Multiwave mixing in semiconductor laser media

Murray Sargent III, Fenglei Zhou, and Stephan W. Koch*
Optical Sciences Center, University of Arizona, Tucson, Arizona 85721
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This paper analyzes the interaction of a strong wave and one or two probe waves with an intrinsic semiconductor medium which is partially inverted by an injection current. The many-body interactions of the electron-hole excitations are described using recently developed generalized Bloch equations for semiconductors. Coupled-mode equations for the different light fields are derived and solved, and predict that the gain and the coupling coefficients experience asymmetric dips. These dips are generated by scattering of the strong field off carrier-density pulsations induced by the pump-probe interference.

I. INTRODUCTION

In this paper we apply the generalized Maxwell-Bloch equations of motion for semiconductors given by Lindberg and Koch[1] to the problem of two- and three-wave mixing in a partially inverted semiconductor medium. These generalized Bloch equations contain the many-body effects of plasma screening, Coulomb enhancement, band filling, and plasma-density-dependent renormalization of the semiconductor band gap. Similar generalized Bloch equations have also been derived in Refs. 2 and 3. Considering fields that vary little in carrier-carrier and carrier-phonon interaction times, we adopt the so-called quasiequilibrium approximation where the intraband carrier distributions are given in terms of quasi-Fermi functions. For room-temperature conditions and sufficiently high carrier densities where the medium is partially inverted, the Coulomb enhancement becomes unimportant in the gain regime and the generalized Bloch equations can be simplified.[4] However, in a consistent description of the high-density electron-hole excitations one always has to include the electronic band-gap renormalization which leads to tuning of the semiconductor gain spectra. Therefore our equations are a generalization of the semiconductor laser equations used by several authors.[5–9]

To analyze the effects of multiwave mixing, we consider the situation where the optical field consists of one or two probe waves with intensities too small to change the carrier density, and a strong wave whose intensity is limited only by the condition that its Rabi-flopping frequency is small compared to the carrier-carrier scattering rate. We use multimode Fourier expansion[10] to derive the equations for the different field components. For the case of one probe wave our analysis shows the appearance of dips in the optical gain and transmission spectra. The situation of two probe waves is relevant for the situation of nondegenerate phase conjugation in semiconductors. Our analysis shows that single-probe propagation is described by Beer's law, and the two-probe propagation is described by coupled-mode equations. The analysis is important for pump-probe spectroscopy, modulation spectroscopy, side-mode buildup in semiconductor lasers, and many other wave-mixing phenomena in semiconductors.

In Sec. II of this paper we present our model for the injection-pumped semiconductor interacting with the multimode light field. We discuss the relevant many-body effects in the electron-hole plasma and derive the coupled equations for the carrier density and the medium polarization. In Sec. III we present the multimode expansion of the equations and compute the coupling equations for the different field components and the probe absorption coefficient. In Sec. IV we evaluate our equations numerically and show that the multiware mixing leads to the occurrence of dips in the probe-absorption spectra.

II. PROBE POLARIZATION OF THE MEDIUM

We consider a semiconductor medium subjected to an arbitrarily intense wave and one or two nonsaturating waves. We assume that the saturating-wave intensity is constant throughout the interaction region and ignore transverse variations. We label the probe waves by the indices 1 and 3 and the saturator wave by 2 as shown in Fig. 1. As discussed in Ref. 10, this scenario can be ap-

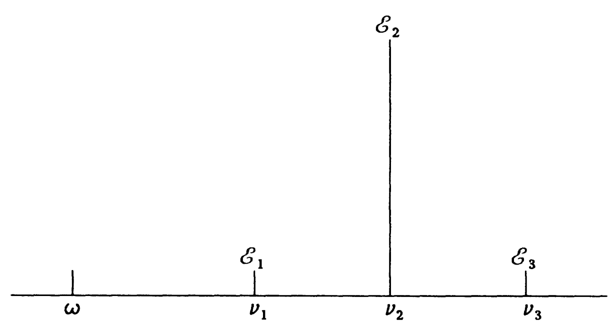

FIG. 1. Spectrum of three-wave field. Waves with frequencies ν_1 and ν_3 are taken to be weak (nonsaturating), while the ν_2 wave is allowed to be arbitrarily intense, provided that its Rabi frequency is still small compared to the carrier-carrier scattering rate.

plied for a large variety of multiwave mixing configurations. Our electric field has the form

$$E(\mathbf{r},t)=\frac{1}{2}\sum_n \mathcal{E}_n(\mathbf{r})e^{i(\mathbf{K}_n\cdot\mathbf{r}-\nu_n t)}+\text{c.c.}\ , \tag{1}$$

where the mode amplitudes $\mathcal{E}_n(\mathbf{r})$ are in general complex and $\mathbf{K}_n$ are the wave propagation vectors. For simplicity we take mode functions appropriate for a unidirectional ring laser. The mode index equals 1, 2, or 3. The field (1) induces the complex polarization

$$P(\mathbf{r},t)=\frac{1}{2}\sum_n \mathcal{P}_n(\mathbf{r})e^{i(\mathbf{K}_n\cdot\mathbf{r}-\nu_n t)}+\text{c.c.}\ , \tag{2}$$

where $\mathcal{P}_n(\mathbf{r})$ is a complex polarization coefficient that yields index and absorption and gain characteristics for the side-mode and saturator waves. The polarization $P(\mathbf{r},t)$ in general has other components, but we are interested only in those given by Eq. (2). In particular, strong wave interactions induce components not only at the frequencies ν_1, ν_2, and ν_3 but at $\nu_1 \pm j(\nu_2-\nu_1)$ as well, where j is an integer.

The problem reduces to determining the probe polarization $\mathcal{P}_1(\mathbf{r})$, which drives the side-mode amplitude $\mathcal{E}_n$ according to the propagation equation

$$\frac{d\mathcal{E}_n}{dz}=i(K/2\epsilon_0)\mathcal{P}_n\ , \tag{3}$$

where ϵ_0 is the background dielectric constant of the host medium. One might guess that the probe polarization is simply a probe Lorentzian multiplied by a probability difference saturated by the saturator wave. However, an additional contribution enters due to probability pulsations. Specifically, the nonlinear probabilities respond to the superposition of the modes to give pulsations at the beat frequency $\Delta=\nu_2-\nu_1$. Since we suppose the probe wave does not saturate, the pulsations occur only at $\pm\Delta$, a point proven below. These pulsations act as modulators (or like Raman "shifters"), putting side bands onto the medium's response to the ν_2 wave. One of these side bands fall precisely at ν_1, yielding a contribution to the probe-absorption coefficient. The other sideband influences the polarization of the probe placed symmetrically on the other side of the strong mode, namely, at the frequency $\nu_2+\nu_2-\nu_1$. This side band provides the coupling in three- and four-wave mixing.

The polarization (2) of the semiconductor medium can be also written as

$$P(z,t)=V^{-1}\sum_k \wp\rho_{cv}+\text{c.c.} \tag{4}$$

where $\wp$ is the electric-dipole matrix element which in general is k-dependent, V is the volume of the medium, ρ_{cv} is the off-diagonal element of the two-band density matrix whose elements are functions of the carrier wave vector k. The subscript c refers to the conduction band and v to the valence band. The volume V cancels out since it appears in the $\sum_k$. Combining Eqs. (2) and (4), we find the slowly varying complex polarization

$$\mathcal{P}=2\wp e^{-i(Kz-\nu t)}V^{-1}\sum_k \rho_{cv}\ . \tag{5}$$

The dynamic equations for ρ_{cv} and the electron and hole densities have been derived in Ref. 1. Here we briefly summarize the results that are relevant for the semiconductor laser. In the two-band approximation the Hamiltonian of the electron-hole system is

$$\mathcal{H}=\sum_k[\epsilon_e^{\text{eff}}(k)a_k^\dagger a_k+\epsilon_h^{\text{eff}}(k)b_{-k}^\dagger b_{-k}] + \frac{1}{2}\sum_{k,k',q\neq 0}V(q)[a_{k+q}^\dagger a_{k'-q}^\dagger a_{k'}a_k+b_{k+q}^\dagger b_{k'-q}^\dagger b_{k'}b_k-2a_{k+q}^\dagger b_{k'-q}^\dagger b_{k'}a_k]+\sum_k(\mathcal{V}_{cv}a_k^\dagger b_{-k}^\dagger+\mathcal{V}_{cv}^* b_{-k}a_k)\ , \tag{6}$$

where

$$\mathcal{V}_{cv}=-\frac{\wp}{2\hbar}\mathcal{E}(\mathbf{r},t)$$

describes the coupling to the multimode electric field $\mathcal{E}(\mathbf{r},t)$. a_k is the electron annihilation operator for the wave vector k and b_{-k} is the hole-annihilation operator for the wave vector $-k$. Note that we use electrons to describe the carriers in the conduction band and holes to describe the missing electrons in the valence band. The energies $\epsilon_e^{\text{eff}}(k)$ and $\epsilon_h^{\text{eff}}(k)$ are the effective single-particle energies of the electron and hole,

$$\epsilon_e^{\text{eff}}(k)=\frac{\hbar^2k^2}{2m_e}+\frac{\epsilon_g}{2}\equiv\epsilon_e(k)\ , \tag{7}$$

$$\epsilon_h^{\text{eff}}(k)=\frac{\hbar^2k^2}{2m_h}+\frac{\epsilon_g}{2}+\sum_{q\neq 0}V(q)\equiv\epsilon_h(k)+\sum_{q\neq 0}V(q)\ , \tag{8}$$

where m_e and m_h are the electron and hole effective masses and ϵ_g is the zero-field band-gap energy. We assume that the Coulomb interaction conserves the number of electrons in each band and we describe the dipole coupling to the laser field $\mathcal{E}(t)$ in the rotating-wave approximation. It is straightforward to generalize this treatment to include the quantum statistics of the laser light, by quantizing the field.

When many electrons and holes exist in the semiconductor, the effect of screening becomes increasingly important. This process can be treated by replacing the unscreened potential $V(q)$ by the screened one, $V_s(q)$, which has a reduced interaction strength especially at long distances. Screening leads to a renormalization of the single-particle energies and of the transition energy

$$\hbar\omega_t=\epsilon_e(k)+\epsilon_h(k)+\delta\epsilon_{\text{Deb}}\ , \tag{9}$$

where

$$\delta\epsilon_{\text{Deb}}=\sum_{q\neq 0}[V_s(q)-V(q)] \tag{10}$$

is the so-called Debye contribution to the electronic band-gap reduction (Debye shift).

To describe laser action in semiconductors, we derive the equations of motion for the expectation values $\langle a_k^\dagger a_k \rangle$, $\langle b_{-k}^\dagger b_{-k} \rangle$, and $\langle a_k^\dagger b_{-k}^\dagger \rangle$, where $\langle a_k^\dagger a_k \rangle = n_e(k)$ is the probability of having electrons with the wave vector k, and $\langle b_{-k}^\dagger b_{-k} \rangle = n_h(k)$ is the probability for holes, and $\langle a_k b_{-k} \rangle = \rho_{cv}$ gives the polarization of the momentum state k. In the language of conventional laser theory, these expressions are the diagonal and off-diagonal terms of the density matrix, where we set $n_e \equiv \rho_{cc}$, which is the probability of having a conduction electron in the state k, and $n_h \equiv 1-\rho_{vv}$, which is the probability of having a hole in the state k. The total carrier density is given by

$$N(t) = V^{-1}\sum_k n_e = V^{-1}\sum_k n_h \;, \tag{11}$$

where V is the volume of the semiconductor. Since the $\sum_k$ is proportional to V, the volume cancels out, leaving a number density.

Using straightforward operator algebra, we obtain equations of motion for the two-operator expectation values in terms of themselves and of expectation values of four-operator products. Reference 1 closes this set of equations by quantum-mechanical projection-operator techniques. In the present paper we are only interested in the limit that the carrier-carrier scattering time γ_r^{-1} and the dipole dephasing time γ^{-1} are much less than other relaxation times and times during which the multimode electric field envelope varies appreciably. For these cases the polarization equation derived in Ref. 1 can be written as

$$\dot{\rho}_{cv}^{(k)} = -(i\omega+\gamma)\rho_{cv}(k) + i\left[\mathcal{V}_{cv}(z,t) + \sum_{q\neq 0} V_s(q)\rho_{cv}(k+q)\right](n_e^{(k)}+n_h^{(k)}-1) \;. \tag{12}$$

The transition energies are again renormalized through the screened exchange contribution, yielding the effective transition energy

$$\hbar\omega = \epsilon_e(k)+\epsilon_h(k)+\delta\epsilon_g \;, \tag{13}$$

with $\delta\epsilon_g$ given by

$$\delta\epsilon_g = \delta\epsilon_{\mathrm{Deb}} - \sum_{q\neq 0} V_s(q)[n_e(q)+n_h(q)] \;, \tag{14}$$

where we have assumed a rigid, i.e., k-independent, band-shift. In Eq. (12) we recognize a variety of many-body contributions that have been discussed extensively in the context of high-excitation semiconductor pump-probe spectroscopy,[11] such as plasma screening (in V_s), band-gap renormalization (in $\delta\epsilon_g$), as well as band and state filling $(1-n_h-n_e)$. As limiting cases, our equations reproduce well-known results from quantum optics and semiconductor physics. If we neglect all Coulomb-interaction terms, we obtain the undamped Bloch equations used in atomic spectroscopy of inhomogeneously broadened systems. In semiconductors, the inhomogeneous broadening is an intrinsic consequence of the dispersion of the single-particle energies. On the other hand, for one electron, one hole, and a vanishing optical field, we regain the Wannier equation for the relative motion of an electron-hole pair.

In the remainder of this paper we simplify the equations by restricting ourselves to the situation of sufficiently high electron-hole densities well above the Mott density, where bound electron-hole pairs (excitons) are already ionized. In this regime it is a reasonable approximation to ignore the term

$$\sum_{q\neq 0} V_s(q)\rho_{cv}(k+q)$$

in comparison to $\mathcal{V}_{cv}(z,t)$ in Eq. (12), since the Coulomb potential is strongly screened. This approximation is equivalent to replacing the Coulomb enhancement factor by unity. It is shown in Ref. 4 that this is indeed a very good approximation for room-temperature bulk semiconductors if the generated carrier densities are high enough for optical gain.

Our set of dynamic equations for the density-matrix elements of momentum state k becomes

$$\dot{\rho}_{cv} = -(i\omega+\gamma)\rho_{cv} + i\mathcal{V}_{cv}(z,t)(n_e+n_h-1) \;, \tag{15}$$

$$\dot{n}_e = \lambda_e - \gamma_{\mathrm{NR}} n_e - \Gamma n_e n_h - \dot{n}_e\,|_{\text{c-c}} - (i\mathcal{V}_{cv}\rho_{vc} + \text{c.c.}) \;, \tag{16}$$

$$\dot{n}_h = \lambda_h - \gamma_{\mathrm{NR}} n_h - \Gamma n_e n_h - \dot{n}_h\,|_{\text{c-c}} - (i\mathcal{V}_{cv}\rho_{vc} + \text{c.c.}) \;, \tag{17}$$

where λ_α, $\alpha = e$ or h, is the pump rate due to an injection current, γ_{NR} is the nonradiative decay constant for the electron and hole probabilities, Γ is the radiative recombination rate constant, and $\dot{n}_\alpha\,|_{\text{c-c}}$ is the carrier-carrier scattering contribution. The rapid carrier-carrier intraband scattering drives the distribution n_α toward the Fermi-Dirac distribution

$$f_\alpha = \frac{1}{e^{\beta[\epsilon_\alpha(k)-\mu_\alpha]}+1} \;, \tag{18}$$

where $\beta = 1/k_B T$, k_B is Boltzmann's constant, T is the absolute temperature, and μ_α is the carrier chemical potential that yields a self-consistent total carrier density N. In fact, the intraband scattering contribution vanishes when n_α is given by a Fermi-Dirac distribution. While rapidly suppressing deviations from the Fermi-Dirac distribution, the scattering does not change the total carrier density N of Eq. (11). Hence summing either Eq. (16) or (17), we find the equation of motion

$$\dot{N} = \lambda - \gamma_{\mathrm{NR}} N - \frac{\Gamma}{V}\sum_k n_e n_h - \left[\frac{i\mathcal{V}_{\mathrm{CV}}}{V}\sum_k \rho_{vc} + \text{c.c.}\right] , \tag{19}$$

where the injection-current pump λ of Eq. (19) is given by

$$\lambda = \eta J/ed \;; \tag{20}$$

η is the efficiency that the injected carriers reach the active region, J is the current density, e is the charge of an electron, and d is the thickness of the active region.

Since the fields we consider vary little in the carrier-carrier scattering time and the Rabi-flopping periods are much larger than this scattering time, we approximate n_α by f_α in the remainder of this paper. The chemical potential μ_α in Eq. (18) is determined from

$$N = V^{-1}\sum_k f_e = V^{-1}\sum_k f_h \; . \tag{21}$$

In our calculations we use the single-plasmon-pole (SPP) approximation to describe the intraband plasma screening of the Coulomb interaction

$$\begin{aligned} V_s(k) &= \frac{4\pi e^2}{\epsilon_0 k^2}\left[1-\left[\frac{\omega_{pl}}{\omega_k}\right]^2\right], \\ \omega_{pl}^2 &= \frac{4\pi e^2 N}{\epsilon_0 m} = 16\pi N a_0^3\left[\frac{E_R}{\hbar}\right]^2, \\ \omega_k^2 &= \omega_{pl}^2\left[1+\frac{k^2}{\kappa^2}\right]+\frac{C}{4}\left[\frac{\hbar k^2}{2m}\right]^2, \end{aligned} \tag{22}$$

where C is a numerical constant usually chosen between 1 and 4 to approximate the continuum of pair excitations in the Lindhard formula by an effective plasmon pole.[4,11] With a_0 and E_R we denote the exciton Bohr radius and Rydberg energy, respectively, and κ is the inverse screening length

$$\kappa = \left[\frac{2\beta a_0 E_R}{\pi^2}\sum_\alpha \int d^3k\, f_\alpha(k)[1-f_\alpha(k)]\right]^{1/2} . \tag{23}$$

Using Eq. (22) one can analytically evaluate the Debye contribution to the band-gap renormalization[4] and the total band-gap shift becomes

$$\delta\epsilon_g = -\frac{2E_R(a_0\kappa)}{\left[1+C^{1/2}\left[\dfrac{E_R}{\hbar\omega_{pl}}\right](a_0\kappa)^2\right]^{1/2}} - \sum_{q\neq 0} V_s(q)[f_e(q)+f_h(q)] \; . \tag{24}$$

III. MULTIMODE EXPANSION

To derive the coupling equations of the multimode field components and to compute the probe absorption we perform now a systematic multimode Fourier expansion of our coupled set of equations. The interaction energy matrix element $\mathcal{V}_{cv}$ for the multimode field of Eq. (1) in the rotating-wave approximation is expanded as

$$\mathcal{V}_{cv} = -\frac{\wp}{2\hbar}\sum_n \mathcal{E}_n(\mathbf{r})e^{i(\mathbf{K}_n\cdot\mathbf{r}-\nu_n t)} , \tag{25}$$

where we assume that the electric-dipole matrix element $\wp$ varies little over the range of k values that interact. To determine the response of the medium to this interaction energy, we Fourier analyze both the polarization component ρ_{cv} of the density matrix as well as the number probabilities. We have

$$\rho_{cv} = e^{i(K_1 z-\nu_1 t)}\sum_m p_{m+1}e^{im[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]} , \tag{26}$$

where we choose the z direction to be along the probe at frequency ν_1. The electron and hole probabilities n_e and n_h have the corresponding Fourier expansions

$$n_\alpha = \sum_j n_{\alpha j}e^{ij[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]}, \quad \alpha=e,h \; . \tag{27}$$

It is also convenient to define the probability difference $D(k,\mathbf{r},t)$ with the expansion

$$\begin{aligned} D(k,\mathbf{r},t) &\equiv n_e(k,\mathbf{r},t)+n_h(k,\mathbf{r},t)-1 \\ &= \sum_j d_j e^{ij[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]} , \end{aligned} \tag{28}$$

where $d_j \equiv n_{ej}+n_{hj}-\delta_{j0}$. The carrier number density $N(t)$ has the expansion

$$N(t) = \sum_j N_j e^{ij[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]} \tag{29}$$

and the Fermi-Dirac distributions have the expansions

$$f_\alpha(k,t) = \sum_j f_{\alpha j}(k)e^{ij[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]}, \quad \alpha=e,h \; . \tag{30}$$

We can approximate the $f_{\alpha j}(k)$ by noting that the Fermi distributions f_α are functions of their respective chemical potentials μ_α, that is, $f_\alpha = f_\alpha[\mu_\alpha(N)]$. Hence we have

$$\begin{aligned} f_\alpha[\mu_\alpha(N_0+\Delta N)] &\cong f_\alpha[\mu_\alpha(N_0)+\Delta N\,\partial\mu_\alpha/\partial N_0] \\ &\cong f_{\alpha 0}[\mu_\alpha(N_0)]+\Delta N\frac{\partial\mu_\alpha}{\partial N_0}\frac{\partial f_{\alpha 0}}{\partial\mu_\alpha} \; . \end{aligned}$$

Taking the small deviation ΔN to be

$$N_1 e^{i[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]} + \text{c.c.}$$

and identifying coefficients of $e^{\pm\Delta t}$ with those in Eq. (29), we find

$$f_{\alpha j} = N_j g_\alpha \; , \tag{31}$$

where the normalized derivative g_α of the Fermi-Dirac distribution f_α is given by

$$g_\alpha = \frac{\partial\mu_\alpha}{\partial N_0}\frac{\partial f_{\alpha 0}}{\partial\mu_\alpha} = \frac{f_{\alpha 0}(f_{\alpha 0}-1)}{V^{-1}\sum_k f_{\alpha 0}(f_{\alpha 0}-1)} \; . \tag{32}$$

Note that here we assume that the carrier temperature does not vary significantly due to the field-envelope modulation caused by the probe waves.

Using Eqs. (23) and (30), the inverse screening length κ is expanded as

$$\kappa^2 = \kappa_0^2 + \frac{2\beta a_0 E_R}{\pi^2}\sum_{j\neq 0}\sum_\alpha \int d^3k\, f_{\alpha j}(k)[1-2f_{\alpha 0}(k)] \times e^{ij[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]} , \tag{33}$$

where

$$\kappa_0 = \left[\frac{2\beta a_0 E_R}{\pi^2}\sum_\alpha \int d^3k\, f_{\alpha 0}(k)[1-f_{\alpha 0}(k)]\right]^{1/2} .$$

Since the band-gap shift $\delta\epsilon_g$, Eq. (24), also depends on the excitation density, it has to be expanded as

$$\delta\epsilon_g(t)=\sum_j \delta\epsilon_{gj}(k)e^{ij[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]} \ . \tag{34}$$

As discussed below, our final results include only the terms with $j=0,\pm1$ of the expansion (34). Therefore we truncate the expansion (34) at $j=\pm1$,

$$\delta\epsilon_g \cong \delta\epsilon_{g0}+(\delta\epsilon_{g1}e^{i[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]}+\text{c.c.}) \ ,$$

where

$$\begin{aligned}
\delta\epsilon_{g0}&=-\frac{2E_R(a_0\kappa_0)}{\left[1+C^{1/2}\left[\frac{E_R}{\hbar\omega_{pl}}\right](a_0\kappa_0)^2\right]^{1/2}} -\sum_q V_s(q)\mid_{N=N_0}[f_{e0}(q)+f_{h0}(q)] \\
&=\delta\epsilon_{g\mathrm{Deb}0}+\delta\epsilon_{g\mathrm{SX}0} \ , \\
\delta\epsilon_{g1}&=\delta\epsilon_{g\mathrm{Deb}1}+\delta\epsilon_{g\mathrm{SX}1}=\hbar\,\delta\omega\,N_1 \ , \\
\delta\epsilon_{g\mathrm{Deb}1}&=\delta\epsilon_{g\mathrm{Deb}0}\frac{1}{2}\frac{\sum_\alpha\int d^3k\, g_\alpha(k)[1-2f_{\alpha0}(k)]}{\sum_\alpha\int d^3k\, f_{\alpha0}(k)[1-f_{\alpha0}(k)]}N_1 \ , \\
\delta\epsilon_{g\mathrm{SX}1}&=\sum_q V_s^2(q)\mid_{N=N_0} \times\frac{[f_{e0}(q)+f_{h0}(q)](a_0\kappa_0)^2}{(8\pi a_0^3E_R)\left[1+\frac{(a_0\kappa_0)^2C\hbar^2q^2}{128\pi mN_0a_0^3E_R}\right]^2} \\
&\quad\times\frac{\sum_\alpha\int d^3k\, g_\alpha(k)[1-2f_{\alpha0}(k)]}{\sum_\alpha\int d^3k\, f_{\alpha0}(k)[1-f_{\alpha0}(k)]}N_1 -\sum_q V_s(q)\mid_{N=N_0}[g_e(q)+g_h(q)]N_1 \ .
\end{aligned} \tag{35}$$

Hence, the effective transition energy of Eq. (13) can be written as

$$\omega\cong\omega_0+\delta\omega(N_1e^{i[(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}-\Delta t]}+\text{c.c.}) \ , \tag{36}$$

with

$$\hbar\omega_0=\epsilon_e(k)+\epsilon_h(k)+\delta\epsilon_{g0} \ . \tag{37}$$

From Eq. (36) we see that the effective transition energy is modulated by the carrier density, thus giving a contribution to the multiwave mixing in semiconductors.

We substitute the expansions (25) through (36) into the density-matrix dipole equation of motion (15) and identify coefficients of common exponential frequency factors. We assume that the probe amplitudes $\mathcal{E}_1$ and $\mathcal{E}_3$ do not saturate, i.e., appear only once. We show that in this approximation only p_1, p_2, and p_3 occur in the polarization expansion (26), and that only $j=0,\pm1$ appear in the probability expansions (27)–(29). Physically this simplification occurs because a product of $\mathcal{E}_1$ and $\mathcal{E}_2$ creates the pulsations like $d_{\pm1}$, and from then on only $\mathcal{E}_2$ can interact. One obtains the polarization side bands of ν_2 at frequencies ν_1 and ν_3, which subsequently combine with ν_2 only to give back $d_{\pm1}$ components.

We calculate the coefficient of $\exp(i\mathbf{K}_2\cdot\mathbf{r}-i\nu_2t)$ for the saturating mode by neglecting the nonsaturating probe fields. We find for the $m=1$ term in Eq. (26)

$$-i\nu_2p_2=-(i\omega_0+\gamma)p_2-i(\wp\mathcal{E}_2/2\hbar)d_0$$

which gives

$$p_2=-(\wp/2\hbar)\mathcal{E}_2\mathcal{D}_2d_0 \ , \tag{38}$$

where for notational simplicity we have defined the complex denominator

$$\mathcal{D}_2=\frac{1}{\gamma+i(\omega_0-\nu_2)} \ . \tag{39}$$

Equation (38) is simply the single-mode density-matrix element in which we include a subscript 2 to specify the saturator wave and have factored out the rapidly varying time-space factor $\exp(i\mathbf{K}_2\cdot\mathbf{r}-i\nu t)$.

In the quasiequilibrium Fermi-Dirac approximation, the dc probability difference d_0 is given by

$$d_0=f_{e0}+f_{h0}-1 \ . \tag{40}$$

The coefficient of $\exp(iK_1z-i\nu_1t)$ for the probe wave [$m=0$ term in Eq. (26)] includes an extra term $\mathcal{E}_2d_{-1}$ and the tuning term $\delta\omega$,

$$-i\nu_1p_1=-(i\omega_0+\gamma)p_1-i(\wp/2\hbar)[\mathcal{E}_1d_0+\mathcal{E}_2d_{-1}] -i\,\delta\omega N_{-1}p_2 \ ,$$

giving

$$p_1=-i(\wp/2\hbar)\mathcal{D}_2[\mathcal{E}_1d_0+\mathcal{E}_2d_{-1}-i\mathcal{E}_2d_0\delta\omega\,\mathcal{D}_2N_{-1}] \ , \tag{41}$$

where in this paper we use $\mathcal{D}_2$ instead of the usual $\mathcal{D}_1$ since the difference $\Delta=\nu_2-\nu_1$ has a much smaller magnitude than the dipole decay constant γ. The $\mathcal{E}_2d_{-1}$ term gives the scattering of $\mathcal{E}_2$ into the $\mathcal{E}_1$ mode by the probability-pulsation component d_{-1}. The polarization component p_0 remains zero when only d_0 and $d_{\pm1}$ are nonzero, since it is proportional to $\mathcal{E}_1d_{-1}$, which involves at least two $\mathcal{E}_1$'s. Similarly, the component p_s has the nonzero value

$$p_3=-i(\wp/2\hbar)\mathcal{D}_2[\mathcal{E}_3d_0+\mathcal{E}_2d_1-i\mathcal{E}_2d_0\delta\omega\,\mathcal{D}_2N_1] \ , \tag{42}$$

while $p_{j>3}$ vanishes since $d_{j>1}$ would be involved.

We have the probability pulsation contributions

$$d_{-1}=f_{e,-1}+f_{h,-1} \tag{43}$$

and $d_1=d^*_{-1}$. Our calculation is self-consistent, since only d_0 and $d_{\pm1}$ can obtain nonzero values from p_1, p_2, p_3, and vice versa.

To find the dc contribution N_0, we seek the steady-state solution of Eq. (19) written in the absence of probes as

$$\begin{aligned}
\dot N&=\lambda-\gamma_{\mathrm{NR}}N-\frac{\Gamma}{V}\sum_k f_{e0}f_{h0}+\left[\frac{i\mathcal{E}_2}{2\hbar V}\sum_k p_2^*+\text{c.c.}\right] \\
&=\lambda-\gamma_{\mathrm{NR}}N-\frac{\Gamma}{V}\sum_k f_{e0}f_{h0}-\frac{|\wp\mathcal{E}_2/\hbar|^2}{2\gamma V}\sum_k \mathcal{L}_2d_0 \ .
\end{aligned} \tag{44}$$

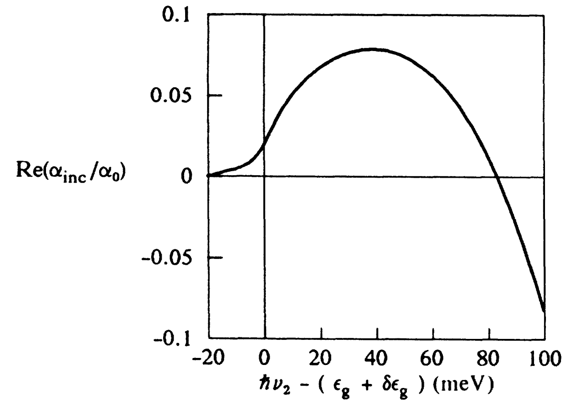

FIG. 2. Real part of $\alpha_{\rm inc}$, in units of α_0 as given by Eq. (55), vs pump detuning $\hbar\nu_2-(\epsilon_g+\delta\epsilon_g)$ for bulk GaAs at $T=293$ K. The carrier density is $N_0=3\times10^{18}$ cm^{-3}, $\hbar\Gamma=0.0007$ meV, $\hbar\gamma_{\rm NR}=0.000\,12$ meV, $\hbar\gamma=4$ meV, $m_e=0.0665m_0$, $m_h=0.52m_0$, where m_0 is the free-electron mass, $\wp\cong3$ Å, the pump rate $\lambda=0.004$ meV$/a_0^3$, $\epsilon_0=12.35$, $a_0=1.243\times10^{-6}$ cm, and $E_R=4.2$ meV. For these parameters, the medium has gain up to $\cong$84.7 meV.

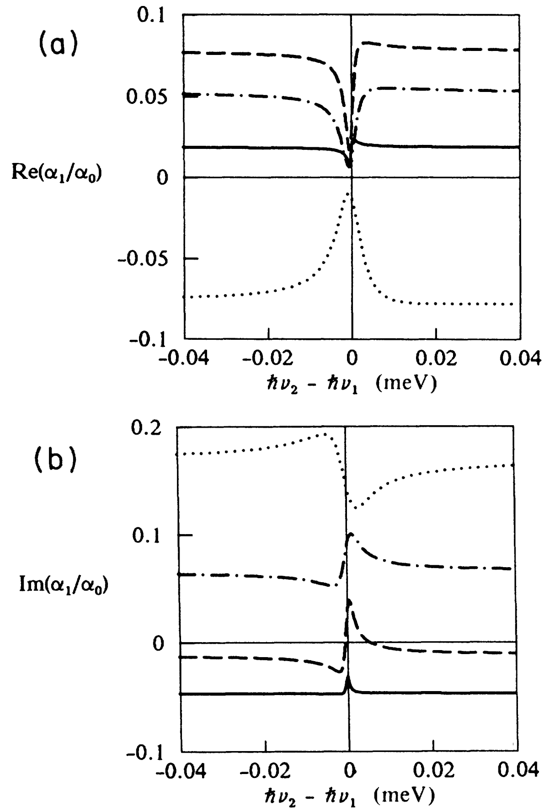

FIG. 3. (a) Real part of α_1, in units of α_0 as given by Eq. (52), vs probe-pump detuning $\hbar\Delta$ for pump detunings $\hbar\nu_2-(\epsilon_g+\delta\epsilon_g)=0$ (solid line, at band gap), 33 meV (dashed line, above band gap), 66 meV (dot-dashed line), and 99 meV (dotted line) for the same parameters as Fig. 2 and $|\wp\mathcal{E}_2|=0.5$ meV, which corresponds to a pump intensity of $\cong1.3$ MW/cm^2. (b) Imaginary part of α_1 corresponding to the real-part curves in (a).

This may be solved numerically using the expansion

$$\dot N(N)\cong\dot N(N_0)+(N-N_0)\frac{d\dot N}{dN}=(N-N_0)\frac{d\dot N}{dN}\ , \tag{45}$$

where $\dot N(N_0)=0$. Calculating $d\dot N/dN$ from Eq. (44) and using the g's defined by Eq. (32), we have

$$\frac{d\dot N_0(N)}{dN}=-\Gamma_1'\ , \tag{46}$$

where the power-broadened carrier-density decay constant Γ_2' is given by

$$\Gamma_1'=\Gamma_1+|\wp\mathcal{E}_2/\hbar|^2(2\gamma V)^{-1}\sum_k\mathcal{L}_2(g_e+g_h) \tag{47}$$

and the carrier-density decay constant Γ_1 is given by

$$\Gamma_1=\gamma_{\rm NR}+\Gamma V^{-1}\sum_k(n_{e0}g_h+n_{h0}g_e)\ . \tag{48}$$

Hence by interating the equation

$$N_0\cong N+(\Gamma_1')^{-1}\dot N \tag{49}$$

we find the steady-state carrier density N_0 and the decay constants Γ_1 and Γ_1'.

The carrier-density pulsation component is given by

$$\begin{aligned}(\gamma_{\rm NR}+i\Delta)N_{-1}&=-\frac{1}{V}\sum_k[\Gamma(f_{e0}f_{h,-1}+f_{h0}f_{e,-1})-i(\wp/2\hbar)(\mathcal{E}_1p_2^*+\mathcal{E}_2p_3^*-\mathcal{E}_2^*p_1-\mathcal{E}_3^*p_2)]\\&=-\frac{1}{V}\sum_k\Bigg\{\Gamma N_{-1}(f_{e0}g_h+f_{h0}g_e)\\&\qquad+\frac{2}{\gamma}\mathcal{L}_2(\wp/2\hbar)^2\{(\mathcal{E}_1\mathcal{E}_2^*+\mathcal{E}_2\mathcal{E}_3^*)d_0+|\mathcal{E}_2|^2N_{-1}(g_e+g_h)[1-\delta\omega\,d_02(\omega_0-\nu_2)\gamma^{-2}\mathcal{L}_2]\}\Bigg\}\ .\end{aligned}$$

Solving for N_{-1}, we get

$$N_{-1}=-\frac{2\gamma^{-1}(\wp/2\hbar)^2(\mathcal{E}_1\mathcal{E}_2^*+\mathcal{E}_2\mathcal{E}_3^*)V^{-1}\sum_{k'}\mathcal{L}_2d_0}{\Gamma_1'+\Gamma_\kappa+i\Delta}\ , \tag{50}$$

where

$$\Gamma_\kappa=|\wp\mathcal{E}_2/\hbar|^2(\gamma V)^{-1}\delta\omega\sum_{k'}(\omega_0-\nu)\gamma^{-2}\mathcal{L}_2^2(g_e+g_h)d_0\ .$$

Substituting Eqs. (50) and (43) into (41), setting $\mathcal{P}_1=2\wp V^{-1}\sum_k p_1$, and using Eq. (3), we find the coupled probe equation of motion

$$\frac{d\mathcal{E}_1}{dz}=\alpha_1\mathcal{E}_1+\chi_1\mathcal{E}_3^* , \tag{51}$$

where the gain coefficient α_1 is given by

$$\alpha_1=\alpha_0\sum_k \gamma\mathcal{D}_2\left\{d_0-\frac{(g_e+g_h)2\gamma^{-1}\,|\wp\mathcal{E}_2/2\hbar|^2V^{-1}\sum_{k'}\mathcal{L}_2d_0(1-i\,\delta\omega\,\mathcal{D}_2d_0)}{\Gamma_1'+\Gamma_\kappa+i\Delta}\right\} , \tag{52}$$

with $d_0\cong f_{e0}+f_{h0}-1$, and the coupling coefficient χ_1 is given by

$$\chi_1=-\alpha_0\sum_k \gamma\mathcal{D}_2\frac{(g_e+g_h)2\gamma^{-1}(\wp\mathcal{E}_2/2\hbar)^2V^{-1}\sum_{k'}\mathcal{L}_2d_0(1-i\,\delta\omega\mathcal{D}_2d_0)}{\Gamma_1'+\Gamma_\kappa+i\Delta} . \tag{53}$$

Here the χ_1 implicitly includes a phase-mismatch factor $\exp[2i(\mathbf{K}_2-\mathbf{K}_1)\cdot\mathbf{r}]$. If this is significant, the χ_1 term in Eq. (51) averages to zero, leaving behind a simple Beer's law equation for the probe wave $\mathcal{E}_1$ alone. For perfect phase matching, the gain coefficient α_1 can be written in terms of χ_1 as

$$\alpha_1=\alpha_{\rm inc}+\chi_1 , \tag{54}$$

where the incoherent gain coefficient is given by

$$\alpha_{\rm inc}=\alpha_0\sum_k \gamma\mathcal{D}_2d_0 . \tag{55}$$

Similar to Eq. (51) the coupled probe amplitude $\mathcal{E}_3$ obeys the equation

$$\frac{d\mathcal{E}_3^*}{dz}=\alpha_3^*\mathcal{E}_3^*+\chi_3^*\mathcal{E}_1 , \tag{56}$$

where the coefficients α_3 and χ_3 are given by α_1 and χ_1, respectively, by interchanging the subscripts 1 and 3 (note this implies replacing Δ by $-\Delta$).

The carrier-density decay constant Γ_1' of Eq. (47) is *not* a function of k. The remaining sums over k produce various multiplicative constants. Hence it is clear from

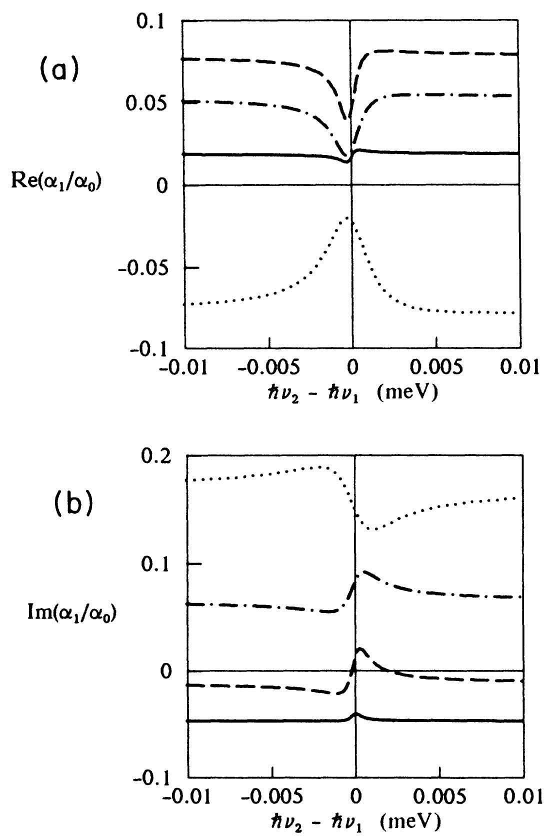

FIG. 4. (a) Same as Fig. 3(a) but for $|\wp\mathcal{E}_2|=0.3$ meV corresponding to a pump intensity $\cong$480 kW/cm^2. (b) Imaginary part of α_1 corresponding to the real-part curves in (a).

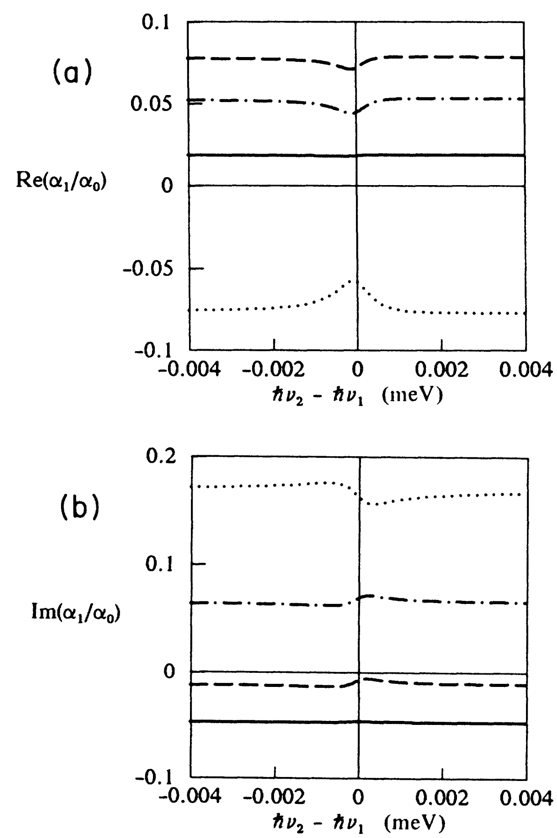

FIG. 5. (a) Same as Fig. 3(a) but for $|\wp\mathcal{E}_2|=0.1$ meV corresponding to a pump intensity $\cong$50 kW/cm^2. (b) Imaginary part of α_1 corresponding to the real-part curves in (a).

the work of Sargent[10,12] that the α_1 and χ_1 of Eqs. (52) and (53) lead to coherent dips in pump-probe and modulation spectroscopy. They furthermore lead to narrow-band retroreflection in phase conjugation by nondegenerate four-wave mixing.[13]

IV. NUMERICAL RESULTS AND DISCUSSION

As illustration, we evaluate our equations for the example of bulk GaAs at room temperature. We concentrate on the situation where we have the strong wave at $\hbar\nu_2$ and only one side mode. The case of two side modes will be analyzed in subsequent publications. For later reference we first evaluate the real part of the incoherent gain coefficient, Eq. (55). Figure 2 shows the resulting gain spectrum for the carrier density $N=3\times10^{18}$ cm^{-3}. We see that the gain regime covers the spectral region between the renormalized band gap and the chemical potential. A closer inspection shows that actually the gain extends somewhat below the renormalized gap as a consequence of the finite linewidth γ.

Figures 3–5 illustrate the real and imaginary parts of the full gain coefficient α_1, Eq. (52), for different intensities of the strong field and for a number of detunings. The comparison with Fig. 2 shows that all of the chosen detunings except one are in the gain regime ($d_0>0$). All curves of Re(α_1) reveal partly asymmetric dips versus probe-pump beat frequency Δ. The magnitude of the dips increases with increasing intensity of the strong field at $\hbar\nu_2$. The asymmetry of the dips is a consequence of the mixing of the real and imaginary parts of the complex Lorentzian $1/(\Gamma_1'+\Gamma_\kappa+i\Delta)$ due to the integrals over the complex Lorentzian $\mathcal{D}_2$. The χ_n have similar probe-tuning dependences, but are displaced uniformly since according to Eq. (54) they do not contain α_{inc}.

Using the parameters specified in the caption of Fig. 2 we estimate the Rabi frequency to vary between $\cong2.4\times10^{11}$ and 2.4×10^{10} Hz when the strong-field intensity varies between 1.3 and 0.05 MW/cm^2, respectively. Hence the corresponding flopping times are still long in comparison to the carrier-carrier intraband scattering time which is less than 100 fs in typical semiconductor lasers.[14]

In conclusion, we presented the semiclassical theory of multiwave mixing in semiconductor media with optical gain. We illustrated the theory numerically for the case of only one side mode to study the development of dips generated by scattering of the strong field off carrier-density pulsations induced by the two-beam interference. These results allow one to measure the power-broadened decay rate constant Γ_1'. They also reveal that modes spaced on the order of Γ_1' are coupled through probability pulsations.[15] We are presently analyzing the case of more than one side mode. Moreover, a related treatment[16] for a quantized probe field shows that the spectrum of resonance fluorescence also features coherent-dip phenomena in the vicinity of the pump wave frequency.

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*Also at the Department of Physics, University of Arizona, Tucson, AZ 85721.

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