

Intersubband optical response of semiconductor quantum wells dressed by strong infrared fields

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A strong infrared field tuned in resonance with dipole-allowed intersubband transitions in quantum-well conduction bands can act to coherently mix or dress the resonant states. The effects of dressing two excited conduction subbands on the linear response of a weak probe field nearly resonant with one of the excited subbands and the partially occupied ground subband is theoretically treated to arbitrary order in the coupling-field strength. Three critical coupling-field strengths are identified that demarcate: (i) the minimum coupling required to observe changes in the linear response, (ii) the onset of a resolvable doublet in the response, and (iii) the strong-coupling limit in which the line doubling is fully developed. Analytic expressions for these three critical fields are obtained in terms of the Rabi frequency and the phenomenological damping parameters associated with states in the three subbands.

I. INTRODUCTION

Since the observation of the very large dipole moment associated with intersubband optical transitions of a quantum well,¹ a great deal of attention has been devoted to electromagnetic interactions with the quantum-well conduction bands. These studies are motivated by both practical applications, such as infrared detectors in different wavelength ranges,^{2,3} and the semiconductor physics, such as excitonic effects,⁴ intersubband electron relaxation times,⁵ and others. The effect of a dc electric field on the resonant frequency and coupling strength of intersubband optical transitions with external optical fields has been considered.⁶ It has also been shown that in a two-conduction-subband system, a strong optical field can produce nonlinear optical effects similar to those observed in atomic two-level systems.⁷

In this study, we address the nonlinear effects, due to a strong optical field nearly in resonance with transitions between two of the conduction subbands. That such a field can mix, or dress the electronic levels with which it is resonant is well known,^{8,9} and the effect of this dressing on the system's response to optical fields that couple the dressed states with the ground state can be considerable, for both the real (refractive) and imaginary (absorptive) components. These concepts have recently been theoretically developed and experimentally examined^{10,11} in the case where the first and second subbands in an empty conduction band are coupled by a strong CO₂ laser-generated field, and this coupling is monitored through its effect on the interband linear excitonic response. Here, we theoretically study a different case where the probe monitors the effect of dressing two excited conduction subbands on the linear intersubband response from the partially occupied ground subband, all within the conduction band. We show that the real and imaginary components of the linear intersubband response are modified for coupling-field strengths greater than a characteristic value. A doubling in the probe spectrum is predicted for coupling fields exceeding a second characteristic value, and a strong-field limit is reached as the

coupling strength increases beyond a third critical level. Expressions for these three critical-field strengths are obtained in terms of the dipole matrix element between the two excited subbands, and the relaxation rates of the states in the three coupled subbands. The field intensity required to observe an effect due to this coherent coupling using CO₂ laser radiation and a conventional *n*-doped quantum well is of the order of ~ 250 kW/cm² at low temperatures.

The remainder of the paper is organized as follows. Section II contains a description of the formalism used to calculate the effects of an arbitrarily strong 10.6 μ m coupling field on the linear response of a doped quantum well system with at least three confined conduction subbands. Due to the crucial role played by the damping rates in this model, Sec. III discusses the underlying physics associated with the population relaxation rates and identifies ranges of realistic values for these parameters in different quantum well configurations. Section IV presents representative linear-response spectra calculated for this system. Section V contains a discussion of the results and concluding remarks are contained in Sec. VI.

II. INTERSUBBAND LINEAR RESPONSE OF THE INFRARED-DRESSED QUANTUM WELL

In symmetric quantum-well heterostructures, such as GaAs/Ga_{1-x}Al_xAs, there can be one or more bound conduction subbands depending on the value of the conduction-band offset.¹² These subbands can be strongly coupled to an infrared field polarized along the growth direction of the quantum well. For this reason the polarizations of both the coupling field, which is used to dress the first and second excited states of the conduction band, and the probe field, which monitors the intersubband transition in the vicinity of the ground to first excited subband, are assumed to be along the growth direction *z*.

The coupling field is near resonance with transitions between the first, $|1, \mathbf{k}\rangle$, and second, $|2, \mathbf{k}\rangle$ excited subband states. Here, \mathbf{k} refers to the in-plane component of the electron wave vector and 1 and 2 are the excited sub-

band indices. The probe field is near resonance with transitions between the ground, $|0, \mathbf{k}\rangle$ and the first excited subband state. Therefore, within the resonance approximation, all but these three subbands are ignored. In the basal plane, a parabolic band structure is assumed with equal in-plane masses for each subband. The detuning of the coupling field from the first to second excited subband states is, therefore, the same for all \mathbf{k} .

The band-structure contributions to the electronic part of the system's Hamiltonian are given by

$$H_0 = \sum_{n_c, \mathbf{k}} E^c(n_c, k) a_{n_c, \mathbf{k}}^\dagger a_{n_c, \mathbf{k}}. \quad (1)$$

Here, $E^c(n_c, k)$ is the energy eigenvalue of the quantum-well wave function. The operator $a_{n_c, \mathbf{k}}^\dagger$ creates an electron in a conduction subband, which is specified by \mathbf{k} and n_c , and $a_{n_c, \mathbf{k}}$ annihilates the same electron. The lowest subband of the quantum well is considered to be partially occupied by free electrons, but all other subbands are assumed to be empty. Due to the necessary presence of these free carriers in this problem, the Coulomb interaction between them should, in principle, also appear in the Hamiltonian. By ignoring this term in the interest of keeping the infrared coupling physics as transparent as possible, we omit effects due to screening, renormalization, correlations, and electron-electron scattering. Electron-electron scattering processes are accounted for phenomenologically below, while the other effects are simply ignored without further justification. However, we note that their neglect in this intersubband geometry should be less serious than in the interband situation, where excitonic correlations clearly must be included for a realistic treatment of the problem near the band edge.

Now let us consider the coupling of this electronic system with an infrared field. Within the rotating-wave and dipole approximations the interaction Hamiltonian of the system involves only the electric field, $E(t) = Ee^{i\omega t}$ and is given by

$$H_1 = - \sum_{\mathbf{k}, n_c, n'_c} \{ \mu_{n_c, n'_c} E(t) a_{n'_c, \mathbf{k}}^\dagger a_{n_c, \mathbf{k}} + \mu_{n_c, n'_c}^* E^*(t) a_{n_c, \mathbf{k}}^\dagger a_{n'_c, \mathbf{k}} \}. \quad (2)$$

Here, n_c and n'_c refer, respectively, to the lower- and higher-energy states of transitions. The μ 's are the electric dipole moments between the coupled conduction subbands, along the growth direction (assumed to \mathbf{k} independent).

Since by assumption the coupling field is in resonance with the first and second excited conduction subbands, the sum over n is omitted in the following. In addition, since the polarization of the coupling field is parallel to the growth direction, the electric-dipole moments in Eq. (2) are real.¹³ We shall work in the interaction picture in which the interaction Hamiltonian has the following form:

$$H_1 = \hbar \sum_{\mathbf{k}} \Omega \{ e^{i\Delta_k t} a_{2, \mathbf{k}}^\dagger a_{1, \mathbf{k}} + e^{-i\Delta_k t} a_{1, \mathbf{k}}^\dagger a_{2, \mathbf{k}} \}. \quad (3)$$

Here, $\Omega = -\mu_{12} E / \hbar$ is the Rabi frequency, and the detuning Δ_k is

$$\Delta_k = \omega_2^k - \omega_1^k - \omega, \quad (4)$$

where ω is the coupling-field frequency, and ω_2^k , and ω_1^k are defined as follows:

$$\omega_2^k = E^c(2, k) / \hbar, \quad (5)$$

$$\omega_1^k = E^c(1, k) / \hbar. \quad (6)$$

Here, the $E^c(1, k)$ and $E^c(2, k)$ are the first and second excited conduction-subband energies, respectively. As noted above, Δ_k is, in fact, independent of k since we assume each subband to have the same dispersion. The equation of motion for the density operator ρ' in the interaction picture is¹⁴

$$\frac{\partial \rho'}{\partial t} = - \frac{i}{\hbar} [H_1', \rho'] - \frac{1}{2} (\Gamma \rho' + \rho' \Gamma), \quad (7)$$

where Γ is a diagonal matrix whose elements Γ_{ii} are the inverse of the relaxation times for the states $|i, \mathbf{k}\rangle$. The term $\frac{1}{2} (\Gamma \rho' - \rho' \Gamma)$ is introduced phenomenologically to account for the damping, due to intrasubband and intersubband relaxation processes, which are assumed to be much faster than radiative relaxation rates. A more rigorous treatment of these dephasing processes would involve adding electron-phonon and electron-electron interaction terms to H_0 . As our purpose here is to elucidate the coupling physics and its general dependence on damping processes, we have purposely omitted specific interactions in H_0 . The proper incorporation of such terms is an area of research in its own right,¹⁵ and their inclusion here would merely complicate the coupling physics. Work towards including electron-phonon and electron-hole interactions in a three-subband quantum well system will be reported elsewhere.¹⁶

In order to obtain the equation of motion for each component of the density matrix, we make the following substitution for each set of the three-subband states at a given \mathbf{k} :

$$X_{00} = \rho'_{00} \quad X_{21} = \rho'_{21} e^{-i\Delta_k t}, \quad (8)$$

$$X_{11} = \rho'_{11} \quad X_{01} = \rho'_{01} e^{i\delta_k t}, \quad (9)$$

$$X_{22} = \rho'_{22} \quad X_{02} = \rho'_{02} e^{i\delta'_k t}, \quad (10)$$

where $\delta_k = \omega_1^k - \omega_0^k$ and $\delta'_k = \omega_2^k - \omega_0^k - \omega$, both of which are k independent for equal subbands dispersions. The transferred equations of motion are

$$\frac{dX_{00}}{dt} = -\Gamma_0 (X_{00} - X_{00}^0), \quad (11)$$

$$\frac{dX_{11}}{dt} = -i\Omega (X_{21} - X_{12}) - \Gamma_1 (X_{11} - X_{11}^0), \quad (12)$$

$$\frac{dX_{22}}{dt} = -i\Omega (X_{12} - X_{21}) - \Gamma_2 (X_{22} - X_{22}^0), \quad (13)$$

$$\frac{dX_{01}}{dt} = [i\delta - \gamma_{01}] X_{01} + i\Omega X_{02}, \quad (14)$$

$$\frac{dX_{02}}{dt} = [i\delta' - \gamma_{02}]X_{02} + i\Omega X_{01}, \quad (15)$$

$$\frac{dX_{12}}{dt} = [i\Delta - \gamma_{12}]X_{12} - i\Omega(X_{22} - X_{11}). \quad (16)$$

Here, we have $X_{ij} = X_{ji}^*$ for which the k indices are suppressed from now on. The X_{ii}^0 refer to the equilibrium probability of finding electrons in the states $|i, \mathbf{k}\rangle$ and, therefore, they are represented by Fermi distributions. Γ_0 is the relaxation constant for slight perturbations of the Fermi-Dirac distribution of carriers in the lowest subband. In this form, Eq. (11) is equivalent to the relaxation time approximation of the Boltzmann equation.¹⁷ The limits of applicability of this equation to treat the dynamics of the kinetic hole left in the Fermi distribution following stimulated emission in an inverted semiconductor has been considered by others.¹⁷ The other relaxation constants Γ_1 and Γ_2 are associated with the decay of carriers excited into the higher subbands, where there is no equilibrium population of free electrons. All three of these rates are typically due to electron-phonon and electron-electron scattering processes. As stated above, microscopic expression for these rates can self-consistently be obtained by including corresponding interaction terms in the Hamiltonian, but here we treat them purely phenomenologically. At this phenomenological level, if polarization scattering is ignored the polarization dephasing constants, γ_{ij} , are related to the population scattering rates as¹⁵

$$\gamma_{ij} = \frac{1}{2}(\Gamma_i + \Gamma_j).$$

Eqs. (11)–(16) can be further reduced to a simple linear matrix equation of motion for a nine element column vector Φ :

$$\frac{d\Phi}{dt} = \mathbf{L}\Phi + \mathbf{K}, \quad (17)$$

where the Φ elements are

$$\begin{aligned} \Phi_1 &= X_{11}, & \Phi_2 &= X_{12}, & \Phi_3 &= X_{02}, \\ \Phi_4 &= X_{21}, & \Phi_5 &= X_{20}, & \Phi_6 &= X_{10}, \\ \Phi_7 &= X_{00}, & \Phi_8 &= X_{01}, & \Phi_9 &= X_{22}. \end{aligned} \quad (18)$$

The elements of the matrix \mathbf{L} and the column vector \mathbf{K} can be easily obtained by inspection of Eqs. (11)–(16), realizing that all of the X_{ii}^0 are zero except X_{00}^0 .

Equation (17) describes a system consisting of a strong-coupling field and three of the conduction subbands of the quantum well. In the following, we use linear-response theory to obtain the response of this system to a weak probe field.¹⁸ The approach, explained in detail in the Appendix, involves using the quantum regression theorem to relate the linear response to the Laplace transform of two-time polarization correlation functions. For simplicity, numerical results are only evaluated in the steady-state limit, which is formally obtained by taking the infinite time limit of the polarization correlation functions. In practice, the system will achieve a quasi-steady-state that these results will adequately de-

scribe so long as the coupling-field pulse duration, τ_p , is such that $1/\tau_p \ll \gamma_{12}$.

The imaginary and real parts of the system susceptibility in linear-response theory are related to the real and imaginary parts, respectively, of the following two-time correlation function characteristic of this system:¹⁹

$$-\sum_{\mathbf{k}} f_{\mathbf{k}} \langle [\hat{p}_{\mathbf{k}}^-(z, t'), p_{\mathbf{k}}^+(t')] \rangle |_{z=i\omega_p}. \quad (19)$$

Here, $f_{\mathbf{k}}$ is the Fermi-Dirac distribution factor (nonzero only for a range of states in the lowest subband) and ω_p refers to the probe frequency. The $p_{\mathbf{k}}^-$ and $p_{\mathbf{k}}^+$ are negative and positive frequency components of the polarization associated with the coherent superpositions of the three subband states at different \mathbf{k} ; they are defined in the Heisenberg picture by

$$p_{\mathbf{k}}^+(t) = \mu_{01} a_{0,\mathbf{k}}^\dagger(t) a_{1,\mathbf{k}}(t) + \mu_{12} a_{1,\mathbf{k}}^\dagger(t) a_{2,\mathbf{k}}(t), \quad (20)$$

$$p_{\mathbf{k}}^-(t) = \mu_{01} a_{1,\mathbf{k}}^\dagger(t) a_{0,\mathbf{k}}(t) + \mu_{12} a_{2,\mathbf{k}}^\dagger(t) a_{1,\mathbf{k}}(t). \quad (21)$$

Here, $\hat{p}_{\mathbf{k}}^-(z, t')$ is the Laplace transform of $p_{\mathbf{k}}^-(t) = p_{\mathbf{k}}^-(t' + \tau)$, with respect to $\tau = t - t'$; $\tau > 0$.

The μ_{01} and μ_{12} are the electric-dipole moments for the transitions from the ground state to the first excited state and from the first to the second excited state, respectively. Because, in practice, we are looking for the linear response to a weak probe tuned near the transition frequency from the ground to the first excited state of the conduction band, the terms including μ_{12} in Eqs. (20)–(21) are off resonant, and can be ignored.

In order to calculate $\langle [\hat{p}_{\mathbf{k}}^-(z, t'), p_{\mathbf{k}}^+(t')] \rangle$ in Eq. (19), we have to find the correlation functions $\langle \hat{p}_{\mathbf{k}}^-(z, t') p_{\mathbf{k}}^+(t') \rangle$ and $\langle p_{\mathbf{k}}^+(t') \hat{p}_{\mathbf{k}}^-(z, t') \rangle$. The details are presented in the Appendix. Briefly, the quantum regression theorem is used to obtain these unequal-time correlation functions from $\langle \hat{p}_{\mathbf{k}}^-(z, t') \rangle$.²⁰

In order to obtain $\langle p_{\mathbf{k}}^-(t) \rangle$, we make use of the density operator as follows:

$$\begin{aligned} \langle p_{\mathbf{k}}^-(t) \rangle &= \text{Tr} \{ \rho(t) \mu_{01} a_{1,\mathbf{k}}^\dagger a_{0,\mathbf{k}} \} \\ &= \mu_{01} \Phi_8(t). \end{aligned} \quad (22)$$

The Laplace transform of Eq. (22) is given by

$$\langle \hat{p}_{\mathbf{k}}^-(z, t') \rangle = \mu_{01} \hat{\Phi}_8(z, t'). \quad (23)$$

Referring to the Laplace transform of Eq. (17), we now obtain

$$\hat{\Phi}(z, t') = \mathbf{R}(z)\Phi(t') + \frac{1}{z}\mathbf{R}(z)\mathbf{K}, \quad (24)$$

with

$$\mathbf{R}(z) = (z\mathbf{I} - \mathbf{L})^{-1},$$

where \mathbf{I} is the identity matrix. Here again $\hat{\Phi}(z, t')$ refers to the Laplace transform of $\Phi(t)$, with respect to τ . We then can calculate $\hat{\Phi}_8(z, t')$ and $\langle \hat{p}_{\mathbf{k}}^-(z, t') \rangle$ as follows:

$$\langle \hat{p}_{\mathbf{k}}^-(z, t') \rangle = \mu_{01} \sum_{j=1}^9 \left\{ R_{8j}(z) \Phi_j(t') + \frac{1}{z} R_{8j}(z) K_j \right\}. \quad (25)$$

The expression for $\langle \hat{p}_k^-(z, t') p_k^+(t') \rangle$ and $\langle p_k^+(t') \hat{p}_k^-(z, t') \rangle$ then follow,

$$\begin{aligned} \langle [\hat{p}_k^-(z), \hat{p}_k^+(\infty)] \rangle = & \mu_{01}^2 \{ R_{83}(z) \Phi_2(\infty) \\ & + R_{88} z [\Phi_1(\infty) - \Phi_7(\infty)] \}. \end{aligned} \quad (26)$$

The detailed deviation of Eq. (26) is given in the Appendix. In order to impose steady-state conditions on the above equation, we put $t' \rightarrow \infty$ (and $t \rightarrow \infty$, $t > t'$). The absorptive and refractive components of the linear response spectra are then obtained by substitution of Eq. (26) in Eq. (19).

III. RELAXATION RATES

The set of equations implicit in Eq. (17) are valid for any system that has three relatively isolated levels within which there are two dipole-allowed transitions. Since the dipole moments act only to scale the fields, the qualitative effects of the coupling field on the linear response are completely determined by the relative values of the dephasing rates, γ_{01} , γ_{02} , γ_{12} . Because the results presented below for the quantum-well case are so dramatically different from those reported for "typical" atomic systems, we devote this section to a discussion of the dephasing rates appropriate for quantum-well samples.

Before discussing these rates it should be noted that the total intersubband linear response is the sum of the individual responses, due to all sets of \mathbf{k} states with nonzero occupation probability [Eq. (19)]. Because we assume that the dispersion of each subband is identical, and that the dipole moments are \mathbf{k} independent, the summation would be trivial if the dephasing rates were also \mathbf{k} independent (i.e., the correlation function would only have to be evaluated once). In most practical situations the dephasing rates will in fact depend on \mathbf{k} (as discussed below), however, it turns out that the qualitative effect of the coupling field on the system's linear response is similar for all realistic sets of γ_{01} , γ_{02} , γ_{12} . For simplicity then, after discussing the origins and typical magnitudes of the \mathbf{k} -dependent relaxation rates, representative values are used to generate numerical results, and no attempt is made to evaluate the complete sum in Eq. (19). All of the pertinent physics of the nonlinear coupling is thus revealed without complications, due to model-specific effects of averaging.

At low temperatures the range of occupied k states is essentially from $k=0$ to k_f , where the Fermi wave vector k_f is given in terms of the density, n , by

$$k_f = (2\pi n)^{0.5}.$$

Γ_0 is the relaxation rate of small perturbations of the electron distribution within the Fermi sea, towards a unity occupation factor. For Fermi energies less than the longitudinal-optical LO-phonon frequency, this relaxation rate of the small perturbations created by optical excitation out of the Fermi sea is determined by electron-electron scattering among the free electrons.^{17,21} Γ_0 is on the order of $\sim 10 \text{ ps}^{-1}$ near the band edge and $\sim 0.1 \text{ ps}^{-1}$

near k_f for effective bulk electron densities of $\sim 5 \times 10^{17} \text{ cm}^{-3}$.¹⁷

The scattering rates of the excited states at a given \mathbf{k} , Γ_1 , and Γ_2 , depend strongly on the intersubband spacings and the carrier density in the ground subband. For excited-state electrons within an LO-phonon energy of the Fermi level, their relaxation rate will be determined by interactions with acoustic phonons²² and free carriers in the lowest subband. Acoustic-phonon scattering rates are on the order of 0.1 ps^{-1} and relatively insensitive to k . Excited-state electrons with energies more than an LO-phonon energy above the Fermi level will scatter primarily via LO phonon emission, with an additional contribution from electron-electron interactions. The LO-phonon scattering rate will be on the order of 1.2 ps^{-1} , due to intersubband scattering if the kinetic energy of the electron is less than an LO-phonon energy above its subband edge, increasing to $\sim 5 \text{ ps}^{-1}$ if it is allowed to undergo intrasubband LO-phonon scattering.

IV. RESULTS

To illustrate the overall effects of optical dressing in a quantum well, we use representative values of $\Gamma_0 = 7 \text{ ps}^{-1}$, $\Gamma_1 = 1.5 \text{ ps}^{-1}$, and $\Gamma_2 = 1.5 \text{ ps}^{-1}$ in the evaluation of the real and imaginary components of Eq. (19). These values correspond to a case in which the energy spacing between the ground and first excited subbands, δE , [$\delta E = E^c(1, k) - E^c(0, k)$] is larger than 50 meV.

Figure 1 shows the linear-response spectra of the quantum well at different values of the coupling-field intensity. Here the Rabi frequency has been allowed to range from zero to the level required to observe well-defined satellite peaks. As Fig. 1 (solid line) shows, the resulting zero-field linewidth with the chosen damping parameters is $\sim 6 \text{ meV}$, similar to experimental absorption spectra of 8.5-nm GaAs wells with $\text{Al}_{0.35}\text{Ga}_{0.65}\text{As}$ barriers.⁵ The absolute CO_2 intensities corresponding to these Rabi frequencies depend of course on the dipole matrix elements. As an estimate, taking $\mu_{12} = e \times 2 \text{ nm}$, $\Omega = 1 \text{ ps}^{-1}$ corresponds to an intensity $I = 0.06 \text{ MW/cm}^{-2}$, and of course I scales as the square of the Rabi frequency.

As illustrated in Fig. 1, the coherent modification of the linear absorption first manifests itself as a broadening of the zero-field line shape, accompanied by a reduction in the peak response. At higher Rabi frequencies the line evolves into a doublet, and eventually each line in the doublet becomes well defined and any further increase in the coupling-field strength merely increases the separation, leaving the individual widths and amplitudes fixed. The total area under the absorption curves (proportional to the imaginary part of the susceptibility) is independent of the coupling field strength in all cases. The evolution of the spectra is qualitatively similar for other realistic combinations of γ_{01} , γ_{02} , γ_{12} ; quantitative differences are found in the widths and the peak heights of the responses.

In order to determine the effect of detuning the coupling field above the 1–2 resonance, we show in Fig. 2 the absorption spectrum for the same parameters as those of Fig. 1 for various detunings. At $\Omega = 25 \text{ ps}^{-1}$, when the

detuning, Δ , is zero, a symmetric splitting is created, as shown by Fig. 1. But with nonzero detuning, the peaks are asymmetric with the lower-energy peak stronger. At $\Delta=6$ meV, the interaction of the coupling field with the quantum well is weak, hence the lower-energy peak approaches the uncoupled absorption peak.

V. DISCUSSION

The values of Ω used to obtain the spectra in Fig. 1 were chosen to demonstrate qualitatively different regimes in the evolution of linear spectra with increasing

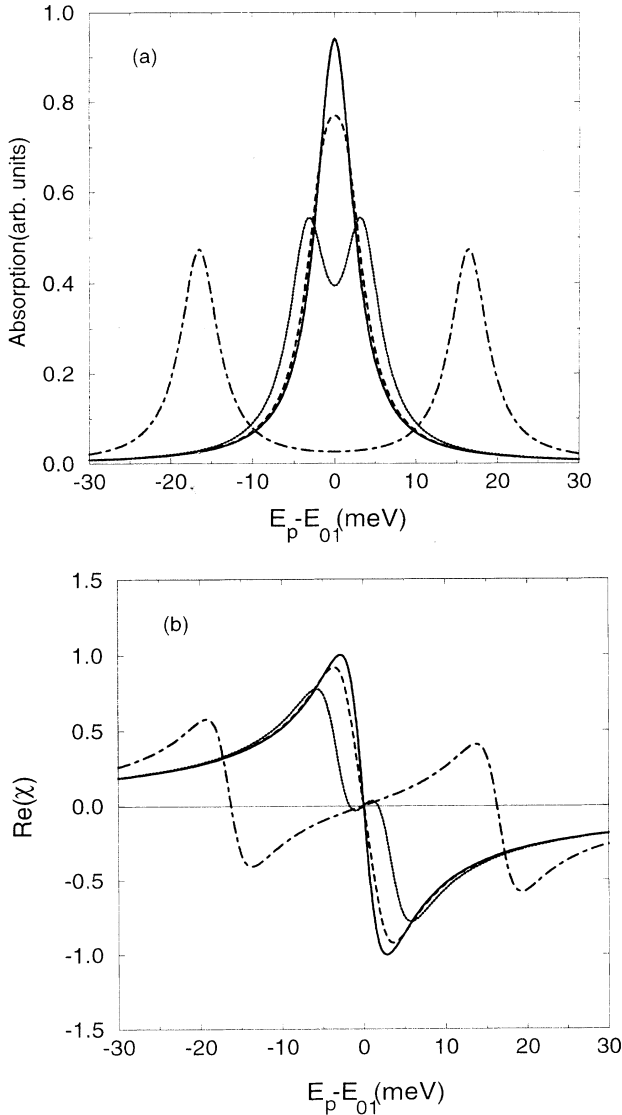


FIG. 1. Intersubband absorption, (a), and refractive response normalized to the peak magnitude of the uncoupled resonance, (b), using average relaxation rates of $\Gamma_0=7$ ps $^{-1}$, $\Gamma_1=1.5$ ps $^{-1}$, $\Gamma_2=1.5$ ps $^{-1}$, and detuning, $\Delta=0$. The Rabi frequencies are $\Omega=0$ ps $^{-1}$ (solid line), $\Omega=2$ ps $^{-1}$ (dashed line), $\Omega=5$ ps $^{-1}$ (dotted line), and $\Omega=25$ ps $^{-1}$ (dashed-dotted line). E_p and E_{01} refer to energies of the probe and ground to first excited state, respectively.

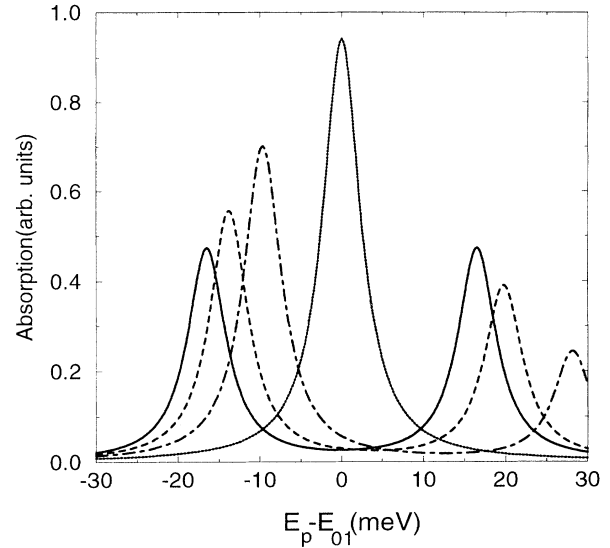


FIG. 2. Intersubband absorption at $\Omega=25$ ps $^{-1}$ for different coupling-field detunings. The solid line corresponds to $\Delta=0$, dashed line to $\Delta=6$ meV, and dashed-dotted line to $\Delta=18$ meV. The dotted line is for $\Omega=0$ ps $^{-1}$ and $\Delta=0$. All relaxation rates are the same as Fig. 1.

coupling-field strength. The effect of relatively weak coupling fields is to broaden the uncoupled absorption peak, thus coherently increasing the transparency at line center. At higher fields the induced spectra develop a doublet structure, and eventually a high-field limit is reached beyond which two well-defined satellite peaks maintain their width as they become further separated. The width of the satellite peaks is at least as large as that of the uncoupled absorption peak.

This evolution is qualitatively different from what has been calculated and observed in “typical” atomic systems.²³ In the atomic case, even weak-coupling fields induce a doublet structure, and further increases of the coupling field merely separate two well-defined lines with widths equal to *half* the uncoupled linewidth, and peak amplitudes unchanged from those in the absence of coupling. Figure 3 illustrates this case by showing the result of solving the present model, using $\Gamma_0=0.001$ ps $^{-1}$, $\Gamma_1=1$ ps $^{-1}$, and $\Gamma_2=0.001$ ps $^{-1}$, to mimic the typical atomic situation where the ground state is stable, and the excited-state broadening is primarily radiative in nature.

These seemingly disparate nonlinear responses of atomic and quantum well systems, and in fact the overall qualitative behavior of both systems, can be understood using the dressed state picture.²⁴ Within the approximations in our model, the coupling field renormalizes the two nearly resonant excited states and leaves the ground state unaffected. The spectra can, therefore, be considered to be due to weakly probing the renormalized excited states from a single, unrenormalized ground state. One immediate consequence is that any structure that may exist within the dressed manifold will only be resolvable from the ground state if its width is of the order of the ground-state width, Γ_0 . Thus, the ground-state width

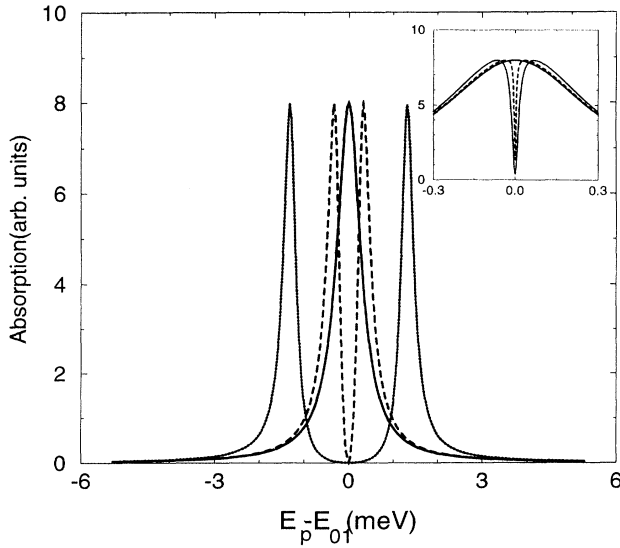


FIG. 3. Intersubband absorption with relaxation rates of $\Gamma_0=0.001 \text{ ps}^{-1}$, $\Gamma_1=1 \text{ ps}^{-1}$, and $\Gamma_2=0.001 \text{ ps}^{-1}$ and detuning, $\Delta=0$ for various Rabi frequencies, $\Omega=0 \text{ ps}^{-1}$ (solid line), $\Omega=0.5 \text{ ps}^{-1}$ (dashed line), and $\Omega=2 \text{ ps}^{-1}$ (dotted line). The inset shows absorption spectra at an early stage of development of the doublet for $\Omega=0 \text{ ps}^{-1}$ (solid line), $\Omega=0.05 \text{ ps}^{-1}$ (dashed line), and $\Omega=0.1 \text{ ps}^{-1}$ (dotted line).

enters the problem as a resolution, or convolution parameter that can be factored into the final spectra after addressing the more interesting issues associated with the dressing of the excited states.

The strong-coupling regime is easiest to understand. Once the Rabi frequency characteristic of the coupling-field strength and the dipole moment between the excited states is much greater than the dephasing rate associated with the transition, γ_{12} , the effect of the coupling field is essentially to lift the degeneracy of the first excited state with N coupling photons present, $|1, N\omega\rangle$, and the second excited state with $(N-1)$ coupling photons present, $|2, (N-1)\omega\rangle$. The well-known result is two non-degenerate superposition states that share the linewidths and oscillator strengths (with respect to the ground state) of the original levels, and that are separated by 2Ω . Including the effect of the ground-state width on the ability to resolve this splitting, the strong-field regime occurs for coupling-field strengths greater than $2(\gamma_{01} + \gamma_{02})$. In both the atomic case and quantum-well regimes, this limit corresponds to the point when the satellite peaks each have essentially a Lorentzian shape, with a full width at half maximum linewidth of $(\gamma_{01} + \gamma_{02})$. Using the fact that the area under the absorption peaks is independent of the coupling field, it is straight forward to give a simple expression for the ratio of the strong-field satellite peak amplitudes (A') to the uncoupled amplitude (A):

$$\frac{A}{A'} = \frac{\gamma_{01} + \gamma_{02}}{\gamma_{01}}.$$

In the atomic case, γ_{02} is usually negligible (Fig. 3), whereas in the quantum-well case it is typically at least as

large as γ_{01} (Fig. 1).

In the weak-field limit, all of the behavior exhibited in the atomic and quantum well regimes can be understood by considering the wider of the two excited states as offering an effective continuum of states that can couple with the narrower of the two states. When the dipole-allowed first excited states, $|1, N\omega\rangle$, is the narrower of the two excited states, it can be thought of as mixing with a continuum of dipole nonallowed states within the linewidth of the second excited state. Borrowing the nomenclature common in the discussion of Fano resonances, this corresponds to the limit where the Fano parameter (in this case the ratio of the dipole matrix elements between the ground state and the narrower state and between the ground and the effective continuum states) is essentially infinite.²⁴ The dressed version of the dipole-allowed state that results after coupling is included then consists of a single Lorentzian peak, broadened in proportion to the coupling field, Ω . The transition to this dressed state is dipole allowed from the ground state and hence the absorption spectrum in this case evolves as a broadened Lorentzian (see Fig. 1).

If the dipole nonallowed state, $|2, (N-1)\omega\rangle$, is the narrower of the two excited states (corresponding to a Fano parameter equal to zero), the dressed state develops as a Lorentzian shaped "hole" driven into the broad, dipole-allowed "effective continuum" associated with the state $|1, N\omega\rangle$.²⁴ This is the situation illustrated in Fig. 3. The inset of Fig. 3 shows that the depth of the hole depends on the intensity of the coupling field.^{25,26} In either of these cases, no significant coupling effects are expected unless the Rabi frequency exceeds the narrower of the two excited-state linewidths. This concept can be generalized to include the resolution limits imposed by the ground state, and to include the intermediate case where the excited state linewidths are comparable. The result is that coupling effects should become observable for Rabi frequencies $\Omega > 1/[(1/\gamma_{01}) + (1/\gamma_{02})]$. In the small Γ_2/Γ_1 limit (the "typical" atomic limit) this effect will manifest itself from the outset as a doublet structure. In the large Γ_2/Γ_1 limit it will first occur as a broadening of a single line, and this will give way to a doublet structure when the width of this dressed state becomes comparable to the width of the broader, second excited state. In general then, the doublet structure will become apparent when $\Omega > \gamma_{02}$. These threshold expressions have been numerically verified by studying a wide range of γ_{01} , γ_{02} , γ_{12} parameter space.

VI. CONCLUSIONS

In summary, we have studied the way in which an arbitrarily strong CO_2 laser field can theoretically modify the linear response of the conduction subbands in a doped quantum well system with three bound subbands. It was shown that when the coupling field is in resonance with the two excited subbands, the magnitude of the absorption peak associated with the ground-to-first excited subband transition can be reduced significantly at the line center when the associated Rabi frequency exceeds $1/[(1/\gamma_{01}) + (1/\gamma_{02})]$, where the γ_{ij} are the polarization

damping rates associated with the subscripted levels. This reduction is followed by a doubling in the absorption spectrum for Rabi frequencies that exceed γ_{02} , and this doublet evolves into a set of well-defined satellite peaks that retain their shape and merely shift further apart for Rabi frequencies that exceed $2(\gamma_{01} + \gamma_{02})$. Due to the presence of the ground-state relaxation rate, Γ_0 , in all of these critical frequencies, the scattering rate of small perturbations of the Fermi distribution in the ground conduction subband plays a crucial role in determining the field strength required to observe coupling effects.

The present formalism puts no limitation on the intensity of the coupling field, and is extendible to include more subbands and/or more than one coupling field. Both of these more general cases offer new opportunities for nonlinear quantum well optics.

The results in this paper are not restricted to continuous excitation conditions. The formalism can be applied in a quasistationary way for pulsed-infrared coupling fields so long as the pulse width, τ_p , is long enough that $1/\tau_p \ll \gamma_{12}$.

APPENDIX

Here, we present some intermediate steps of the deviation of Eq. (26) from Eq. (25) by the use of the quantum regression theorem. Before proceeding let us recall that the restriction $t > t'$ in the unequal-time correlation functions $\langle [p_k^-(t), p_k^+(t')] \rangle$ can be expressed explicitly by putting $t = t' + \tau$ for $\tau > 0$. The linear susceptibility of the system can then be related to the Laplace transform of these correlation functions:

$$\int_0^\infty \langle p_k^-(t' + \tau), p_k^+(t') \rangle e^{-z\tau} d\tau \quad (\text{A1})$$

taken at $z = i\omega_p$ [as stated in Eq. (19)].^{19,27} In this paper, all Laplace transforms with respect to τ depend both on t' and z .

The quantum regression theorem describes the motion of a system operator M in terms of a set of system operators, $\{M_\mu\}$. Here M could be one member of the set $\{M_\mu\}$ or a linear combination of them. The time evolution of such an operator can be written as follows:²⁰

$$\langle M(t) \rangle = \sum_\mu O_\mu(t, t') \langle M_\mu(t') \rangle + \lambda_\mu, \quad (\text{A2})$$

where $t > t'$. If the numerical coefficients $O_\mu(t, t')$ are known for a single set of t and t' , then the mean of a two-time operator can be obtained by

$$\langle M(t)N(t') \rangle = \sum_\mu O_\mu(t, t') \langle M_\mu(t')N(t') \rangle + \lambda_\mu \langle N(t') \rangle, \quad (\text{A3})$$

where N can be any system operator and t' must precede the time t .

In order to use this theorem for calculation of the unequal-time correlation functions in Eq. (19), we first identify a complete set of operators, $\{M_\mu\}$ which fully describes our system. A suitable choice is the set of operators $\{a_i^\dagger a_j\}$ that are related to the averages Φ_i of

Eq. (18). Here, i and j can take values of 0, 1, or 2. The next step is to find the O_μ by writing an expression for the one-time average analogous to Eq. (A2). With respect to the correlation functions we are looking for, we need only to set $M = p_k^-(t)$ and then compare Eq. (25) with Eq. (A2) to specify O_μ .

We proceed next by introducing the $p_k^+(t')$ as $N(t')$ in Eq. (25). In this way the left-hand side of Eq. (25) is converted to $\langle \hat{p}_k^-(z, t') p_k^+(t') \rangle$, one of our desired results. On the right-hand side, we must then evaluate the expectation values of $M_\mu(t') p_k^+(t')$ and $p_k^+(t')$. This has the effect of replacing the $\{\Phi\}$ in Eq. (25) by

$$\begin{aligned} \Phi_1(t') &\rightarrow \mu_{12} e^{-i\omega t'} \Phi_4(t'), \\ \Phi_2(t') &\rightarrow \mu_{12} e^{-i\omega t'} \Phi_9(t'), \quad \Phi_3(t') \rightarrow \mu_{01} \Phi_2(t'), \\ \Phi_4(t') &\rightarrow 0, \quad \Phi_5(t') \rightarrow 0, \\ \Phi_6(t') &\rightarrow \mu_{12} e^{-i\omega t'} \Phi_5(t'), \quad \Phi_7(t') \rightarrow \mu_{01} \Phi_6(t'), \\ \Phi_8(t') &\rightarrow \mu_{01} \Phi_1(t'), \quad \Phi_9(t') \rightarrow 0, \end{aligned}$$

and the substitution of K_j by $K_j \langle p_k^+(t') \rangle$. The result after putting $z = i\omega_p$ is a general result for the linear-response theory for the susceptibility of the system. But since in this paper we are interested in the response of the system in the steady state, the condition $t' \rightarrow \infty$ ($t \rightarrow \infty$) is imposed. Therefore, the $\{\Phi\}$'s in Eq. (17) take the following simple forms:

$$\Phi_i(\infty) = - \sum_j (\mathbf{L}^{-1})_{ij} \mathbf{K}_j. \quad (\text{A4})$$

Also because of the steady-state condition, the terms in $\langle \hat{p}_k^-(z, t') p_k^+(t') \rangle$ which correspond to $e^{-i\omega t}$ and $e^{-i\omega t'}$, make no contribution to the correlation functions and only the terms which evolve as $e^{-i\omega(t-t')}(e^{i\omega\tau})$ remain effective.

The result, after removing the vanishing terms in the steady-state limit and retaining only nonzero R_{ij} is

$$\langle \hat{p}_k^-(z) p_k^+(\infty) \rangle = \mu_{01}^2 \left\{ R_{83}(z) \Phi_2(\infty) + R_{88}(z) \Phi_1(\infty) + \sum_{j=1}^9 \frac{1}{z} R_{8j}(z) \Phi_6(\infty) K_j \right\}. \quad (\text{A5})$$

Following the same procedure, one can obtain an expression for $\langle p_k^+(\infty) \hat{p}_k^-(z) \rangle$ as follows:

$$\begin{aligned} \langle p_k^+(\infty) \hat{p}_k^-(z) \rangle &= \mu_{01}^2 \left\{ R_{88}(z) \Phi_7(\infty) + \sum_{j=1}^9 \frac{1}{z} R_{8j}(z) \Phi_6(\infty) K_j \right\}, \end{aligned} \quad (\text{A6})$$

and, therefore, Eq. (26) is obtained as

$$\begin{aligned} \langle [\hat{p}_k^-(z), \hat{p}_k^+(\infty)] \rangle &= \mu_{01}^2 \{ R_{83}(z) \Phi_2(\infty) \\ &\quad + R_{88}(z) [\Phi_1(\infty) - \Phi_7(\infty)] \}. \end{aligned} \quad (\text{A7})$$

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