Effective Bloch equations for semiconductors

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Generalized Bloch equations for laser-excited semiconductors are derived applying quantum-mechanical projection-operator techniques. The equations include phase-space filling and the many-body Coulomb effects. The coherent part of the equations is evaluated for the regime of ultrafast-pump-probe excitation and shown to reduce to the inhomogeneously broadened two-level Bloch equations for the different momentum states if the proper Coulomb enhancement in the density of states is taken into account. For the high-excitation quasithermal regime a generalization of the Elliott formula for the absorption spectrum is derived which is valid not only for bulk semiconductors, but also for quantum-well structures and other systems with reduced spatial dimensions.

I. INTRODUCTION

A resonant laser field applied to a semiconductor generates electron-hole pairs by exciting electrons over the energy gap from the valence band into the conduction band. The electron-hole pairs exist for a characteristic lifetime before they recombine radiatively or nonradiatively. Typically, the electron-hole-pair recombination time is long compared to the time scale of intraband carrier-scattering processes which cause a thermalization of the electron and hole populations within the bands. The time regime after the quasiequilibration and before recombination defines the so-called quasiequilibrum or hydrodynamic regime. The optical properties of semiconductors in this regime have been investigated extensively.[1-3]

Quite recently, the development in ultrafast laser-pulse techniques made it possible to study semiconductors also in states where the dynamics of the excitation is still relevant, i.e., on time scales which are shorter than or at least comparable to the carrier-carrier scattering times. Examples for the observed phenomena are the optical Stark effect,[4,5] the spectral hole burning[6-8] and transient oscillations in semiconductor transmission spectra.[9,10]

It is known from the spectroscopy of atomic systems that the optical Bloch equations are well suited to describe the interaction between light and matter, including coherent effects and relaxation phenomena. A similar description for semiconductors has been obtained recently by Haug *et al.*[11] These authors present a set of equations which can be considered as a generalization of the optical Bloch equations for semiconductors. The equations contain the most important many-body effects which renormalize the band gap of the unexcited system, they contain the effects of electron-hole attraction giving rise to excitonic effects, and they include the contributions of phase-space filling. Haug *et al.*[11] derive these effective Bloch equations with the help of nonequilibrium Green's-function techniques.

In this paper, we show how these equations can be derived and generalized in a simple way by using quantum-mechanical projection-operator techniques.[12] Furthermore, we discuss some limiting cases and applications of the effective Bloch equations. After the Hamiltonian formulation of our problem (Sec. II) we derive the equations of motion in Sec. III. In Sec. IV we show how the equations can be used to study ultrafast effects in semiconductor pump-probe spectroscopy. For the case of not too strong excitation, when screening and band-gap renormalization can still be neglected, but Coulomb attraction between electrons and holes is important, our equations for the population and polarization of the different momentum states become formally identical to the inhomogeneously broadened Bloch equations known from quantum optics. However, when we compute the total polarization and sum over the continuum of states, the Coulomb attraction of electrons and holes gives rise to a weighting factor (Coulomb enhancement) which is responsible for the replacement of the square-root absorption shape of noninteracting particles by the more or less structureless continuum absorption in the case with Coulomb attraction.

In Sec. V we study the high-excitation quasiequilibrium regime where the carrier distributions are assumed to be quasi-Fermi functions within the bands. The consistent inclusion of phase-space filling factors makes the problem non-Hermitian. We derive a generalization of the Elliott formula for the excitation-dependent absorption coefficient, which is valid independent of the dimensionality of the studied semiconductor system. For the case of bulk semiconductors, when it is justified to neglect phase-space filling terms in the Wannier equation, we show how the generalized Elliott formula reduces to the result obtained in Ref. 13.

Some mathematical details of the quantum-mechanical projection-operator technique are summarized in the Appendix.

II. HAMILTONIAN FORMULATION

The optical properties of a semiconductor are mainly determined by the conduction band and the uppermost valence bands. To make the analysis as simple as possible we assume a nondegenerate situation described by the Hamiltonian

$$H=\sum_k \varepsilon_c(k)a^\dagger_{c,k}a_{c,k}+\sum_k \varepsilon_v(k)a^\dagger_{v,k}a_{v,k}+V\ , \tag{1a}$$

where

$$V=\tfrac{1}{2}\sum_{k,k';q\,(\neq 0)} V(q)(a^\dagger_{c,k+q}a^\dagger_{c,k'-q}a_{c,k'}a_{c,k} + a^\dagger_{v,k+q}a^\dagger_{v,k'-q}a_{v,k'}a_{v,k} + 2a^\dagger_{c,k+q}a^\dagger_{v,k'-q}a_{vk'}a_{ck})\ . \tag{1b}$$

The energies $\varepsilon_c(k)$ and $\varepsilon_v(k)$ are defined for a single electron in the lattice. In addition, we have assumed that the Coulomb interaction conserves the number of carriers in each band. The dipole coupling to a laser field is described in the rotating-wave approximation

$$H_f=-\sum_k [\mu_k E(t)a^\dagger_{c,k}a_{v,k}+\mu^*_k E^*(t)a^\dagger_{v,k}a_{c,k}]\ . \tag{2}$$

It is often convenient to transform the Hamiltonian $H+H_f$ into the electron-hole picture. Using the notation

$$\begin{aligned} a_{c,k}&\rightarrow a_k\ ,\\ a^\dagger_{v,k}&\rightarrow b_{-k}\end{aligned} \tag{3}$$

where a_k and b_{-k} are the electron and hole annihilation operators, respectively, and introducing normal ordering of the new operators, we obtain

$$H+H_f=\sum_k [\varepsilon^s_e(k)a^\dagger_k a_k+\varepsilon^s_h(k)b^\dagger_{-k}b_{-k}]+\tfrac{1}{2}\sum_{k,k';q\,(\neq 0)} V(q)(a^\dagger_{k+q}a^\dagger_{k'-q}a_{k'}a_k+b^\dagger_{k+q}b^\dagger_{k'-q}b_{k'}b_k-2a^\dagger_{k+q}b^\dagger_{k'-q}b_{k'}a_k) - \sum_k [\mu_k E(t)a^\dagger_k b^\dagger_{-k}+\mu^*_k E^*(t)b_{-k}a_k]+\sum_k \varepsilon_v(k)-\tfrac{1}{2}\sum_{k;q\,(\neq 0)} V(q)\ , \tag{4}$$

where the effective single-particle energies are

$$\begin{aligned} \varepsilon^s_e(k)&=\varepsilon_c(k)\ ,\\ \varepsilon^s_h(k)&=-\varepsilon_v(k)+\sum_{q\,(\neq 0)} V(q)\ .\end{aligned} \tag{5}$$

These energies are often approximated in effective-mass theory, assuming the bands to be parabolic near the extrema. The transformation of the Hamiltonian into the electron-hole picture shows that the low-intensity interband excitation spectrum, which is determined by the energy difference between the states with zero and one electron-hole pairs, is given by

$$\Delta\varepsilon^s(k)=\varepsilon_c(k)-\varepsilon_v(k)+\sum_{q\,(\neq 0)} V(q)\ . \tag{6}$$

The last two terms in the Hamiltonian (4) are constants giving the total energy of the full valence band. They have no dynamical consequences and are left out.

III. EQUATIONS OF MOTION

In this section, we derive the equations of motion for the expectation values $\langle a^\dagger_k a_k\rangle=n_{e,k}$, $\langle b^\dagger_{-k}b_{-k}\rangle=n_{h,k}$, and $\langle a^\dagger_k b^\dagger_{-k}\rangle=p^*_k$. The first two expressions are simply the populations of the electrons and holes at the wave vector k and the third gives the polarization of the medium, which becomes macroscopic because of the applied external field. The Hamiltonian equation of motion for the expectation value of operator A in the density-matrix formalism is ($\hbar\equiv 1$)

$$\frac{\partial}{\partial t}\langle A\rangle=-i\,\mathrm{tr}([A,H_{\mathrm{tot}}]\rho)\ . \tag{7}$$

Straightforward operator-algebra manipulations yield

$$\frac{\partial}{\partial t}p^*_k=i[\varepsilon^s_e(k)+\varepsilon^s_h(k)]p^*_k+i\mu^*_k E^*(t)(n_{e,k}+n_{h,k}-1) + i\sum_{k';q\,(\neq 0)} V(q)(\langle a^\dagger_{k-q}a^\dagger_{k'+q}a_{k'}b^\dagger_{-k}\rangle-\langle a^\dagger_k a^\dagger_{k'+q}a_{k'}b^\dagger_{-k-q}\rangle + \langle a^\dagger_k b^\dagger_{-k-q}b^\dagger_{k'+q}b_{k'}\rangle-\langle a^\dagger_{k+q}b^\dagger_{k'-q}b_{k'}b^\dagger_{-k}\rangle)\ , \tag{8a}$$

$$\frac{\partial}{\partial t}n_{e,k}=-2\,\mathrm{Im}[\mu_k E(t)p^*_k]+i\sum_{k';q\,(\neq 0)} V(q)(\langle a^\dagger_k a^\dagger_{k'-q}a_{k-q}a_{k'}\rangle-\langle a^\dagger_{k+q}a^\dagger_{k'-q}a_k a_{k'}\rangle + \langle a^\dagger_k a_{k-q}b^\dagger_{k'-q}b_{k'}\rangle-\langle a^\dagger_{k+q}a_k b^\dagger_{k'-q}b_{k'}\rangle)\ , \tag{8b}$$

and

$$\frac{\partial}{\partial t}n_{h,k}=-2\,\mathrm{Im}[\mu_k E(t)p^*_k]+i\sum_{k';q\,(\neq 0)} V(q)(\langle b^\dagger_{-k}b^\dagger_{k'-q}b_{-k-q}b_{k'}\rangle-\langle b^\dagger_{-k+q}b^\dagger_{k'-q}b_{-k}b_{k'}\rangle + \langle a^\dagger_{k'+q}a_k b^\dagger_{-k}b_{-k+q}\rangle-\langle a^\dagger_{k'+q}a_k b^\dagger_{-k-q}b_{-k}\rangle)\ . \tag{8c}$$

As usual, the equations of motion include couplings to higher-order correlations. In this paper we derive systematic approximations to these correlation terms using projection-operator techniques.[12] As a result, the equations of motion are separated into two parts. In the Appendix we briefly summarize the main steps of the projection method. Here we only notice that the basic idea is that—without approximation—the density matrix of the system can be projected into two parts, σ and $\delta\rho$, describing the "coherent contributions" and the "scattering contributions," respectively,

$$\begin{aligned}\frac{\partial}{\partial t}\langle A\rangle &= -i\,\mathrm{tr}([A,H_{\mathrm{tot}}]\rho)\\ &= -i\,\mathrm{tr}([A,H_{\mathrm{tot}}]\sigma)-i\,\mathrm{tr}([A,H_{\mathrm{tot}}]\delta\rho)\\ &= \frac{\partial}{\partial t}\langle A\rangle_{\mathrm{coh}}+\frac{\partial}{\partial t}\langle A\rangle_{\mathrm{scat}}\ . \end{aligned} \tag{9}$$

In the following, we first compute the coherent contributions by taking the expectation values with the part σ of the density matrix,

$$\langle A\rangle_{\mathrm{coh}}=\mathrm{tr}(A\sigma)\ .$$

It turns out that the coherent part of the equations of motion is equivalent to the result obtained in time-dependent Hartree-Fock approximation, since the four-operator expectation values simply factorize into products of two-operator expectation values. With our choice of σ, Eq. (A6), we keep only those products in the factorization which contain "macroscopic" expectation values, i.e., which are diagonal in wave-vector indices such as

$$\langle a_k^\dagger a_{k'}^\dagger a_p a_{p'}\rangle_{\mathrm{coh}}=(\delta_{k,p}\delta_{k',p}-\delta_{k,p}\delta_{k',p'})n_{e,k}n_{e,k'}\ . \tag{10}$$

As a result we obtain the coherent part of the equations of motion in the form

$$\begin{aligned}\frac{\partial}{\partial t}p^*_{k,\mathrm{coh}} &= i\left\{\varepsilon_e^s(k)+\varepsilon_h^s(k)\right.\\ &\quad\left.-\sum_{q\,(\neq 0)}V(q)(n_{e,k+q}+n_{h,k+q})\right\}p_k^*\\ &\quad -i\left\{\mu_k^*E^*(t)+\sum_{q\,(\neq 0)}V(q)p^*_{k+q}\right\}\\ &\quad\times(1-n_{e,k}-n_{h,k})\ , \end{aligned} \tag{11a}$$

$$\frac{\partial}{\partial t}n_{e,k,\mathrm{coh}}=-2\,\mathrm{Im}\left[\left\{\mu_k E(t)+\sum_{q\,(\neq 0)}V(q)p_{k+q}\right\}p_k^*\right]\ , \tag{11b}$$

and

$$\frac{\partial}{\partial t}n_{h,k,\mathrm{coh}}=\frac{\partial}{\partial t}n_{e,k,\mathrm{coh}}\ . \tag{11c}$$

Since there are no scattering contributions in Eqs. (11), Eq. (11c) gives the constant of motion

$$n_{e,k}-n_{h,k}=0\quad\text{for all } k\ , \tag{12}$$

showing that the electron and hole distributions remain equal.

The scattering parts of the equations of motion have to be computed from

$$\mathrm{tr}([\hat{\alpha}_i,H]\delta\rho)\ ,$$

where $\{\hat{\alpha}_i\}=\{a_k^\dagger a_k, b^\dagger_{-k}b_{-k}, a_k^\dagger b^\dagger_{-k}, b_{-k}a_k\}$. Using the definitions of H and $\delta\rho$ we see that

$$\mathrm{tr}([\hat{\alpha}_i,H]\delta\rho)=\mathrm{tr}([\hat{\alpha}_i,V]\delta\rho)\ , \tag{13}$$

where V is the Coulomb interaction part (1b) of the Hamiltonian. To get explicit results we have to approximate $\delta\rho$. Following the argumentation in the Appendix, we find that the properties of H, Q, and $\delta\rho$ give the Eq. (A12) in the form

$$i\frac{\partial}{\partial t}\delta\rho=[H,\delta\rho]-\sum_i\frac{\partial\sigma}{\partial\alpha_i}\mathrm{tr}([\hat{\alpha}_i,V]\delta\rho)+Q([H,\sigma])\ . \tag{14}$$

The last term on the right-hand side (rhs) of Eq. (14) is a source term which is known. To be able to solve Eq. (14) we have to make some approximations. First, we neglect the second term on the rhs and, in the first term, we replace H by the effective Hamiltonian H_{eff}. This effective Hamiltonian is linear in the operators $\hat{\alpha}_i$ but it is also a functional of the expectation values α_i. It is determined such that it yields the coherent Eqs. (11a)–(11c) as result of

$$i\frac{\partial}{\partial t}\alpha_i=\mathrm{tr}([\hat{\alpha}_i,H_{\mathrm{eff}}]\rho)\ . \tag{15}$$

In our case the effective Hamiltonian has the form

$$\begin{aligned}H_{\mathrm{eff}} &= \sum_k[\varepsilon_e^s(k)a_k^\dagger a_k+\varepsilon_h^s(k)b^\dagger_{-k}b_{-k}]\\ &\quad-\sum_{k;q\,(\neq 0)}[V(q)(n_{e,k-q}a_k^\dagger a_k+n_{h,k-q}b^\dagger_{-k}b_{-k}\\ &\qquad+p^*_{k-q}b_{-k}a_k+p_{k-q}a_k^\dagger b^\dagger_{-k})]\ . \end{aligned} \tag{16}$$

The Coulomb interaction is responsible for the shift of the single-particle energies. Additionally, the last two terms on the rhs of (16) represent an effective dipole interaction of the material with its own macroscopic polarization (self-field coupling of the polarization). The effective Hamiltonian is time dependent. The analysis shows that keeping this time dependence of H_{eff}, Q, and σ causes the solution to depend on the history of the expectation values α_i. In the following, we therefore assume that only the rapid time dependence at optical frequencies is important in H_{eff}, $Q(t)$, and $\sigma(t)$. This assumption is equivalent to neglecting the memory effects.

We then have to solve the equation

$$i\frac{\partial}{\partial t}\delta\rho=[H_{\mathrm{eff}},\delta\rho]+Q([H,\sigma])\ . \tag{17}$$

We furthermore assume that the polarization is small so that we are justified to neglect the self-field terms proportional to p_k in H_{eff} and in the final scattering terms we keep at most terms linear in p_k. This leads to the situation that the scattering rate of the populations will be independent of the polarization components. The general

case can also be solved but is slightly more tedious. We want to emphasize that the scattering takes place between states which have as energies the *Hartree-Fock energies* which are shifted as a consequence of the Coulomb interaction between the particles.

As a consequence of our approximations, the effective Hamiltonian is diagonal and independent of time. In the adiabatic limit, when initial conditions become irrelevant, the solution of Eq. (17) therefore has the form

$$\delta\rho = -i\int_{-\infty}^{\infty} dt'\, e^{-\eta(t-t')} e^{-iH_{\rm eff}(t-t')} Q(t') \times [V, \sigma(t')] e^{iH_{\rm eff}(t-t')} . \tag{18}$$

Since the memory effects are neglected, it is now straightforward to evaluate (18) and we suppress the details of the algebra.

In deriving the explicit form of the Coulomb matrix element entering the scattering rates we only take the most divergent contribution of the unscreened Coulomb potential. This approximation is equivalent to ignoring the exchange term $V(q)V(k-k')$ from the Born approximation[14] of the scattering matrix element $V(q)[V(q)-V(k-k')]$.

The scattering term for the electron population $n_e(k)$ is given by

$$\begin{aligned}\frac{\partial}{\partial t} n_{e,k,\rm scat} = &- \sum_{k';q\,(\neq 0)} 2\pi V^2(q)\delta(\tilde{\varepsilon}_e(k)+\tilde{\varepsilon}_e(k'+q)-\tilde{\varepsilon}_e(k')-\tilde{\varepsilon}_e(k+q)) \\ &\times [n_{e,k} n_{e,k'+q}(1-n_{e,k'})(1-n_{e,k+q}) - n_{e,k'} n_{e,k+q}(1-n_{e,k})(1-n_{e,k'+q})] \\ &- \sum_{k';q\,(\neq 0)} 2\pi V^2(q)\delta(\tilde{\varepsilon}_e(k)+\tilde{\varepsilon}_h(k')-\tilde{\varepsilon}_e(k+q)-\tilde{\varepsilon}_h(k'+q)) \\ &\times [n_{e,k} n_{h,k'}(1-n_{e,k+q})(1-n_{h,k'+q}) - n_{e,k+q} n_{h,k'+q}(1-n_{e,k})(1-n_{h,k'})] ,\end{aligned} \tag{19}$$

where we have denoted the shifted single-particle energies by

$$\tilde{\varepsilon}_i(k) = \varepsilon_i^s(k) - \sum_{q\,(\neq 0)} V(q) n_i(k), \quad i = e,h . \tag{20}$$

The scattering term for the hole population $n_h(k)$ is obtained from Eq. (19) by changing everywhere $e \leftrightarrow h$.

Finally, we have to compute the scattering terms entering into the dynamic equation for the polarization. Keeping again only the contributions which are linear in p_k, we find that the resulting scattering rate is of the general form

$$\frac{\partial}{\partial t} p_{k,\rm scat} = -A(k) p_k + \sum_{q\,(\neq 0)} B(k,q) p_{k+q} , \tag{21}$$

where A and B are complex functions. Inserting (21) into (11a) shows that the imaginary parts of these terms give rise to additional energy renormalizations in the first term and to potential corrections in the second term on the rhs of Eq. (11a), both of which are proportional to the square of the Coulomb potential. We neglect these higher-order corrections and evaluate only the contributions proportional to the real parts of A and B. This way we find the decay rate of the polarization as

$$\begin{aligned}{\rm Re}A(k) = \Gamma(k) = \sum_{k';q\,(\neq 0)} &\pi V^2(q)\{\delta(\tilde{\varepsilon}_e(k'-q)+\tilde{\varepsilon}_e(k+q)-\tilde{\varepsilon}_e(k')-\tilde{\varepsilon}_e(k)) \\ &\times [n_{e,k'-q} n_{e,k+q}(1-n_{e,k'}) + n_{e,k'}(1-n_{e,k'-q})(1-n_{e,k+q})] \\ &+ \delta(\tilde{\varepsilon}_e(k'-q)+\tilde{\varepsilon}_h(k-q)-\tilde{\varepsilon}_e(k')-\tilde{\varepsilon}_h(k)) \\ &\times [n_{e,k'-q} n_{h,k-q}(1-n_{e,k'}) + n_{e,k'}(1-n_{h,k-q})(1-n_{e,k'-q})]\} \\ &+ \text{same terms with } e \leftrightarrow h .\end{aligned} \tag{22}$$

The coefficient ${\rm Re}B(k,q)$ describes the rate of polarization transfer between the states k and q due to the carrier-carrier collisions. Our result for B is

$$\begin{aligned}{\rm Re}B(k,q) = \Lambda(k,q) = \pi V^2(q) \sum_{k'} &\{\delta(\tilde{\varepsilon}_e(k'+q)+\tilde{\varepsilon}_e(k)-\tilde{\varepsilon}_e(k')-\tilde{\varepsilon}_e(k+q)) \\ &\times [n_{e,k'+q} n_{e,k}(1-n_{e,k'}) + n_{e,k'}(1-n_{e,k'+q})(1-n_{e,k})] \\ &+ \delta(\tilde{\varepsilon}_e(k'-q)+\tilde{\varepsilon}_h(k)-\tilde{\varepsilon}_h(k+q)+\tilde{\varepsilon}_e(k')) \\ &\times [n_{e,k'-q} n_{h,k}(1-n_{e,k'}) + n_{e,k'}(1-n_{e,k'-q})(1-n_{h,k})]\} \\ &+ \text{same terms with } e \leftrightarrow h .\end{aligned} \tag{23}$$

Adding the scattering terms (19) and (21) to Eqs. (11a)–(11c) we have a set of equations which describes both the coherent and incoherent processes taking place in laser-excited semiconductors. Our approximations restrict the validity of the equations to situations where it is justified to neglect higher-order terms of the polarization. Our approximation scheme shows that the coherent effects, like frequency shifts, contribute in first order in the Coulomb interaction potential, but the incoherent effects contribute in second order. In both cases, these are the lowest orders where these effects contribute at all. Equations (11a)–(11c) plus the scattering terms (19) and (21) are the *effective Bloch equations.* The coherent parts of the equations have also been derived by Haug *et al.*[11] As limiting cases, the effective Bloch equations reproduce well-known results which will be discussed in the following sections of this paper together with some generalizations.

IV. LOW-EXCITATION COHERENT REGIME

To study the situation of not too strong ultrafast optical excitation, we assume that it is justified to neglect the effects of phase-space filling and band-gap renormalization. Hence, from the Coulomb interaction we only keep the mutual attraction between electrons and holes. These approximations are best justified if the density of generated electron-hole pairs is well below the Mott density. Under this condition the coherent equations (11) reduce to

$$\frac{\partial}{\partial t}p_k^* = i[\varepsilon_e^s(k)+\varepsilon_h^s(k)]p_k^* - i\sum_{q\,(\neq 0)} V(q)p_{k+q}^* - i\mu_k^* E(t)^*(1-n_{e,k}-n_{h,k}) , \tag{24a}$$

$$\frac{\partial}{\partial t}n_{e,k} = i[\mu_k E(t)p_k^* - \mu_k^* E^*(t)p_k] , \tag{24b}$$

and

$$n_{h,k} = n_{e,k} . \tag{24c}$$

As initial conditions we assume

$$n_{h,k} = n_{e,k} = p_k = 0 .$$

For the following analysis, it is advantageous to work in real space. Fourier transformation of Eqs. (24a) and (24b) yields

$$\frac{\partial}{\partial t}\phi(x) = i[H_e(x)+H_h(x)-V(x)]\phi(x) - i\mu^* E^*(t)[\delta(x)-2n(x)] \tag{25a}$$

and

$$\frac{\partial}{\partial t}n(x) = i\mu E(t)\phi(x) - i\mu^* E^*(t)\phi^*(-x) , \tag{25b}$$

where we have defined

$$\phi(x) = \sum_k e^{ikx}p_k^* ,$$
$$n(x) = \sum_k e^{ikx}n_{e,k} = \sum_k e^{ikx}n_{h,k} . \tag{26}$$

Note that under the present conditions the equations yield local charge neutrality, i.e., $n_e(x)=n_h(-x)=n(x)$. In the following we use the notation

$$\int dx\ \psi_\lambda^*(x)\phi(x) = \phi_\lambda ,$$
$$\int dx\ \psi_\lambda^*(x)\phi^*(-x) = \bar{\phi}_\lambda , \tag{27}$$
$$\int dx\ \psi_\lambda^*(x)n(x) = n_\lambda ,$$

where $\psi_\lambda(x)$ are the eigenfunctions of the problem

$$[H_e(x)+H_h(x)-V(x)]\psi_\lambda(x) = \varepsilon_\lambda\psi_\lambda(x) . \tag{28}$$

With the help of Eqs. (27) and (28) we obtain from Eqs. (25)

$$\frac{\partial}{\partial t}\phi_\lambda = i\varepsilon_\lambda\phi_\lambda - i\mu^* E^*(t)[\psi_\lambda^*(0)-2n_\lambda] , \tag{29a}$$

$$\frac{\partial}{\partial t}\bar{\phi}_\lambda = -i\varepsilon_\lambda\bar{\phi}_\lambda + i\mu E(t)[\psi_\lambda^*(0)-2n_\lambda] , \tag{29b}$$

and

$$\frac{\partial}{\partial t}n_\lambda = i\mu E(t)\phi_\lambda - i\mu^* E^*(t)\bar{\phi}_\lambda . \tag{29c}$$

This set of equations is closed for each λ. The source terms in (29a) and (29b) are proportional to $\psi_\lambda^*(0)$, i.e., to the wave function in the origin. From the Coulomb problem of two oppositely charged particles we know that $\psi_\lambda^*(0)\neq 0$ only for s waves. In this case $\psi_\lambda(x)$ is a real function and depends only on $|x|$. Hence, $\bar{\phi}_\lambda=\phi_\lambda^*$ and $n_\lambda^*=n_\lambda$. If we now introduce

$$\phi_\lambda = \psi_\lambda^*(0)\hat{\phi}_\lambda ,$$
$$n_\lambda = \psi_\lambda^*(0)\hat{n}_\lambda , \tag{30}$$

we obtain the simplified equations of motion

$$\frac{\partial}{\partial t}\hat{\phi}_\lambda = i\varepsilon_\lambda\hat{\phi}_\lambda - i\mu^* E^*(t)(1-2\hat{n}_\lambda) ,$$
$$\frac{\partial}{\partial t}\hat{n}_\lambda = i\mu E(t)\hat{\phi}_\lambda - i\mu^* E^*(t)\hat{\phi}_\lambda^* . \tag{31}$$

These equations are identical to the well-known Bloch equations for the off-diagonal, $\hat{\phi}_\lambda$, and diagonal, $\hat{n}_\lambda$, elements of the density matrix of a two-level atom. The total polarization is obtained as

$$P = \mu\sum_k \langle a_k^\dagger b_{-k}^\dagger\rangle + \text{c.c.}$$
$$= \mu\phi(0) + \text{c.c.}$$
$$= \mu\sum_\lambda |\psi_\lambda(0)|^2\hat{\phi}_\lambda + \text{c.c.} , \tag{32}$$

where we have used the completeness of the functions $\psi_\lambda(x)$.

Equations (31) and (32) show that for the case of not too high excitation, when phase-space filling, screening,

and band-gap renormalization can be neglected, the optical response of the semiconductor can be computed as solution of an inhomogeneously broadened two-level system. The inhomogeneous broadening in semiconductors is an intrinsic consequence of the energy dispersion. However, when summing over the energies, the density of states has to be weighted by a factor $|\psi_\lambda(0)|^2$. This factor is known as Coulomb or Sommerfeld enhancement and is responsible for the replacement of the square-root absorption shape of noninteracting particles by the more or less structureless continuum absorption in the case with Coulomb attraction.

In Ref. 15 we have used the two-level Bloch equations to model the individual k states of a semiconductor for our analysis of the coherent transients and optical Stark effect. Even though we do not consider Coulomb attraction explicitly in Ref. 15, we still use a constant effective density of states when summing over the continuum of k states. Equations (31) and (32) provide the microscopic justification of this approach and the explicit results and figures of Ref. 15, as well as the comparison with femtosecond experiments in Ref. 10 may serve as illustrations.

V. HIGH-EXCITATION QUASIEQUILIBRIUM REGIME

In the following we show that Eqs. (11a)–(11c) *plus the scattering terms* can be used to derive a generalization of the Elliott formula[16] for the absorption spectrum of a semiconductor to include screening, phase-space filling, and band-gap shifts. All these effects become important when the system is near the quasithermal equilibrium. Typically this situation is reached in resonant pump-probe experiments on the time scale of a few hundred femtoseconds up to a few picoseconds. On this time scale, the carrier-carrier scattering caused the evolution of the originally nonthermal distributions of electrons and holes within their bands into quasi-Fermi distributions. However, the respective chemical potentials are defined within each band and the carriers are at an electronic temperature which is generally higher than the lattice temperature. The chemical potentials are determined by the numbers of excited electrons and holes and the plasma temperature is mainly determined through the electron-hole excess energy with respect to the band gap. The details of the electron-hole excitation process are considered unimportant, and the optical nonlinearities do not depend on the exciting light field directly, but only on the number and distributions of the generated carriers. The semiconductor state is probed with a spectrally broad low-intensity probe beam.

For the considered quasi-steady-state high-excitation situation it is important to include the effect of plasma screening of the Coulomb interaction between the charged particles. The screening may be described in a self-consistent way by replacing the unscreened potential $V(q)$ by a screened one, $V_s(q)$, which has a reduced interaction strength especially at long distances. Because this replacement must be done in the original Hamiltonian (1) and (2), the single-particle energies are in this case

$$\begin{aligned}\varepsilon_e^{sc}(k)&=\varepsilon_c(k)=\varepsilon_e^s(k)\ ,\\ \varepsilon_h^{sc}(k)&=-\varepsilon_v(k)+\sum_{q\ (\neq 0)} V_s(q)\\ &=\varepsilon_h^s(k)+\sum_{q\ (\neq 0)}[V_s(q)-V(q)]\\ &=\varepsilon_h^s(k)+\delta\varepsilon_{\text{Debye}}\end{aligned} \tag{33}$$

where the last term in $\varepsilon_h^{sc}(k)$, which is independent of the wave vector k, is usually considered as a band-gap shift due to screening (the Debye shift).

To obtain the explicit form of the screening potential is a problem in itself. A reasonable encounter is to apply a self-consistent linear-response theory with Poisson equation or to use Green's-function techniques.[14] If one ignores interband contributions to the screening, the polarization induced by the charge distribution change may be approximated by the Lindhard formula. This formula is determined by the populations of the k states. To describe the dynamics of the screening, one has to consider the spatial rearrangement of electrons and holes and the local population fluctuations. These effects are neglected in our present treatment, but for the discussed situation near quasiequilibrium, it is a reasonable approximation to include screening in the dynamic equations simply by replacing $V\rightarrow V_s$ and $\varepsilon^s\rightarrow\varepsilon^{sc}$.

In order to compute the semiconductor susceptibility for a given distribution of electron-hole pairs, we solve the polarization equation in first order in the external probe field. For the sake of compactness of notation we again work in position representation. The equation of motion for the polarization is given by

$$\begin{aligned}\frac{i\partial}{\partial t}\phi(x)=&-[H_e(x)+H_h(x)]\phi(x)\\ &+\int dr V_s(r)N(r)\phi(x-r)+V_s(x)\phi(x)\\ &-\int dr V_s(r)\phi(r)N(x-r)\\ &-\mu^* E^*(t)[\delta(x)-N(x)]\ ,\end{aligned} \tag{34}$$

where $N(x)=n_e(x)+n_h(-x)$ with $n_i(x)$, $i=e,h$, denoting the quasithermal carrier distribution.

The first two terms on the right-hand side of Eq. (34) are the nonlocal Hartree problem of electron and holes,

$$(H_H^i y_\lambda^i)(x)=E_\lambda^i y_\lambda^i(x),\quad i=e,h \tag{35}$$

where H_H is defined as

$$(H_H^i y_\lambda^i)(x)=H_i(x)y_\lambda^i(x)-\int dr\ V_s(r)n_i(r)y_\lambda^i(x-r)\ . \tag{36}$$

In our approximation this problem is linear in y_λ^i. However, since the energy arguments in the quasithermal distributions in the Hamiltonian (36) are given by the eigenenergies of (35), the eigenvalue problem is already nonlinear.

It has been shown in Refs. 1–3 that a k-independent (rigid)band-gap shift is a good approximation when dealing with the nonlinear optical spectra of laser-excited

semiconductors. This approximation is equivalent to making the Hartree Hamiltonians local, i.e.,

$$\int dr\, V_s(r)N(r)\phi(x-r)\cong\int dr\, V_s(r)N(r)\phi(x)\ . \tag{37}$$

To obtain a formal solution of Eq. (36) in steady state, it turns out to be very helpful using a coordinate independent representation. We write

$$i\frac{\partial}{\partial t}\,|\,\phi\rangle=-H_H\,|\,\phi\rangle+SV_s\,|\,\phi\rangle-\mu^*E^*(t)\,|\,f\rangle\ , \tag{38}$$

where the operator S is defined as

$$\langle x\,|\,S\,|\,\xi\rangle=\xi(x)-\int dr\,\xi(r)N(x-r)\ . \tag{39}$$

The Hilbert-space vector $|\,f\rangle$ in Eq. (38) in position representation is given by

$$\langle x\,|\,f\rangle=f(x)=\delta(x)-N(x)\ .$$

The solution of Eq. (34) is complicated by the fact, that even though both operators S and V_s are Hermitian, their product is not. To be able to continue, we make the following assumptions: (i) the operator H_H-SV_s has complete sets of both right-hand and left-hand eigenvectors: (ii) all eigenvalues are real. We have not proved these assumptions but there are cases where a proof can be given. For example, the problem can easily be mapped to a Hermitian problem if no gain occurs in the parameters region of interest, i.e., if the all Fourier components of $f(x)$ are positive. A second, less trivial case is when the system has no bound states.[17,18]

We now assume that we know the solutions to the eigenvalue problems

$$(H_H-SV_s)\,|\,\xi_\lambda\rangle=\varepsilon_\lambda\,|\,\xi_\lambda\rangle \tag{40}$$

and

$$\langle\eta_\lambda\,|\,(H_H-SV_s)=\varepsilon_\lambda\langle\eta_\lambda\,|\ . \tag{41}$$

The solutions are normalized such that

$$\langle\eta_\lambda\,|\,\xi_\mu\rangle=\delta_{\lambda\mu}\ . \tag{42}$$

Since the operator S commutes with H_H, we can rewrite Eq. (41) in the form

$$(H_H-SV_s)S\,|\,\eta_\lambda\rangle=\varepsilon_\lambda S\,|\,\eta_\lambda\rangle\ . \tag{43}$$

A comparison of Eqs. (40) and (43) shows that it is possible to choose the left-hand and right-hand eigenvectors such that

$$S\,|\,\eta_\lambda\rangle\propto|\,\xi_\lambda\rangle\ . \tag{44}$$

Using the completeness of the right-hand eigenvectors, we can write

$$|\,\phi\rangle=\sum_\lambda a_\lambda\,|\,\xi_\lambda\rangle$$

and

$$a_\lambda=\langle\eta_\lambda\,|\,\phi\rangle\ . \tag{45}$$

Inserting (45) into (37) yields

$$i\frac{\partial}{\partial t}a_\lambda=-\varepsilon_\lambda a_\lambda-\mu^*E^*(t)\langle\eta_\lambda\,|\,f\rangle\ . \tag{46}$$

Because the field has the time dependence $E^*(t)=E^*\exp(i\omega t)$, the steady-state solution of Eq. (46) is

$$\phi(x)=-\mu^*E^*(t)\sum_\lambda\frac{\langle\eta_\lambda\,|\,f\rangle\xi_\lambda(x)}{i\gamma+\varepsilon_\lambda-\omega}\ . \tag{47}$$

The left-hand and right-hand eigenvectors can be normalized independently allowing us to normalize η such that $|\langle\eta_\lambda\,|\,S\,|\,\eta_\lambda\rangle|=1$ and

$$\langle\eta_\lambda\,|\,S\,|\,\eta_\mu\rangle=\mathrm{sgn}\langle\eta_\lambda\,|\,S\,|\,\eta_\lambda\rangle\delta_{\lambda\mu}\ . \tag{48}$$

The sign function is necessary because the operator S is not positive definite. Physically, the sign describes gain (minus) or absorption (plus) at the corresponding states. As in Eq. (32), we obtain the polarization as

$$\begin{aligned}P&=\mu\phi(x=0)+\text{c.c.}\\&=-|\,\mu\,|^2E^*(t)\sum_\lambda\mathrm{sgn}\langle\eta_\lambda\,|\,S\,|\,\eta_\lambda\rangle\ |\langle x=0\,|\,S\,|\,\eta_\lambda\rangle|^2\\&\quad\times\frac{1}{i\gamma+\varepsilon_\lambda-\omega}+\text{c.c.}\ ,\end{aligned} \tag{49}$$

and the absorption coefficient as

$$\begin{aligned}\alpha(\omega)=C\sum_\lambda&\mathrm{sgn}(\langle\eta_\lambda\,|\,S\,|\,\eta_\lambda\rangle)\\&\times|\langle x=0\,|\,S\,|\,\eta_\lambda\rangle|^2\delta(\varepsilon_\lambda-\omega)\ ,\end{aligned} \tag{50}$$

where C is a constant. This result can be considered a generalization of the Elliott formula for highly excited semiconductors. It is valid independent of the dimensionality of the system. Hence, it can be applied not only for bulk semiconductors but also for quantum-well structures and for other systems with reduced dimensions.

Equation (50) has several well-known limiting cases. For the unexcited situation when the electron and hole populations are zero, S is the unit operator and the eigenfunctions η_λ are the eigenfunctions of the pure Coulomb problem (Wannier equation). Under this condition, Eq. (50) reduces to the original Elliott formula.[16] On the other hand, when we ignore the Coulomb potential V the problem is diagonalized in a plane-wave basis, and carefully making the algebra of the normalization, one can show that

$$\alpha(\omega)=C\sum_k[1-n_e(k)-n_h-k]\delta(\varepsilon_e(k)+\varepsilon_h-k-\omega)\ , \tag{51}$$

which is the free-particle result.

The absorption formula used by Banyai and Koch[13] is obtained by ignoring the phase-space-filling corrections in the eigenvalue equations (40) and (41). Formally, this approximation is equivalent to replacing S by the unit operator without changing the source term f. In this case Eq. (50) cannot be used because it was derived using Eq. (51) and the explicit relation between $f(x)$ and the operator S. To obtain the absorption formula of Banyai

and Koch, we start from Eq. (47) and replace S by the unit operator. The population factors are manipulated as discussed in Ref. 13. We assume that the energies in the arguments of the δ function are close to the free-particle energies. The resulting absorption coefficient is

$$\alpha(\omega)=C\tanh\left[(\omega-\mu_e-\mu_h)\frac{\beta}{2}\right]\times\sum_{\lambda}|\eta_\lambda(x=0)|^2\delta(\varepsilon_\lambda-\omega)\ . \tag{52}$$

This result has been used successfully to analyze the nonlinear absorption spectra of many different laser-excited semiconductor materials.[19]

VI. SUMMARY AND CONCLUSIONS

Within the Hartree-Fock approximation, we have derived a set of equations for the electron-hole population and for the polarization of a semiconductor using a Hamiltonian formalism. These equations may be regarded as generalized Bloch equations. They include the important many-body Coulomb effects, band-gap renormalization, and phase-space filling. We show how the derived equations can be used to study semiconductors both under ultrafast optical excitation as well as close to quasithermal equilibrium.

Since our theory is independent of the dimensionality of the system of electronic excitations, our results are applicable not only for bulk semiconductors but also for quantum wells, quantum wires, and the like. We are presently investigating whether it is possible to obtain a simplified description of optical nonlinearities in systems with reduced space dimensions in a similar spirit as the plasma theory of Ref. 13. This task, however, is considerably more complicated than in bulk systems, since in lower-dimensional structures it is not justified to neglect the phase-space-filling terms in the generalized Wannier equation, and one has to deal with the nonlocal potential term.

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APPENDIX

Projector techniques are often used when it becomes necessary to exactly separate two subspaces. In the following we summarize the steps used in the main part of this paper. Generally, a projector P and its conjugate projector Q have to fulfill the equations

$$P^2=P,\quad Q^2=Q,\quad PQ=QP=0,\quad P+Q=\hat{1}\ , \tag{A1}$$

where $\hat{1}$ is the unit operator. The actual form of P and Q is determined by the specific physical problem under consideration. Generally, the projector P should be chosen such that it isolates the subspace which contains the essential elements of the discussed problem.

In time-dependent problems it is possible to use time-dependent projectors. An extensive study of the projector methods both in classical and quantum mechanics is given by Grabert.[12] We basically follow his approach. The key assumption is that the dynamics of the electron-hole system can be described with expectation values of a restricted set of operators denoted by $\{\hat{\alpha}_i\}$. We choose these operators to be

$$\hat{n}_{e,k}=a_k^\dagger a_k,\quad \hat{n}_{h,k}=b_{-k}^\dagger b_{-k}\ ,\qquad \hat{p}_k^\dagger=a_k^\dagger b_{-k}^\dagger,\quad \hat{p}_k=b_{-k}a_k\ . \tag{A2}$$

The so-called *relevant density matrix* σ is that part of the total density matrix ρ which contains the information of the expectation values of the operators (A2). This relevant density matrix is parametrically time dependent through the expectation values of the operators. We assume that, if chosen appropriately, the relevant density matrix contains the essential part of the dynamics of the problem under consideration. Hence, we request the following properties:

$$\mathrm{tr}(\sigma)=1\ , \tag{A3}$$

$$\mathrm{tr}(\hat{\alpha}_i\sigma)=\mathrm{tr}(\hat{\alpha}_i\rho)\equiv\alpha_i\ . \tag{A4}$$

To uniquely determine σ we additionally demand that σ is separable in k space and that

$$\mathrm{tr}[\hat{n}_e(k)\hat{n}_h(k)\sigma]=n_e(k)n_h(k)+|p(k)|^2\ . \tag{A5}$$

This gives the relevant density matrix σ the form

$$\sigma=\prod_k\sigma_k \tag{A6}$$

with

$$\begin{aligned}\sigma_k=&(1-n_{e,k}-n_{h,k}+n_{e,k}n_{h,k}+|p_k|^2)\hat{1}_k-(1-2n_{e,k}-n_{h,k}+2n_{e,k}n_{h,k}+2|p_k|^2)\hat{n}_{e,k}\\&-(1-2n_{h,k}-n_{e,k}+2n_{e,k}n_{h,k}+2|p_k|^2)\hat{n}_{h,k}+p_k\hat{p}_k^\dagger+p_k^*\hat{p}_k\\&+(1-2n_{e,k}-2n_{h,k}+4n_{e,k}n_{h,k}+4|p_k|^2)\hat{n}_{e,k}\hat{n}_{h,k}\ .\end{aligned} \tag{A7}$$

We arrived at the explicit result (A6) by noting that all operators constructed from an even number of the electron and hole creation and destruction operators

$$a_k^\dagger,\ a_k,\ b_{-k}^\dagger,\ b_{-k}$$

can be reduced to a linear combination of the operators

$$\hat{1}_k,\ \hat{n}_{e,k},\ \hat{n}_{h,k},\ \hat{p}_k^\dagger,\ \hat{p}_k\ .$$

Hence, σ is given as a linear combination of the six operators with the coefficients determined from (A3)–(A5), leading to the result (A7). Chosen this way, the relevant density matrix has the property that it factorizes all many-operator expectation values in terms of the expectation values α_i.

We now need a projector with the following properties:

$$P(\rho)=\sigma\ , \tag{A8}$$

$$P\left[\frac{\partial\rho}{\partial t}\right]=\frac{\partial}{\partial t}P(\rho)\ . \tag{A9}$$

The second property is important because we want that both ρ and its equation of motion separate into two parts. Following Grabert[12] we see that the projector needed is given by the equation

$$P(\mu)=\sigma\ \mathrm{tr}(\mu)+\sum_i\frac{\partial\sigma}{\partial\alpha_i}[\mathrm{tr}(\hat{\alpha}_i\mu)-\alpha_1\mathrm{tr}(\sigma)]\ , \tag{A10}$$

where μ is an arbitrary element in the space of density matrices, With the help of this projector we obtain the equations of motion

$$i\frac{\partial}{\partial t}\sigma=P([H,\sigma])+P([H,\delta\rho])\ , \tag{A11}$$

$$i\frac{\partial}{\partial t}\delta\rho=Q([H,\delta\rho])+Q([H,\sigma])\ , \tag{A12}$$

where $\delta\rho=Q(\rho)$. Equations (A11) and (A12) yield the equations of motion for α_i's in the form

$$i\frac{\partial}{\partial t}\alpha_i=\mathrm{tr}([\hat{\alpha}_i,H]\sigma)+\mathrm{tr}([\hat{\alpha}_i,H]\delta\rho)\ . \tag{A13}$$

The first term is the factorized, "coherent" contribution and the second term is the scattering part. The division in Eq. (A5) is exact and can be used as a basis of approximations for $\delta\rho$ and hence also to Eq. (A13).

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