Nonlinear optical rectification in parabolic quantum wells with an applied electric field

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Optical rectification in parabolic quantum wells, with an applied electric field, due to resonant intersubband transitions is analyzed using a compact density-matrix approach. The large dipolar matrix elements obtained in such structures are partly due to the small effective masses of the host materials and are interpreted in terms of the participation of the whole band structure in the optical transitions. The other origin of the large optical rectification coefficient lies in the possibility of tuning independently the frequency ω_0 of the parabolic potential and the applied electric field F.

I. INTRODUCTION

Wide parabolic quantum wells (PQW's) have been proposed as structures in which a high-mobility quasi-three-dimensional electron gas can be realized. These wide PQW's have recently been grown[1,2] by tailoring the conduction-band edge of a graded $Ga_{1-x}Al_xAs$ semiconductor. Quantum confinement of carriers in a semiconductor parabolic well leads to the formation of discrete energy levels and the drastic change of optical susceptibilities.[3] One of the most remarkable properties of these quasi-two-dimensional electronic systems is that the optical transitions between the size-quantized subbands are feasible. The electrons are quantized into subbands where their wave functions in the growth direction have the form of envelope functions with an extension equal to the effective well width, i.e., in the range of a few nanometers. Electromagnetic waves may induce electronic transitions between these subbands. The dipole matrix elements associated with these intersubband transitions have the same order of magnitude as the effective well width leading to extremely large absorption.

The dipole matrix elements are thus in the nanometer range instead of the few picometers obtained in usual molecular or ionic systems.[4] Since second-order optical susceptibilities have a cubic dependence relative to the dipole matrix elements, strong second-order optical nonlinearities are expected in multiple quantum wells (MQW's) insofar as inversion symmetry is broken. In their work, Gurnick and DeTemple have suggested obtaining this asymmetry by growing $Al_xGa_{1-x}As$ MQW's with asymmetric composition gradients of Al in the growth direction.[5] In their paper, these authors have considered an asymmetric Morse potential and have shown that nonlinearities of 10–100 times larger than in bulk materials could be theoretically possible. Khurgin later suggested using asymmetric coupled quantum wells.[6] Ahn and Chuang proposed to bias a symmetric QW electrically to obtain this asymmetry.[7] This has been realized by Fejer *et al.*, who obtained a second-generation coefficient more than 70 times higher than in bulk GaAs.[8] In a more recent work, Yuh and Wang suggested that the use of a step-quantum-well structure, which consists of a small well inside a larger one, would be easier to fabricate and could yield also large second-harmonic nonlinearities.[9] Rosencher *et al.* have shown that these step QW's could be designed so that the absorption could be doubly resonant, leading to second-harmonic-generation coefficients more than three orders of magnitude higher than in bulk GaAs.[10] These latter authors have realized different step QW's and observed indeed extremely large second-order optical nonlinearities.[11] These huge nonlinearities in step QW's have been confirmed by Karunasiri, Mii, and Wang, who measured linear Stark effects as high as 0.44 meV/kV.[12]

In this paper, we study the optical rectification of a parabolic quantum well with an applied electric field. In Sec. II we shall present the theoretical development of perturbation theory and density motion, and give the simple expressions of nonlinear optical rectification. In Sec. III we will give our results and discussions. We find that in the PQW's with an applied electric field the dipole matrix elements $\mu_{01}^2\Delta$ increase with the enhancement of the electric field F, but increase with the decrease of the frequency ω_0 of the parabolic potential (see Fig. 1). We also find that the optical rectification $\chi_0^{(2)}$ is six orders of magnitude higher than in bulk GaAs.

II. THEORY

Electrons in PQW's with an applied electric field are described by the effective-mass Hamiltonian

$$H=-\frac{\hbar^2}{2m^*}\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+\frac{\partial^2}{\partial z^2}\right]+\tfrac{1}{2}m^*\omega_0^2z^2+qFz\ , \tag{1}$$

where z represents the growth direction, $\hbar$ is Planck's constant, $\frac{1}{2}m^*\omega_0^2z^2$ is the parabolic confining potential in the quantum well, and F is the applied electric field. The conduction-band effective mass m^* will be taken to be constant in the rest of the paper. The eigenfunctions

$\Psi_{n,\mathbf{k}}(\mathbf{r})$ and the eigenenergies $\varepsilon_{n,\mathbf{k}}$ are solutions of the Schrödinger equation $H\Psi_{n,\mathbf{k}}(\mathbf{r})=\varepsilon_{n,\mathbf{k}}\Psi_{n,\mathbf{k}}(\mathbf{r})$ and are given by

$$\Psi_{n,\mathbf{k}}(\mathbf{r})=\Phi_n(\mathbf{z})\mathbf{U}_c(\mathbf{r})e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel} , \tag{2}$$

and

$$\varepsilon_{n,\mathbf{k}}=E_n+\frac{\hbar^2}{2m^*}|\mathbf{k}_\parallel|^2 . \tag{3}$$

Here, $\mathbf{k}_\parallel$ and $\mathbf{r}_\parallel$ are the wave vector and coordinate in the xy plane and $U_c(\mathbf{r})$ is the periodic part of the Bloch function in the conduction band at $\mathbf{k}=0$. Φ_n and E_n are, respectively, the envelope wave function and the transverse energy of the nth subband, solutions of the one-dimensional Schrödinger equation $H_0\Phi_n(z)=E_n\Phi_n(z)$, where H_0 is the z part of the Hamiltonian H in Eq. (1), i.e., $H_0=-(\hbar^2/2m^*)(\partial^2/\partial z^2)+\frac{1}{2}m^*\omega_0^2z^2+qFz$. We know that the Schrödinger equation can be solved exactly.

If the structure is doped, at sufficiently low temperature most of the electrons are located in the ground subband ε_0. Let us consider an electromagnetic field of frequency ω which is incident with a polarization vector normal to the parabolic wells. The polar interaction is given by $qE_0z\cos(\omega t)$, where q is the electronic charge. Time-dependent perturbation theory shows that the electrons are then in the steady state:[13]

$$|\Psi(t)\rangle=|\Psi_0\rangle+\sum_{n\neq0}\frac{qE_0}{2\hbar}\langle\Psi_n|z|\Psi_0\rangle\frac{e^{-i\omega t}}{(\omega_{0n}-\omega)+\frac{\hbar}{2m^*}|\mathbf{k}_\parallel^n-\mathbf{k}_\parallel^0|^2+i\Gamma_{n0}}|\Psi_n\rangle , \tag{4}$$

where $\omega_{0n}=E_{0n}/\hbar=(E_n-E_0)/\hbar$ is Bohr's frequency, and $\mathbf{k}_\parallel^n$ and $\mathbf{k}_\parallel^0$ are the parallel wave vectors in subbands n and 0, respectively. We have made the near-resonant approximation [neglecting the $(\omega+\omega_{0n})$ terms] and neglected transient behavior [$\exp(\pm i\omega_{0n}t)$ terms] by introducing the lifetimes Γ_{n0}. Symmetry considerations on Bloch states show that

$$\langle\Psi_n|z|\Psi_0\rangle=\delta_{\mathbf{k}_\parallel^n,\mathbf{k}_\parallel^0}\langle\Phi_n|z|\Phi_0\rangle , \tag{5}$$

where δ is the Kronecker delta function.

Now we will present a formalism for the derivation of nonlinear optical rectification in parabolic quantum wells with an applied electric field. Let us consider the system described by the Hamiltonian

$$H_0=-(\hbar^2/2m^*)(\partial^2/\partial z^2)+\tfrac{1}{2}m^*\omega_0^2z^2+qFz .$$

At thermal equilibrium, the density matrix $\rho^{(0)}$ is a diagonal one, in which the diagonal elements $\rho_{ii}^{(0)}$ are the surface thermal population ρ_i of level E_i given by the Fermi level in the parabolic quantum well. The system is excited by an internal electromagnetic field

$$E(t)=\tilde{E}e^{i\omega t}+\tilde{E}e^{-i\omega t} . \tag{6}$$

The evolution of the density matrix is given by the time-dependent Schrödinger equation

$$\frac{\partial\rho_{ij}}{\partial t}=\frac{1}{i\hbar}[H_0-qzE(t),\rho]_{ij}-\Gamma_{ij}(\rho-\rho^{(0)})_{ij} . \tag{7}$$

For simplicity, we will assume in the following only two different values of the relaxation rates: $\Gamma_1=1/T_1$ for $i=j$ is the diagonal relaxation rate and $\Gamma_2=1/T_2$ is the off-diagonal relaxation rate. Equation (7) is solved using the usual iterative method:[4,7]

$$\rho(t)=\sum_n\rho^{(n)}(t) , \tag{8}$$

with

$$\frac{\partial\rho_{ij}^{(n+1)}}{\partial t}=\frac{1}{i\hbar}\{[H_0,\rho^{(n+1)}]_{ij}-i\hbar\Gamma_{ij}\rho_{ij}^{(n+1)}\}-\frac{1}{i\hbar}[qz,\rho^{(n)}]_{ij}E(t) . \tag{9}$$

The electronic polarization of the PQW will also be a series expansion as in Eq. (8). We shall limit ourselves to the first two orders, i.e.,

$$P(t)=(\epsilon_0\chi^{(1)}\tilde{E}e^{i\omega t}+\epsilon_0\chi_{2\omega}^{(2)}\tilde{E}^2e^{2i\omega t})+\text{c.c.}+\epsilon_0\chi_0^{(2)}\tilde{E}^2 , \tag{10}$$

where $\chi^{(1)}$, $\chi_{2\omega}^{(2)}$, and $\chi_0^{(2)}$ are the linear, second-harmonic generation, and optical rectification coefficients, respectively. The electronic polarization of the nth order is given by

$$P^{(n)}(t)=\frac{1}{s}\text{Tr}(\rho^{(n)}qz) , \tag{11}$$

where s is the area of interaction.

We finally find the optical rectification coefficient per unit surface:

$$\chi_0^{(2)}=4\frac{q^3\rho_s}{\epsilon_0\hbar^2}\mu_{01}^2\Delta\frac{\omega_{01}^2\left[1+\frac{\Gamma_2}{\Gamma_1}\right]+(\omega^2+\Gamma_2^2)\left[\frac{\Gamma_2}{\Gamma_1}-1\right]}{[(\omega_{01}-\omega)^2+\Gamma_2^2][(\omega_{01}+\omega)^2+\Gamma_2^2]} . \tag{12}$$

It is clear that optical rectification will occur in our model where the mean electron displacement Δ does not equal zero (from the calculation, we get $\Delta=qF/m^*\omega_0^2$). We can see from Eq. (12) that the enhancement of the optical rectification $\chi_0^{(2)}$ originates from the contribution of $\mu_{01}^2\Delta$. We can tune independently the frequency of the parabolic potential and the applied electric field to optimize $\mu_{01}^2\Delta$. An examination of Eq. (12) shows different ways to enhance the $\chi_0^{(2)}$: the geometrical factor $\mu_{01}^2\Delta$, the doping concentration ρ_s, and the time constant product T_1T_2.

T_2 is certainly governed by intrinsic mechanisms such as electron-electron interaction or optical-phonon emission for an excitation energy, without clear possibilities to act upon it. On the other hand, T_1 is a population relaxation time and can be enhanced by storing the excited electrons on a metastable level.

III. RESULTS AND DISCUSSIONS

In Fig. 1, we show that the dipolar matrix elements $\mu_{01}^2\Delta$ increase with the enhancement of the electric field F and increase with the decrease of the frequency ω_0 of the parabolic potential. Since we know that if the electric field F is too strong, it will break down the semiconductors, that means there is a superior limit to F. Of course, there is also a limit to ω_0. We get the optimum values $F=2.0\times10^7$ V/m and $\omega_0=3.6\times10^{14}$ s^{-1}.

Figure 2 shows the maximum second-order nonlinear susceptibility (optical rectification) as a function of photon energy obtained in this model. We have assumed usual relaxation times of $T_2=0.2$ ps and $T_1=1$ ps.[11] We optimize $\omega=10^{12}$ s^{-1}, $\rho_2=5\times10^{24}$ m^{-3}, $F=2.0\times10^7$ V/m, and $\omega_0=3.6\times10^{14}$ s^{-1}. In our calculation, the effective parabolic width $W=4000$ Å and the height $V=150$ meV.[14] Our curve is compared with the one given by the classical polarizable sphere limit of Gurnick and DeTemple.[5] In their paper, these authors estimate that the classical model leads to limit values which are higher than the ones obtained using the quantum Morse potential model. This is not so in our case. The huge enhancement in the theoretical values of the maximum susceptibility in our model comes from the possibility of independently tuning the applied electric field F and the frequency ω_0 of the parabolic potential. Comparing the order of magnitude for optical rectification with two other possibilities—biased quantum well[7] and asymmetric quantum well[5]—we know that our results have the same order of magnitude for optical rectification as the results obtained in Refs. 5 and 7.

In conclusion, we have used a compact density-matrix approach to analyze the optical nonlinearities in parabolic quantum wells with an applied electric field due to resonant intersubband transitions. The origin of the large oscillator strengths in GaAs parabolic QW's is analyzed in terms of the contribution of the optical transition over the whole band structure of GaAs. The dipolar matrix elements which lead to the maximum values of the second-order susceptibility are gained. We thus show that the huge nonlinearities observed in these structures are due to (i) the effect of the small effective mass (i.e., $m_0=14.9m^*$ in GaAs), and mainly (ii) the possibility of tuning independently the frequency of the parabolic potential and the applied electric field. Values of optical rectification coefficients as high as 1.6×10^{-3} m/V at 10.6 μm (i.e., six orders of magnitude higher than in bulk

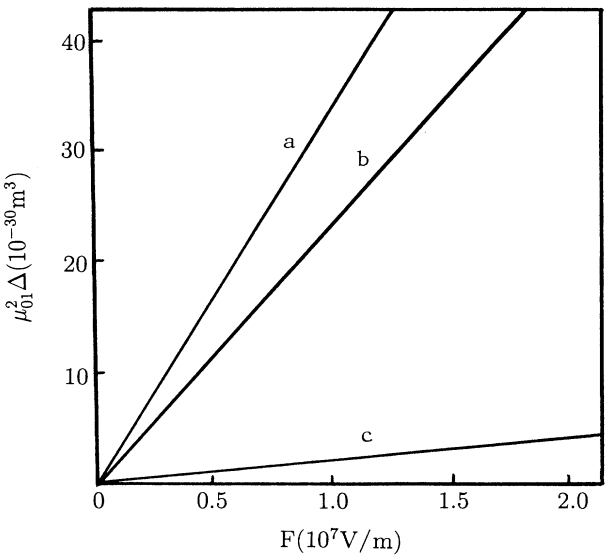

FIG. 1. Variation of the product of dipolar matrix elements $\mu_{01}^2\Delta$ as a function of the electric field F. It is plotted for three different frequencies ω_0 of the parabolic potential: (a) $\omega_0=9.0\times10^{14}$ s^{-1}; (b) $\omega_0=1.04\times10^{15}$ s^{-1}; and (c) $\omega_0=3\times10^{15}$ s^{-1}. It shows that the dipolar matrix elements $\mu_{01}^2\Delta$ increase with the enhancement of the applied electric field F and increase with the decrease of the frequency ω_0 of the parabolic potential.

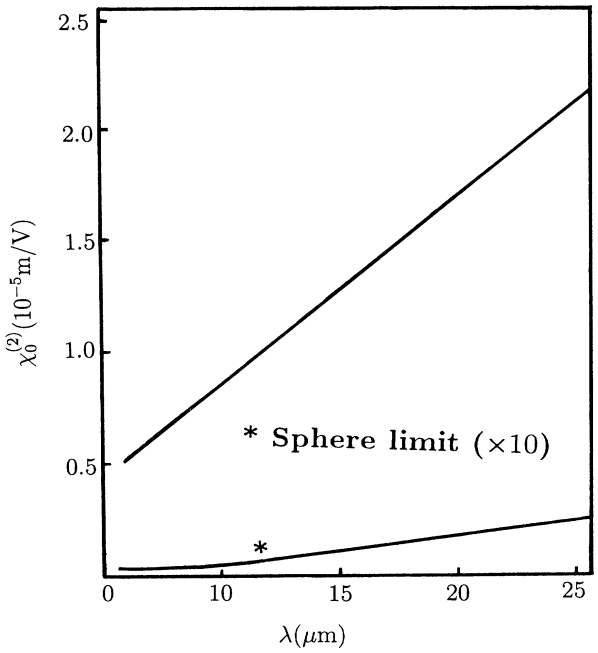

FIG. 2. Quantum limitation of $\chi_0^{(2)}$ in parabolic quantum wells. The diagonal and off-diagonal relaxation times are 0.2 and 1 ps, respectively; $\omega=10^{12}$ s^{-1}, $\rho_s=5\times10^{24}$ m^{-3}, $\omega_0=3.6\times10^{14}$ s^{-1}, and $F=2.0\times10^7$ V/m. The effective parabolic well width W is 4000 Å and the height V is 150 meV (Ref. 14). The results are compared with the polarizable sphere limit of Gurnick and DeTemple (Ref. 5).

GaAs) may be obtained in optimized structures. Finally, it is hoped that this paper would stimulate more experimental work, which could be helpful in an understanding of the optical nonlinearities in the parabolic quantum well with an applied electric field.

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[1]E. G. Gwinn, R. M. Westervelt, P. F. Hopkins, A. J. Rimberg, M. Sundaram, and A. G. Gossard, Phys. Rev. B **39**, 6260 (1989); A. J. Rimberg and R. M. Westervelt, *ibid.* **40**, 3970 (1989).

[2]T. Sajoto, J. Jo, L. Engel, M. Santos, and M. Shayegan, Phys. Rev. B **39**, 10 464 (1989).

[3]R. Dingle, W. Wiegman, and C. H. Henry, Phys. Rev. Lett. **33**, 827 (1974).

[4]Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

[5]M. K. Gurnick and T. A. DeTemple, IEEE J. Quantum Electron. **QE-19**, 791 (1983).

[6]J. Khurgin, *Second-Order Intersubband Nonlinear Optical Susceptibilities of Asymmetric Quantum Well Structures* (Optical Society of America, Washington, DC, 1989), pp. 69–72.

[7]D. Ahn and S. L. Chuang, IEEE J. Quantum Electron. **23**, 2196 (1987).

[8]M. M. Fejer, S. J. B. Yoo, R. L. Byer, A. Harwit, and J. S. Harris, Phys. Rev. Lett. **62**, 1041 (1989).

[9]P. F. Yuh and K. L. Wang, J. Appl. Phys. **65**, 4377 (1989).

[10]E. Rosencher, P. Bois, J. Nagle, and S. Delaître, Electron. Lett. **25**, 1063 (1989).

[11]E. Rosencher, P. Bois, and J. Nagle, SPIE Proc. **138**, 1273 (1990).

[12]R. P. Karunasiri, Y. J. Mii, and K. L. Wang, IEEE Electron Dev. Lett. **11**, 227 (1990).

[13]C. Cohen-Tannoudji, B. Diu, and F. Laloë, *Mécanique Quantique* (Hermann, Paris, 1973).

[14]L. Brey, N. F. Johnson, and B. I. Halperin, Phys. Rev. B **40**, 10 647 (1989).