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Second Harmonic Generation at the Quantum-Interference Induced Transparency in Semiconductor Quantum Wells: The Influence of Permanent Dipole Moments

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***Abstract*—The influence of permanent dipole moments of quantized states on intersubband second harmonic generation based on quantum-interference induced transparency in semiconductor quantum wells is explored using the harmonic balance method. The permanent moments are found to be quite important: they affect the transparency condition, especially at larger pump intensities. Hence, both the conversion efficiency and the optimal interaction path length change significantly when accounting for the permanent moments, and the conversion efficiency is reduced.**

***Index Terms*—Fano-interference, quantum interference, transparency.**

I. Introduction

THE USE of quantum interference of lifetime-broadened resonances for obtaining radiation amplification without inversion, initially proposed in [1], has been the subject of numerous papers on this topic, including the proposals of potential devices using this effect (see, e.g., [2] for a recent review). In a situation of having two close, strongly coupled upper states, which decay to the same continuum, the existence of different interaction paths leading from the initial state to the continuum induces the asymmetry in the absorption-emission profile due to Fano-type interference. States coupling via the electromagnetic field in a four-level atomic system was suggested for obtaining high conversion efficiencies in four-wave mixing, by resonantly enhanced third-order susceptibility, while simultaneously meeting the condition for transparency of the medium at the generated frequency [3]. Second-harmonic generation in atomic hydrogen with reduced absorption, by using this scheme, has been reported [4]. Among more recent proposals for utilization of quantum interference in free atoms, we mention photonic switching [5], parametric self-oscillation [6], and a huge reduction of the speed of light [7].

In contrast to optical control schemes (commonly used in atomic systems for nonlinear optical effects) which usually employ one or more coherent light sources to control the transition rates from the initial to the final state, in semiconductor quantum well (QW) systems it is the states coupling via tunneling or phonon scattering which may bring about useful effects. QW systems are particularly attractive since the parameters of interest (dipole-matrix elements, dephasing and lifetime broadening rates, having essentially fixed values in free atoms) can be readily controlled by an appropriate choice of growth parameters. Together with rather large values of dipole matrix elements, all this makes QWs generally suitable candidates for observing quantum interference effects. To obtain the Fano-type interference, however, one needs two close interaction paths with about equal probability amplitudes, and achieving this in semiconductor QWs requires quite some care in their design, due to separation of initial and resonance states from the continuum via the potential barrier [8]. Furthermore, the nonlifetime-related mechanisms of line broadening, adversely influencing the quantum intererence, are much more prominent in QWs than in gases, and a considerable effort is necessary to produce appropriate high-quality QWs. For these reasons, the advances toward observing and using quantum interference based effects in QWs lagged somewhat behind those in gases. Quantum interference has first been experimentally verified in interband transitions [9], while the reduced intersubband absorption feature has been demonstrated in three-level asymmetric or symmetric QWs [10]–[12], and was given detailed theoretical consideration in [13]. Among the quantum interference related effects in QWs, we further note reduced-absorption second-harmonic generation [14], photonic switching [15], and probe beam amplification [16].

In this paper, we discuss the impact of permanent dipole moments of quantized states on the transparency enhanced intersubband second harmonic generation (SHG) in three-level semiconductor QWs. The idea of using the resonant enhancement of the nonlinear susceptibility while simultaneously eliminating the absorption in quantum interference based semiconductor structures was exploited in [14]. We have previously considered the influence of permanent moments in resonant SHG [17], but only in classical (i.e., not transparency based) structures. There are two ways in which the permanent moments influence optical processes in QWs. The electrostatic aspect of this influence is the effect of levels shifting due to charge redistribution, related to the permanent moments, and has been rather thoroughly studied in the literature. There is, however, another as-

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pect: the direct interaction of permanent moments with the optical field(s). Here we show that this may be quite important, especially when high conversion efficiency is required, i.e., at large pump intensities. The permanent moments degrade the efficiency, as a consequence of the fact that the transparency condition becomes dependent on the pump and harmonic fields intensities, i.e., the full transparency cannot be achieved along the whole interaction path.

II. Theoretical Considerations

We consider a three-level asymmetric semiconductor QW, with the energy spacing between the two upper states much smaller than the LO phonon energy. The coupling of these states occurs via tunneling and/or the the emission/reabsorption of LO phonons, with the ground state as the intermediate state. The equations used to describe the system dynamics are obtained from the density-matrix approach. Accounting for the permanent dipole moments, we arrive to a somewhat extended set of equations used in [18]

$$\begin{aligned}
& i\,\frac{\partial \rho_{22}}{\partial t} + \frac{i}{T_2}(\rho_{22}-\rho_{22}^0) + (\Omega_{12}+\Omega_j)(\rho_{12}-\rho_{21}) \\
&\quad - i\kappa(\rho_{23}+\rho_{32}) \\
&\quad = 0 \\
& i\,\frac{\partial \rho_{33}}{\partial t} + \frac{i}{T_3}(\rho_{33}-\rho_{33}^0) + (\Omega_{13}+\Omega_k)(\rho_{13}-\rho_{31}) \\
&\quad - i\kappa(\rho_{23}+\rho_{32}) \\
&\quad = 0 \\
& \rho_{11}+\rho_{22}+\rho_{33} \\
&\quad = 1 \\
& \frac{1}{i}\,\frac{\partial \rho_{12}}{\partial t} - \left(\omega_{21}+\frac{i}{\tau_{21}}\right)\rho_{12} - \Pi_{12}\rho_{12} + (\Omega_{12}+\Omega_j) \\
&\quad (1-2\rho_{22}-\rho_{33}) - (\Omega_{13}+\Omega_k)\rho_{32} + i\kappa\rho_{13} \\
&\quad = 0 \\
& \frac{1}{i}\,\frac{\partial \rho_{13}}{\partial t} - \left(\omega_{31}+\frac{i}{\tau_{31}}\right)\rho_{13} - \Pi_{13}\rho_{13} + (\Omega_{13}+\Omega_k) \\
&\quad \cdot (1-\rho_{22}-2\rho_{33}) - (\Omega_{12}+\Omega_j)\rho_{23} + i\kappa\rho_{12} \\
&\quad = 0 \\
& \frac{1}{i}\,\frac{\partial \rho_{23}}{\partial t} - \left(\omega_{32}+\frac{i}{\tau_{32}}\right)\rho_{23} - \Pi_{23}\rho_{23} - (\Omega_{12}+\Omega_j)\rho_{13} \\
&\quad + (\Omega_{13}+\Omega_k)\rho_{21} + i\kappa(\rho_{22}+\rho_{33}) \\
&\quad = 0.
\end{aligned} \tag{1}$$

Here, T_2 and T_3 denote the diagonal, and τ_{ij} the off-diagonal relaxation times

$$\Omega_{1j} = d_{1j}E_h/2\hbar + d_{1j}E_p/2\hbar$$

and

$$\Omega_{k(j)} = \frac{E_p^2}{4\hbar^2}\sum_s \frac{d_{1s}d_{s2(3)}}{\omega_{s1}-\omega_p}$$

are the effective Rabi frequences, dependent on the transition dipole moments, which take into account all possible intermediate states since the pump photon energy is assumed to be far from the resonance. E_p and E_h are the complex amplitudes of the pump and harmonic fields, respectively, which become coordinate-dependent when considering propagation. $\Pi_{ij} = \Delta d_{ij}E/2\hbar$ are the permanent-moment analogs of Rabi frequencies, proportional to the differences of permanent dipole moments ($\Delta d_{ij} = d_{ii} - d_{jj}$). Finally, the important coupling term, κ, is defined as $\kappa = \sqrt{\Gamma_{2l}\Gamma_{3l}}$, where Γ_{2l} and Γ_{3l} are the lifetime broadening relaxation rates of states 2 and 3. Total relaxation rates of states 2 and 3 are given as $\Gamma_j = \Gamma_{jl} + \Gamma_{jdph}$, where Γ_{jdph} is the dephasing rate, and

$$\Gamma_j = \frac{\hbar}{2T_j} + \frac{\hbar}{\tau_{j1}}, \qquad j = 2, 3.$$

To get insight into the way the permanent dipole moments affect the transparency condition, given by $\mathrm{Im}(\chi^{(1)}(\omega_h)) = 0$ [14], we make some simple algebraic manipulations, assuming that the pump field is weak and that all electrons reside in state 1 only (no depletion limit). In the steady state [$\partial/\partial t = 0$ in (1)], we find

$$\begin{aligned}
& \left(\omega_{21}+\frac{i}{\tau_{21}}+\Pi_{12}\right)\rho_{12} - i\kappa\rho_{13} \\
&\quad = (\Omega_{12}+\Omega_j) - (\Omega_{13}+\Omega_k)\rho_{32} \\
& -i\kappa\rho_{12} + \left(\omega_{31}+\frac{i}{\tau_{31}}+\Pi_{13}\right)\rho_{13} \\
&\quad = (\Omega_{13}+\Omega_k) - (\Omega_{12}+\Omega_j)\rho_{23}
\end{aligned} \tag{2}$$

where ω_p and $\omega_h = 2\omega_p$ are the pump and harmonic frequencies. Neglecting the terms proportional to ρ_{23} (ρ_{32}) on the right-hand side and setting $\Pi_{ij} = 0$, solving for ρ_{12} and ρ_{12} yields the equations for linear and nonlinear susceptibilities at harmonic frequency as given in [14]. In that case, the coefficients on the left-hand side of (2) depend only on the system parameters but not on the field intensity. Therefore, by adjusting the parameters appropriately, one obtains the transparency condition which (in the limit of no dephasing) has a simple form [14]

$$(\omega_{21}-\omega_h)d_{13}\sqrt{\Gamma_{3l}} - (\omega_{31}-\omega_h)d_{12}\sqrt{\Gamma_{2l}} = 0. \tag{3}$$

With the dephasing present, the transparency is no longer perfect, but the absorption at the generated harmonic frequency is nevertheless reduced. With finite permanent dipole moments, however, the situation is quite different, as can be seen from (2). The transparency condition is modified due to the Π_{ij} term, which is proportional to Δd_{ij}. It is no longer possible to find an explicit expression showing how the permanent moments affect the transparency, because one would have to solve the system of density matrix equations along with Maxwell's equations describing the propagation of pump/harmonic fields. Besides, at large pump field intensities, carriers are redistributed over the available states, thus further complicating the situation. Apart from these difficulties, one can expect, firstly, that the permanent moments induce a shift of the pump photon energy at which the maximum conversion would occur, and secondly, that the transparency condition varying along the interaction path will result in a reduced conversion efficiency. Quantitative results may be obtained only by numerical calculations.

Due to strong fields involved in the interaction, we choose the harmonic-balance method for solving (1). The following components of the density matrix are taken: ρ_{12} and ρ_{13} (and their complex conjugates) are assumed to have a dc component (to account for Δd_{ij}), a component oscillating at pump frequency ω_a, and a component oscillating at the generated harmonic frequency $\omega_h = 2\omega_p$. The ρ_{23} (ρ_{32}) and diagonal elements of the density matrix are taken to have only the dc term, but, since this assumption is not quite justified at large fields, we also add terms at both the pump and harmonic frequency for very strong pumps. Upon substituting these in (1), and equating the terms at appropriate frequencies, we obtain a system of equations which is solved numerically. The polarization components are calculated from $P = Tr(d\rho)$, and the evolution of fields follows from Maxwell's equations.

III. Numerical Results and Discussion

For numerical calculations we have designed the following AlGaAs based QW, matched for the CO_2 laser as the pump source: 55 Å/35 Å/55 Å $Al_{0.3}Ga_{0.7}As/Al_{0.5}Ga_{0.5}As/GaAs$, in bulk $Al_{0.5}Ga_{0.5}As$. The central region of the shallow well was doped (sheet density $N_s = 5 \times 10^{11}$ cm^{-2}) and an external electric field (47 kV/cm) was applied across the structure to provide favorable alignment of levels, and enable strong coupling of levels 2 and 3 ($E_{32} = 12$ meV). The dipole moments were calculated using the self-consistent procedure [19], and amount to $d_{12} = -11.3$Å, $d_{13} = 10.6$ Å, $d_{23} = -45$ Å, $\Delta d_{12} = 46$ Å, $\Delta d_{13} = 45.5$ Å. An average electron concentration is taken to be 2×10^{17} cm^{-3}.

The quantities of prime importance for the efficiency of SHG are the states' relaxation rates. These were investigated in detail in [20]. For quantum interference to be observed, it was found necessary to have the dephasing rates 3–4 times smaller than the lifetime broadening rates. Indeed, the numerical calculations we performed show an increasing conversion efficiency as the diagonal and off-diagonal relaxation times become comparable. The smallest reported value of the total linewidth [21] of 2.5 meV (full width) was used in the calculations (and $\Gamma_{2dph} = \Gamma_{3dph}$ is assumed). Since the coupling of levels 2 and 3 depends on κ, and hence on lifetime broadening rates, two values of the diagonal relaxation time, also assumed to be the same for states 2 and 3, were used ($T = 1$ ps and $T = 2$ ps) to demonstrate the influence of lifetime broadening. It follows from (3) that, with the stated values of parameters, the transparency will occur at the harmonic photon energy very close to $(E_{21} + E_{31})/2$.

In Fig. 1, we show the conversion efficiency as it depends on the pump wavelength for two different values of T, with permanent dipole moments neglected or accounted for. The permanent moments are clearly very important in this system, reducing the conversion efficiency by almost a factor of 2, as a consequence of the light intensity (and therefore the coordinate)-dependent transparency condition. The pump photon energy which delivers the maximum conversion will vary somewhat with the the pump intensity (by $\sim$2 meV for the intensities in the tens of megawatts per cm^2 range). In fact, the system is very sensitive to the precise tuning of the pump wavelength to achieve transparency, a property itself not related to the existence of permanent moments, but which clearly translates into the intensity-specific optimal pump wavelength.

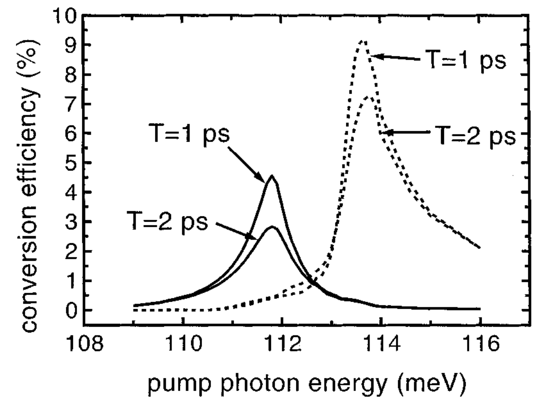

Fig. 1. The conversion efficiency versus the pump photon energy dependence for two values of the diagonal relaxation time $T = 1$ ps and $T = 2$ ps, with the permanent moments included (solid lines) or neglected (dashed lines). The maximum conversion is reduced and takes place at lower photon energies if the permanent moments are accounted for.

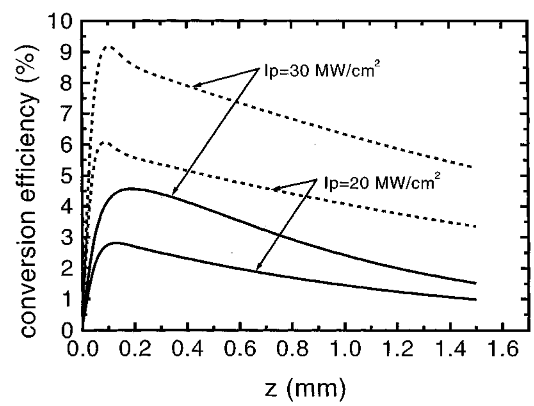

Fig. 2. Intensity of the harmonic (arbitrary units) along the interaction path at different pump intensities. In a case when the permanent moments are neglected (dashed lines), both curves are obtained for the photon energy 113.8 meV. With the permanent moments included, the upper solid line is obtained for the photon energy 111.8 meV, and the lower solid line for 112.2 meV. With increasing the pump intensity the optimal photon energy (i.e., necessary for the maximal conversion) decreases.

Examples of the harmonic field propagation through the medium are given in Fig. 2, as calculated for different pump intensities and with the permanent dipole moments neglected or accounted for. The pump is not displayed because it is almost constant (less than 10% drop along the interaction path), since it is far off resonance. Clearly, the permanent moments affect the optimal conversion length, though not so drastically as the conversion efficiency. We may also note the increase of the optimal conversion length with increasing pump intensity, a feature also known in conventional resonant SHG, and not specific to the existence of permanent moments. What is different, however, is a long interaction path of $\sim$1.5 mm, obtained under the best-matched-to-transparency conditions.

IV. Conclusion

In conclusion, the influence of the permanent dipole moments of QW quantized states on intersubband SHG based on the quantum-interference induced transparency was explored. The analytical model describing the intersubband transitions at large pump intensities was expanded to account for permanent moments. It is shown that the permanent moments induce a shift of the pump photon energy at which the conversion efficiency is maximum. Since this shift depends on the pump and harmonic fields, it cannot be maintained constant along the interaction path, hence the efficiency has to be lower than would be the case if permanent moments were neglected. Due to a high sensitivity of transparency on photon frequency, a considerable drop in efficiency is found in realistic structures.

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