Maxwell-Bloch formulation for semiconductors: Effects of coherent Coulomb exchange

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A generalized Bloch-Maxwell formulation for laser-field-coupled semiconductors is derived from a two-band model which includes direct Coulomb interactions. The momentum-dependent, microscopic, electron-hole equations of motion in the time-dependent Hartree-Fock approximation and neglecting interband exchange interactions form the starting point for the formulation. A self-consistent set of coupled equations in four dynamical variables for the medium, together with the electric-field amplitude coupled through the Maxwell wave equation in a semiclassical approximation, are obtained to lowest order in the coherent Coulomb exchange interaction and the density-of-states distribution. Intrinsic optical bistability is predicted in a steady state due to a carrier density-dependent redshift of the band edge due explicitly to coherent Coulomb exchange. Integration of the dynamical equations for conditions which correspond to the ultrafast time regime exhibit intrinsic adiabatic inversion, adiabatic following, anomalous Rabi cycling, and unique, as well as fast, optical switching, all of which depend upon the coherent Coulomb exchange interaction.

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I. INTRODUCTION

For more than a decade, much interest has been expressed in the formulation of the dynamics of semiconductor lasers in a representation similar to the Maxwell-Bloch equations of nonlinear optics [1]. The root of this interest is essentially twofold: (i) The expanding importance of semiconductor lasers in pure and applied science and the device applications in optoelectronics, optical communications, and optical data processing [2]; and (ii) the relative ease with which a Bloch-Maxwell-like formulation can predict and analyze new physical phenomena and characteristics, as has been the case in nonlinear and quantum optics of gases and vapors [3,4]. Until recently [5–7], attempts to arrive at an analogous formulation of reduced equations of motion, *ab initio,* have not been particularly successful [1].

The phenomenological approach, on the other hand, based upon the linear variation of gain and refractive index with carrier density, has resulted in a variety of important and interesting results [8]. Attempts have been made to improve the phenomenological description by using the density-matrix formulation in which a semiconductor laser is modeled as a collection of two-level atoms with a range of transition frequencies, similar to an inhomogeneously broadened, two-level systems [9–13]. Such an approach has led to fundamental understanding and interpretation of gain suppression (so-called nonlinear gain) in semiconductor lasers [9,12], the description of gain and mixing susceptibilities in amplifiers [10,11], and analysis of spectral hole-burning and gain saturation [12,13] in semiconductor lasers. A formulation similar to that of the optical Bloch equations [3] has only recently been presented to lowest order in the density-of-states distribution [5–7], and has been shown to be extremely useful in the rate-equation limit in the demonstration that the index saturates because of intraband relaxation effects, whereas the gain saturates mainly due to interband transitions, and that the origin of phase-amplitude coupling, manifested through the linewidth enhancement factor, lies in the asymmetric nature of the carrier distribution within the conduction and valence bands [5]. In addition, the dynamical equations were used to clarify the concept of the linewidth enhancement factor in semiconductor lasers and its usefulness as well as limitations under conditions for subpicosecond pulse excitation [6].

There has been a recent surge of interest in laser field-induced many-body coherence and cooperative effects in semiconductors [14–17]. This is due in part to the rather recent widespread experimental capability to generate intense subpicosecond laser pulses. Recent experiments using quantum-well materials and bulk semiconductors under intense, subpicosecond, nonresonant, below-gap pulse excitation have revealed light-induced shifts of excitonic resonances and corresponding changes in oscillator strengths [18–20], whereas under resonant conditions, spectral hole-burning was observed. These phenomena have been interpreted in terms of many-body collective and coherence effects in the ultrashort-time, collisionless regime [21]. Qualitatively, these coherence effects have counterparts in nonlinear optics, as the ac Stark effect and hole-burning in inhomogeneously broadened systems [3]. More recently, numerical experiments conducted by integrating the Hartree-Fock equations for a many-body, electron-hole model of a two-band semiconductor dipole coupled to an externally applied laser field have demon-

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strated remarkable coherence and cooperative dynamical behavior in the short pulse time regime [14–17]. Many-body Coulomb effects cause doubling of the effective Rabi frequency [3] of the applied field in the absence of detuning, whereas transient adiabatic following [3] is obtained for large detunings [14]. Strong and weak excitations of the semiconductor by the applied laser field exhibit properties of atomic systems within the traditional Maxwell-Bloch formulation, and complete Rabi oscillations of the electron-hole density is observed for area-2π [3] applied field pulses [17] due to coherent Coulomb exchange effects. Cooperative resonance fluorescence [16] and many-body effects in superfluorescence, including subradiance and energy trapping [22], may be present in semiconductors in the subpicosecond time regime. All of these phenomena have strong qualitative similarities to well-known phenomena in nonlinear optics described within the optical Bloch-Maxwell formulation.

In spite of its potential for prediction and physical interpretation, a formulation similar to that of the optical Bloch-Maxwell equations, reduced from first principles, and including coherent Coulomb exchange effects as well as the distribution over the density of states, has yet to be formulated. Our previous work [5] addressed this issue to lowest order in the density-of-states distribution. There, we introduced a density-matrix formulation of the equations of motion for carriers, dipole-coupled to a single mode of the electromagnetic field in the semiclassical approximation, which accounts for the band structure and intraband and interband relaxation processes. We used a perturbative procedure, similar to that introduced by Graham and Cho [23], to reduce the set of equations for an inhomogeneously broadened medium to a set of Bloch-like equations which do not require explicit treatment of the band-structure details, as in previous, *ab initio* treatments [1,24]. In our formulation, all of the information concerning the joint density-of-states distribution is contained in a single expansion parameter. We showed that the equations, to lowest order in the density-of-states distribution, reduce to the usual carrier density equation of motion in the rate-equation limit [5], but all the parameters have explicit fundamental physical meaning.

Here, we extend our previous results [5] where we developed a generalized Bloch-Maxwell formulation for semiconductors to lowest order in the density-of-states distribution. In the present development we include coherent Coulomb exchange effects to lowest order as well. Thus, our purpose here is to develop a generalized Bloch-Maxwell formulation for semiconductors to lowest order in the coherent Coulomb exchange interaction and to lowest order in the density-of-states distribution as well. The following section is used to present the laser field dipole-coupled, two-band semiconductor model in the form of conventional Hamiltonian formulation, from which the hierarchy of equations of motion for the carriers and polarizations for the various momentum states are obtained within the time-dependent Hartree-Fock decorrelation approximation. The electromagnetic field is treated classically in the dipole-coupling approximation. From the set of density-matrix equations of motion, we develop the density-of-states-averaged, generalized Bloch-Maxwell equations of motion to lowest order in the coherent Coulomb exchange interaction and to lowest order in the density-of-states distribution in Sec. III. Section IV is used to discuss results for the rate-equation limit and circumstances for a first-order phase transition in steady state and intrinsic optical bistability which stem from the model. Ultrafast phenomena are discussed in Sec. V and predictions for novel optical switching, Rabi oscillations, strong self-phase modulation, and intrinsic adiabatic inversion are presented. The final section is used for summary and conclusion.

II. THE SEMICONDUCTOR MODEL

A two-band model for laser-driven semiconductors is considered which includes the direct Coulomb interactions. The exciton binding energy is assumed small compared to the band gap, thus the interband exchange interactions are neglected. The Hamiltonian from which we will derive the microscopic Bloch equations is [24]

$$\mathcal{H}=\sum_k \epsilon_c(k)a^\dagger_{c,k}a_{c,k}+\sum_k \epsilon_v(k)a^\dagger_{v,k}a_{v,k}+V+\mathcal{H}_F\ , \tag{2.1}$$

$$\begin{aligned}V=\tfrac{1}{2}\sum_{k,k';q\neq 0} v(q)[&a^\dagger_{c,k+q}a^\dagger_{c,k'-q}a_{c,k'}a_{c,k}\\&+a^\dagger_{v,k+q}a^\dagger_{v,k'-q}a_{v,k'}a_{v,k}\\&+2a^\dagger_{c,k+q}a^\dagger_{v,k'-q}a_{v,k'}a_{c,k}]\ ,\end{aligned} \tag{2.2}$$

$$\mathcal{H}_F=-\sum_k [\mu_k E_L(t)a^\dagger_{c,k}a_{v,k}+\mu^*_k E^*_L(t)a^\dagger_{v,k}a_{c,k}]\ , \tag{2.3}$$

where E_L is the microscopic field that couples to the dipoles,

$$\begin{aligned}&v(q)=\hbar\omega_x\frac{8\pi a_0}{\kappa_b q^2}\ ,\quad \epsilon_c(k)=E_g+\frac{\hbar^2k^2}{2m_e}\ ,\\&\epsilon_v(k)=-\frac{\hbar^2k^2}{2m_e}\ .\end{aligned} \tag{2.4}$$

The energies $\epsilon_c(k)$ and $\epsilon_v(k)$ in Eq. (2.4) are the momentum-specific, single-electron energies in the conduction and valence bands, respectively, and E_g is the unperturbed energy of the band gap. The Coulomb-exchange interaction coefficients $v(q)$ in Eq. (2.4) are given in terms of the exciton energy, $\hbar\omega_x$, and the Bohr radius of the exciton, a_0, and κ_b is the background dielectric constant of the material. The laser field is coupled to the lattice electrons in the electric dipole and rotating-wave approximations as shown in Eq. (2.3), where μ_k is the matrix element of the interband transition dipole moment. Here, it is emphasized that E_L, which appears in Eq. (2.3), is the microscopic local field. The macroscopic Maxwell field E is defined below [Eqs. (2.14)–(2.16)]. The positive-frequency component of the macroscopic field is included by using (in the plane-wave limit)

$$E(z,t)=\tfrac{1}{2}\mathcal{E}(z,t)e^{-i(\omega_L t-k_L z)}\ , \tag{2.5}$$

together with a similar representation for E_L. The corresponding induced polarization can be written as

$$P(z,t)=\tfrac{1}{2}\mathcal{P}(z,t)e^{-i(\omega_L t-k_L z)} . \tag{2.6}$$

Here, we have neglected dipole coupling to the quantized field modes, and hence, ignore spontaneous emission effects. Also, $\mathcal{E}$ and $\mathcal{P}$ are slowly varying complex amplitudes in space z and time t, and ω_L and k_L are the carrier frequency of the laser field and associated wave vector, respectively. In Eqs. (2.1)–(2.3), $a^{\dagger}_{c,k}$ and $a_{c,k}$ are the creation and annihilation operators for conduction-band electrons. The corresponding operators for the valence band are $a^{\dagger}_{v,k}$ and $a_{v,k}$.

From Eqs. (2.1)–(2.3), we obtain the equations of motion for the expectation values for the occupation probability n_k and transition probability p_k, defined as

$$n_k=\langle a^{\dagger}_{c,k}a_{c,k}\rangle , \quad p_k=\langle a^{\dagger}_{c,k}a_{v,k}\rangle . \tag{2.7}$$

The equation of motion for valence-band electrons is not needed if we assume

$$\langle a^{\dagger}_{c,k}a_{c,k}\rangle=1-\langle a^{\dagger}_{v,k}a_{v,k}\rangle . \tag{2.8}$$

Within the time-dependent Hartree-Foch approximation [24], the microscopic equations of motion are [14]

$$i\hbar\frac{\partial p_k}{\partial t}=\left\{\Delta_k-\frac{i\hbar}{T_2}-2\sum_{k'\neq k}v(k-k')n_{k'}\right\}p_k -(1-2n_k)\left\{\sum_{k'\neq k}v(k-k')p_{k'}+\mu_k E_L(t)\right\} , \tag{2.9}$$

$$\hbar\frac{\partial n_k}{\partial t}=-\hbar\frac{(n_k-n_k^{(0)})}{T_1} +2\,\mathrm{Im}\{\mu_k^* E_L^*(t)p_k+\sum_{k\neq k'}v(k-k')p_{k'}^*p_k\} , \tag{2.10}$$

where

$$\Delta_k=\frac{\hbar^2k^2}{2m_{\mathrm{eff}}}+E_g , \tag{2.11}$$

$n_k^{(0)}$ is the quasiequilibrium value of n_k, and m_{eff} is the effective mass for electrons and holes. In (2.9) and (2.10), we have combined the scattering contributions into the terms involving T_2 and T_1, and retained coherent Coulomb exchange contributions explicitly. The hierarchy of equations in the momentum k, Eqs. (2.9) and (2.10), have been termed the "effective Bloch equations for semiconductors," [1]. To apply these equations to the physics of laser-driven semiconductors requires considerable numerical computation.

These equations, Eqs. (2.9) and (2.10), can be written more nearly in the form of Bloch equations if we let

$$\mu_k E_T(k)=\mu_k E_L+\sum_{k'\neq k}v(k-k')p_{k'} . \tag{2.12}$$

In Eq. (2.12), the expectation value of the resultant renormalized field, E_T, which couples to an electron of momentum k, is the linear superposition of the microscopic field E_L, together with the reaction field, due to coherent Coulomb-exchange interaction, the second term on the right-hand side of Eq. (2.12), due to all the other dipole moments. The macroscopic Maxwell field E is the average of $E_T(k)$ over the density of states, whereas

$$E_L=E+\mathcal{J}(E) , \tag{2.13}$$

$$\langle E_T\rangle=E+\mathcal{J}(E)+\left\langle\mu_k^{-1}\sum_{k'\neq k}v(k-k')p_{k'}\right\rangle , \tag{2.14}$$

where angular brackets denote average over the density of states. Since $\langle E_T\rangle$ is just the macroscopic field E, we obtain

$$\mathcal{J}(E)=-\left\langle\mu_k^{-1}\sum_{k'\neq k}v(k-k')p_{k'}\right\rangle , \tag{2.15}$$

$$\langle E_T\rangle=E . \tag{2.16}$$

We have included here the Coulomb-exchange contribution to the local field. The dipole-dipole interaction contribution, which is normally much smaller for semiconductors, is not included, but is additive and is treated in the Appendix.

It is useful to introduce an inversion variable $w_k=2n_k-1$ and rewrite the microscopic equations of motion in the form

$$\frac{\partial p_k}{\partial t}=-\frac{i}{\hbar}\left\{\bar{\Delta}_k-\frac{i\hbar}{T_2}-\sum_{k'\neq k}v(k-k')w_{k'}\right\}p_k+\frac{w_k\mu_k E_T(k)}{i\hbar} , \tag{2.17}$$

$$\frac{\partial w_k}{\partial t}=-\frac{(w_k-w_k^{(0)})}{T_1}+\frac{4}{\hbar}\mathrm{Im}\{\mu_k^*E_T^*(k)p_k\} , \tag{2.18}$$

where

$$\bar{\Delta}_k=\Delta_k-\hbar\epsilon_k=\frac{\hbar^2k^2}{2m_{\mathrm{eff}}}+(E_g-\epsilon_k) , \tag{2.19}$$

$w_k^{(0)}$ is the quasiequilibrium value of w_k, and

$$\epsilon_k\equiv\frac{1}{\hbar}\sum_{k'\neq k}v(k-k') . \tag{2.20}$$

Equation (2.19) shows that ϵ_k can be interpreted as the Debye shift resulting from band-gap renormalization due to Coulomb-exchange interaction. To proceed further, we make the following ansatz:

$$\sum_{k'\neq k}v(k-k')w_{k'}\approx\hbar\epsilon_k\langle w_{k'}\rangle . \tag{2.21}$$

Consistent with the ansatz (2.21) and the form of $v(q)$, Eq. (2.4), we assume that ϵ_k can be taken as independent of k, and thus, write $\epsilon_k=\epsilon$ in what follows. This is justified since the main contribution to ϵ_k stems from a narrow region in k space in the neighborhood of $k=k'$, Eq. (2.4). We also replace the microscopic field $E_T(k)$ by its average $\langle E_T(k)\rangle$. This amounts to making the local field correction [5,25], Eqs. (2.13)–(2.16), and from here on, we use

$$W\equiv\langle w_k\rangle , \quad P\equiv\langle p_k\rangle . \tag{2.22}$$

Also, in what follows we take ϵ as k independent. In addition, we let

$$\tilde{\Omega}\equiv\frac{\mu\langle E_T(k)\rangle}{\hbar}\ , \tag{2.23}$$

$$\frac{\Delta_k}{\hbar}=(\omega_k-\omega_0)+(\omega_0-\omega_L)\ , \tag{2.24}$$

where

$$\omega_k=\frac{1}{\hbar}\left[\frac{\hbar^2k^2}{2m_{\text{eff}}}+E_g-\epsilon\right] \tag{2.25}$$

and

$$\Delta\equiv\frac{\omega_0-\omega_L}{\gamma_T}\ ,\quad \delta(k)=\frac{\omega_k-\omega_0}{\gamma_T}\ ,\quad \gamma_T=\frac{1}{T_2}\ . \tag{2.26}$$

Making use of (2.21) in Eq. (2.17) and using the relations (2.19) and (2.23)–(2.26), Eqs. (2.17) and (2.18) take the form

$$\frac{\partial\tilde{p}_k}{\partial t}=-\gamma_T(1+i\Delta)\tilde{p}_k-i\gamma_T\delta(k)\tilde{p}_k+i\epsilon W\tilde{p}_k-\frac{i}{2}\Omega w_k\ , \tag{2.27}$$

$$\frac{\partial w_k}{\partial t}=-\gamma_L(w_k-w_k^{(0)})-i[\Omega^*\tilde{p}_k-\tilde{p}_k^*\Omega]\ , \tag{2.28}$$

where we have made the rotating-wave approximation and $\tilde{p}_k$ and Ω are slowly varying, defined similar to Eqs. (2.5) and (2.6). We emphasize that we have taken

$$\sum_{k'\neq k}v(k-k')w_{k'}=\langle w_{k'}\rangle\sum_{k'\neq k}v(k-k')=\hbar\epsilon W\ . \tag{2.29}$$

It should be noted that $W=\langle w_{k'}\rangle$ is an average over the density of states and its appearance in Eq. (2.27) constitutes a carrier density-dependent renormalization of the band gap, i.e., a redshift with increasing carrier concentration. The strength of the shift is determined by the Debye factor ϵ, Eq. (2.20). For low carrier density the term that appears on the right-hand side (rhs) of Eq. (2.27) proportional to ϵ, then, is just the linear Debye shift of the band edge due to coherent Coulomb exchange interaction, Eq. (2.19). Equations (2.27) and (2.28) are the main result of this section.

It is instructive to note here that qualitatively a similar renormalization occurs because of the Lorenz-Lorentz local-field correction due to dipole-dipole interactions [5,25]. This is demonstrated in the Appendix. As shown there, a term of the form of the third term on the rhs of Eq. (2.27) is generated, but of opposite sign. The strength of the near dipole-dipole coupling, $\tilde{\epsilon}$, is proportional to μ^2 and the density of electrons but is normally quite small compared to ϵ. Also to note is the fact that the Lorenz-Lorentz correction involves the average $\langle p_k\rangle$ rather than $\langle w_k\rangle$ as for the coherent Coulomb exchange. Consequently, a term proportional to $\tilde{\epsilon}$ will appear on the rhs of microscopic Eq. (2.28) as well. Although this term vanishes in the averaged equation for a homogeneously broadened medium [25], it can make an important contribution for inhomogeneously broadened systems [5].

III. GENERALIZED BLOCH-MAXWELL EQUATIONS

This section is used to develop the macroscopic generalized Bloch equations from the microscopic equations of motion, Eqs. (2.27) and (2.28). The perturbative method of Graham and Cho [23] will be used in a similar development as presented in Ref. [5]. Here, we shall neglect the dipole-dipole contribution (see the Appendix) which was treated explicitly earlier [5], but, of course, retain the coherent Coulomb contribution, Eq. (2.27), which is normally much stronger. Since the two contributions, the Lorenz-Lorentz and the coherent Coulomb interaction, are additive, the local-field correction can be added at the end. To proceed unencumbered, we ignore the Lorenz-Lorentz contribution in this development.

If Eqs. (2.27) and (2.28) are averaged with respect to the density of states, the resulting macroscopic equations are

$$\frac{\partial P}{\partial t}=-\gamma_T(1+i\Delta)P-\gamma_T\langle i\delta(k)\tilde{p}_k\rangle+i\epsilon WP-\frac{i}{2}W\Omega\ , \tag{3.1}$$

$$\frac{\partial W}{\partial t}=-\gamma_L(W-W^{(0)})-i[\Omega^*P-P^*\Omega]\ . \tag{3.2}$$

Here, we have written the averaged variables in terms of slowly varying components:

$$P=\tilde{P}e^{-i\omega_Lt}\ ,\quad \Omega_k=\tilde{\Omega}_ke^{-i\omega_Lt}\ ,\quad \gamma_L=\frac{1}{T_1}\ .$$

Apart from the third term on the rhs of Eq. (3.1), these equations are precisely of the form for the optical Bloch equations for an inhomogeneously broadened system. This term, however, leads to profound and novel consequences.

The usual problem with regard to inhomogeneously broadened systems is embodied in the second term on the rhs of Eq. (3.1), which involves the first moment of the microscopic polarization with respect to the density-of-states distribution. Following the scheme of Graham and Cho [23], we introduce a new variable S and expansion parameter $\mathcal{S}^2$, to be specified later, such that

$$\mathcal{S}^2S\equiv\langle i\delta\tilde{p}_k\rangle \tag{3.3}$$

and the corresponding additional macroscopic equation of motion

$$\mathcal{S}^2\frac{\partial S}{\partial t}=\left\langle i\delta\frac{\partial\tilde{p}_k}{\delta t}\right\rangle\ . \tag{3.4}$$

Thus, inserting the indicated macroscopic equation of motion, Eq. (2.27), into (3.4) and using the definition, (3.3), Eq. (3.4) can be written as

$$\mathcal{S}^2\frac{\partial S}{\partial t}=-\gamma_T\mathcal{S}^2S-i\Delta\mathcal{S}^2S+\gamma_T\langle\delta^2\tilde{p}_k\rangle+i\epsilon W\mathcal{S}^2S+\frac{\Omega}{2}\langle\delta w_k\rangle\ . \tag{3.5}$$

Here, the second moment of the microscopic polarization, p_k, appears in the third term on the rhs and the first moment of the carrier distribution appears in the last

term. This suggests the introduction of additional new variables, U and P_0:

$$U \equiv \langle \delta w_k \rangle / \mathcal{S}^2 ,$$
$$P_0 \equiv \left\langle \left[1 - \frac{\delta^2}{\mathcal{S}^2} \right] \tilde{p}_k \right\rangle , \tag{3.6}$$

where the form of P_0 in terms of the second moment has been chosen to facilitate explicit determination of $\mathcal{S}^2$ upon termination of the heirarchy of the new macroscopic equations to lowest order. It is seen from (3.6) that the variable U is a dynamical measure of the asymmetry of the carrier density distribution with respect to the density of states. Thus, in terms of (3.6), Eq. (3.5) becomes

$$\frac{\partial S}{\partial t} = -\gamma_T(1+i\Delta)S + i\epsilon W S + \frac{\Omega}{2}U + \gamma_T(P - P_0) , \tag{3.7}$$

and we have, from (3.6), another additional equation:

$$\frac{\partial U}{\partial t} = \frac{1}{\mathcal{S}^2}\left\langle \delta \frac{\partial w_k}{\partial t} \right\rangle . \tag{3.8}$$

Upon inserting Eq. (2.28) into (3.8), we obtain the equation

$$\frac{\partial U}{\partial t} = -\gamma_c(U - \bar{U}) - [\Omega^* S + S^* \Omega] , \tag{3.9}$$

where $\bar{U}$ is the quasiequilibrium value of U established by the intraband relaxation processes. The intraband relaxation rate γ_c has been introduced in Eq. (3.9). At this point we terminate the heirarchy to lowest order in the perturbation by choosing $P_0 = 0$. From (3.6), this gives an evaluation of the expansion parameter $\mathcal{S}^2$,

$$\mathcal{S}^2 = \frac{\langle \delta^2 \tilde{p}_k^{ss} \rangle}{\langle \tilde{p}_k^{ss} \rangle} , \quad P_0 = 0 , \tag{3.10}$$

where we have chosen to evaluate the normalized second moment at steady state.

Collecting the macroscopic equations together, Eqs. (3.1), (3.2), (3.7), and (3.9) with (3.3) and condition (3.10), we have the closed set of generalized Bloch-Maxwell equations:

$$\frac{\partial W}{\partial t} = -\gamma_L(W - W^{(0)}) - i[\Omega^* P - P^* \Omega] + \langle \Lambda \rangle , \tag{3.11}$$

$$\frac{\partial P}{\partial t} = -\gamma_T(1+i\Delta)P - \mathcal{S}^2\gamma_T S + i\epsilon W P - \frac{i}{2}W\Omega , \tag{3.12}$$

$$\frac{\partial S}{\partial t} = -\gamma_T(1+i\Delta)S + i\epsilon W S + \frac{\Omega}{2}U + \gamma_T P , \tag{3.13}$$

$$\frac{\partial U}{\partial t} = -\gamma_c(U - \bar{U}) - [\Omega^* S + S^* \Omega] , \tag{3.14}$$

$$\frac{\partial \Omega}{\partial z} + \frac{1}{V_g}\frac{\partial \Omega}{\partial t} = \frac{i\omega_L \mu^2 \rho}{\epsilon_0 \hbar c n} P . \tag{3.15}$$

A pump term $\langle \Lambda \rangle$ was added as the last term in (3.11). Equation (3.15) is the Maxwell wave equation in the slowly varying envelope and phase approximation (SVEA), which couples the field Ω with the macroscopic polarization P. Parameters in Eq. (3.15) are the group velocity of the field propagation V_g, ρ is the number of density of electrons, and n is the linear index of refraction for the medium. Equations (3.11)–(3.15) constitute a self-consistent set of generalized Bloch-Maxwell equations to lowest order in the density-of-states distribution and to lowest order in the coherent Coulomb-exchange interaction. All of the details of the density-of-states distribution are contained in the single, time-independent expansion parameter, Eq. (3.10). These equations are the main result of this paper. The next several sections are used to present and discuss several results which stem from this formulation.

IV. RATE-EQUATION AND STEADY-STATE LIMITS

A. Rate-equation limit

The rate-equation limit is achieved under conditions for adiabatic elimination:

$$\left| \frac{\partial P}{\partial t} \right| \ll \gamma_T |P| , \quad \left| \frac{\partial S}{\partial t} \right| \ll \gamma_T |S| ,$$
$$\frac{\partial P}{\partial t} = \frac{\partial S}{\partial t} = \frac{\partial U}{\partial t} = 0 . \tag{4.1}$$

This leads to [5]

$$\frac{\partial W}{\partial t} = \langle \Lambda \rangle - \gamma_L(W - W^{(0)}) - \frac{|\Omega|^2 W}{\gamma_T(1+\mathcal{S}^2)} . \tag{4.2}$$

Since $W = 2\langle n_k \rangle - 1$, and if ρ is the volume density of electrons, and if we use the carrier density $N \equiv \rho \langle n_k \rangle$, we get

$$\frac{\partial N}{\partial t} = \frac{I_{IN}}{qV} - \gamma_L N - \frac{\gamma_L}{(1+\mathcal{S}^2)}\frac{|\varepsilon|^2}{I_S}(N - N_0) , \tag{4.3}$$

which is the standard form for the rate equation for the carrier concentration, except that all parameters are now well defined in terms of fundamental physical quantities. In the derivation of Eq. (4.3) we have taken $\Delta/\gamma_T \ll 1$ and $\varepsilon/\gamma_T \ll 1$ for simplicity. Here, N_0 is the thermal equilibrium value, $I_{IN} = qV\rho\langle \Lambda \rangle / 2$ and $I_S = (\hbar^2\mu^2)/(\gamma_T\gamma_L)$ is the interband saturation intensity, q is the magnitude of the elementary charge, and V is the Fermi velocity. (I_{IN} is thus the injection current.) Equation (4.3) is isomorphic to the simplest standard rate equation for semiconductors [9]. As noted elsewhere [5], the usual phenomenological parameters that appear in (4.3) can now be given precise physical meaning in terms of material parameters. The usual gain parameter a,

$$a = \frac{\mu^2}{\hbar^2 \gamma_T (1+\mathcal{S}^2)} , \tag{4.4}$$

is given explicitly in terms of the dipole moment matrix element μ, the dipole relaxation rate γ_T, and the inhomogeneous distribution parameter $\mathcal{S}^2$. Using the constitutive relation $p = \epsilon_0 \chi \varepsilon$, the susceptibility χ is

$$\chi = -\frac{\mu^2 \rho}{\epsilon_0 \hbar \gamma_T (1+\mathcal{S}^2)} \left[iW + \frac{\mathcal{S}^2 \bar{U}}{1 + |\varepsilon|^2 / \tilde{I}_S} \right] , \tag{4.5}$$

which depends upon the intraband saturation intensity $\bar{I}_S=\hbar^2\gamma_L\gamma_T/\mu^2(1+\mathcal{S}^2)$ and the asymmetry term $\bar{U}$ which involves redistribution of electrons within the conduction band. The real and imaginary parts of (4.5) are related to the index change $\Delta n=\mathrm{Re}\chi/(2n)$ and the optical gain $g=-2k\,\mathrm{Im}\chi$. The important linewidth enhancement factor β_c is given by

$$\beta_c=\mathrm{Re}\chi/\mathrm{Im}\chi=\frac{\mathcal{S}^2(\bar{U}/W)}{1+|\varepsilon|^2/I_S}$$

and is manifestly proportional to the asymmetry factor $\bar{U}$. The term β_c has been interpreted for short pulse excitation [6].

B. Steady state: Intrinsic optical bistability

If conditions of steady state are considered,

$$\frac{\partial W}{\partial t}=\frac{\partial P}{\partial t}=\frac{\partial S}{\partial t}=\frac{\partial U}{\partial t}=0\ ,$$

the solution for the carrier density W as a function of the field Ω is

$$\frac{\gamma_L}{\gamma_T}(W-W^{(0)})[\gamma_T^2(1+\mathcal{S}^2)+(\Delta-\epsilon W)^2]=-W|\Omega|^2\ . \tag{4.6}$$

In arriving at (4.6) we have neglected asymmetry contributions for the distribution of carriers in the conduction band, i.e., we have taken $\bar{U}=0$.

Equation (4.6) is cubic in W and leads to intrinsic optical bistability [27] for the carrier concentration W, as a function of the field Ω, for suitable values of the parameters. This is entirely a local condition and does not require optical feedback, as would be provided by cavity mirrors. For conditions such that three real distinct roots exist for the solution, Eq. (4.6), then represents a first-order phase transition for the system far from thermodynamic equilibrium. The cubic condition stems explicitly from the coherent Coulomb-exchange interaction contribution determined by the parameter ϵ, the Debye shift.

Equation (4.6) is represented in Fig. 1 for zero detuning and various values for ϵ/γ_T. The threshold for the phase transition is approximately the condition that the Debye shift ϵ equal the homogeneous linewidth γ_T. Larger values of the ratio lead to larger contrast between the upper and lower states for the same intensity $|\Omega|^2$, which leads to reversible hysteresis if $|\Omega|^2$ is varied adiabatically. A linearized stability analysis about the steady state indicates that the upper and lower states for the same $|\Omega|^2$ are stable, whereas the intermediate state is always unstable and corresponds to a saddle condition. Equation (4.6) depicts a renormalization (shrinkage) of the band gap with increase in carrier density, as explicitly noted by the term in brackets on the left-hand side which contains ϵ. Such redshifts with regard to the band edge and bistability have been observed [27].

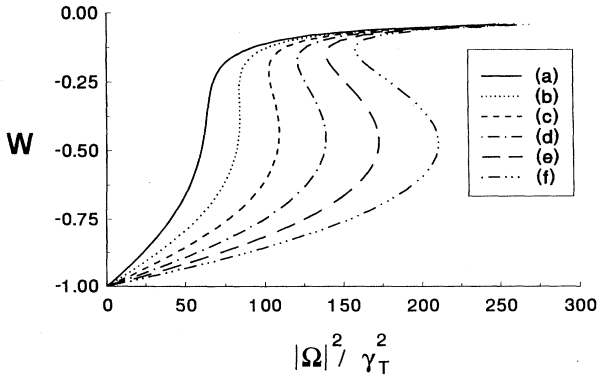

FIG. 1. Equation (4.6), W vs $|\Omega|^2/\gamma_T^2$ for $\gamma_L/\gamma_T=2$, $\mathcal{S}^2=4$, $W^{(0)}=-1$, and $\Delta=0$, for different values of ϵ/γ_T; ϵ/γ_T = (a) 10, (b) 12, (c) 14, (d) (16), (e) 18, (f) 20.

V. ULTRAFAST PHENOMENA

Ultrafast dynamical processes in semiconductors can be characterized under conditions for subpicosecond pulse excitation where the temporal behavior of the excitation pulse $\varepsilon=\varepsilon(t/\tau_P)$ is such that

$$\tau_P\ll\gamma_T^{-1}\ ,\ \gamma_L^{-1}\ . \tag{5.1}$$

Then, Eqs. (3.11) and (3.12) decouple from the hierarchy (3.11)–(3.14), and the result is

$$\frac{\partial W}{\partial t}=-i[\Omega^*P-P^*\Omega]+\langle\Lambda\rangle\ , \tag{5.2}$$

$$\frac{\partial P}{\partial t}=-i[\Delta-\epsilon W]P-\frac{i}{2}W\Omega\ . \tag{5.3}$$

So, in the short-pulse regime, explicit effects of the inhomogeneous distribution over the density of states are completely decoupled from the equations of motion. However, the coherent Coulomb interaction contribution is manifestly present in the nonlinear term in Eq. (5.3). This contribution can have novel and profound effects in the system dynamics.

As an example, we set the gain term $\langle\Lambda\rangle=0$ and analyze the dynamics for short-pulse-induced excitation. For convenience, the polarization P is written in terms of real and imaginary components, v and u, respectively.

$$P\equiv\tfrac{1}{2}(v+iu)\ . \tag{5.4}$$

Thus, Eqs. (5.2) and (5.3) are equivalent to

$$\frac{\partial u}{\partial t}=-(\Delta-\epsilon W)v\ , \tag{5.5}$$

$$\frac{\partial v}{\partial t}=(\Delta-\epsilon W)u+\Omega W\ , \tag{5.6}$$

$$\frac{\partial W}{\partial t}=-\Omega v\ . \tag{5.7}$$

Equations (5.5)–(5.7) are exactly equivalent to the J model presented in Ref. [17] as the dynamical equivalent to Eqs. (2.9) and (2.10) in the regime where $|\Omega|/\epsilon\approx1$. As pointed out in Ref. [17], novel ultrafast phenomena such as intrinsic adiabatic inversion and fast optical switching discussed in Ref. [28] for short-pulse excitation dynamics and interpreted in terms of adiabatic following [29] per-

tain directly to time integration results for Eqs. (5.5)–(5.7). Much of the results of simulation studies of Eqs. (2.9) and (2.10) [14,15,17] can now be qualitatively interpreted on the basis of Eqs. (3.11)–(3.15) and Eqs. (5.5)–(5.7).

VI. SUMMARY AND CONCLUSION

A widely used, two-band, laser-driven, many-body semiconductor model was used to obtain the microscopic Bloch-like equations to lowest order in the coherent Coulomb-exchange interaction, Eqs. (2.17) and (2.18). The macroscopic equations were generated from these equations by averaging over the density-of-states distribution using a perturbative scheme to obtain a closed set of Maxwell-Bloch-like equations to lowest order in the density-of-states distribution, as well as the coherent Coulomb-exchange interaction, Eqs. (3.11)–(3.15). All of the information concerning the density-of-states distribution is self-contained in the single expansion parameter, Eq. (3.10). Equations (3.11)–(3.15) are the main result of this paper.

Equations (3.11)–(3.14) were shown to reduce in the adiabatic limit to the form for the simplest, commonly used rate equation for the carrier density, Eq. (4.3), where all parameters from our model equations are expressive of definite materials properties, Eqs. (4.4) and (4.5). The steady-state conditions lead to a first-order phase transition far from thermodynamic equilibrium, Eq. (4.6), which is a manifestation of the coherent Coulomb-exchange interaction which causes a carrier density-dependent redshift of the band gap.

The limit of ultrafast pulse excitation was shown to result in Eqs. (5.5)–(5.7) which are isomorphic with the J-model equations of Ref. [17]. There, the equations were shown to adequately reproduce the dynamics of Eqs. (2.9) and (2.10) in the short excitation pulse regime within a large region of the parameter space. The inclusion of the effects of the density of states in the perturbative treatment to obtain Eqs. (3.11)–(3.15), the main results of this paper, should extend the parameter region of validity of the generalized Maxwell-Bloch equations. As cited in Ref. [17], the results in the ultrafast regime reported in Refs. [28] and [29] carry over completely with regard to Eqs. (5.5)–(5.7).

Future work will focus upon validation of the dynamics and regions of validity of Eqs. (3.11)–(3.15) and (5.5)–(5.7) with regard to Eqs. (2.9) and (2.10). We will also focus upon the analysis of fundamental processes and phenomena associated with these equations to formulate interesting experiments and provide analysis. It is anticipated that the formulation presented here contains a plethora of new and interesting fundamental physics of semiconductors as yet untapped.

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APPENDIX

We develop the contribution to the local field, E_L, which stems from the coupling of the dipoles with the field modes [26],

$$\tilde{E}_L = E + \frac{4\pi}{3}\langle p_k \rangle \ . \tag{A1}$$

Here, we ignore for the moment the Coulomb-exchange effects. Elimination of the local field from Eqs. (2.17) and (2.18) using (A1) yields

$$\frac{\partial p_k}{\partial t} = -\gamma_T(1+i\bar{\Delta})p_k - i\gamma_T \delta p_k - i\tilde{\epsilon} w_k \langle p_k \rangle - \frac{i}{2}\Omega w_k \ , \tag{A2}$$

$$\frac{\partial w_k}{\partial t} = -\gamma_L(w_k - w_k^{(0)}) - i[\Omega^* p_k - p_k^* \Omega] - 2i\tilde{\epsilon}[\langle p_k \rangle^* p_k - p_k^* \langle p_k \rangle] \ , \tag{A3}$$

where

$$\tilde{\epsilon} = \frac{4\pi\mu^2\rho}{3} \ , \quad \Omega = \frac{\mu\mathcal{E}}{\hbar} \ . \tag{A4}$$

We notice that these equations, (A2) and (A3) as well as the coupling Eq. (A4), are qualitatively different from Eqs. (2.17) and (2.18). This is due to the nature of the coupling in the two cases. However, the effect of combining the two interactions in the averaged equations, Eqs. (3.1) and (3.2), is additive. The result for the macroscopic generalized Maxwell-Bloch equations, however, is qualitatively different. We therefore present here the result using (A1). Using the identical perturbative procedure as used in Sec. III, the generalized Maxwell-Bloch equations are

$$\frac{\partial W}{\partial t} = -\gamma_L(W - W^{(0)}) - i[\Omega^* P - P^* \Omega] \ , \tag{A5}$$

$$\frac{\partial P}{\partial t} = -\gamma_T(1+i\Delta)P - \mathcal{S}^2\gamma_T S - i\tilde{\epsilon} P W - \frac{i}{2}\Omega W \ , \tag{A6}$$

$$\frac{\partial S}{\partial t} = -\gamma_T(1+i\Delta)S + \gamma_T P + \tilde{\epsilon} P U + \tfrac{1}{2}\Omega U \ , \tag{A7}$$

$$\frac{\partial U}{\partial t} = -\gamma_c(U - \bar{U}) - [\Omega^* S + \Omega S^*] - 2\tilde{\epsilon}(P^* S + S^* P) \ , \tag{A8}$$

$$\frac{\partial \Omega}{\partial z} + \frac{1}{V_g}\frac{\partial \Omega}{\partial t} = \frac{i\omega_L \mu^2 \rho P}{\epsilon_0 \hbar c n} \ . \tag{A9}$$

The above equations are qualitatively distinct from Eqs. (3.11)–(3.15) in the terms in $\tilde{\epsilon}$. A sign change by comparison occurs in Eq. (A6). The third term on the rhs, Eq. (A7), is in qualitative discrepancy with its counterpart, Eq. (3.13), while the last term on the rhs, Eq. (A8), is in contradistinction to the complete absence of an ϵ-dependent term in Eq. (3.14). The net response of the system is the combined effect which can be trivially obtained by combining the respective contributions from Eqs. (3.11)–(3.15) and (A5)–(A9). These contributions can have important effects upon the dynamics as well as the steady state of the system.

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