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Interaction of ultrashort light pulses with semiconductors: Effective Bloch equations with relaxation and memory effects

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Within the framework of the Keldysh formalism, a simple prescription for the derivation of quantum kinetic equations is given, which makes use of a real-time, multiband generalization of the Kadanoff-Baym ansatz. Such equations are given explicitly for the case of a two-band semiconductor interacting with classic optical pulses of very short duration, while the carriers can also interact with phonons and/or with one another via the screened or unscreened Coulomb potential. They take on the form of optical Bloch equations for interband polarization and intraband carrier distributions, with relaxation terms containing memory effects. In the initial coherent regime the influence of scattering events involving nondiagonal components of the two-band density matrix is shown to be of the same order as that of the more traditional diagonal scattering. Memory effects in collision integrals, which account for the time-energy uncertainty principle in scattering events, also change drastically the energy dependence of the scattering rates in this regime. Therefore, these two effects must be taken into account to achieve a correct description of femtosecond thermalization of optically excited semiconductor plasmas.

I. INTRODUCTION

With the advent of femtosecond lasers, the ultrafast spectroscopy of semiconductors has experienced an explosive growth in recent years. The availability of light pulses with duration shorter than or comparable to the longitudinal and transverse relaxation times of the medium has made it possible to observe in semiconductors coherent optical effects, such as the optical Stark effect (OSE), and, moreover, to obtain unique information on carrier relaxation processes.

Although the coherent optical phenomena as such have been extensively studied for atomic systems, where due to longer relaxation times the coherent limit (pulse duration much shorter than relaxation times) is easily achieved experimentally, experimental observations of the OSE in semiconductors[1,2] have attracted much attention as prospective ultrafast nonlinear-optical devices.[3] Later experiments,[4–6] however, made it clear that the excitonic OSE has some important peculiarities compared with the OSE in pure two-level systems, and a number of theoretical approaches[7–17] have been presented to explain these features.

Although the issue of how best to describe the excitonic OSE seems somewhat controversial (Refs. 8–11, Refs. 12–16, and Ref. 17 give three different physical pictures), a common feature of all these approaches is that they treat the OSE in the steady-state regime. This is apparently justified by the fact that, in principle, the OSE is a strong-field, and not necessarily a short-pulse effect, but in practice the extremely rapid relaxation in semiconductors compels the experimentalists to use such short pulses that the time-energy uncertainty principle starts to come into effect (which is definitely not the case with atomic spectroscopy).

Experimentally this shows up in the appearance of the oscillatory structures in probe transmission spectra, which are superimposed on the Stark shift.[18,19] A convenient basis for a theoretical description of such transient effects is the set of effective Bloch equations,[20] which, in contrast to Refs. 7–17, do not rely on the concept of light-renormalized quasiparticles[21] and therefore work in nonstationary situations as well. Although these equations have been shown to give good qualitative understanding of pump-and-probe experiments,[22–25] they use a very simplistic description of the relaxation processes, and to achieve quantitative agreement with the experiment one has to introduce different relaxation constants for the pump and the probe,[26] which cannot be understood in terms of the Markovian relaxation paradigm. This explains recent efforts to treat the non-Markovian relaxation of polarization,[27–30] which, however, are far from being comprehensive. One can say, therefore, that at present the "coherent" aspects of femtosecond optical processes in semiconductors have become reasonably clear (although there are still questions to be answered),[31–33] while there is a need for more systematic approaches to the description of relaxation processes.

The study of the relaxation processes has until now been focused on the so-called "photoexcited hot carriers" and has an interesting 20-years-long history.[34] When an interband light pulse creates electrons and holes in the bands, they are initially very far from equilibrium. The evolution of their energy distribution towards equilibrium is known to consist of several stages;[35] femtosecond optical techniques can provide invaluable information on the first and fastest of them, that of thermalization of carriers, during which the initial nonequilibrium distributions transform into Maxwell-Boltzmann ones. Recent experiments[36,37] show that the thermalization of photoexcited carriers is very fast indeed and takes just 100–10 fs. The theoretical description of this process is based on the

Boltzmann equation, which is solved either straightforwardly,[38–40] or using elaborate Monte Carlo techniques.[37,41] Although these calculations claim very good agreement with experiments, they ignore the peculiarities of femtosecond excitation conditions and therefore leave out at least two important effects.

For one thing, the distribution functions (DF) of the electrons and the holes, which enter the Boltzmann equation, are just the diagonal components of the density matrix (DM) of these particles. The DM also has nondiagonal (over the band indices) components, which describe the phase coherence between the electron and the hole created by the same photon; a statistical average of these nondiagonal elements gives the optical polarization.[9,20] These components can and should be neglected in the description of relaxation processes, if the relevant time scales are much longer than the polarization decay time. The experimental dephasing times,[42–45] however, prove to be of the same order (10 ps to 10 fs) as the thermalization times, so that in realistic situations there is no reason why the nondiagonal components of the DM should be smaller than the diagonal ones. This should lead to an additional scattering of carriers on the optical polarization.

Thus, the correct theoretical description of the carrier dynamics immediately after photoexcitation should include not only the equations for the diagonal density-matrix components (i.e., Boltzmann equations with additional terms describing such scattering on the polarization), but also the equations for the polarization dynamics. In essence, such a set of equations is nothing but the optical Bloch equations,[20] which have to be complemented with corresponding relaxation terms. The carrier dynamics cannot be solved independently of the dephasing processes, which, in spite of the substantial number of experiments,[42–45] at present are not very well understood theoretically.[20] Although recently quite a few approaches to the nonequilibrium optical properties[46–48] and the kinetics of light-renormalized quasiparticles[49–53] have been developed, none of them treats this intermingling of carrier kinetics with the dephasing explicitly.

There is also another, and rather more serious, problem that needs to be taken care of in this situation. The time-energy uncertainty principle states that an electron created by a light pulse with duration τ cannot have its energy defined better than with an accuracy of order h/τ; for τ in the subpicosecond range this uncertainty can amount to some tens of meV. When such electrons scatter, their energy cannot be conserved with the accuracy exceeding this quantum limit, and therefore the use of the semiclassic Boltzmann equations with energy-conserving δ functions is not quite appropriate here. Thus, one has to replace these equations with their quantum generalizations, which contain integrals over the system's past instead of δ functions in the scattering terms. At present such quantum kinetic equations are abundant in the literature,[54–62] but they are mostly formulated for the case of strong dc electric fields, and apparently cannot be used as they are to treat the specifics of ultrafast optical excitation.

This brief review of the state of the art shows the rationale for the present work. In this paper we present a first-principles derivation of the set of equations for the carrier DF's and for the interband polarization (the effective Bloch equations), which includes the influence of the polarization on the particle scattering, as well as the non-Markovian nature of the relaxation processes.

The procedure we use to do that can be seen as yet another way to obtain the equations for the one-time density matrix using the two-time nonequilibrium Green's functions. It is described in Sec. II, where we also discuss the underlying approximations. This procedure is used in Sec. III to obtain the explicit form of the Bloch equations for different interaction mechanisms; in this section the emphasis is mainly on the origins of the polarization scattering and therefore the equations of this section are Markovian, which allows comparison with the results of other authors. In Sec. IV these equations are extended to the non-Markovian case, and the impact of the memory effects on the carrier dynamics is discussed qualitatively. Section V contains concluding remarks.

II. DENSITY-MATRIX EQUATIONS WITH NON-MARKOVIAN RELAXATION

The aim of this section is to present a procedure for the derivation of quantum kinetic equations with the use of Keldysh Green's functions.[63] To make the paper more self-contained, we had to also include some standard steps, which can be found elsewhere[56,63] but are necessary at least to fix the notations.

The physical situation we are going to deal with in the present paper is that of a two-band semiconductor with conduction and valence (c and v) bands; Heisenberg creation operators for an electron in the band α with a definite wave vector $\mathbf{k}$ shall be denoted $a^{\dagger}_{\alpha\mathbf{k}}$. The electrons can interact with an exciting laser field, with each other, with phonons, etc.; the interaction Hamiltonians will be made explicit below.

In the *spatially homogeneous case,* to which we restrict ourselves throughout the paper, this situation can be comprehensively described by means of the density matrix:[9,11,20]

$$N^{\alpha\beta}_{\mathbf{k}}(t) \equiv \begin{bmatrix} \langle a^{\dagger}_{c\mathbf{k}}(t)a_{c\mathbf{k}}(t)\rangle & \langle a^{\dagger}_{v\mathbf{k}}(t)a_{c\mathbf{k}}(t)\rangle \\ \langle a^{\dagger}_{c\mathbf{k}}(t)a_{v\mathbf{k}}(t)\rangle & \langle a^{\dagger}_{v\mathbf{k}}(t)a_{v\mathbf{k}}(t)\rangle \end{bmatrix}_{\alpha\beta} \equiv \begin{bmatrix} n_{c\mathbf{k}}(t) & p_{\mathbf{k}}(t) \\ p^{*}_{\mathbf{k}}(t) & n_{v\mathbf{k}}(t) \end{bmatrix}_{\alpha\beta} \tag{1}$$

(where $\langle \cdots \rangle$ denotes the statistical average), whose diagonal components give the distribution functions of carriers within the bands, while the nondiagonal ones describe the quantum-mechanical coherence between the bands; $\sum_{\mathbf{k}} p_{\mathbf{k}}$ is known to be proportional to the optical polarization[47] [see Eq. (42)].

In this section we will obtain the equations of motion for $N^{\alpha\beta}$ from the corresponding equations for the nonequilibrium Green's functions (GF) $G^{pq}_{\alpha\beta}(tt')$, which are immediate two-time generalizations of (1):

$$iG^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')\equiv i\begin{bmatrix} G^{++} & G^{+-} \\ G^{-+} & G^{--} \end{bmatrix}_{pq} \equiv \begin{bmatrix} \langle \mathcal{T} a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle & -\langle a^{\dagger}_{\beta\mathbf{k}}(t') a_{\alpha\mathbf{k}}(t)\rangle \\ \langle a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle & \langle \tilde{\mathcal{T}} a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle \end{bmatrix} \tag{2}$$

(there are almost as many conventions about the superscripts $\{p,q\}=\{+,-\}$ as there are authors; here we will follow the notations of Ref. 47). The quantity (2) is a $(2\times2\times2\times2)$ matrix, whose components depend on the two times t,t'; the symbols $\mathcal{T}$ and $\tilde{\mathcal{T}}$ denote time ordering and anti-time-ordering, respectively:

$$iG^{++}_{\alpha\beta_{\mathbf{k}}}(tt')\equiv\langle \mathcal{T} a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle = \begin{cases} \langle a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle, & t>t' \\ -\langle a^{\dagger}_{\beta\mathbf{k}}(t') a_{\alpha\mathbf{k}}(t)\rangle, & t<t', \end{cases} \tag{3}$$

$$iG^{--}_{\alpha\beta_{\mathbf{k}}}(tt')\equiv\langle \tilde{\mathcal{T}} a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle \equiv \begin{cases} -\langle a^{\dagger}_{\beta\mathbf{k}}(t') a_{\alpha\mathbf{k}}(t)\rangle, & t>t' \\ \langle a_{\alpha\mathbf{k}}(t) a^{\dagger}_{\beta\mathbf{k}}(t')\rangle, & t<t' . \end{cases} \tag{4}$$

Comparing (2) with (3) and (4), one sees that all the pq components of G^{pq} are made of just two correlation functions:

$$G^{++}=\begin{cases} G^{-+}, & t>t' \\ G^{+-}, & t<t' , \end{cases} \qquad G^{--}(tt')=\begin{cases} G^{+-}, & t>t' \\ G^{-+}, & t<t' \end{cases} \tag{5}$$

so that the pq components are linearly dependent:

$$G^{++}+G^{--}=G^{+-}+G^{-+} . \tag{6}$$

From these components one can also construct the retarded and advanced Green's functions:

$$G^{R}\equiv G^{++}-G^{+-}=0 \text{ for } t<t' , \qquad G^{A}\equiv G^{++}-G^{-+}=0 \text{ for } t>t' . \tag{7}$$

To finish this account of Green's-functions formal properties, let us note the time-reversal symmetry:

$$G^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')=-[G^{\bar{q}\bar{p}}_{\beta\alpha_{\mathbf{k}}}(t't)]^{*} , \tag{8}$$

where $\{p,q\}=\{+,-\}$, and $\mp=-$, $=\!=+$; in particular, it means that $G^{R}(tt')=G^{A}(t't)^{*}$. The properties (5)–(8) are valid in the presence of any interactions and will be used extensively later on.

The Heisenberg operators a obey the equations of motion

$$\frac{\partial a_{\alpha\mathbf{k}}}{\partial t}=i[\hat{H},a_{\alpha\mathbf{k}}] . \tag{9}$$

In the absence of interactions the Hamiltonian $\hat{H}$ of the semiconductor has the form

$$\hat{H}=\hat{H}_0=\sum_{\alpha\mathbf{k}}\varepsilon_{\alpha\mathbf{k}}a^{\dagger}_{\alpha\mathbf{k}}a_{\alpha\mathbf{k}} , \tag{10}$$

so that, according to (9), without interaction the operators $a,a^{\dagger}$ just oscillate with time:

$$a^{\dagger}_{\alpha\mathbf{k}}(t)=a^{\dagger}_{\alpha\mathbf{k}}(0)e^{i\varepsilon_{\alpha\mathbf{k}}t} , \qquad a_{\alpha\mathbf{k}}(t)=a_{\alpha\mathbf{k}}(0)e^{-i\varepsilon_{\alpha\mathbf{k}}t} , \tag{11}$$

which allows us to find the time dependence of $N^{\alpha\beta}$ [(1)] for the noninteracting system

$$N^{\alpha\beta(0)}_{\mathbf{k}}(t)=N^{\alpha\beta}_{\mathbf{k}}(0)\exp[(\varepsilon_{\beta\mathbf{k}}-\varepsilon_{\alpha\mathbf{k}})t] \tag{12}$$

[its diagonal components do not change with time, while the off-diagonal ones oscillate with the interband frequency $(\varepsilon_{c\mathbf{k}}-\varepsilon_{v\mathbf{k}})$]. The same can be done, of course, for G as well:

$$iG^{pq(0)}_{\alpha\beta_{\mathbf{k}}}(tt')=\begin{bmatrix} \delta_{\alpha\beta}\Theta(t-t')-N^{\alpha\beta}_{\mathbf{k}}(0) & -N^{\alpha\beta}_{\mathbf{k}}(0) \\ \delta_{\alpha\beta}-N^{\alpha\beta}_{\mathbf{k}}(0) & \delta_{\alpha\beta}\Theta(t'-t)-N^{\alpha\beta}_{\mathbf{k}}(0) \end{bmatrix}_{pq}\exp[i(\varepsilon_{\beta\mathbf{k}}t'-\varepsilon_{\alpha\mathbf{k}}t)] . \tag{13}$$

From (12) and (13) it is clear that N and G are closely connected; in general, any pq component of G at coincident times can be used to obtain N:

$$i\lim_{t\to t'+0}G^{pq}_{\alpha\beta}(tt')=\begin{bmatrix} \delta_{\alpha\beta}-N^{\alpha\beta}(t) & -N^{\alpha\beta}(t) \\ \delta_{\alpha\beta}-N^{\alpha\beta}(t) & -N^{\alpha\beta}(t) \end{bmatrix}_{pq} \tag{14}$$

[which follows directly from definitions (1) and (2)].

Although G is a much more complex object compared to N, sometimes it proves convenient[62] to use the Green's functions in order to obtain quantum kinetic equations for N. One of the reasons for this is that one can write down a set of formally exact equations for G, which include interaction to all orders, namely, the Dyson equations:[47,63]

$$G(tt')=G^{(0)}(tt')+\int_{-\infty}^{+\infty}d1\,d2\,G^{(0)}(t1)\Sigma(12)G(2t') \tag{15}$$

(here and later on 1,2 stand for t_1,t_2, etc.; G and the self-energies Σ are understood as matrices $G\equiv G^{pq}_{\alpha\beta}$ and are multiplied by the corresponding rules). The self-energy $\Sigma^{pq}_{\alpha\beta}(tt')$, which enters Eq. (15), has the following formal properties:[63]

$$\Sigma^{++}(tt')=-\begin{cases}\Sigma^{-+}, & t>t'\\ \Sigma^{+-}, & t<t',\end{cases} \tag{16}$$

$$\Sigma^{++}+\Sigma^{--}=-(\Sigma^{+-}+\Sigma^{-+}) , \tag{17}$$

$$\Sigma^{R}\equiv\Sigma^{++}+\Sigma^{+-},\quad \Sigma^{pq}_{\alpha\beta}(tt')=[\Sigma^{\bar{q}\bar{p}}_{\beta\alpha}(tt')]^{*} , \tag{18}$$

which practically coincide with the corresponding GF properties (5)–(8). Properties (5)–(8) and (16)–(18) enable one to write down the "conjugate" Dyson equation:

$$G(tt')=G^{(0)}(tt')+\int_{-\infty}^{+\infty}d1\,d2\,G(t1)\Sigma(12)G^{(0)}(2t') . \tag{15'}$$

The idea behind the use of the Dyson's equations (15) as the starting point is that well-defined diagrammatic rules[63] for calculating Σ exist. These equations are integral equations, which does not matter much in steady-state (e.g., equilibrium) conditions when they can be straightforwardly solved by a Fourier transform.[47,63] However, for nonstationary problems it is advantageous to make them differential equations.

Taking the time derivatives of (13), one can see that the "free" GF $G^{(0)}$ obeys the following equations of motion:

$$\frac{\partial G^{(0)}}{\partial t}=-i\hat{\sigma}_z\delta(t-t')-i\hat{\varepsilon}G^{(0)} , \tag{19a}$$

$$\frac{\partial G^{(0)}}{\partial t'}=+i\hat{\sigma}_z\delta(t-t')+i\hat{\varepsilon}G^{(0)} , \tag{19b}$$

where we have introduced the energy matrix

$$\hat{\varepsilon}_{\alpha\beta}=\begin{bmatrix}\varepsilon_{c\mathbf{k}} & 0\\ 0 & \varepsilon_{v\mathbf{k}}\end{bmatrix}_{\alpha\beta}\delta_{pq} \tag{20}$$

and the Pauli matrix

$$\hat{\sigma}_z=\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}_{pq}\delta_{\alpha\beta} , \tag{21}$$

which of course has nothing to do with the spin here. Now, differentiating (15) and (15') with respect to t and t', we get

$$\frac{\partial G}{\partial t}=-i\hat{\sigma}_z\delta(t-t')-i\hat{\varepsilon}G^{(0)}+\int d1\,d2[-i\hat{\sigma}_z\delta(t1)-i\hat{\varepsilon}G^{(0)}]\Sigma G=-i\hat{\sigma}_z\delta(t-t')-i\hat{\varepsilon}G-i\int d2\,\hat{\sigma}_z\Sigma(t2)G(2t') , \tag{22a}$$

$$\frac{\partial G}{\partial t'}=+i\hat{\sigma}_z\delta(t-t')+iG\hat{\varepsilon}+i\int d1\,G(t1)\Sigma(1t')\hat{\sigma}_z . \tag{22b}$$

Equations (22) are merely the differential form of the Dyson equations and are therefore also formally exact. To obtain kinetic equations out of (22), one has to introduce new time variables: the "absolute" time

$$T\equiv\frac{t+t'}{2} \tag{23}$$

and the relative time

$$\tau=t-t' . \tag{24}$$

With respect to these variables, Eq. (22) reads

$$\frac{\partial G}{\partial T}=i(G\hat{\varepsilon}-\hat{\varepsilon}G)-i\int_{-\infty}^{+\infty}d1[\hat{\sigma}_z\Sigma(t1)G(1t')-G(t1)\Sigma(1t')\hat{\sigma}_z] , \tag{25a}$$

$$\frac{\partial G}{\partial\tau}=-i\hat{\sigma}_z\delta(\tau)-\frac{i}{2}(G\hat{\varepsilon}+\hat{\varepsilon}G)-\frac{i}{2}\int d1(\hat{\sigma}_z\Sigma G+G\Sigma\hat{\sigma}_z). \tag{25b}$$

One can loosely say that now (25a) contains information on the kinetics of the system, while (25b) describes its spectral properties. The existence of relations (5)–(8) between the pq components of the GF makes this set of equations redundant; in particular, four equations (25b) are equivalent to a single equation for the retarded GF:

$$i\frac{\partial G^{R}}{\partial\tau}=\delta(\tau)+\tfrac{1}{2}(G^{R}\hat{\varepsilon}+\hat{\varepsilon}G^{R})+\tfrac{1}{2}\int_{-\infty}^{+\infty}d1(\Sigma^{R}G^{R}+G^{R}\Sigma^{R}). \tag{25c}$$

Similarly, any of the four pq components of (25a) can now be used to obtain the equation for the DM in the limit of coincident times [(14)]; e.g., for the $(++)$ component we have

$$\begin{aligned}\frac{\partial N(T)}{\partial T}&=-i\frac{\partial}{\partial T}\lim_{\tau\to+0}G^{++}(T,\tau)\\ &=i(N\hat{\varepsilon}-\hat{\varepsilon}N)-\lim_{\substack{t'\to t+0\\ t\to T}}\int_{-\infty}^{+\infty}d1[(\hat{\sigma}_z\Sigma G)^{++}-(G\Sigma\hat{\sigma}_z)^{++}]\\ &=i[N,\hat{\varepsilon}]+\lim_{\substack{t'\to t+0\\ t\to T}}\int_{-\infty}^{+\infty}d1[G^{++}(t1)\Sigma^{++}(1t')+G^{+-}(t1)\Sigma^{-+}(1t')\\ &\qquad-\Sigma^{++}(t1)G^{++}(1t')-\Sigma^{+-}(t1)G^{-+}(1t')] .\end{aligned} \tag{26}$$

Equation (26) represents the general form of the kinetic equation for the system in question (the "generalized Boltzmann equation")[56] and is still formally exact. Its first term is responsible for the free oscillations of the polarization (12), while the second describes the effects of interaction. It is, however, quite useless in this form, since it is not closed—its right-hand side contains G and Σ, which have to be expressed through the DM.

Moreover, Eq. (26) has two major shortcomings: first, it involves taking the limit, which is simply inconvenient, and second, the evolution of the system at a time T is determined by the integral over the system's future as well as over its past, which is physically unacceptable.

In fact, however, the future of course does not contribute to the integral in (26), since due to the properties (5) of G and (16) of Σ the integrand strictly vanishes when the integration time variable t_1 becomes greater than $\tilde{t} \equiv \max(t,t')+0$, so that we can replace the upper integration limit in (26) by $\tilde{t}$. This leaves us with a set of time variables in this equation: T, t, t', and $\tilde{t}$, of which only T is directly relevant to the problem. The reason that we cannot just set them all equal to T as a result of the transition to the limit in (26) is that the self-energy Σ may have a δ-shaped singularity at coincident times (such singularity is produced by any mean-field contributions to Σ, e.g., by an external classical field); hence, the prescription for taking the limit in (26) consists in processing this singularity first, and setting all these times equal to T only after that.

Thus, let us extract from Σ the "instantaneous" part

$$\Sigma^{pq}_{\alpha\beta}(tt') \equiv \Delta^{pq}_{\alpha\beta}(t)\delta(t-t')\delta_{pq} + \tilde{\Sigma}^{pq}_{\alpha\beta}(tt') , \tag{27}$$

where $\tilde{\Sigma}$ is the regular part of the self-energy. Substitution of (27) into (26) yields for the contribution of the singular term

$$\left[\frac{\partial N}{\partial t}\right]_{\text{sing}} = \lim_{\substack{t'\to t+0 \\ t\to T}} [G^{++}(tt')\Delta^{++}(t') - \Delta^{++}(t)G^{++}(tt')] . \tag{28}$$

Taking the limit in accordance with (14) and considering that $\delta_{\alpha\beta}$ commutes with Δ, we get for the singular term contribution

$$\left[\frac{\partial N}{\partial t}\right]_{\text{sing}} = iN(T)\Delta^{++}(T) - i\Delta^{++}(T)N(T) \tag{29}$$

(note that N and Δ are matrices with respect to band indices). Introducing the renormalized energy matrix

$$\tilde{\varepsilon}_{\alpha\beta} \equiv \hat{\varepsilon}_{\alpha\beta} + \Delta^{++}_{\alpha\beta} = \begin{bmatrix} \varepsilon_{c\mathbf{k}} + \Delta^{++}_{cc_{\mathbf{k}}} & \Delta^{++}_{cv_{\mathbf{k}}} \\ \Delta^{++}_{vc_{\mathbf{k}}} & \varepsilon_{v\mathbf{k}} + \Delta^{++}_{vv_{\mathbf{k}}} \end{bmatrix}_{\alpha\beta} \tag{30}$$

we can combine the contribution of the instantaneous interactions with the first term of (26):

$$\left[\frac{\partial N^{\alpha\beta}_{\mathbf{k}}(T)}{\partial T}\right]_{\text{coh}} = i[N_{\mathbf{k}},\tilde{\varepsilon}_{\mathbf{k}}] \equiv i\sum_{\gamma}(N^{\alpha\gamma}_{\mathbf{k}}\tilde{\varepsilon}_{\gamma\beta} - \tilde{\varepsilon}_{\alpha\gamma}N^{\gamma\beta}_{\mathbf{k}}) . \tag{31}$$

The remaining interactions are represented by the regular part of the self-energy $\tilde{\Sigma}$; it is this part that causes relaxation, i.e., it gives rise to what should be called the collision term. The absence of singularity makes the limit in (26) trivial, so that we can easily rewrite this part as the generalized collision integral:[56]

$$\left[\frac{\partial N^{\alpha\beta}}{\partial t}\right]_{St} = \int_{-\infty}^{T} d1 \left\{ \sum_{\gamma} [G^{++}_{\alpha\gamma}(T1)\tilde{\Sigma}^{++}_{\gamma\beta}(1T) + G^{+-}_{\alpha\gamma}(T1)\tilde{\Sigma}^{-+}_{\gamma\beta}(1T) - \tilde{\Sigma}^{++}_{\alpha\gamma}(T1)G^{++}_{\gamma\beta}(1T) - \tilde{\Sigma}^{+-}_{\alpha\gamma}(T1)G^{-+}_{\gamma\beta}(1T)] \right\} . \tag{32}$$

Now the transformed equation for the DM reads

$$\left[\frac{\partial N}{\partial T}\right] = \left[\frac{\partial N}{\partial T}\right]_{\text{coh}} + \left[\frac{\partial N}{\partial T}\right]_{St} , \tag{33}$$

where the first term ("coherent part")[20] [(31)] describes the evolution of the carriers distributions ($\alpha=\beta$) and of the polarization ($\alpha\neq\beta$) under the action of instantaneous interactions (the contribution of the present), while the second, integral term gives rise to relaxation that, as suggested by its explicitly non-Markovian form [(32)], depends on the past history of the system (the contribution of the past).

However, in order to use the kinetic equation in the form (33), we need to specify the procedure for evaluation of the relaxation term, that is, to establish some relations between the density matrix and the GF, which enter the right-hand side of (32).

At this stage some assumptions have to be made. The central approximation of the present work consists in relating these two quantities by the following *Ansätze:*

$$iG^{pq}_{\alpha\beta_{\mathbf{k}}}(tt') = \begin{bmatrix} -N^{\alpha\beta}_{\mathbf{k}}(t)+\delta_{\alpha\beta}\Theta(t-t') & -N^{\alpha\beta}_{\mathbf{k}}(t) \\ -N^{\alpha\beta}_{\mathbf{k}}(t)+\delta_{\alpha\beta} & -N^{\alpha\beta}_{\mathbf{k}}(t)+\delta_{\alpha\beta}\Theta(t'-t) \end{bmatrix} \exp[i\varepsilon_{\beta\mathbf{k}}(t'-t)] , \tag{34a}$$

$$iG^{pq}_{\alpha\beta_{\mathbf{k}}}(tt') = \begin{bmatrix} -N^{\alpha\beta}_{\mathbf{k}}(t')+\delta_{\alpha\beta}\Theta(t-t') & -N^{\alpha\beta}_{\mathbf{k}}(t') \\ -N^{\alpha\beta}_{\mathbf{k}}(t')+\delta_{\alpha\beta} & -N^{\alpha\beta}_{\mathbf{k}}(t')+\delta_{\alpha\beta}\Theta(t'-t) \end{bmatrix} \exp[i\varepsilon_{\alpha\mathbf{k}}(t'-t)] , \tag{34b}$$

which expresses G in terms of $N(t)$ and $N(t')$, respectively. The physical content of (34) becomes clear from the following facts: (i) $G(tt')$ of (34) satisfies the equations of motion (19) for the free GF; (ii) to ensure that (34a) and (34b) define the same function, the DM on the right-hand side must obey the equation

$$\left[\frac{\partial N}{\partial T}\right]=i[N,\hat{\varepsilon}] , \tag{35}$$

i.e., the "free" part of the kinetic equation (26); (iii) the retarded GF that corresponds to (34),

$$G^R_{\alpha\beta}(tt')=\delta_{\alpha\beta}\Theta(t-t')e^{i\varepsilon_\alpha(t'-t)} , \tag{36}$$

satisfies Eq. (25c) without interaction; (iv) *Ansätze* (34) are consistent with all the symmetries (5)–(8) and the property (14). To put it simply, (34) means that *locally* G is supposed to look exactly like it used to in the absence of interaction.

Ansätze like (34) are inherent to GF treatments of kinetic phenomena. However, Lipavsky, Spička, and Velický[56] have pointed out that care should be taken in choosing the time arguments of N in (34)—these should be either t or t', and not the absolute time $(t+t')/2$, as has been originally suggested by Kadanoff and Baym. This choice does not matter much in a truly semiclassic limit of a slowly varying DM, but it becomes crucial if one wants to go beyond the semiclassic regime.

These particular *Ansätze* (34) can be seen as a multiband extension of the "generalized Kadanoff-Baym *Ansatz*" of Ref. 56 combined with the quasiparticle approximation (36) for G^R. It becomes exact in the limit of vanishing internal interactions and can handle arbitrarily fast variations of external fields. Although one might be led to think that it neglects the influence of the interaction on the energy spectrum, in fact, as we will see below, this approximation corresponds to the usual procedure for the calculation of many-body renormalizations.

However, these *Ansätze* leave out some important effects. These are (i) the collisional broadening,[58,59,62] which to include we should have first solved Eq. (25c) for the retarded GF and then inserted in (34) the resulting $G^R(\tau)$ instead of $e^{i\varepsilon\tau}$ (in this way the generalized Kadanoff-Baym *Ansatz*[56] would be completely recovered); we will not do that in the present paper in order to avoid confusing this effect with what we want to obtain here; (ii) light-induced renormalizations in strong optical fields,[21,49–53] that is, the presence of nondiagonal terms in the renormalized energy matrix $\tilde{\varepsilon}$ (30); it means that (34) can only be valid in relatively weak fields (which may, however, be strong enough to produce other nonlinear effects).

According to Ref. 56, when evaluating the relaxation terms [(32)] one has to choose that of the two expressions (34) which contains the DM as a function of integration variable t_1, i.e., one must replace $G(T1)$ with (34b) and $G(1T)$ with (34a)—this prescription ensures that in the resulting non-Markovian kinetic equation the retardation factors are correct. This will be done explicitly in Sec. IV.

In order to get back to the semiclassic regime, in principle one has to assume that DM components in the general non-Markovian expression are slowly varying and take them out of the integral—this would result in a collision term containing only $N(T)$. *Ansätze* (34), however, offer a shortcut to these Markovian results: inserting (34a) instead of $G(T1)$ and (34b) for $G(1T)$ in (32), we will immediately arrive at relaxation terms containing only $N(T)$, which coincide with the results of the abovementioned general procedure. This prescription will be explored in the following section.

III. EFFECTIVE BLOCH EQUATIONS WITH COHERENT EFFECTS IN RELAXATION

In this section we are going to evaluate the collision terms of the quantum kinetic equations (32) for a number of interaction models. To do that, we need to specify the interaction Hamiltonians first.

A. Interaction Hamiltonians

We will consider the following types of interaction: (i) with an external optical field; (ii) the Coulomb interaction between carriers; (iii) the electron-phonon interaction. Consequently, the full Hamiltonian of the problem reads

$$\hat{H}=\hat{H}_0+\hat{H}_{\rm opt}+\hat{H}_{\rm Coul}+\hat{H}_{\rm ph} , \tag{37}$$

where $\hat{H}_0$ [(10)] describes noninteracting carriers.

The interaction with a classic uniform electric field $E(t)$ in the dipole approximation has the form[20,21]

$$\hat{H}_{\rm opt}=-\sum_{\substack{\alpha\beta\\ \mathbf{k}}}\mu_{\alpha\beta}(\mathbf{k})a^\dagger_{\alpha\mathbf{k}}a_{\beta\mathbf{k}}\cdot\mathbf{E}(t) . \tag{38}$$

In what follows we will neglect intraband optical transitions, i.e., we will assume that the transition matrix elements $\mu_{\alpha\beta}$ have the form

$$\mu_{\alpha\beta}(\mathbf{k})=\begin{bmatrix}0 & \mu_{cv}\\ \mu^*_{cv} & 0\end{bmatrix}_{\alpha\beta} \tag{39}$$

and the $\mathbf{k}$ dependence of the interband matrix elements $\mu_{cv}=\mu^*_{vc}$ will also be neglected, which is justified for direct-gap semiconductors.[64]

Comparing the expression resulting from (38) and (39),

$$\hat{H}_{\rm opt}=-\sum_{\mathbf{k}}[\mu_{cv}a^\dagger_{c\mathbf{k}}a_{v\mathbf{k}}\cdot\mathbf{E}^*(t)+\mu^*_{cv}a^\dagger_{v\mathbf{k}}a_{c\mathbf{k}}\cdot\mathbf{E}(t)] , \tag{40}$$

with the macroscopic form of the interaction energy,

$$H=-\mathbf{P}^*\cdot\mathbf{E}-\mathbf{P}\cdot\mathbf{E}^* , \tag{41}$$

we see that the quantity

$$\mathbf{P}=\sum_{\mathbf{k}}\mu_{cv}(\mathbf{k})\langle a^\dagger_{c\mathbf{k}}a_{v\mathbf{k}}\rangle\equiv\sum_{\mathbf{k}}\mu_{cv}(\mathbf{k})\cdot p_{\mathbf{k}} \tag{42}$$

is nothing but the macroscopic optical polarization, so that the knowledge of the nondiagonal elements of the DM, $p_{\mathbf{k}}\equiv N^{cv}_{\mathbf{k}}$, is all we need to describe the optical properties of the system.[20,47] The form (38) ignores quantum fluctuations of the field and leaves out effects such as radiative recombination, which are supposed to have much

longer time scales. For us it is important that the "mean-field" interaction of (38) is instantaneous.

The Hamiltonian of the Coulomb interaction in the standard (long-wavelength) approximation reads[20,64]

$$\hat{H}_{\text{Coul}}=\frac{1}{2}\sum_{\substack{\mathbf{k}\mathbf{k}'\mathbf{q}\\ \alpha\beta}} V(q)a^{\dagger}_{\alpha\mathbf{k}+\mathbf{q}}a^{\dagger}_{\beta\mathbf{k}'-\mathbf{q}}a_{\beta\mathbf{k}'}a_{\alpha\mathbf{k}}\ , \tag{43}$$

where $V(\mathbf{q})$ is the Fourier transform of the unscreened Coulomb potential:

$$V(\mathbf{q})=\frac{4\pi e^2}{\epsilon\mathbf{q}^2}\ . \tag{44}$$

Here, ϵ is the dielectric constant; see Ref. 64 for the discussion of the proper choice of this quantity.

Finally, the phonons give the following contribution to the Hamiltonian:

$$\hat{H}_{\text{ph}}=\sum_{\mathbf{q}}\omega_{\mathbf{q}}b^{\dagger}_{\mathbf{q}}b_{\mathbf{q}}+\sum_{\mathbf{k}\mathbf{q}\alpha}M_{\alpha\mathbf{q}}(b_{\mathbf{q}}+b^{\dagger}_{-\mathbf{q}})a^{\dagger}_{\alpha\mathbf{k}}a_{\alpha\mathbf{k}-\mathbf{q}}\ , \tag{45}$$

where a single phonon mode with the dispersion $\omega_{\mathbf{q}}$ is taken into consideration; $M_{\alpha\mathbf{q}}$ is its coupling to the carriers of the band α and depends on the type of phonons.

B. The coherent part of the Bloch equations

To evaluate the coherent part (31) of the Bloch equations we have to single out the singular part (27) of the full self-energy. There are only two sources of such singularities in our case, namely the interaction with the external optical field (38),

$$\begin{aligned}&[\Sigma^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')]_{\text{opt}}=[\Delta^{pq}_{\alpha\beta}(t)]_{\text{opt}}\delta(t-t')\ ,\\ &[\Delta^{pq}_{\alpha\beta}(t)]_{\text{opt}}=-\mu_{\alpha\beta}(\mathbf{k})\mathbf{E}(t)\begin{bmatrix}1&0\\0&-1\end{bmatrix}_{pq}\end{aligned} \tag{46}$$

(this is the *exact* self-energy), and the Hartree-Fock part of the Coulomb self-energy, which is given by the diagram in Fig. 1. Analytically this diagram reads

$$\begin{aligned}-i[\Sigma^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')]_{\text{HF}}&=\sum_{\mathbf{q}}iG^{pq}_{\alpha\beta}(tt')iV^{pq}(\mathbf{q})\delta(t-t')\\ &\equiv-i(\Delta^{pq}_{\alpha\beta_{\mathbf{k}}})_{\text{Coul}}\delta(t-t')\ ,\end{aligned} \tag{47}$$

where

$$V^{pq}\equiv\begin{bmatrix}-V(\mathbf{q})&0\\0&V(\mathbf{q})\end{bmatrix} \tag{48}$$

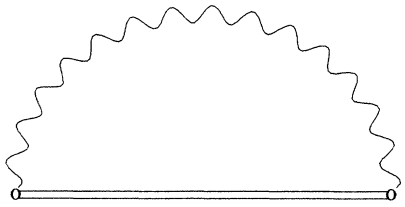

FIG. 1. Hartree-Fock self-energy, Eq. (47). Wavy line, bare Coulomb potential [(48)]; double solid line, full electronic Green's function [(2)].

(the singularity here is due to the fact that the unscreened Coulomb interaction is supposed to be instantaneous). Inserting (48) and (14) into (47), we can express Δ_{Coul} in terms of the DM:

$$\begin{aligned}&[\Delta^{pq}_{\alpha\beta_{\mathbf{k}}}(t)]_{\text{Coul}}\\ &\quad=\sum_{\mathbf{q}}\begin{bmatrix}-V(\mathbf{q})N^{\alpha\beta}_{\mathbf{k}-\mathbf{q}}(t)+V(\mathbf{q})\delta_{\alpha\beta}&0\\0&V(\mathbf{q})N^{\alpha\beta}_{\mathbf{k}-\mathbf{q}}(t)\end{bmatrix}.\end{aligned} \tag{49}$$

From (46) and (49) we get for the renormalized energy matrix (30)

$$\tilde{\varepsilon}_{\alpha\beta}=\varepsilon_{\alpha\mathbf{k}}\delta_{\alpha\beta}-\mu_{\alpha\beta}\mathbf{E}(t)-\sum_{\mathbf{q}}V(\mathbf{q})N^{\alpha\beta}_{\mathbf{k}-\mathbf{q}}(t) \tag{50}$$

(the term $V\delta_{\alpha\beta}$ is omitted because it commutes with $N^{\alpha\beta}$). Now we are ready to evaluate each component of the coherent part (31) of the kinetic equation (33). We have

$$\begin{aligned}\frac{\partial n_{c\mathbf{k}}}{\partial T}&=i(p_{\mathbf{k}}\tilde{\varepsilon}_{vc}-\tilde{\varepsilon}_{cv}p^{*}_{\mathbf{k}})\\ &=-ip_{\mathbf{k}}\left[-\mu_{vc}\mathbf{E}^{*}-\sum_{\mathbf{q}}V(\mathbf{q})p^{*}_{\mathbf{k}-\mathbf{q}}\right]+\text{c.c.}\\ &=-2\,\text{Im}\left[p^{*}_{\mathbf{k}}\left[\mu_{cv}\mathbf{E}+\sum_{\mathbf{q}}V(\mathbf{q})p_{\mathbf{k}-\mathbf{q}}\right]\right]\ ,\end{aligned} \tag{51a}$$

$$\frac{\partial n_{v\mathbf{k}}}{\partial T}=i(\tilde{\varepsilon}_{cv}p_{\mathbf{k}}-p^{*}_{\mathbf{k}}\tilde{\varepsilon}_{vc})\equiv-\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]\ , \tag{51b}$$

$$\begin{aligned}\frac{\partial p_{\mathbf{k}}}{\partial T}&=i[p_{\mathbf{k}}(\tilde{\varepsilon}_{vv}-\tilde{\varepsilon}_{cc})+\tilde{\varepsilon}_{cv}(n_{c\mathbf{k}}-n_{v\mathbf{k}})]\\ &=-i\left[\varepsilon_{c\mathbf{k}}-\varepsilon_{v\mathbf{k}}-\sum_{\mathbf{q}}V(\mathbf{q})(n_{c\mathbf{k}-\mathbf{q}}-n_{v\mathbf{k}-\mathbf{q}})\right]p_{\mathbf{k}}\\ &\quad+i\left[\mu_{cv}\mathbf{E}+\sum_{\mathbf{q}}V(\mathbf{q})p_{\mathbf{k}-\mathbf{q}}\right](n_{v\mathbf{k}}-n_{c\mathbf{k}})\ ,\end{aligned} \tag{51c}$$

$$\frac{\partial p^{*}_{\mathbf{k}}}{\partial T}\equiv\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]^{*}\ . \tag{51d}$$

Equations (51) are the effective Bloch equations, which were derived by other methods in Refs. 9 and 20. They

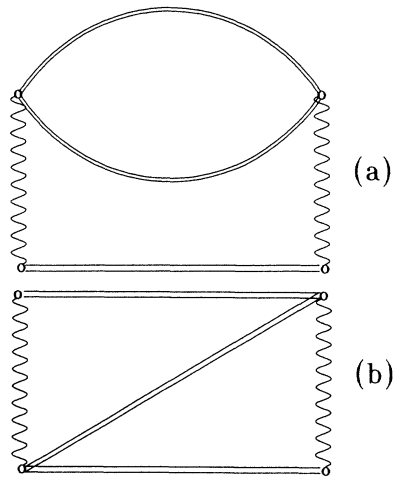

FIG. 2. Diagrammatic representation of (a) direct and (b) exchange Coulomb scattering in the Born approximation.

contain many physical effects, such as the Stark shift of the exciton,[9,11,26] coherent transients,[22,23,26] as well as other more traditional coherent-optical phenomena,[31,32] but they do not and cannot contain relaxation, which has to be added "by hand" in order to achieve a satisfactory description of realistic situations.[22–27] Our procedure enables us not only to obtain these "coherent" equations, but also to supplement them with corresponding microscopic expressions for relaxation terms.

C. Unscreened Coulomb relaxation

The simplest process that contributes to the relaxation part (32) of the kinetic equation (33) is that of pair collisions between charged particles. They are described by two self-energy diagrams shown in Fig 2, which represent direct and exchange particle-particle scattering in the Born approximation. We will evaluate here only the direct contribution, which reads

$$i\Sigma^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')=-\sum_{\substack{\mathbf{k}'\mathbf{q}\\ \alpha_1\beta_1p_1q_1}}\int_{-\infty}^{T}d1\,d2[iV^{qq_1}(\mathbf{q})\delta(t'2)iV^{pp_1}(\mathbf{q})\delta(t1)iG^{pq}_{\alpha\beta_{\mathbf{k}-\mathbf{q}}}(tt')iG^{p_1q_1}_{\alpha_1\beta_{1\mathbf{k}'}}(12)iG^{q_1p_1}_{\beta_1\alpha_{1\mathbf{k}'-\mathbf{q}'}}(21)] \tag{52}$$

or, according to the expression (48) for V^{pq},

$$\Sigma^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')=-i\sum_{\substack{\mathbf{k}'\mathbf{q}\\ \alpha_1\beta_1}}\begin{bmatrix}-1 & 1\\ 1 & -1\end{bmatrix}_{pq}V^2(\mathbf{q})[iG^{pq}_{\alpha\beta_{\mathbf{k}-\mathbf{q}}}(tt')][iG^{pq}_{\alpha_1\beta_{1\mathbf{k}'}}(tt')][iG^{qp}_{\beta_1\alpha_{1\mathbf{k}'-\mathbf{q}}}(t't)]\ . \tag{52a}$$

Our task here is to evaluate the relaxation part (32) with the self-energy (52). In order to illustrate the procedure that we use, let us consider only the first term in the expression (32) for $(\partial N/\partial T)_{St}$:

$$I_1\equiv\int_{-\infty}^{T}d1\sum_{\gamma}G^{++}_{\alpha\gamma_{\mathbf{k}}}(T1)\Sigma^{++}_{\gamma\beta_{\mathbf{k}}}(1T)\ . \tag{53}$$

The first step consists in eliminating GF from expression (52) for the self-energy by using *Ansätze* (34): we insert (34a) instead of $G(T1)$ and (34b) instead of $G(1T)$. Remembering that $T>1$, we get

$$\Sigma^{++}_{\gamma\beta_{\mathbf{k}}}(1T)=i\sum_{\substack{\mathbf{k}'\mathbf{q}\\ \alpha_1\beta_1}}V^2(\mathbf{q})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}N^{\alpha_1\beta_1}_{\mathbf{k}'}(\delta_{\beta_1\alpha_1}-N^{\beta_1\alpha_1}_{\mathbf{k}'-\mathbf{q}})\exp[i(1-T)(\varepsilon_{\alpha_1\mathbf{k}'-\mathbf{q}}-\varepsilon_{\alpha_1\mathbf{k}'}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}})] \tag{54}$$

[$N\equiv N(T)$ throughout this section]. This way one can express Σ in terms of $N(T)$ for any particular self-energy diagram. Furthermore, (34a) gives for $G^{++}(T1)$ in (53),

$$G^{++}_{\alpha\gamma_{\mathbf{k}}}(T1)=-i(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})e^{i\varepsilon_{\gamma\mathbf{k}}(1-T)}\quad(T>1)\ . \tag{55}$$

Combining (54) with (55), we obtain for (53)

$$I_1=\sum_{\substack{\mathbf{k}'\mathbf{q}\\ \alpha_1\beta_1\gamma}}V^2(\mathbf{q})(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}N^{\alpha_1\beta_1}_{\mathbf{k}'}(\delta_{\beta_1\alpha_1}-N^{\beta_1\alpha_1}_{\mathbf{k}'-\mathbf{q}})\int_{-\infty}^{T}d1\exp[i(1-T)\Delta\varepsilon_{\gamma\alpha_1}]\ , \tag{56}$$

where

$$\Delta\varepsilon_{\gamma\alpha_1}\equiv\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}+\varepsilon_{\alpha_1\mathbf{k}'-\mathbf{q}}-\varepsilon_{\alpha_1\mathbf{k}'}\ . \tag{57}$$

In the same way one can process the remaining terms in (32), which finally yield

$$\begin{aligned}\left[\frac{\partial N^{\alpha\beta}_{\mathbf{k}}}{\partial T}\right]_{St}=\sum_{\substack{\mathbf{k}'\mathbf{q}\\ \alpha_1\beta_1\gamma}}V^2(\mathbf{q})\Big\{&[(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}N^{\alpha_1\beta_1}_{\mathbf{k}'}(\delta_{\alpha_1\beta_1}-N^{\beta_1\alpha_1}_{\mathbf{k}'-\mathbf{q}})\\ &-N^{\alpha\gamma}_{\mathbf{k}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}})(\delta_{\alpha_1\beta_1}-N^{\alpha_1\beta_1}_{\mathbf{k}'})N^{\beta_1\alpha_1}_{\mathbf{k}'-\mathbf{q}}]\int_{-\infty}^{0}\exp(i\tau\Delta\varepsilon_{\gamma\alpha_1})d\tau\\ &+[N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}})(\delta_{\alpha_1\beta_1}-N^{\alpha_1\beta_1}_{\mathbf{k}'-\mathbf{q}})N^{\beta_1\alpha_1}_{\mathbf{k}'}\\ &-(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}})N^{\gamma\beta}_{\mathbf{k}}N^{\alpha_1\beta_1}_{\mathbf{k}'-\mathbf{q}}(\delta_{\beta_1\alpha_1}-N^{\beta_1\alpha_1}_{\mathbf{k}'})]\int_{0}^{+\infty}\exp(i\tau\Delta\varepsilon_{\gamma\alpha_1})d\tau\Big\}\ .\end{aligned} \tag{58}$$

The integrals in (58) are equal to

$$\int_{-\infty}^{0}d\tau\,e^{i\tau\varepsilon}=\pi\delta(\varepsilon)-i\lim_{\delta\to0}\frac{\varepsilon}{\varepsilon^2+\delta^2}\equiv\pi\delta_-(\varepsilon)\ , \tag{59a}$$

$$\int_{0}^{+\infty}d\tau\,e^{i\tau\varepsilon}=\pi\delta(\varepsilon)+i\lim_{\delta\to0}\frac{\varepsilon}{\varepsilon^2+\delta^2}\equiv\pi\delta_+(\varepsilon)\ , \tag{59b}$$

$$\delta_+(\varepsilon)=\delta^*_-(\varepsilon)=\delta_-(-\varepsilon)\ . \tag{59c}$$

To gain insight into the physical contents of Eq. (58), let us first extract the usual Boltzmann terms out of it. This is achieved by retaining only diagonal (over band indices) terms in the equation for n_{ck}, which follows from (58) at $\alpha=\beta=c$. Handling the integrals in (58) according to (59) when performing the summation over band indices, we obtain

$$\left[\frac{\partial n_{ck}}{\partial T}\right]_{\text{Boltzmann}}=2\pi\sum_{k'q}V^2(\mathbf{q})\{(1-n_{ck})n_{ck-q}[n_{ck'}(1-n_{ck'-q})\delta(\Delta\varepsilon_{cc})+n_{vk'}(1-n_{vk'-q})\delta(\Delta\varepsilon_{cv})]$$
$$-n_{ck}(1-n_{ck-q})[(1-n_{ck'})n_{ck'-q}\delta(\Delta\varepsilon_{cc})+(1-n_{vk'})n_{vk'-q}\delta(\Delta\varepsilon_{cv})]\}\ . \tag{60a}$$

This expression has a characteristic gain-loss structure and is quite clear in meaning: the particles scatter on one another conserving their total energy. The structure of the remaining terms in Eq. (58), however, is far less obvious. Performing the band indices summation in (58), we obtain, on the one hand, the following term:

$$\left[\frac{\partial n_{ck}}{\partial T}\right]_{p1}=\pi\sum_{k'q}V^2(\mathbf{q})(n_{ck}-n_{ck-q})p_{k'}p^*_{k'-q}[\delta_-(\Delta\varepsilon_{cc})+\delta_+(\Delta\varepsilon_{cv})]+\text{c.c.} \tag{60b}$$

This term can be said to describe the effect of scattering events whereby an electron scatters out of the state $\mathbf{k}$ due to interaction with the *polarization wave* $\sum(p_{k'}p^*_{k'-q}+\text{c.c.})$ with the wave vector $\mathbf{q}$. Finally, the term

$$\left[\frac{\partial n_{ck}}{\partial T}\right]_{p2}=\pi\sum_{k'q}V^2(\mathbf{q})p_kp^*_{k-q}[(n_{ck'-q}-n_{ck'})\delta_-(\Delta\varepsilon_{vc})+(n_{vk'-q}-n_{vk'})\delta_+(\Delta\varepsilon_{vv})]+\text{ c.c.} \tag{60c}$$

corresponds to the reverse process, namely the appearance (or disappearance) of electrons in the state $\mathbf{k}$ during scattering of another electron or hole ($\mathbf{k}'\rightarrow\mathbf{k}'-\mathbf{q}$) on the polarization wave $p_kp^*_{k-q}$. The overall scattering rate is therefore

$$\left[\frac{\partial n_{ck}}{\partial T}\right]_{St}=\left[\frac{\partial n_{ck}}{\partial T}\right]_{\text{Boltzmann}}+\left[\frac{\partial n_{ck}}{\partial T}\right]_{p1}+\left[\frac{\partial n_{ck}}{\partial T}\right]_{p2}. \tag{61}$$

Terms (60b) and (60c) deserve some comment. First, they can be expected to be of importance in the initial coherent regime of the optical excitation,[9] i.e., at times shorter than the dephasing time. Indeed, in this regime and in the low-field limit there is an approximate relation[9,11] between p_k and $n_{\alpha k}$:

$$|p_k|^2\approx n_{ck}\approx(1-n_{vk})\ . \tag{62}$$

Equation (62) shows clearly that in this regime all terms in (61) are *of the same order*— there are no *a priori* reasons why the last two terms, which represent "polarization scattering," should be neglected in comparison with the Boltzmann term. Let us note that this scattering mechanism can, in somewhat different language, be incorporated into kinetic equations for light-renormalized quasiparticles in the form of "coherent factors."[21] This, however, implies the existence of a relation like (62) between n and p, i.e., the absence of relaxation. Equation (61), on the other hand, is valid also beyond the coherent regime, when some relaxation has already occurred, and does not require a steady-state optical field so that it is more general than previous results.[21]

The second important observation is that processes like (60b) and (60c) *do not conserve the energy* because of imaginary parts in δ_+ and δ_-, which do not have δ-function structure and do not cancel out as they did in (60a). In fact, there is nothing strange about it, since when $p\neq0$ the scattering electrons are not in definite-energy eigenstates anymore—their wave functions are quantum-mechanical superpositions of conduction- and valence-band states. This makes polarization scattering nontrivial, and below we will discuss the underlying physics in more detail.

To be complete, let us also consider the dephasing; the corresponding equation is obtained from (58) by setting $\alpha=c$ and $\beta=v$. There are the "Boltzmann-like" terms as well:

$$\left[\frac{\partial p_k}{\partial T}\right]_{\text{Boltzmann}}=\pi\sum_{k'q}V^2(\mathbf{q})(-p_k\{n_{ck-q}[n_{ck'}(1-n_{ck-q})\delta_+(\Delta\varepsilon_{cc})+n_{vk'}(1-n_{vk'-q})\delta_+(\Delta\varepsilon_{cv})]$$
$$+(1-n_{ck-q})[n_{ck'-q}(1-n_{ck'})\delta_+(\Delta\varepsilon_{cc})+n_{vk'-q}(1-n_{vk'-q})\delta_+(\Delta\varepsilon_{cv})]$$
$$+n_{vk-q}[n_{ck'}(1-n_{ck'-q})\delta_-(\Delta\varepsilon_{vc})+N_{vk'}(1-n_{vk'-q})\delta_-(\Delta\varepsilon_{vv})]$$
$$+(1-n_{vk-q})[n_{ck'-q}(1-n_{ck'})\delta_-(\Delta\varepsilon_{vc})+n_{vk'-q}(1-n_{vk'})\delta_-(\Delta\varepsilon_{vv})]\} \tag{63a}$$
$$+p_{k-q}\{n_{ck}[n_{ck'-q}(1-n_{ck'})\delta_-(\Delta\varepsilon_{cc})+n_{vk'-q}(1-n_{vk'})\delta_-(\Delta\varepsilon_{cv})]$$
$$+(1-n_{ck})[n_{ck'}(1-n_{ck'-q})\delta_-(\Delta\varepsilon_{cc})+n_{vk'}(1-n_{vk'-q})\delta_-(\Delta\varepsilon_{cv})]$$
$$+n_{vk}[n_{ck'-q}(1-n_{ck'})\delta_+(\Delta\varepsilon_{vc})+n_{vk'-q}(1-n_{vk'})\delta_+(\Delta\varepsilon_{vv})]$$
$$+(1-n_{vk})[n_{ck'}(1-n_{ck'-q})\delta_+(\Delta\varepsilon_{vc})+n_{vk'}(1-n_{vk'-q})\delta_+(\Delta\varepsilon_{vv})]\})\ . \tag{63b}$$

These terms, whose real parts have been obtained also in Ref. 20, describe the decay [(63a)] and the diffusion of polarization [(63b)] in $\mathbf{k}$ space as a result of "Boltzmann" scattering of particles on one another. The remaining terms account for the effect of scattering on polarization waves:

$$\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_p = \pi \sum_{\mathbf{k}'\mathbf{q}} V^2(\mathbf{q})(p_{\mathbf{k}}\{p_{\mathbf{k}'-\mathbf{q}}p^*_{\mathbf{k}'}[\delta_+(\Delta\varepsilon_{cc})+\delta_-(\Delta\varepsilon_{vv})]+p_{\mathbf{k}'}p^*_{\mathbf{k}'-\mathbf{q}}[\delta_+(\Delta\varepsilon_{cv})+\delta_-(\Delta\varepsilon_{vc})]\}$$
$$-p_{\mathbf{k}-\mathbf{q}}\{p_{\mathbf{k}'}p^*_{\mathbf{k}'-\mathbf{q}}[\delta_-(\Delta\varepsilon_{cc})+\delta_+(\Delta\varepsilon_{vv})]+p_{\mathbf{k}'-\mathbf{q}}p^*_{\mathbf{k}'}[\delta_-(\Delta\varepsilon_{cv})+\delta_+(\Delta\varepsilon_{vc})]\}) \tag{63c}$$

so that the total dephasing rate is given by

$$\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_{\text{total}} = \left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_{\text{Boltzmann}} + \left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_p . \tag{64}$$

It is easy to see that in the low-field limit the factors beside the δ functions are of the order of $n \sim \mathbf{E}^2$ in (63a) and (63b), and of the order of $|p|^2 \sim \mathbf{E}^2$ in (63c); in higher fields both terms are of the same order as well. This shows that the polarization scattering is very important to the correct description of dephasing processes. To demonstrate this for other interaction mechanisms, and to gain further understanding of the physical origins of this process, we now proceed to the case of the screened Coulomb interaction.

D. Screened Coulomb relaxation

One of the reasons that the results of the preceding subsection are of little practical value is that in Coulomb systems it is crucial to ensure that screening is properly accounted for. A brief account of screening theory within the framework of Keldysh's GF formalism is given in the Appendix.

Our aim here is to investigate how screening affects particle kinetics and dephasing, using the concept of the Bloch equations. We will restrict ourselves to the so-called screened Hartree-Fock (SHF) approximation[64,65] for the self-energy, which is represented diagrammatically in Fig. 3.

Let us note from the start that the diagram of Fig. 3 formally contains the pair Coulomb scattering diagram of Fig. 2, as well as the unscreened Hartree-Fock self-energy of Fig. 1. The latter has a singularity at coincident times [Eq. (47)], which has been accounted for when deriving the coherent part of the Bloch equations (51). Hence, to evaluate the relaxation term we should use

$$\tilde{\Sigma}^{pq}_{\alpha\beta_{\mathbf{k}}}(tt') = -i \sum_{\mathbf{q}} G^{pq}_{\alpha\beta_{\mathbf{k}-\mathbf{q}}}(tt')\Delta V^{pq}_S(tt') , \tag{65}$$

where

$$\Delta V^{pq}_S \equiv V^{pq}_S - V^{pq} . \tag{66}$$

For illustrative purposes we will again consider only the first term (53) of the relaxation part (32). Proceeding exactly as above with the self-energy (65), we obtain

$$\int_{-\infty}^{T} G^{++}_{\alpha\gamma}\Sigma^{++}_{\gamma\beta} \equiv I_1 = -i \sum_{\mathbf{q}\gamma} (\delta_{\alpha\gamma} - N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}} \int_{-\infty}^{T} [V_S^{++}(1T)+V]\exp[i(1-T)(\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}})_{d1}] \tag{67}$$

(other terms are treated in the same way). Noting that, due to properties (A4) of the screened potential, we have

$$V_S^{++}(T1)+V = -V_S^{-+}(T1), \quad V_S^{++}(1T)+V = -V_S^{+-}(1T) , \tag{68}$$

and introducing shorthand notation

$$\int_{-\infty}^{T} V_S^{+-}(1T)\exp[i(1-T)(\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}})]d1 \equiv V^{+-}_{SA}(\varepsilon_\gamma) , \tag{69a}$$

$$\int_{-\infty}^{T} V_S^{+-}(T1)\exp[i(T-1)(\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}})]d1 \equiv V^{+-}_{SR}(\varepsilon_\gamma) \tag{69b}$$

(and similarly for V^{-+}), after some algebra we will get for the relaxation term in the screened Hartree-Fock approximation

$$\left[\frac{\partial N^{\alpha\beta}_{\mathbf{k}}}{\partial T}\right]_{St} = i \sum_{\mathbf{q}\gamma} [-(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}})N^{\gamma\beta}_{\mathbf{k}}V^{-+}_{SR}(\varepsilon_\gamma)+N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}})V^{+-}_{SR}(\varepsilon_\gamma)$$
$$+(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}V^{+-}_{SA}(\varepsilon_\gamma)-N^{\alpha\gamma}_{\mathbf{k}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}})V^{-+}_{SA}(\varepsilon_\gamma)] . \tag{70}$$

Let us concentrate on the carrier kinetics first. The summation over γ yields for $\alpha=\beta=c$

$$\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{St} = i \sum_{\mathbf{q}} \{n_{c\mathbf{k}-\mathbf{q}}(1-n_{c\mathbf{k}})[V^{+-}_{SR}(\varepsilon_c)+V^{+-}_{SA}(\varepsilon_c)]-n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})[V^{-+}_{SR}(\varepsilon_c)+V^{-+}_{SA}(\varepsilon_c)]$$
$$+p^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}[V^{-+}_{SR}(\varepsilon_v)+V^{+-}_{SR}(\varepsilon_v)]+p^*_{\mathbf{k}-\mathbf{q}}p_{\mathbf{k}}[V^{-+}_{SA}(\varepsilon_v)+V^{+-}_{SA}(\varepsilon_v)]\} . \tag{71}$$

From (68), (69), and (A4) it follows that

$$V_{SR}^{+-}+V_{SA}^{+-}\equiv V_S^{+-}(\omega=\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}) , \tag{72a}$$

$$V_{SR}^{-+}-V_{SR}^{+-}\equiv -V+V_S^R(\omega=\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}),\quad V_{SA}^{-+}-V_{SA}^{+-}\equiv V_S^A-V \tag{72b}$$

so that (71) can be transformed further:

$$\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{St}=(1-n_{c\mathbf{k}})\left[\sum_{\mathbf{q}} n_{c\mathbf{k}-\mathbf{q}} iV_S^{+-}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})\right]-n_{c\mathbf{k}}\sum_{\mathbf{q}}[(1-n_{c\mathbf{k}-\mathbf{q}})iV_S^{-+}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})]$$

$$+ip_{\mathbf{k}}^*\sum_{\mathbf{q}} p_{\mathbf{k}-\mathbf{q}}[V_S^R(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})-V]-ip_{\mathbf{k}}\sum_{\mathbf{q}} p_{\mathbf{k}-\mathbf{q}}^*[V_S^A(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})-V] . \tag{73}$$

Combining (73) with the coherent part (51a), we finally get

$$\frac{\partial n_{c\mathbf{k}}}{\partial T}=-2\,\mathrm{Im}\left[p_{\mathbf{k}}^*\left[\mu_{cv}\mathbf{E}(t)+\sum_{\mathbf{q}} V_S^R(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})p_{\mathbf{k}-\mathbf{q}}\right]\right]$$

$$+(1-n_{c\mathbf{k}})\left[\sum_{\mathbf{q}} n_{c\mathbf{k}-\mathbf{q}} iV_S^{+-}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})\right]-n_{c\mathbf{k}}\left[\sum_{\mathbf{q}}(1-n_{c\mathbf{k}-\mathbf{q}})iV_S^{-+}\right] . \tag{74}$$

This is our basic equation for particle kinetics in the SHF approximation. The second line in (74) describes scattering of electrons in and out of the state $\mathbf{k}$. The first term of the first line is merely the generation of electrons by an external optical field $\mathbf{E}(t)$. The polarization scattering is explicitly given by the second term, which is proportional to $p_{\mathbf{k}}^* p_{\mathbf{k}-\mathbf{q}}$ [note that the presence of a nonzero interband polarization also distorts pq components of V_S (see the Appendix), so that even the traditional Boltzmann terms are affected by it].

Equation (74) gives us an opportunity to see where the polarization scattering comes from: alongside the external electric field $\mathbf{E}(t)$, there also is a *local field* $\sum_{\mathbf{q}} V_S^R p_{\mathbf{k}-\mathbf{q}}$ [it is often said that the large parentheses in the first term of (74) contain the "renormalized Rabi frequency"].[9,11,20] This local field interferes with $p_{\mathbf{k}}^*$ and thereby creates carriers in some portions of $\mathbf{k}$ space, destroying them elsewhere at the same time. In order to check that this is indeed scattering, and not some extra generation source, let us consider the evolution of the total number of carriers due to this process:

$$\sum_{\mathbf{k}}\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_p=i\sum_{\mathbf{kq}} p_{\mathbf{k}}^* p_{\mathbf{k}-\mathbf{q}} V_S^R(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})-i\sum_{\mathbf{kq}} p_{\mathbf{k}} p_{\mathbf{k}-\mathbf{q}}^* V_S^A(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})$$

$$=i\sum_{\mathbf{kq}} p_{\mathbf{k}}^* p_{\mathbf{k}-\mathbf{q}}[V_S^R(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})-V_S^A(\varepsilon_{v\mathbf{k}-\mathbf{q}}-\varepsilon_{v\mathbf{k}})]\equiv 0 \tag{75}$$

since

$$V_S^R(\omega)=V_S^A(-\omega) . \tag{76}$$

As has been noted, in the initial coherent regime, i.e., as long as Eq. (62) holds, this scattering is as important as the traditional Boltzmann one, and therefore should not be omitted in calculations of carrier dynamics, as was the case in Refs. 38–41. However, to take this process into account, one has to monitor the polarization dynamics as well.

The equation for polarization is obtained from Eq. (70) by setting $\alpha=c$ and $\beta=v$. After the summation over γ, this gives

$$\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_{St}=-ip_{\mathbf{k}}\sum_{\mathbf{q}}[(1-n_{c\mathbf{k}-\mathbf{q}})V_{SR}^{-+}(\varepsilon_c)+n_{c\mathbf{k}-\mathbf{q}}V_{SR}^{+-}(\varepsilon_c)+n_{v\mathbf{k}-\mathbf{q}}V_{SA}^{+-}(\varepsilon_v)+(1-n_{v\mathbf{k}-\mathbf{q}})V_{SA}^{-+}(\varepsilon_v)]$$

$$+i\sum_{\mathbf{q}} p_{\mathbf{k}-\mathbf{q}}[n_{v\mathbf{k}}V_{SR}^{-+}(\varepsilon_v)+(1-n_{v\mathbf{k}})V_{SR}^{+-}(\varepsilon_v)+n_{c\mathbf{k}}V_{SA}^{-+}(\varepsilon_c)+(1-n_{c\mathbf{k}})V_{SA}^{+-}(\varepsilon_c)] . \tag{77}$$

The factor next to $p_{\mathbf{k}}$ can be transformed to the form

$$\sum_{\mathbf{q}}[(1-n_{c\mathbf{k}-\mathbf{q}})V_{SR}^{-+}(\varepsilon_c)+n_{c\mathbf{k}-\mathbf{q}}V_{SR}^{+-}(\varepsilon_c)]\equiv\tilde{\Sigma}_{cc}^R(\omega=\varepsilon_{c\mathbf{k}}) , \tag{78a}$$

$$\sum_{\mathbf{q}}[n_{v\mathbf{k}-\mathbf{q}}V_{SA}^{+-}(\varepsilon_v)+(1-n_{v\mathbf{k}-\mathbf{q}})V_{SA}^{-+}(\varepsilon_v)]\equiv\tilde{\Sigma}_{vv}^A(\omega=\varepsilon_{v\mathbf{k}}) , \tag{78b}$$

i.e., it is the sum of electron and hole correlation energies.[64] Performing simple transformations with the term containing $p_{\mathbf{k}-\mathbf{q}}$, and combining (77) with the coherent part (51c), we finally get

$$\frac{\partial p_{\mathbf{k}}}{\partial T}=-ip_{\mathbf{k}}\left\{\left[\varepsilon_{c\mathbf{k}}+\left[-\sum_{\mathbf{q}}V_{\mathbf{q}}n_{c\mathbf{k}-\mathbf{q}}+\tilde{\Sigma}^{R}_{cc}(\varepsilon_{c\mathbf{k}})\right]\right]-\left[\varepsilon_{v\mathbf{k}}+\left[-\sum_{\mathbf{q}}V_{\mathbf{q}}n_{v\mathbf{k}-\mathbf{q}}+\tilde{\Sigma}^{A}_{vv}(\varepsilon_{v\mathbf{k}})\right]\right]\right\}$$
$$+i\mu_{cv}\mathbf{E}(t)(n_{v\mathbf{k}}-n_{c\mathbf{k}})+in_{v\mathbf{k}}\sum_{\mathbf{q}}V_S^R(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})p_{\mathbf{k}-\mathbf{q}}-in_{c\mathbf{k}}\sum_{\mathbf{q}}V_S^A(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})p_{\mathbf{k}-\mathbf{q}}$$
$$+i\sum_{\mathbf{q}}p_{\mathbf{k}-\mathbf{q}}[V_{SA}^{+-}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})+V_{SR}^{+-}(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})]\ . \tag{79}$$

The curly brackets in (79) contain the renormalized transition energy: the renormalization of, say, electron energy consists of the exchange self-energy, $-\sum_{\mathbf{q}}V_{\mathbf{q}}n_{c\mathbf{k}-\mathbf{q}}$, and of correlation energy $\tilde{\Sigma}^{R}$ (Ref. 47). The latter has a real part, which gives the energy shift proper, and an imaginary part that gives rise to the polarization decay. The correlation energies enter Eq. (70) in combination $\tilde{\Sigma}^{R}_{cc}-\tilde{\Sigma}^{A}_{vv}$, which ensures the "right" sign of dephasing. From (79) one can see that, although at first sight *Ansätze* (34) neglect renormalizations of the spectrum, the energy of the optical transitions (which is the only source of information on the spectrum in this case) becomes properly renormalized.

The set of the Bloch equations (74) and (79) in the SHF approximation is formulated in terms of frequency-dependent pq components of the screened potential V_S which have to be calculated beforehand. In principle, one can use the results of the Appendix to this effect, i.e., one can use expressions (A6) for V_S^{pq} with Π^{pq} given by (A10) and (A11). Although this would make the problem completely self-contained, the resulting expressions would be too complicated to make any practical sense. That is why one has to resort to some approximation schemes for the screened potential.

The simplest and by far the most widely used approximation for V_S is the static-screening approximation (SSA),[47,64] which neglects the frequency dependence of V_S^{pq}:

$$V_S^{pq}(\omega)\approx\begin{bmatrix}-V_S^R(\omega=0) & 0\\ 0 & V_S^R(\omega=0)\end{bmatrix}_{pq}\ , \tag{80}$$

where the statically screened potential V_S^R can be defined in a number of ways.[47,64,65] Comparing (80) with (48), we see that this is equivalent to the replacement of V by V_S in the initial Hamiltonian (43) (as was actually done in Ref. 20). Noting that within the SSA

$$\tilde{\Sigma}^{R}_{cc}(\varepsilon_{c\mathbf{k}})\approx\sum_{\mathbf{q}}[V_{\mathbf{q}}-V_S^R(\mathbf{q})]n_{c\mathbf{k}-\mathbf{q}}\ , \tag{81a}$$

$$\tilde{\Sigma}^{R}_{vv}(\varepsilon_{v\mathbf{k}})\approx\sum_{\mathbf{q}}[V_{\mathbf{q}}-V_S^R(\mathbf{q})]n_{v\mathbf{k}-\mathbf{q}} \tag{81b}$$

(since $V_S^R=V_S^A$ in this case), we obtain from (79)

$$\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_{\substack{\text{Static}\\ \text{screening}}}=-ip_{\mathbf{k}}\left[\left[\varepsilon_{c\mathbf{k}}-\sum_{\mathbf{q}}V_S^R(\mathbf{q})n_{c\mathbf{k}-\mathbf{q}}\right]-\left[\varepsilon_{v\mathbf{k}}-\sum_{\mathbf{q}}V_S^R(\mathbf{q})n_{v\mathbf{k}-\mathbf{q}}\right]\right]+i\left[\mu_{cv}\mathbf{E}+\sum_{\mathbf{q}}V_S^R(\mathbf{q})p_{\mathbf{k}-\mathbf{q}}\right](n_{v\mathbf{k}}-n_{c\mathbf{k}}) \tag{82}$$

i.e., the result of Ref. 20. The comparison of (82) with (79) makes clear the consequences of leaving the dynamic screening effects out: (i) it leads to the omission of the Coulomb-hole part[47,65] of the correlation energy [Eq. (82) contains only "screened exchange" contributions,[20,47] which have no imaginary part and therefore do not produce polarization decay]; (ii) one can combine terms with V^R and V^A from Eq. (79) in the form (82) only ignoring the difference in their frequency arguments; (iii) the last term in (79) has completely dropped out in the SSA.

The static-screening approximation gives the following result for the carrier kinetics:

$$\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{\substack{\text{Static}\\ \text{screening}}}=-2\,\text{Im}\left[p_{\mathbf{k}}^{*}\left[\mu_{cv}\mathbf{E}+\sum_{\mathbf{q}}V_S^R(\mathbf{q})p_{\mathbf{k}-\mathbf{q}}\right]\right]\ , \tag{83}$$

i.e., it leaves out all scattering mechanisms except the polarization scattering.

Another prescription for neglecting the dynamic effects in screening that gives less disastrous results for carrier kinetics consists in replacing the bare Coulomb potential $V_{\mathbf{q}}$ with the statically screened one directly in the Boltzmann equation (60a) for particle-particle scattering.[38–40] Let us consider how it can be understood within the GF formalism.

First of all, it is clear that simple replacement of $V_{\mathbf{q}}$ by V_S in the Coulomb scattering self-energy diagram of Fig. 2, which would have given the desired result, does not make much sense since it would contradict the definition

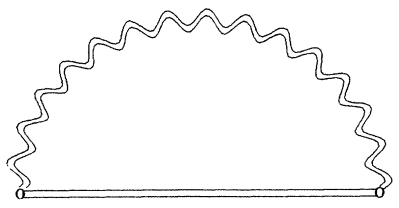

FIG. 3. Screened Hartree-Fock self-energy [(65)]; the double wavy line represents the screened Coulomb potential.

of the screened potential (see the Appendix).

However, one can recover the kinetic equations used in Refs. 38–40 from (74) by making the following approximation for V_S^{+-} and V_S^{-+} in Eq. (74):

$$V_S^{pq}(\omega)=-\frac{V^2\Pi^{pq}(\omega)}{[1-V\Pi^R(\omega)][1-V\Pi^A(\omega)]} \approx -V_S^2\Pi^{pq}(\omega) \tag{84a}$$

[see Eqs. (A6b) and (A6c)], where

$$V_S=V_S^R(\omega=0)=\frac{V}{1-V\Pi^R} \tag{84b}$$

(i.e., we neglect the frequency dependence of V_S^R but retain it in V_S^{+-} and V_S^{-+}). Now, e.g., the "gain" term in (74) takes on the form

$$(1-n_{c\mathbf{k}})\left[\sum_{\mathbf{q}} n_{c\mathbf{k}-\mathbf{q}} iV_S^{+-}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})\right] \approx -i\sum_{\mathbf{q}} n_{c\mathbf{k}-\mathbf{q}}(1-n_{c\mathbf{k}})V_S^2\Pi^{+-}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}) . \tag{85}$$

Inserting the expression for Π^{+-} (A11) in (85) and taking into account that

$$\frac{1}{\omega\pm i\delta}=\mp i\pi\delta_{\pm}(\omega) \tag{86}$$

[and doing the same with the loss term of Eq. (74)], we will obtain Eq. (61) with $V(\mathbf{q})$ replaced by the screened potential (84); this replacement has to be done of course in the coherent part (51) as well. This allows us to include the polarization scattering in the approach of Refs. 38–40.

However, in spite of a good agreement of the numerical results of Refs. 38–40 with experiment, this approach does not seem to be very consistent: the possibility of neglecting dynamic effects in some of the pq components of V_S while keeping them in the others [which is what (84) amounts to] apparently needs some more profound justification than just numerical convenience. Moreover, approximation (84) applied to the polarization equation (79) does not achieve anything at all: it leaves out polarization decay completely, which is clearly unphysical.

These considerations, as well as some recent discussions on the role of dynamic screening in relaxation processes,[66,67] urge caution with the use of static-screening-type approximations, and demonstrate the need for more elaborate approaches to ensure proper understanding of relaxation in Coulomb systems.

E. The equilibrium limit

The results of the preceding subsection are valid regardless of whether the system is in equilibrium or not. However, in the quasiequilibrium case they should reduce to well-known results of the quasiequilibrium theory of highly excited semiconductors.[64] In this subsection we will carry out this reduction explicitly.

Quasiequilibrium means, first, that $p_{\mathbf{k}}\to 0$; in addition, the carriers have Fermi distributions within the bands:

$$n_{\alpha\mathbf{k}}=\{1+\exp[\beta(\varepsilon_{\alpha\mathbf{k}}-\mu_\alpha)]\}^{-1} , \tag{87}$$

where β is the inverse temperature; physically it means that the system has stayed in a weak steady-state field for long enough.[64]

In such cases we have to regard $p_{\mathbf{k}}$ in the corresponding equation as the linear response to a weak perturbation $\mu_{cv}\mathbf{E}$, neglecting all terms of the order of $|p|^2$, e.g., polarization contributions to V_S. Let us note from the start that in quasiequilibrium the pq components of V_S and Π are related by the Kubo-Martin-Schwinger condition:[47,64]

$$\Pi^{-+}(\tau)=\Pi^{+-}(\tau+i\beta) , \tag{88a}$$

$$V_S^{-+}(\tau)=V_S^{+-}(\tau+i\beta) , \tag{88b}$$

which in the frequency space reads

$$V_S^{+-}(\omega)=e^{-\beta\omega}V_S^{-+}(\omega) . \tag{88c}$$

From the properties of V_S [(A4)] it follows that

$$V_S^R-V_S^A=2i\,\mathrm{Im}V_S^R\equiv V_S^{-+}-V_S^{+-} \tag{89}$$

so that we can express V_S^{+-} and V_S^{-+} through the imaginary part of the retarded screened potential:

$$V_S^{-+}(\omega)=-ig(-\omega)[2\,\mathrm{Im}V_S^R(\omega)] , \tag{90a}$$

$$V_S^{+-}(\omega)=ig(\omega)[2\,\mathrm{Im}V_S^R(\omega)] , \tag{90b}$$

where

$$g(\omega)=\frac{1}{e^{\beta\omega}-1} \tag{91}$$

is the Bose distribution function.

Let us now consider what happens in quasiequilibrium to the self-energies that enter Eq. (79) for the optical polarization. According to expression (78a), the retarded self-energy is equal to

$$\Sigma_{cc}^R(\varepsilon_{c\mathbf{k}})=\sum_{\mathbf{q}}[(1-n_{c\mathbf{k}-\mathbf{q}})V_{SR}^{-+}(\varepsilon_c)+n_{c\mathbf{k}-\mathbf{q}}V_{SR}^{+-}(\varepsilon_c)] =\sum_{\mathbf{q}}\{V_{SR}^{-+}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}) +n_{c\mathbf{k}-\mathbf{q}}[V_{\mathbf{q}}-V_S^R(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})]\} . \tag{92}$$

Both V_{SR}^{-+} and $V-V_S^R$ are equal to zero as functions of τ at $\tau<0$, so that their Fourier transforms are analytical in the upper-half-plane and therefore admit a spectral representation:[64,65]

$$V_S^R(\omega)=V-\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\frac{\mathrm{Im}V_S^R(\omega')}{\omega-\omega'+i\delta} \tag{93}$$

(and a similar one for V_{SR}^{+-}), so that (92) can be cast into the form

$$\Sigma_{cc}^R(\varepsilon_{c\mathbf{k}})=\sum_{\mathbf{q}}\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi}\frac{n_{c\mathbf{k}-\mathbf{q}}\mathrm{Im}V_S^R(\omega')-\mathrm{Im}V_{SR}^{-+}(\omega')}{\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega'+i\delta} . \tag{94}$$

Taking into account (90a) and the fact that

$$2i\,\mathrm{Im}V_{SR}^{-+}(\omega)=V_S^{-+}(\omega) , \tag{95}$$

in the quasiequilibrium limit the correlation energy takes on the form

$$[\Sigma^R_{cc}(\varepsilon_{c\mathbf{k}})]_{\mathrm{Eq}} = \sum_{\mathbf{q}} \int_{-\infty}^{+\infty} \frac{d\omega'}{\pi} \frac{[g(-\omega')+n_{c\mathbf{k}-\mathbf{q}}]\mathrm{Im}V_S^R(\omega')}{\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega'+i\delta} , \quad (96)$$

which coincides with the Montroll-Ward result for the correlation energy within the RPA [Eq. (27) of Ref. 47].

Hence, the first term in (79) now contains the quasiequilibrium values of the renormalized quasiparticle energies, $\tilde{\varepsilon}_{\alpha k}$. If we make the static-screening approximation (80) at this stage, we will obtain instead of (79)

$$\frac{\partial p_{\mathbf{k}}}{\partial T} = -i(\tilde{\varepsilon}_{c\mathbf{k}}-\tilde{\varepsilon}_{v\mathbf{k}})p_{\mathbf{k}} + i\left[\mu_{cv}\mathbf{E}+\sum_{\mathbf{q}} V_S(\mathbf{q})p_{\mathbf{k}-\mathbf{q}}\right](n_{v\mathbf{k}}-n_{c\mathbf{k}}) . \quad (97)$$

We have to determine from this equation the linear response of $p_{\mathbf{k}}$ to a harmonic perturbation, i.e., we must set

$$\mathbf{E}=\mathbf{E}_0 e^{-i\omega t}, \quad p_{\mathbf{k}}=\tilde{p}_{\mathbf{k}} e^{-i\omega t} , \quad (98)$$

which allows us to rewrite (97) as

$$\tilde{p}_{\mathbf{k}}(\tilde{\varepsilon}_{c\mathbf{k}}-\tilde{\varepsilon}_{v\mathbf{k}}-\omega) = \left[\mu_{cv}E_0+\sum_{\mathbf{q}} V_S(\mathbf{q})\tilde{p}_{\mathbf{k}-\mathbf{q}}\right](n_{v\mathbf{k}}-n_{c\mathbf{k}}) , \quad (99)$$

which is Eq. (58) of Ref. 47. Defining the susceptibility

$$\chi_{\mathbf{k}}(\omega)\equiv\frac{\tilde{p}_{\mathbf{k}}}{\mathbf{E}_0} , \quad (100)$$

from (98) we obtain the following result for this quantity:

$$\chi_{\mathbf{k}}(\omega)=\frac{(n_{v\mathbf{k}}-n_{c\mathbf{k}})}{\tilde{\varepsilon}_{c\mathbf{k}}-\tilde{\varepsilon}_{v\mathbf{k}}-\omega}\left[\mu_{cv}+\sum_{\mathbf{q}} V_S(\mathbf{q})\chi_{\mathbf{k}-\mathbf{q}}(\omega)\right] . \quad (101)$$

This is the Bethe-Salpeter equation (BSE) in the static-screening approximation [Eq. (60) of Ref. 47]. The macroscopic optical polarization is expressed through $\chi_{\mathbf{k}}$ as

$$\mathbf{P}(\omega)=\sum_{\mathbf{k}}\mu_{cv}\tilde{p}_{\mathbf{k}}(\omega)=\left[\sum_{\mathbf{k}}\mu_{cv}\chi_{\mathbf{k}}(\omega)\right]\mathbf{E}_0=\chi(\omega)\mathbf{E}_0 , \quad (102)$$

where $\chi(\omega)$ is the total susceptibility.

Now let us consider what becomes of Eq. (74) for carrier kinetics in quasiequilibrium. Its second line (gain-loss terms) vanishes in quasiequilibrium, as it should be, and as one can easily check by inserting the quasiequilibrium relations (87) and (88) into (74). As for the first line in (74), we are interested only in its effect on the total number of carriers, since the distribution functions stay Fermi-like anyway; let us sum it up over the whole $\mathbf{k}$ space:

$$\frac{dn}{dT}=\sum_{\mathbf{k}}\frac{\partial n_{\mathbf{k}}}{\partial T}=-2\,\mathrm{Im}\sum_{\mathbf{k}} p^*_{\mathbf{k}}\mu_{cv}\mathbf{E}-2\,\mathrm{Im}\sum_{\mathbf{kq}} V_S^R p^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}} . \quad (103)$$

The second term, which accounts for the polarization scattering, vanishes according to (75), while the first one yields

$$\frac{dn}{dT}=-2\,\mathrm{Im}\left[\left(\sum_{\mathbf{k}} p^*_{\mathbf{k}}\mu_{cv}\right)\mathbf{E}\right]=-2\,\mathrm{Im}\chi^*(\omega)|\mathbf{E}|^2 = 2\,\mathrm{Im}\chi(\omega)|\mathbf{E}|^2 , \quad (104)$$

which is just the optical generation rate.

Hence, this subsection shows that in the quasiequilibrium limit we recover the results of the quasiequilibrium theory;[47,64] Eq. (74) for carrier kinetics reduces in this limit to the rate equation for carrier density[64] (without the recombination terms, however), while Eq. (79) for the polarization takes on the form of the BSE and allows us to determine linear susceptibility and hence all linear optical properties of a highly excited semiconductor.[64]

F. Interaction with phonons and other collective excitations

In this subsection we are going to derive the Bloch equations for the case when the carriers interact with a collective mode. To start with, let us consider a phonon mode with dispersion $\omega_{\mathbf{q}}$. The Hamiltonian of such a system is the sum of (10) and (45):

$$\hat{H}=\sum_{\mathbf{k}\alpha}\varepsilon_{a\mathbf{k}}a^\dagger_{a\mathbf{k}}a_{a\mathbf{k}}+\sum_{\mathbf{q}}\omega_{\mathbf{q}}b^\dagger_{\mathbf{q}}b_{\mathbf{q}} + \sum_{\mathbf{qk}\alpha} M_{\alpha\mathbf{q}}(b_{\mathbf{q}}+b_{-\mathbf{q}})a^\dagger_{a\mathbf{k}}a_{a\mathbf{k}} . \quad (105)$$

The phonons are described by their own Green's functions:

$$iD^{pq}_{\mathbf{q}}(tt')\equiv\begin{bmatrix}\langle \mathcal{T}b_{\mathbf{q}}(t)b^\dagger_{\mathbf{q}}(t')\rangle & \langle b^\dagger_{\mathbf{q}}(t')b_{\mathbf{q}}(t)\rangle \\ \langle b_{\mathbf{q}}(t)b^\dagger_{\mathbf{q}}(t')\rangle & \langle \tilde{\mathcal{T}}b_{\mathbf{q}}(t)b^\dagger_{\mathbf{q}}(t')\rangle\end{bmatrix}_{pq} . \quad (106)$$

In a noninteracting system the GF (106) is related to the phonon density $\mathcal{N}_{\mathbf{q}}$ in the following way:

$$iD^{pq}_{\mathbf{q}}(tt')=\begin{bmatrix}\mathcal{N}_{\mathbf{q}}(t)+\Theta(t-t') & \mathcal{N}_{\mathbf{q}}(t) \\ \mathcal{N}_{\mathbf{q}}(t)+1 & \mathcal{N}_{\mathbf{q}}+\Theta(t'-t)\end{bmatrix}_{pq} \times\exp[i\omega_{\mathbf{q}}(t'-t)] . \quad (107)$$

According to the above, we will use (107) as a phonon counterpart of *Ansatze* (34) further on. Note that with phonons it does not matter which time (t or t') is used as the argument of $\mathcal{N}_{\mathbf{q}}$ in (107), since it does not have any oscillating components.

The electron-phonon interaction [the last term in (105)] gives rise to an effective Fröhlich interaction between carriers:

$$V^{pq}_{\mathrm{eff}}(tt')=M^2_{\mathbf{q}}[\hat{\sigma}_z\cdot\tilde{D}(tt')\cdot\hat{\sigma}_z]_{pq} \quad (108)$$

(for simplicity we have assumed $M_{c\mathbf{q}}=M_{v\mathbf{q}}=M_{\mathbf{q}}$), with

$$\tilde{D}^{pq}_{\mathbf{q}}(tt')\equiv D^{pq}_{\mathbf{q}}(tt')+D^{qp}_{-\mathbf{q}}(t't) , \quad (109)$$

which enters the diagrams for the electron Green's func-

tions in the same way as the Coulomb interaction. Its physical origin is an exchange of phonons with the wave vector $\pm\mathbf{q}$ between carriers.

To obtain the Bloch equations we need to specify the self-energy; let us take the lowest-order diagram, which describes single-phonon processes and is obtained by replacing the screened potential in the screened Hartree-Fock self-energy of Fig. 3 with the Fröhlich interaction (108):

$$-i\Sigma^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')=-\sum_{\mathbf{q}} G^{pq}_{\alpha\beta_{\mathbf{k}-\mathbf{q}}}(tt')V^{pq}_{\text{eff}}(tt') \ . \tag{110}$$

Using *Ansätze* (34) for G and (107) for V_{eff}, we can easily evaluate the relaxation term of the Bloch equations with the self-energy (110):

$$\begin{aligned}\left[\frac{\partial N^{\alpha\beta}_{\mathbf{k}}}{\partial T}\right]_{St}=\sum_{\mathbf{q}\gamma} M^2_{\mathbf{q}}\{&\delta_-(\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})[(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}\mathcal{N}_{\mathbf{q}}-N^{\alpha\gamma}_{\mathbf{k}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}})(1+\mathcal{N}_{\mathbf{q}})]\\&+\delta_-(\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}+\omega_{-\mathbf{q}})[(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}(1+\mathcal{N}_{-\mathbf{q}})-N^{\alpha\gamma}_{\mathbf{k}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}})\mathcal{N}_{-\mathbf{q}}]\\&+\delta_+(\varepsilon_{\gamma k}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})[N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}})\mathcal{N}_{\mathbf{q}}-(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}})N^{\gamma\beta}_{\mathbf{k}}(1+\mathcal{N}_{\mathbf{q}})]\\&+\delta_+(\varepsilon_{\gamma\mathbf{k}}-\varepsilon_{\gamma\mathbf{k}-\mathbf{q}}+\omega_{-\mathbf{q}})[N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}})(1+\mathcal{N}_{\mathbf{q}})-(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}})N^{\gamma\beta}_{\mathbf{k}}\mathcal{N}_{\mathbf{q}}]\} \ .\end{aligned} \tag{111}$$

Performing the summation over γ, we obtain for $n_{c\mathbf{k}}$

$$\begin{aligned}\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{\text{ph}}=\sum_{\mathbf{q}} M^2_{\mathbf{q}}\Bigg[&2\pi\delta(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})[\mathcal{N}_{\mathbf{q}}n_{c\mathbf{k}-\mathbf{q}}(1-n_{c\mathbf{k}})-(1+\mathcal{N}_{\mathbf{q}})n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})]\\&+2\pi\delta(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})[(1+\mathcal{N}_{\mathbf{q}})n_{c\mathbf{k}-\mathbf{q}}(1-n_{c\mathbf{k}})-\mathcal{N}_{\mathbf{q}}n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})]\\&+(p^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}+p_{\mathbf{k}}p^*_{\mathbf{k}-\mathbf{q}})[\pi\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})-\pi\delta(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})]\\&+i(p^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}-p_{\mathbf{k}}p^*_{\mathbf{k}-\mathbf{q}})\frac{2\omega_{\mathbf{q}}}{(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})^2-\omega^2_{\mathbf{q}}}\Bigg]\end{aligned} \tag{112}$$

(here we have taken into account that $\omega_{\mathbf{q}}=\omega_{-\mathbf{q}}$). The first two terms in (112) correspond to the usual Boltzmann scattering, while the last two describe scattering on the phonon-assisted polarization wave. These latter two terms can be transformed further:

$$\begin{aligned}\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{p}&=\sum_{\mathbf{q}} M^2_{\mathbf{q}}\left[ip^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}\frac{2\omega_{\mathbf{q}}}{(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}+i\delta)^2-\omega^2_{\mathbf{q}}}-\text{c.c.}\right]\\&=-2\,\text{Im}\sum_{\mathbf{q}} p^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}V^R_{\text{eff}}(\omega=\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}) \ ,\end{aligned} \tag{113}$$

i.e., to the same form as with the Coulomb screened potential (74); it means, in particular, that (113) also has the particle-conserving property (75).

The equation for the polarization that follows from (111) can be cast into the form

$$\begin{aligned}\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_{\text{ph}}=&-ip_{\mathbf{k}}[\Sigma^R_{cc}(\varepsilon_{c\mathbf{k}})-\Sigma^A_{vv}(\varepsilon_{v\mathbf{k}})]\\&+in_{v\mathbf{k}}\sum_{\mathbf{q}} V^R_{\text{eff}}(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})p_{\mathbf{k}-\mathbf{q}}-in_{c\mathbf{k}}\sum_{\mathbf{q}} V^A_{\text{eff}}(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})p_{\mathbf{k}-\mathbf{q}}\\&+\sum_{\mathbf{q}} M^2_{\mathbf{q}}p_{\mathbf{k}-\mathbf{q}}\pi\{\mathcal{N}_{\mathbf{q}}[\delta_+(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})+\delta_-(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})]\\&+(1+\mathcal{N}_{\mathbf{q}})[\delta_+(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})+\delta_-(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})]\} \ ,\end{aligned} \tag{114}$$

which also closely resembles the corresponding result (79) for the case of the screened Coulomb interaction. In the two preceding equations, V^R_{eff} denotes the retarded Fröhlich interaction:

$$V^R_{\text{eff}}(\omega)=-V^{++}_{\text{eff}}-V^{+-}_{\text{eff}}=M^2_{\mathbf{q}}\frac{2\omega_{\mathbf{q}}}{(\omega+i\delta)^2-\omega^2_{\mathbf{q}}}=M^2_{\mathbf{q}}\tilde{D}^R_{\mathbf{q}}(\omega) \tag{115}$$

(i.e., it is the retarded phonon propagator multiplied by the coupling constant);[64] the energy renormalizations are given by

$$\Sigma^R_{cc}(\varepsilon_{c\mathbf{k}})=\sum_{\mathbf{q}} M^2_{\mathbf{q}}\left[\frac{1+\mathcal{N}_{\mathbf{q}}-n_{c\mathbf{k}-\mathbf{q}}}{\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}}+i\delta}+\frac{\mathcal{N}_{\mathbf{q}}+n_{c\mathbf{k}-\mathbf{q}}}{\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}}+i\delta}\right], \tag{116a}$$

$$\Sigma^A_{vv}(\varepsilon_{v\mathbf{k}})=\sum_{\mathbf{q}} M^2_{\mathbf{q}}\left[\frac{1+\mathcal{N}_{\mathbf{q}}-n_{v\mathbf{k}-\mathbf{q}}}{\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}}-i\delta}+\frac{\mathcal{N}_{\mathbf{q}}+n_{v\mathbf{k}-\mathbf{q}}}{\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}}-i\delta}\right]. \tag{116b}$$

In the quasiequilibrium limit, expressions (116) reduce to the results of Refs. 64 and 68 and give the polaron shift in lowest order; the poles in (116) correspond to phonon sidebands.[69]

The above expressions contain the phonon DF, $\mathcal{N}_{\mathbf{q}}$, which also has to be determined from an appropriate kinetic equation. This equation can be derived by the same argument that has brought us from the Dyson's equations (15) to the kinetic equation (32); the result reads

$$\left[\frac{\partial\mathcal{N}_{\mathbf{q}}}{\partial T}\right]=\int_{-\infty}^{T} d1[D^{++}_{\mathbf{q}}(T1)\Sigma^{++}_{\mathbf{q}}(1T)+D^{+-}_{\mathbf{q}}(T1)\Sigma^{-+}_{\mathbf{q}}(1T)-\Sigma^{++}_{\mathbf{q}}(T1)D^{++}_{\mathbf{q}}(1T)-\Sigma^{+-}_{\mathbf{q}}(T1)D^{-+}_{\mathbf{q}}(1T)], \tag{117}$$

which differs from the respective result (32) for the carriers only by the absence of the band indices in Σ and D.

To evaluate (117) we again have to specify the self-energy, this time the phonon one, $\Sigma_{\mathbf{q}}$. The phonon self-energy that corresponds to single-particle processes described by the carriers self-energy (110) reads

$$-i\Sigma^{pq}_{\mathbf{q}}(tt')=(M^2_{\mathbf{q}})^{pq}\sum_{\mathbf{k}\alpha\beta} G^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')G^{qp}_{\beta\alpha_{\mathbf{k}-\mathbf{q}}}(t't), \tag{118}$$

where

$$(M^2_{\mathbf{q}})^{pq}\equiv\hat{\sigma}_z\cdot M^2_{\mathbf{q}}\cdot\hat{\sigma}_z=M^2_{\mathbf{q}}\begin{bmatrix}1 & -1\\ -1 & 1\end{bmatrix}_{pq} \tag{119}$$

[cf. Eq. (109)].

The diagrammatic representation of (118), which describes changes in the phonon subsystem produced by the scattering of carriers from $\mathbf{k}$ to $\mathbf{k}-\mathbf{q}$, is just the RPA polarization bubble shown in Fig. 5 in the Appendix. Comparing (118) with (A7), we see that the phonon self-energy practically coincides with the polarization operator in the RPA:

$$\Sigma^{pq}_{\mathbf{q}}(tt')=\Pi^{pq}_{\mathbf{q}}(tt')(M^2_{\mathbf{q}})^{pq}, \tag{120}$$

so that we can use the results of the Appendix for Π to evaluate (118).

Expressing D with the help of (107), and Σ through (A8), we obtain for the collisional term (117)

$$\left[\frac{\partial\mathcal{N}_{\mathbf{q}}}{\partial T}\right]=M^2_{\mathbf{q}}\sum_{\mathbf{k}\alpha\beta}\pi\delta_+(\varepsilon_{\alpha\mathbf{k}}-\varepsilon_{\alpha\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})[(1+\mathcal{N}_{\mathbf{q}})N^{\alpha\beta}_{\mathbf{k}}(\delta_{\beta\alpha}-N^{\beta\alpha}_{\mathbf{k}-\mathbf{q}})-\mathcal{N}_{\mathbf{q}}(\delta_{\alpha\beta}-N^{\alpha\beta}_{\mathbf{k}})N^{\beta\alpha}_{\mathbf{k}-\mathbf{q}}]+\text{c.c.} \tag{121}$$

The diagonal (with $\alpha=\beta$) terms in (121) give rise to Boltzmann scattering:

$$\left[\frac{\partial\mathcal{N}_{\mathbf{q}}}{\partial T}\right]_{\text{Boltzmann}}=M^2_{\mathbf{q}}\sum_{\mathbf{k}}2\pi\delta(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})[(1+\mathcal{N}_{\mathbf{q}})n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})-\mathcal{N}_{c\mathbf{k}-\mathbf{q}}(1-n_{c\mathbf{k}})n_{c\mathbf{k}-\mathbf{q}}]+(c\rightarrow v), \tag{122a}$$

while the off-diagonal ones describe the influence of polarization scattering on the phonons:

$$\left[\frac{\partial\mathcal{N}_{\mathbf{q}}}{\partial T}\right]_p=-2\,\text{Im}\sum_{\mathbf{k}} M^2_{\mathbf{q}}p^*_{\mathbf{k}}\{p_{\mathbf{k}-\mathbf{q}}[D^R_{\mathbf{q}}(\omega=\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})+D^A_{\mathbf{q}}(\varepsilon_{v\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}})]\}, \tag{122b}$$

where

$$D^R_{\mathbf{q}}(\omega)=\frac{1}{\omega-\omega_{\mathbf{q}}+i\delta} \tag{123}$$

[according to (107)]; a comparison of (122b) with (113) shows that every time an electron or a hole scatters from $\mathbf{k}$ to $\mathbf{k}-\mathbf{q}$ on the polarization wave, one phonon is destroyed. Equations (112), (114), and (122) form a closed set, which can be used as a basis for the analysis of the role the phonons play in relaxation phenomena.

The similarity between the equations for the electron-phonon interaction and those for the screened Coulomb interaction is not incidental: screening, like phonons, is also a collective effect. To explore this analogy further, let us recall some facts of the plasma theory here.

The spectrum of the elementary excitations of the plasma is given by zeros of the retarded dielectric function, $\epsilon^R(\mathbf{q}\omega)$, which is defined by[65]

$$V^R_S(\mathbf{q}\omega)\equiv\frac{V(\mathbf{q})}{\epsilon^R(\mathbf{q}\omega)} \tag{124}$$

and consists of single-particle excitations and of collective

plasmon modes[65] [within the RPA, ϵ^R is given by the Lindhard formula (A12)]. For many practical purposes this spectrum can be replaced with a single plasmon mode [the single-plasmon-hole approximation (SPPA)],[64,65] so that the retarded screened potential takes on the form

$$V_S^R(\mathbf{q}\omega)\approx V(\mathbf{q})+V(\mathbf{q})\frac{\omega_{\rm pl}^2}{(\omega+i\delta)^2-\omega_{\mathbf{q}}^2}\ . \tag{125}$$

The SPPA replaces the rather formidable task of solving the basic equation (A2), which determines V_S, within the RPA (see the Appendix), by the much simpler problem of finding suitable plasmon dispersion $\omega_{\mathbf{q}}$. If the latter is chosen properly, the SPPA yields reliable results for self-energies and other quantities of interest in quasiequilibrium systems.[64,54]

In nonequilibrium situations, however, we need approximations not only for V_S^R, but also for all pq components of V_S. Let us show that the central concept of the SPPA—the existence of a single collective mode—can be carried over to the nonequilibrium case in a trivial way.

To start with, let us note that the standard SPPA [(125)] can be rewritten as

$$\begin{aligned}V_S^R(\mathbf{q}\omega)&=V(\mathbf{q})+V(\mathbf{q})\frac{\omega_{\rm pl}^2}{2\omega_{\mathbf{q}}}\left[\frac{2\omega_{\mathbf{q}}}{(\omega+i\delta)^2-\omega_{\mathbf{q}}^2}\right]\\&=V+W_{\mathbf{q}}^2\tilde{B}_{\mathbf{q}}^R(\omega)\ ,\end{aligned} \tag{126}$$

where

$$\tilde{B}_{\mathbf{q}}^R(\omega)=\frac{2\omega_{\mathbf{q}}}{(\omega+i\delta)^2-\omega_{\mathbf{q}}^2} \tag{127}$$

is the retarded plasmon propagator, and

$$W_{\mathbf{q}}^2\equiv V(\mathbf{q})\frac{\omega_{\rm pl}^2}{2\omega_{\mathbf{q}}} \tag{128}$$

is the electron-plasmon coupling constant. Comparing (126) with (115), we see that the retarded screened potential is expressed through the plasmon propagator in exactly the same way as the retarded Fröhlich interaction (115) is related to the phonon propagator (109). If we return to the time variables and introduce plasmon Green's functions B^{pq},

$$\begin{aligned}iB_{\mathbf{q}}^{pq}(tt')=&\begin{bmatrix}\mathcal{N}_{\mathbf{q}}^{\rm pl}(t)+\Theta(t-t') & \mathcal{N}_{\mathbf{q}}^{\rm pl}(t)\\ \mathcal{N}_{\mathbf{q}}^{\rm pl}(t)+1 & \mathcal{N}_{\mathbf{q}}^{\rm pl}+\Theta(t'-t)\end{bmatrix}_{pq}\\&\times\exp[i\omega_{\mathbf{q}}(t'-t)]\end{aligned} \tag{129}$$

[cf. (107), $\mathcal{N}_{\mathbf{q}}^{\rm pl}$ is the plasmon distribution function], through which the propagator (127) is expressed as

$$\tilde{B}_{\mathbf{q}}^{pq}(tt')=B_{\mathbf{q}}^{pq}(tt')+B_{-\mathbf{q}}^{qp}(t't) \tag{130}$$

[cf. (109)], we can extend the SPPA (126) to cover all the pq components of the screened potential:

$$V_S^{pq}(tt')=V^{pq}(\mathbf{q})+W_{\mathbf{q}}^2[\hat{\sigma}_z\cdot\tilde{B}(tt')\cdot\hat{\sigma}_z]_{pq} \tag{131}$$

[cf. (108)].

Taking the retarded component, $-(V_S^{++}+V_S^{+-})$, of (131), we recover the SPPA [(126)] and the Pauli matrices $\hat{\sigma}_z$ [(21)] ensure that all symmetries (A4) of the screened potential are kept intact. Equation (131) is the required nonequilibrium generalization of the SPPA in the sense that it reduces to the standard SPPA in quasiequilibrium, and self-consistently simulates nonequilibrium screening by means of (nonequilibrium) plasmon distributions $\mathcal{N}_q^{\rm pl}(t)$.

Approximation (131) allows us to reformulate the Bloch equations for the screened Coulomb interaction [(74) and (79)] in terms of plasmons. The complete analogy between definitions (129) and (130) and their phonon counterparts (107) and (109) ensures that within the "generalized SPPA" (131) the carrier kinetics and dephasing are governed by the "phonon" equations (112) and (114), respectively, with the phonon distribution function and coupling, $\mathcal{N}_{\mathbf{q}}$ and $M_{\mathbf{q}}$, being replaced by plasmon ones, $\mathcal{N}_{\mathbf{q}}^{\rm pl}$ and $W_{\mathbf{q}}$. With the same minor changes, Eq. (122) describes the kinetics of plasmons and, hence, via (131), the evolution of the screened potential; it serves as a substitute for the basic equation (A2) for the screened potential, which had to be solved together with (74) and (79) in the full RPA calculation scheme.

The approximation scheme for V_S based on (131) does the opposite to the SSA in the form (84): whereas the SSA neglects all dynamic effects but retains particle-particle collisions, Eq. (131), on the contrary, focuses on the dynamic effects at the expense of direct carrier-carrier scattering. It is interesting from a fundamental point of view, because it invokes such nontrivial concepts as nonequilibrium plasmons and electron-plasmon energy exchange.

From the vast numerical experience accumulated for the case when the dominant relaxation mechanism is the interaction of carriers with LO phonons,[34] we may be reasonably sure that if the plasmons are supposed to play the role of phonons, the basic physics will remain the same: at first, carriers created by the exciting pulse will relax their kinetic energy by emitting plasmons; this will lead to a buildup of the nonequilibrium population of plasmons ("hot-plasmon effect"), which will later give back their energy to carriers, slowing down their energy relaxation.

This physical picture is, however, likely to cause objections of the kind expressed by Collet,[67] who argues that the total plasma energy cannot change as a result of internal interactions, and therefore that the electron-plasmon interaction cannot lead to energy relaxation. Let us note, however, that the energy of plasmons usually is not included in the plasma energy, which is normally understood as the sum of kinetic energies of noninteracting electrons. The energy of plasmons is part of the Coulomb interaction energy, and there is no reason why there should be no energy exchange between single-particle and collective excitations of the plasma, i.e., between its kinetic and potential energy. There are some indications[66] that the energy loss to plasmons can be about an order of magnitude greater than that due to other interactions, so that it certainly would be interesting to explore the role of collective modes further, and the proposed procedure seems to be a convenient starting point

for doing that.

Let us finally note that the plasmon-phonon modes[64] can be included in our treatment in exactly the same way.

IV. BLOCH EQUATIONS WITH MEMORY EFFECTS

The relaxation terms in the Bloch equations that have been obtained in Sec. III contain only values of the DM at the same time instant T, and are therefore Markovian: the system's past does not affect its evolution. Physically it is equivalent to the semiclassic approximation, which holds true as long as the particles have well-defined energy, and changes of their distributions over this energy are slow on the time scale of $\hbar/\Delta\varepsilon$, where $\Delta\varepsilon$ is the characteristic energy exchange in a scattering event.[63]

This approximation, however, can be expected to fail whenever the duration of the excitation pulse (which at present can be as short as 10 fs)[43] becomes comparable to the inverse LO-phonon frequency (of the order of 10–100 fs). In such femtosecond excitation conditions one has to account for the fact that each optically created carrier is spread over a large portion of energy space due to large spectral width of the exciting pulse. In order to describe scattering of such carriers with ill-defined energy, non-Markovian description of relaxation is called for, and our formalism can be used to obtain the corresponding generalizations of the relaxation terms considered above.

A. Interaction with phonons

In order to incorporate memory effects in our description of electron-phonon scattering, we need to replace the relaxation term (111), which contains only $N^{\alpha\beta}(T)$, with an expression containing an integral over the system's past. Because the basic equation (32) does have such an integral, this goal proves to be relatively easily achieved: when using *Ansätze* (34) and (107) to evaluate (32), we should now select that of the two expressions (34) that contains $N^{\alpha\beta}(t_1)$. When the interaction is described by the self-energy (110), this prescription yields instead of (111)

$$\begin{aligned}\frac{\partial N^{\alpha\beta}_{\mathbf{k}}}{\partial T}=\sum_{\mathbf{q}} M_{\mathbf{q}}^2\int_{-\infty}^{T} d1\{&\exp[i(1-T)(\varepsilon_{\alpha\mathbf{k}}-\varepsilon_{\beta\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})][(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}\mathcal{N}_{\mathbf{q}}-N^{\alpha\gamma}_{\mathbf{k}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}})(1+\mathcal{N}_{\mathbf{q}})]\\&+\exp[i(1-T)(\varepsilon_{\alpha\mathbf{k}}-\varepsilon_{\beta\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})][(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}})N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}}(1+\mathcal{N}_{\mathbf{q}})-N^{\alpha,\gamma}_{\mathbf{k}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}-\mathbf{q}})\mathcal{N}_{\mathbf{q}}]\\&+\exp[i(1-T)(\varepsilon_{\alpha\mathbf{k}-\mathbf{q}}-\varepsilon_{\beta\mathbf{k}}+\omega_{\mathbf{q}})][N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}})\mathcal{N}_{\mathbf{q}}-(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}})N^{\gamma\beta}_{\mathbf{k}}(1+\mathcal{N}_{\mathbf{q}})]\\&+\exp[i(1-T)(\varepsilon_{\alpha\mathbf{k}-\mathbf{q}}-\varepsilon_{\beta\mathbf{k}}-\omega_{\mathbf{q}})][N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}}(\delta_{\gamma\beta}-N^{\gamma\beta}_{\mathbf{k}})(1+\mathcal{N}_{\mathbf{q}})-(\delta_{\alpha\gamma}-N^{\alpha\gamma}_{\mathbf{k}-\mathbf{q}})N^{\gamma\beta}_{\mathbf{k}}\mathcal{N}_{\mathbf{q}}]\}\ ,\end{aligned}\tag{132}$$

where all N and $\mathcal{N}$ are understood as functions of the integration variable, t_1. It can be easily shown by inserting the steady-state temporal dependence of N [(12)] into (132) that in a steady state, (132) reduces to the result (111) and is therefore its non-Markovian generalization. Introducing the notations

$$\varepsilon_{\alpha}^{+}\equiv\varepsilon_{\alpha\mathbf{k}}-\varepsilon_{\alpha\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}},\quad \varepsilon_{\alpha}^{-}\equiv\varepsilon_{\alpha\mathbf{k}}-\varepsilon_{\alpha\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}}\ ,\quad \tau=T-1\ ,\tag{133}$$

we can extract the equation for $n_{c\mathbf{k}}$ from (132):

$$\begin{aligned}\frac{\partial n_{c\mathbf{k}}}{\partial T}=\sum_{\mathbf{q}} M_{\mathbf{q}}^2\int_{-\infty}^{T} d1\{&-n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})[(1+\mathcal{N}_{\mathbf{q}})e^{-i\tau\varepsilon_c^-}+\mathcal{N}_{\mathbf{q}}e^{-i\tau\varepsilon_c^+}+(1+\mathcal{N}_{\mathbf{q}})e^{i\tau\varepsilon_c^-}+\mathcal{N}_{q}e^{i\tau\varepsilon_c^+}]\\&+(1-n_{c\mathbf{k}})n_{c\mathbf{k}-\mathbf{q}}[(1+\mathcal{N}_{\mathbf{q}})e^{-i\tau\varepsilon_c^+}+\mathcal{N}_{\mathbf{q}}e^{-i\tau\varepsilon_c^-}+(1+\mathcal{N}_{\mathbf{q}})e^{i\tau\varepsilon_c^+}+\mathcal{N}_{\mathbf{q}}e^{i\tau\varepsilon_c^-}]\\&+p_{\mathbf{k}}p^{*}_{\mathbf{k}-\mathbf{q}}(e^{-i\tau\varepsilon_c^-}-e^{-i\tau\varepsilon_c^+})+p^{*}_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}(e^{+i\tau\varepsilon_c^-}-e^{+i\tau\varepsilon_c^+})\}\ ,\end{aligned}\tag{134}$$

which can be further transformed to the form

$$\begin{aligned}\frac{\partial n_{c\mathbf{k}}}{\partial T}=\sum_{\mathbf{q}} M_{\mathbf{q}}^2\int_{-\infty}^{T} d1\{&-n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})[\mathcal{N}_{\mathbf{q}}2\cos(\tau\varepsilon_c^+)+(1+\mathcal{N}_{\mathbf{q}})2\cos(\tau\varepsilon_c^-)]\\&+(1-n_{c\mathbf{k}})n_{c\mathbf{k}-\mathbf{q}}[(1+\mathcal{N}_{\mathbf{q}})2\cos(\tau\varepsilon_c^+)+\mathcal{N}_{\mathbf{q}}2\cos(\tau\varepsilon_c^-)]\\&+i(p_{\mathbf{k}}p^{*}_{\mathbf{k}-\mathbf{q}}e^{-i\tau(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})}-p^{*}_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}e^{+i\tau(\varepsilon_{c\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}})})2\sin(\tau\omega_{\mathbf{q}})\}\ .\end{aligned}\tag{135}$$

In the Markovian case, when n and $\mathcal{N}$ in (135) do not depend on τ, the integration in (135) picks out of the $\mathbf{q}$ sum only those components for which the integrands do not oscillate at all, which assures energy conservation. When the occupation numbers do depend on time, in general all terms in the sum over $\mathbf{q}$ space begin to contribute to the scattering rate. For example, if the carriers are created by an optical pulse with the duration τ_p, then the integration in (135) will conserve the energy only with the accuracy $\hbar/\tau_p$, so that the carriers can scatter to a broader region of $\mathbf{k}$ space out of a given state, with correspondingly smaller amplitude.

In order to clarify the impact of the memory effects, let us consider the weak-excitation limit: $n\ll 1$, $\mathcal{N}_{\mathbf{q}}(t)=\text{const}$.

In this case the rate of scattering out of a given $\mathbf{k}$ state can be cast into the form

$$\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{\text{out}} = -\int_{-\infty}^{T} d1\, n_{c\mathbf{k}}(1) \sum_{\mathbf{q}} M_{\mathbf{q}}^2 [2\mathcal{N}_{\mathbf{q}} \cos(\tau\varepsilon_c^+) + 2(1+\mathcal{N}_{\mathbf{q}})\cos(\tau\varepsilon_c^-)]$$
$$= -\int_{-\infty}^{T} dt' n_{c\mathbf{k}}(t') \sigma_{c\mathbf{k}}^{\text{out}}(T-t') , \tag{136}$$

where the kernel

$$\sigma_{c\mathbf{k}}^{\text{out}}(\tau) = \sum_{\mathbf{q}} M_{\mathbf{q}}^2 [\mathcal{N}_{\mathbf{q}}(e^{i\tau\varepsilon_c^+} + e^{-i\tau\varepsilon_c^+}) + (1+\mathcal{N}_{\mathbf{q}})(e^{i\tau\varepsilon_c^-} + e^{-i\tau\varepsilon_c^-})] \tag{137}$$

describes the response of the semiconductor. At $\tau \to 0$ it diverges, since

$$\sigma_{c\mathbf{k}}^{\text{out}}(\tau=0) = 2(1+2\mathcal{N}_{\mathbf{q}}) \sum_{\mathbf{q}} M_{\mathbf{q}}^2 \propto V_{\text{eff}}(r=0) = \infty \tag{138}$$

(cf. Ref. 28), and tends to zero when $\tau \to \infty$. Since (136) has the form of a convolution, it is advantageous to make the Fourier transform

$$\left[\frac{\partial n_{c\mathbf{k}}}{\partial T}\right]_{\text{out}} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega\, n_{c\mathbf{k}}^r(\omega) \sigma_{c\mathbf{k}}^{\text{out}}(\omega) e^{-i\omega t} , \tag{139}$$

where the retarded Fourirer transform of the density is defined as

$$n^r(\omega) = \int_{-\infty}^{t} n(t') e^{i\omega t'} dt' \tag{140}$$

and the Fourier transform $\sigma(\omega)$ can be evaluated explicitly. For dispersionless polar-optical phonons with[64]

$$M_{\mathbf{q}}^2 = \frac{C}{\mathbf{q}^2} \tag{141}$$

we have, e.g., for the first term of (137)

$$(\sigma_{c\mathbf{k}}^{\text{out}}(\omega))_{\text{I}} = \int_{-\infty}^{+\infty} d\tau\, e^{i\omega\tau} \left[\sum_{\mathbf{q}} M_{\mathbf{q}}^2 \mathcal{N}_{\mathbf{q}} \exp[i\tau(\varepsilon_{c\mathbf{k}} - \varepsilon_{c\mathbf{k}-\mathbf{q}} + \omega_{\mathbf{q}})]\right]$$
$$= 2\pi \sum_{\mathbf{q}} M_{\mathbf{q}}^2 \mathcal{N}_{\mathbf{q}} \delta(\varepsilon_{c\mathbf{k}} - \varepsilon_{c\mathbf{k}-\mathbf{q}} + \omega_{\mathbf{q}} + \omega) = C_{c\mathbf{k}} \mathcal{N}_{\mathbf{q}} g_{\text{LO}}\left[\frac{\omega_{\mathbf{q}}+\omega}{\varepsilon_{c\mathbf{k}}}\right] , \tag{142}$$

where

$$g_{\text{LO}}(x) \equiv \ln\left[\frac{1+\sqrt{1-x}}{|1-\sqrt{1-x}|}\right] \tag{143}$$

and

$$C_{c\mathbf{k}} = \frac{Cm_c}{\pi|\mathbf{k}|} . \tag{144}$$

Carrying out such a summation over $\mathbf{q}$ in the remaining terms of (137), we arrive at

$$\sigma_{c\mathbf{k}}^{\text{out}}(\omega) = C_{c\mathbf{k}} \left\{(1+\mathcal{N}_{\mathbf{q}})\left[g\left[\frac{\omega_{\mathbf{q}}-\omega}{\varepsilon_{c\mathbf{k}}}\right] + g\left[\frac{\omega_{\mathbf{q}}+\omega}{\varepsilon_{c\mathbf{k}}}\right]\right] + \mathcal{N}_{\mathbf{q}}\left[g\left[\frac{-\omega_{\mathbf{q}}-\omega}{\varepsilon_{c\mathbf{k}}}\right] + g\left[\frac{\omega-\omega_{\mathbf{q}}}{\varepsilon_{c\mathbf{k}}}\right]\right]\right\} . \tag{145}$$

The response function $\sigma(\omega)$ is shown schematically in Fig. 4. For pulsed excitation the real part of the retarded Fourier transform $n^r(\omega)$ of the density has the form of a peak centered around $\omega=0$ with width of the order of $\hbar/\tau_p$ (see Fig. 4). Hence, for very long pulses the scattering rate (139) is simply $n(t)\sigma(\omega=0)$ (Markovian result), but when $(\tau_p\omega_{\mathbf{q}})^{-1}$ becomes comparable to unity, all Fourier components of σ begin to contribute to the scattering rate (139), and since $\sigma(\omega)$ is not always a flat function, the resulting scattering rate can significantly differ from its Markovian value (the results of a model evaluation of scattering rates will be presented elsewhere[70]).

These considerations show that to achieve the correct description of photoexcited carriers relaxation it is not enough to just broaden the initial distribution $n(\varepsilon)$ by the amount of $\hbar/\tau_p$ and then solve a Markovian Boltzmann equation,[37–41] because such broadening occurs in every

scattering event, and that makes carriers spread over $\mathbf{k}$ space more rapidly, and thus speeds up thermalization.

The last line of (135), which represents polarization scattering, also reduces to the respective Markovian result (122b) only provided $p_{\mathbf{k}} \propto \exp[i(\varepsilon_{v\mathbf{k}}-\varepsilon_{c\mathbf{k}})t]$ [see Eq. (12)], i.e., in a steady state. In the low-excitation limit this term can be dealt with along similar lines.[70] Here we will only note that if, as is the case under femtosecond excitation, the combination $|p^*_{\mathbf{k}}p_{\mathbf{k}-\mathbf{q}}|$ is nonzero only at very short times before the present moment T, i.e., at $\tau<\tau_p\leq\omega_{\mathbf{q}}^{-1}$, when $\sin(\tau\omega_{\mathbf{q}})$ is still small, the contribution of the polarization terms will be significantly lower than its Markovian value (120b), and tends to zero at $\tau_p\to 0$.

From (132) we can also extract the equation for the polarization:

$$\frac{\partial p_{\mathbf{k}}}{\partial T}=\int_{-\infty}^{T}d1\left[-p_{\mathbf{k}}(1)\left\{\sum_{\mathbf{q}}M_{\mathbf{q}}^2\{[(1+\mathcal{N}_{\mathbf{q}}-n_{v\mathbf{k}-\mathbf{q}})\exp[-i\tau(\varepsilon_{c\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})]+(\mathcal{N}_{\mathbf{q}}+n_{v\mathbf{k}-\mathbf{q}})\exp[-i\tau(\varepsilon_{c\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})]\right.\right.\right.$$
$$\left.\left.\left.+(1+\mathcal{N}_{\mathbf{q}}-n_{c\mathbf{k}-\mathbf{q}})\exp[i\tau(\varepsilon_{v\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})]+(\mathcal{N}_{\mathbf{q}}+n_{c\mathbf{k}-\mathbf{q}})\exp[i\tau(\varepsilon_{v\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})]\}\right\}\right] \quad (146a)$$

$$+\sum_{\mathbf{q}}M_{\mathbf{q}}^2p_{\mathbf{k}-\mathbf{q}}(1)\{[(1+\mathcal{N}_{\mathbf{q}}-n_{c\mathbf{k}})\exp[-i\tau(\varepsilon_{c\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})]+(\mathcal{N}_{\mathbf{q}}+n_{c\mathbf{k}})\exp[-i\tau(\varepsilon_{c\mathbf{k}}-\varepsilon_{v\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})]$$
$$+(1+\mathcal{N}_{\mathbf{q}}-n_{v\mathbf{k}})\exp[i\tau(\varepsilon_{v\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}+\omega_{\mathbf{q}})]+(\mathcal{N}_{\mathbf{q}}+n_{c\mathbf{k}})\exp[i\tau(\varepsilon_{v\mathbf{k}}-\varepsilon_{c\mathbf{k}-\mathbf{q}}-\omega_{\mathbf{q}})]\}\Big]$$
$$\equiv\int_{-\infty}^{T}dt'\left[p_{\mathbf{k}}(t')\sigma_{\mathbf{k}}^{\mathrm{I}}(T-t')+\sum_{\mathbf{q}}p_{\mathbf{k}-\mathbf{q}}(t')\sigma_{\mathbf{k}}^{\mathrm{II}}(T-t')\right], \quad (146b)$$

where the term (146a) describes transition energy renormalization and dephasing proper, while the term (146b) accounts for the diffusion of the polarization over $\mathbf{k}$ space.

In a steady state, when $p_{\mathbf{k}}$ oscillates with time according to (12), the integration in (146) turns the exponents into the corresponding energy denominators that enter the Markovan expressions (114)–(116). In the general case, however, Eqs. (146) show that each Fourier component of $p_{\mathbf{k}}$ has its own shift and broadening:

$$\left[\frac{\partial p_{\mathbf{k}}}{\partial T}\right]_{\mathrm{I}}=-\frac{i}{2\pi}\int_{-\infty}^{+\infty}d\omega\,p_{\mathbf{k}}(\omega)i\sigma_{\mathbf{k}}^{\mathrm{I}}(\omega)\,, \quad (147)$$

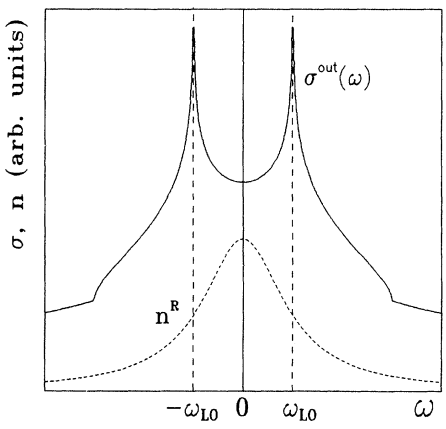

FIG. 4. Fourier-transformed response function (145) and the retarded density $n^r(\omega)$ [(140)] (schematically).

which, in particular, may be considered as a justification for the use of different damping constants for the pump and the probe[26] provided they have different frequencies. Equations (146) can be seen as a generalization of the result of Ref. 28 for the case when not only $|p|$ but also $\mathcal{N}$ and n change rapidly with time.

Under femtosecond excitation, when the pulse excites polarization in a broad region of $\mathbf{k}$ space, the second, diffusion term of (146) [(146b)], which is usually omitted,[27,28] starts to play an important role, too. In contrast to the few-level systems where the polarization cannot diffuse effectively through energy gaps between the levels, in semiconductors the excited $\mathbf{k}$ states form a continuum, and this diffusion term turns out to be as important[70] as the direct dephasing term (146a).

B. Other interactions

The procedure of obtaining non-Markovian expressions for relaxation terms, which has been discussed in the preceding subsections, is quite general and can be applied in a straightforward way to all sets of Bloch equations derived above (we do not present the resulting non-Markovian equations here because they are all very much alike).

It must be noted, however, that a fundamental difficulty arises when this procedure is applied to the case of the screened Coulomb interaction. There is no problem in writing down the non-Markovian generalizations of Eqs. (74) and (79), but there is the vexing problem of how to determine the time-dependent screened potential $V_S(tt')$ in these equations. The procedure described in

the Appendix relies substantially on the assumption of stationarity, which is not likely to hold true when memory effects become important. In order to be consistent, in the non-Markovian case we should solve the basic integral equation (A2) for the screened potential directly in the time domain, which is an almost unaccomplishable feat (although some encouraging attempts in this direction have been reported).[71]

In short, we are faced here with the fundamental problem of screening in nonequilibrium and nonstationary situations,[72,73] which is definitely still far from being resolved. Our approach is too simplistic to overcome this difficulty, which apparently needs a conceptual breakthrough.

In the meantime we suggest using the idea of the electron-plasmon interaction as a substitute for the full nonstationary solution for V_S. However, unlike the case of phonons, the plasmon spectrum is likely to experience dramatic changes during femtosecond excitation due to the rapid growth of the number of carriers, which makes the *Ansatz* (129) for plasmons not very convincing. When such changes are not very pronounced (as may be the case with modulation-doped quantum wells,[37] where there is a high density of carriers prior to excitation), this procedure may work reasonably well, but in the general case it is not even clear whether the very concept of plasmon oscillations still makes sense on a femtosecond time scale (the results of Ref. 71 suggest that it probably does).

V. CONCLUSIONS

In the present paper we have used the Keldysh GF formalism in order to derive quantum generalizations of the semiclassic equations that are currently used to describe coherent and relaxation phenomena in semiconductors. Our procedure is based on the Kadanoff-Baym-like *Ansatz* in the time domain, which makes the derivation of non-Markovian kinetic equations easy and transparent. This procedure can be improved in a straightforward way to include collisional broadening, which has not been done here mainly to avoid confusing this effect with the one we wanted to emphasize in this paper.

We were focusing on two quantum effects that emerge under conditions of femtosecond optical excitation. The first of them is due to the presence of a nonzero interband polarization, which provides carriers with an additional scattering channel. This polarization scattering is nontrivial in a number of ways and, at least in some intermediate range of excitation durations, is as important as the traditional Boltzmann scattering.

The second effect is due to the energy uncertainty of carriers created by a very short light pulse, which makes them less constrained in choosing final states in scattering events. Note that this effect is different from the collisional broadening,[58,59] because it persists even in the limit of vanishing interaction, although both effects are essentially due to the same time-energy uncertainty principle. The role of such memory effects in the optical polarization is acknowledged at present;[27–30] here we wished to stress that they are of importance for the carrier kinetics as well.

A model numerical example[70] shows that both effects are not only of academic interest; they also significantly enhance the carriers' ability to spread over $\mathbf{k}$ space, which leads to much shorter thermalization times. It would be interesting to attempt a more-realistic calculation of carrier dynamics with these effects included, although this will apparently involve tremendous numerical difficulties.

Finally, let us note that another interesting field where these results can be applied is that of the dynamics of many-body renormalizations, where some conceptual difficulties are now apparent.[37] We intend to address this problem in our future work.

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APPENDIX: SCREENING WITHIN THE KELDYSH FORMALISM

The concept of the screened interaction takes into account an important class of many-body processes in Coulomb systems. Formally, it is introduced by summing up all diagrams that begin and end by a bare-interaction line, which gives for the screened potential V_S an equation that is similar to Dyson's equation (15):

$$V_S^{pq}(\mathbf{q},t_1,t_2)=V^{pq}(\mathbf{q})\delta(t_1t_2)+\sum_{p_q q_1}\int_{-\infty}^{+\infty}d3\,d4\,V^{pq}(\mathbf{q})\delta(13)\times\Pi_{\mathbf{q}}^{p_1q_1}(34)V_S^{q_1q}(\mathbf{q},42) \quad \text{(A1)}$$

(see Fig. 5 for its diagrammatic representation). Using the explicit form of unscreened interaction V^{pq} [(48)], and suppressing the $\mathbf{q}$ dependence further on, we can rewrite (A1) as

$$V_S^{pq}(tt')=V^{pq}\delta(tt')+\sum_{q_1}\int_{-\infty}^{+\infty}d1\,V^{pp}\Pi^{pq_1}(t1)V_S^{q_1q}(1t')\ . \quad \text{(A2)}$$

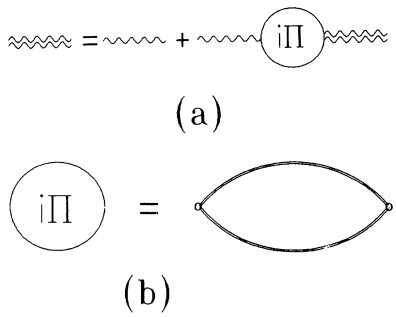

FIG. 5. (a) Diagrammatic representation of the basic equation (A2) for the screened potential (double wavy line), and (b) the random-phase approximation (RPA) for the polarization operator (A7).

Here, Π is the polarization operator (the sum of all irreducible polarization diagrams).[65]

The quantities V_S^{pq} and Π^{pq} possess symmetry properties similar to those of G[(5)–(8)] and Σ[(16)–(18)]:

$$\Pi^{++}+\Pi^{--}=\Pi^{+-}+\Pi^{-+}\ , \tag{A3a}$$

$$\Pi^{++}(tt')=\begin{cases}\Pi^{+-}, & t<t'\\ \Pi^{-+}, & t>t'\ ,\end{cases} \tag{A3b}$$

$$\Pi^{R}(tt')=\Pi^{+-}-\Pi^{++}=0 \text{ at } t<t\ , \tag{A3c}$$

$$V_S^{++}+V_S^{--}=-V_S^{+-}-V_S^{-+}\ , \tag{A4a}$$

$$V_S^{++}(tt')=\begin{cases}-V\delta(tt')-V_S^{+-}, & t<t'\\ -V\delta(tt')-V_S^{-+}, & t>t'\ ,\end{cases} \tag{A4b}$$

$$V_S^{R}=-V_S^{++}-V_S^{+-},\quad V_S^{A}=-V_S^{++}-V_S^{-+}\ . \tag{A4c}$$

One of the consequences of (A3) and (A4) is that the integrand in (A2) becomes zero at $t_1>\max(t,t')$, so that (A2) does not break causality.

In a steady state, Π and V_S depend only on the difference of their time arguments, so that one can Fourier transform (A2):

$$V_S^{pq}(\omega)=V^{pq}+\sum V^{pp}\Pi^{pq_1}(\omega)V^{q_1q}(\omega)\ . \tag{A5}$$

Equation (A5) is in fact a set of four algebraic equations for pq components of V_S, which can be solved in a straightforward way:

$$V_S^{++}(\omega)=-\frac{V[1-V\Pi^{--}(\omega)]}{(1-V\Pi^{R})(1-V\Pi^{A})}\ , \tag{A6a}$$

$$V_s^{+-}(\omega)=-\frac{V^2\Pi^{+-}(\omega)}{(1-V\Pi^{R})(1-V\Pi^{A})}\ , \tag{A6b}$$

$$V_S^{\mp}(\omega)=-\frac{V^2\Pi^{-+}(\omega)}{(1-V\Pi^{R})(1-V\Pi^{A})}\ , \tag{A6c}$$

$$V_S^{--}(\omega)=-\frac{V[1+V\Pi^{++}(\omega)]}{(1-V\Pi^{R})(1-V\Pi^{A})}\ , \tag{A6d}$$

$$V_S^{R}(\omega)=\frac{V}{(1-\Pi^{R})},\quad V_S^{A}(\omega)=\frac{V}{(1-V\Pi^{A})}\ . \tag{A6e}$$

Equation (A6) links the pq components of the screened potential to the Fourier transform of the polarization operator Π. The most widely used approximation for Π is the well-known random-phase approximation (RPA), which replaces Π with the polarization bubble shown in Fig. 5:

$$-i\Pi_{\mathbf{q}}^{pq}(tt')=-\sum_{\alpha\beta\mathbf{k}} iG^{pq}_{\alpha\beta_{\mathbf{k}}}(tt')iG^{qp}_{\beta\alpha_{\mathbf{k}-\mathbf{q}}}(t't)\ . \tag{A7}$$

Using *Ansätze* (34), we can readily express (A7) through the density matrix

$$\Pi^{--}(T1)=\Pi^{+-}(T1)=i\sum_{\alpha\beta_{\mathbf{k}}} N_{\mathbf{k}}^{\alpha\beta}(\delta_{\alpha\beta}-N_{\mathbf{k}-\mathbf{q}}^{\beta\alpha})\exp[i(1-T)(\varepsilon_{\beta\mathbf{k}}-\varepsilon_{\beta\mathbf{k}-\mathbf{q}})]\quad (T>1) \tag{A8}$$

(other pq components of Π can be dealt with in exactly the same way). Expressions like (A8) allow us to determine Laplace transforms:

$$\begin{aligned}\Pi_R^{pq}(\omega)&\equiv\int_0^{+\infty}d\tau\,\Pi^{pq}(\tau=T-1)e^{i\omega\tau}\ ,\\ \Pi_A^{pq}(\omega)&\equiv\int_{-\infty}^{0}d\tau\,\Pi^{pq}(\tau=T-1)e^{i\omega\tau}\ ,\end{aligned} \tag{A9}$$

which read, according to (A8),

$$\Pi_R^{-+}=\sum_{\mathbf{k}}\left[\frac{p_{\mathbf{k}}^{*}p_{\mathbf{k}-\mathbf{q}}-n_{c\mathbf{k}-\mathbf{q}}(1-n_{c\mathbf{k}})}{\omega+i\delta-\varepsilon_{c\mathbf{k}}+\varepsilon_{c\mathbf{k}+\mathbf{q}}}+\frac{p_{\mathbf{k}}^{*}p_{\mathbf{k}-\mathbf{q}}-n_{v\mathbf{k}-\mathbf{q}}(1-n_{v\mathbf{k}})}{\omega+i\delta-\varepsilon_{v\mathbf{k}}+\varepsilon_{v\mathbf{k}-\mathbf{q}}}\right], \tag{A10a}$$

$$\Pi_R^{+-}=\sum_{\mathbf{k}}\left[\frac{p_{\mathbf{k}}^{*}p_{\mathbf{k}-\mathbf{q}}-n_{c\mathbf{k}}(1-n_{c\mathbf{k}-\mathbf{q}})}{\omega+i\delta-\varepsilon_{c\mathbf{k}}+\varepsilon_{c\mathbf{k}-\mathbf{q}}}+\frac{p_{\mathbf{k}}^{*}p_{\mathbf{k}-\mathbf{q}}-n_{v\mathbf{k}}(1-n_{v\mathbf{k}-\mathbf{q}})}{\omega+i\delta-\varepsilon_{v\mathbf{k}}+\varepsilon_{v\mathbf{k}-\mathbf{q}}}\right]. \tag{A10b}$$

These quantities allow us to express the Fourier transforms of Π that enter (A6):

$$\begin{aligned}&\Pi^{+-}(\omega)=\Pi_R^{+-}+\Pi_A^{+-},\quad \Pi^{-+}(\omega)=\Pi_R^{-+}+\Pi_A^{-+}\ ,\\ &\Pi^{++}(\omega)=\Pi_R^{-+}+\Pi_A^{+-},\quad \Pi^{--}(\omega)=\Pi_R^{+-}+\Pi_A^{-+}\ ,\end{aligned} \tag{A11}$$

where $\Pi_A^{pq}=(\Pi_R^{pq})^*$. Let us finally note that, in contrast to (A10), the retarded polarization operator

$$\Pi^{R}(\omega)=\sum_{\mathbf{k}\alpha}\frac{n_{\alpha\mathbf{k}-\mathbf{q}}-n_{\alpha\mathbf{k}}}{\omega+i\delta-\varepsilon_{\alpha\mathbf{k}}+\varepsilon_{\alpha\mathbf{k}-\mathbf{q}}} \tag{A12}$$

does not contain optical polarization terms $p_{\mathbf{k}}^{*}p_{\mathbf{k}-\mathbf{q}}$: the presence of nonzero optical polarization affects all the pq components of V_S and Π, but the corresponding retarded functions are still given by Lindhard formula (A12).[64]

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