ and $\omega_{v'v}$, respectively, in the two parentheses, according to $\omega_{cv}=\omega_{cv'}+\omega_{v'v}=\omega_{c'v}+\omega_{cc'}$ and using the fact that $\omega,\ \omega_{cv}\gg\omega_{cc'},\ \omega_{v'v}$.

A swap of the indices gives the following expression:

$$\chi_{xzx}(2\omega)=+\frac{q^3}{\varepsilon_0 V}\sum_{\mathbf{k}_\parallel}\sum_{v,c}\left[\frac{x_{cv}\mu_{vc}N_v(\mathbf{k}_\parallel)}{(2\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)}\right]-\sum_{c,v}\left[\frac{x_{cv}\mu_{vc}N_v(\mathbf{k}_\parallel)}{(2\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma)}\right], \tag{12}$$

where

$$\mu_{vc}=\sum_{c'}z_{c'c}x_{vc'}=\sum_{v'}z_{vv'}x_{v'c}=\langle \mathrm{hh}_v|z|e_c\rangle\frac{P}{m_0\omega_g}\ . \tag{13}$$

P is the volume valence to conduction-band momentum matrix element, and here we have again applied the effective-mass approximation, so that the matrix elements are written as the product of envelope functions and the Bloch bulk wave functions at the center of the Brillouin zone. Further simplification has been achieved by using the completeness of each of the two-band envelope wave-function bases. Thus, the two terms in the parentheses of Eq. (12) are equal so that the optical second-order susceptibility is equal to zero (Ref. 3) to the first order in $\omega_{cc'}$ and $\omega_{v'v}$.

One may now continue the development to the second order and obtain

$$\chi_{xzx}(2\omega)=\frac{q^3}{\varepsilon_0 V}\left[\sum_{\mathbf{k}_\parallel,v,c'}N_V(\mathbf{k}_\parallel)\frac{x_{c'v}\nu_{c'v}}{(2\hbar\omega-\hbar\omega_{c'v}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{c'v}-i\hbar\gamma)^2}\right.$$
$$\left.-\sum_{\mathbf{k}_\parallel,c,v'}N_{v'}(\mathbf{k}_\parallel)\frac{x_{cv'}\nu_{cv'}}{(2\hbar\omega-\hbar\omega_{cv'}-i\hbar\gamma)(\hbar\omega-\hbar\omega_{cv'}-i\hbar\gamma)^2}\right], \tag{14}$$

where

$$\nu_{c'v}=\sum_c z_{cc'}x_{vc}\hbar\omega_{cc'}=-\frac{P}{m_0\omega_g}\frac{i\hbar}{m_{c'}}\langle \mathrm{hh}_v|P^z|e_{c'}\rangle\ ,$$
$$\nu_{cv'}=\sum_v z_{v'v}x_{vc}\hbar\omega_{v'v}=+\frac{P}{m_0\omega_g}\frac{i\hbar}{m_{v'}}\langle \mathrm{hh}_{v'}|P^z|e_c\rangle\ , \tag{15}$$

have been computed using the hermiticity of H_e (respectively, H_{hh}), the commutator of H_e and z (respectively, H_{hh} and z), and the sum rules over the two bases. Here we have assumed that the matrix elements are independent of the wave vector $\mathbf{k}_\parallel$, as are the energy separations between subbands. We then perform the integration, keeping only the terms that are resonant near midgap

$$\chi_{xzx}(2\omega)=\frac{q^3P^2}{\pi\varepsilon_0 m_0^2 L(\hbar\omega)^2(\hbar\omega_g)^2}$$
$$\times\sum_{cv}i\hbar\langle e_c|\mathrm{hh}_v\rangle\langle \mathrm{hh}_v|P^z|e_c\rangle$$
$$\times\ln\left[\frac{\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma}{2\hbar\omega-\hbar\omega_{cv}-i\hbar\gamma}\right], \tag{16}$$

where from now on the sum is only over transitions between subband extrema, i.e.,

$$\hbar\omega_{ij}=\hbar\omega_{ij}(\mathbf{k}_\parallel=0)\ .$$

The terms no longer cancel each other. The formulation of Eq. (16) avoids a triple sum in Eq. (14) and assures a reliable convergence of a reasonable number of (mainly continuum) levels in the numerical calculations. In our formulation, the need for asymmetry is manifest in the second matrix element $\langle \mathrm{hh}_v|P^z|e_c\rangle$, which would be zero for allowed interband transitions in the symmetric case. According to this expression, the second-order susceptibility near midgap has its physical origin in the transition between a ground electron of the conduction band and a ground heavy hole of the valence band. Therefore, on the contrary to Khurgin, who uses virtual transitions within one band, a QW with only one subband in each band can be of use. The sum rules effectively include all virtual transitions, also to the continuum states.

If we consider a two-band model with only one subband in each band contributing to the second-order susceptibility, we derive the very simple expression

$$\chi_{xzx}(2\omega)\approx\frac{q^3P^2i\hbar}{\pi\varepsilon_0 m_0^2 L(\hbar\omega)^2(\hbar\omega_g)^2}\langle e_1|\mathrm{hh}_1\rangle\langle \mathrm{hh}_1|P^z|e_1\rangle \times\ln\left[\frac{\hbar\omega-\hbar\omega_{e_1\mathrm{hh}_1}-i\hbar\gamma}{2\hbar\omega-\hbar\omega_{e_1\mathrm{hh}_1}-i\hbar\gamma}\right] . \quad (17)$$

RESULTS

In order to obtain quantitative results, we first need the wave functions in order to evaluate the relevant matrix elements. These are found by solving the Schrödinger Eqs. (2) and (3) numerically for a potential representing a single quantum well between two large barriers subjected to an external electric field F. We are interested in studying the effect of the QW on the nonlinear susceptibility. As we shall see, the logarithmic dependence on frequency in expression (16) is quite weak, so transitions between states above the well give an important contribution to the susceptibility even for frequencies close to and below the lowest transition $\omega_{e_1\mathrm{hh}_1}/2$. *Rigorously speaking, this means that the polarization depends not only on the well but on the whole structure.* On the other side, under applied bias, the barriers contribute an effective second-order nonlinearity, which comes from odd-order nonlinear coefficients (the adopted model does not take into account the quadratic susceptibility of the bulk). To distinguish between the two contributions and to obtain a convenient operational definition of the susceptibility due only to the quantum well, we recall that the physically meaningful entity, from a microscopical point of view, is the total induced dipole. The polarization is then obtained by dividing by the interaction volume, which in our case is given by the product between the area of the sample and the total length of the system well barriers (L in the preceding expressions). To compare the contributions of the well and of the box, we assume that the induced dipole *per unit area* D_S can be written as

$$D_S=D_S^{\mathrm{QW}}+P^{\mathrm{BULK}}L^{\mathrm{BOX}} , \quad (18)$$

where P^{BULK} and L^{BOX} are, respectively, the polarization contributed by the barriers and their total length. We again stress that, since the well is a two-dimensional system, its contribution to the polarization is best expressed by the dipole per unit area D_S^{QW}, while the same is not true for the bulk. An effective second-order susceptibility for the well $\chi^{(2)}$ (QW) can then be defined, following the definition (7), by dividing D_S^{QW} by the product of the electric fields, by ε_0, and by an interaction length L, which is the sum of the well width and the length of barrier, which is necessary for the isolation of the well. We assume L to be given by L_w/f, where L_w is the well thickness and f is a filling factor, taken equal to $\frac{1}{2}$ and independent of L_w. We, therefore, define $\chi^{(2)}$ (QW) as

$$\chi_{xzx}^{(2)}(\mathrm{QW})=\frac{(D_S^{\mathrm{QW}})_x f}{\varepsilon_0 E_z E_x L_w} . \quad (19)$$

This quantity differs from what is directly calculated from Eq. (16) for the contribution $P^{\mathrm{BULK}}L^{\mathrm{BOX}}$ due to the bulk. To overcome this difficulty, we shall subtract from the total susceptibility of Eq. (16) the same expression calculated in a big box without the well. We emphasize that the $\chi_{xzx}^{(2)}$ (QW) so defined is the appropriate one in a structure composed of many quantum wells, where each well occupies a minimum length L.

We now show results for symmetric wells of GaAs and barriers of $Al_{0.4}Ga_{0.6}As$, for which we have taken the conduction-band offset ΔE_c to be 334 meV and the valence-band offset ΔE_v to be 167 meV. The electron and heavy-hole effective masses are taken equal to 0.067 and

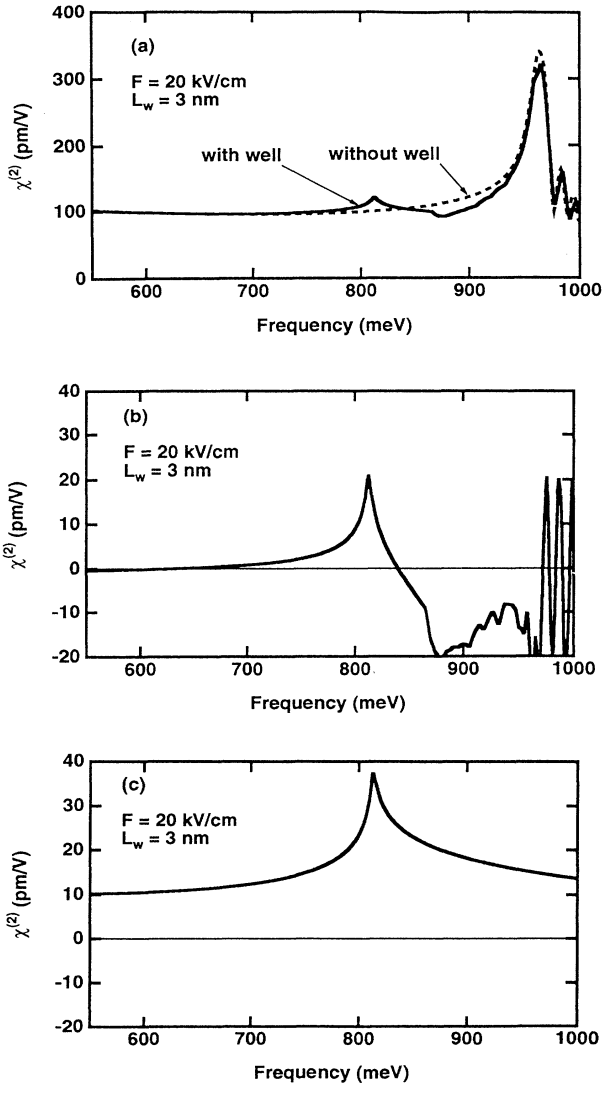

FIG. 1. (a) Absolute value of the calculated second-order susceptibility for second-harmonic generation vs pump frequency with and without a 3-nm quantum well in a 20 kV/cm field perpendicular to the QW interfaces; (b) the susceptibility contribution of the quantum well; (c) the susceptibility contribution of the quantum well using Eq. (17).

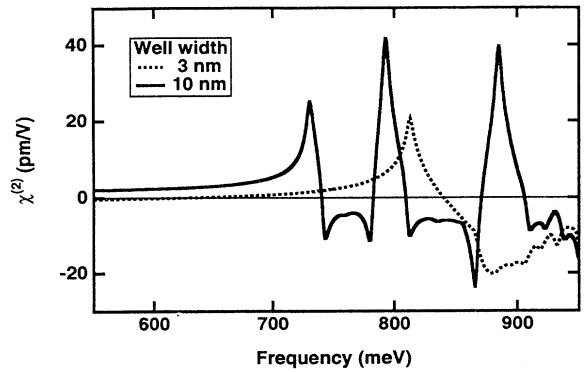

FIG. 2. Calculated quantum-well second-order susceptibilities (absolute value) vs pump frequency for two well widths.

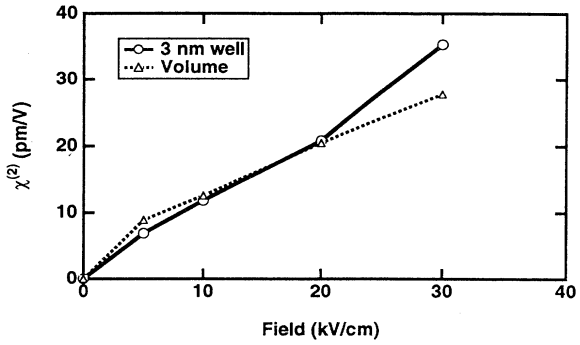

FIG. 3. Maximum second-order susceptibility (absolute value) for a 3-nm quantum well vs applied electric field compared with field-dependent part of the bulk susceptibility.

0.377 times the free-electron mass, respectively. All the line broadenings are taken equal: $\gamma = 3$ meV.

In Fig. 1 we show results for a 3-nm quantum well in an applied field of 20 kV/cm. We have used 50 electron and 50 heavy-hole subbands. In accordance with the definition Eq. (19), the length L entering Eq. (16) has been taken as L_w/f. It can be seen in Fig. 1(a) that the numerous continuum transitions contribute a large background volume polarization. In Fig. 1(b), the background has been subtracted. It can be seen that a peak susceptibility of about 20 pm/V is found at the frequency 813 meV corresponding to half the fundamental hh1-e1 transition. If we use the simple Eq. (17) involving only the fundamental well levels, we obtain the result shown in Fig. 1(c). Comparing to the many-subband result, we observe that the transitions to and from continuum subbands interfere destructively with the fundamental transition and reduce the peak susceptibility. The negative well susceptibilities above the peak are an artifact of our definition of the $\chi^{(2)}$ (QW) as the difference between the absolute values of the $\chi^{(2)}$ with and without well. The spurious peaks in the high-frequency part of the spectra are a consequence of the artificial quantization of the continuum levels due to the finite box length.

If we increase well width, we obtain typical results as displayed in Fig. 2. The 10-nm well contains three bound electron levels, which give rise to three peaks and a more complicated spectral dependence. As a function of well width, the peak susceptibilities do not show a simple dependence but remain of the same order of magnitude between 3 and 20 nm.

Finally, if we study the field dependence of the susceptibility peak, we find as expected the linear relation shown in Fig. 3. Our model in which we assume all matrix elements independent of $\mathbf{k}_\parallel$ yields a vanishing polarizability if the applied field is zero. On the other hand, we can use our model to estimate the field-dependent part of that susceptibility from the results without well. If we take the peak susceptibility corresponding to the gap of $Al_{0.4}Ga_{0.6}As$ seen on Fig. 1(a), we find the field dependence of the bulk susceptibility shown in Fig. 3. The same approximately linear increase as in the case of the quantum well is found with the same order of magnitude susceptibility.

CONCLUSION

We have developed a theory within the effective-mass approximation of the second-order susceptibility of a quantum well subject to an applied electric field. We have shown that close to the midgap of the structure, it is possible within that approximation to reduce the normally derived triple sums over subbands to a sum over all the optically allowed transitions. This not only reduces the number of calculations to perform, but also assures a much better convergence rate with increasing number of subbands taken into account. Moreover, this sum rule allows us to get rid of the unnecessary intersubband transitions, which complexify the work of Khurgin.[3]

For very deep wells, our expression (17) only involving the ground subbands is very compact and easy to study. On the other hand, we have demonstrated that in *realistic structures* higher transitions play a strong role for quantitative results. It, therefore, seems that models as those of Ref. 9 in which only three subband levels are included and in which the sum rule (13) is not used lead to erroneous results.

We have pointed out that the strong influence of the higher-energy transitions on the second-order susceptibility has the consequence that it is not generally possible to define a quantum-well susceptibility independent of the structure in which the well is inserted. On the other hand, the results obtained by our operational definition compared with the field-dependent part of the bulk susceptibility indicate that the quantum wells should not lead to spectacular enhancements of the second-harmonic-generation susceptibility.

It has been recently argued[11,16] that the $\chi^{(2)}$ tensor should have a nonvanishing (zzz) component due to light-hole–electron virtual transitions. However, away from exciton resonances, an expression analogous to (16) can be derived for the $\chi^{(2)}_{zzz}$. As it is easily seen,[13] the product $\langle e_c|\mathrm{hh}_v\rangle\langle \mathrm{hh}_v|P^z|e_c\rangle$ vanishes if the product between the effective mass and the well potential profile is equal for electrons and holes. Since this is almost the case for electrons and light holes, this cancellation effect makes the light holes contribution to the $\chi^{(2)}$ negligible. We, therefore, expect the $\chi^{(2)}_{zzz}$ value to be negligible relative to the calculated $\chi^{(2)}_{xzx}$ values.

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