Role of exchange interaction in Coulomb quantum kinetics

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(Received 29 August 2002; published 13 March 2003)

In a quantum-kinetic approach based on density-matrix formulation we study the role of exchange interaction for ultrafast carrier relaxation. We find that the exchange contributions to scattering and screening tend to compensate each other. With very short times, however, due to the retarded buildup of the screening the exchange contribution to scattering prevails resulting in a reduced relaxation.

DOI: 10.1103/PhysRevB.67.115311 PACS number(s): 71.10.−w, 71.45.Gm, 73.21.Fg, 78.47.+p

One of the most fundamental properties of quantum mechanics is the indistinguishability of particles which gives rise to the existence of two different types of particles: Bosons with totally symmetric and fermions with totally antisymmetric wave functions. For charged particles interacting via the Coulomb interaction, as a result of the symmetry or antisymmetry, the direct Coulomb interaction is complemented by an additional term, the exchange interaction. The exchange interaction often plays a dominant role for the existence of bound states, but also modifies scattering processes between identical particles because the indistinguishability in the final states gives rise to an interference between direct and exchange terms in the scattering rate.

The quantum kinetics of a gas of charged particles on ultrashort-time scales has been a field of very active research in past years first because the description of modern optical experiments using pulses in the range of a few to a few tens of femtoseconds requires a detailed modeling of the carrier dynamics and second because here fundamental phenomena such as memory effects, the energy-time uncertainty, or the buildup of screening can be studied. Indeed it has been found that at very early times the dynamics is well described by scattering processes via an unscreened potential[1,2] while with increasing time the screening is built up[3–5] resulting in a reduced scattering efficiency. This buildup of the screening has been studied in sophisticated quantum-kinetic calculations based on fully time-dependent random-phase approximation (RPA) dielectric functions[4] and good agreement with experiments in the ultrafast regime has been obtained.[6,7] In these calculations, however, which are mostly based on a nonequlibrium Green's-function approach, exchange effects have been neglected because they correspond to a different class of diagrams beyond RPA.

An alternative approach that has proven to be particularly useful for the description of quantum-kinetic phenomena on ultrashort-time scales is density-matrix formalism. It has been extensively applied to electron-phonon quantum kinetics[8–11] as well as to Coulomb quantum kinetics in the low-density regime.[12–16] It is well known already since the early 1960s that also here, if two-particle correlations are included, the full dynamical RPA screening of the scattering rates is obtained[17–20] as on the corresponding level of Green's-function theory. Besides Green's-function and density-matrix theories there is a variety of other approaches which have been developed and used in the past for the study of different aspects of the nonequilibrium dynamics in semiconductors. Among these are projection operator techniques,[21–26] quantum-kinetic Langevin equations,[27–29] path-integral formulations,[30] and time-dependent density-functional theory.[31–33] In principle, all these approaches provide exact descriptions of the quantum dynamics and, therefore, they are equivalent as long as no further approximations are applied. However, such exact treatments are not possible for typical many-body systems and approximations have to be made. Different approaches, in general, suggest different approximation strategies. Examples for such strategies are the selection of a certain class of diagrams within Green's-function theory, correlations up to a certain number of particles in the density-matrix approach, or the selection of the ''relevant'' subspace in a projection operator formalism. An approximation level, which in one approach is ''natural'' within the strategy o the respective approach, does not necessarily coincide with an equally natural approximation level in another approach. It is therefore not a trivial finding that the RPA dynamics appears as a natural level of approximation within different approaches such as Green's-function and density-matrix theories.

In recent years several authors have addressed the treatment of screening within density-matrix formalism,[8,34–36] however, it has scarcely been applied to numerical investigations. Interestingly, in this approach the exchange contributions appear in a natural way on the same level as the direct contributions. In fact, it is obvious from the equations that they are necessary to obtain two-particle density matrices that fulfill the correct Fermi antisymmetry. Exchange terms modify both the scattering and the screening contributions to the dynamics of two-particle correlations and it is the interplay between these two phenomena which is the focus of our investigations. Exchange contributions to the carrier relaxation have been studied in terms of semiclassical scattering rates.[37–39] This approach, however, does not hold at ultrashort times where quantum-kinetic effects are important. Exchange contributions have also been included in molecular-dynamics-type simulations of carrier relaxation.[40,41] This, however, required an *ad hoc* introduction of wave packets with properties that cannot be strictly derived from the quantum-mechanical equations of motion.

In this paper we study the role of exchange contributions for the ultrafast relaxation of a two-dimensional electron gas due to Coulomb interaction processes based on density-matrix formalism. By selectively switching on and off the contributions corresponding to RPA screening, exchange scattering, and exchange screening we can clearly identify the role of these contributions for the relaxation dynamics.

We find that, as expected for fermion systems, exchange terms both reduce the scattering and the screening efficiency because of their opposite sign with respect to the direct contributions. On ultrafast time scales, however, they exhibit a remarkably different dynamical behavior. The scattering terms are efficient already from the beginning while screening takes some time to be built up. Consequently, on very short-time scales the relaxation is reduced with respect to calculations, taking into account only direct-scattering terms. With increasing time this reduction is strongly compensated by the reduced screening efficiency resulting finally in a behavior similar to the RPA dynamics where the exchange contributions for both phenomena are neglected.

An example for a physical situation where the relaxation and screening dynamics studied in the present paper is expected to play a prominent role is the case of an intrinsic semiconductor quantum well excited by an ultrashort intense optical pulse. In this case a plasma is generated in the conduction as well as in the valance band on such a short-time scale that initially the generated particles interact by means of an unscreend Coulomb potential because there was not yet enough time for the carriers to rearrange and provide the screening. However, in a real experiment of the above type the effects of scattering and screening are hard to separate from other contributions to the dynamics such as the kinetics of the optical generation process and the influences of interband transition densities. In theory we are in a position to study these processes separately. Thus, in order to concentrate on relaxation and screening processes we limit ourselves to a simple one-band model for the conduction band of a GaAs quantum well. Within this model the complications induced by details of the carrier generation process are bypassed by considering an initial distribution of carriers far from equilibrium. In fact, in this way our study concentrates on the relaxation and screening dynamics, which are generic for a two-dimensional electron gas, that by some means is brought into a nonequilibrium state.

For this one-component plasma the Hamiltonian is given by

$$H = H^0 + H^{cc} \tag{1}$$

with the kinetic-energy part

$$H^0 = \sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} \tag{2}$$

and the Coulomb interaction part

$$H^{cc} = \frac{1}{2} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q},\sigma,\sigma'} V_{\mathbf{q}} c^\dagger_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}'\sigma'} c_{\mathbf{k}'+\mathbf{q}\sigma'} c_{\mathbf{k}-\mathbf{q}\sigma}. \tag{3}$$

Here, $c^\dagger_{\mathbf{k}\sigma}$ and $c_{\mathbf{k}\sigma}$ denote creation and annihilation operators for an electron with in-plane momentum $\mathbf{k}$ and spin index σ, $\varepsilon_{\mathbf{k}}$ is the free-carrier dispersion relation assumed to be parabolic, and $V_{\mathbf{q}}$ is the Coulomb matrix element screened only by the background dielectric constant. In density-matrix theory Coulomb scattering and screening are described by a coupled set of equations for the one-particle density matrix

$$f^\sigma_{\mathbf{k}} := \langle c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} \rangle \tag{4}$$

and the two-particle density matrix

$$K^{\sigma,\sigma',\sigma',\sigma}_{\mathbf{k},\mathbf{k}',\mathbf{k}'+\mathbf{q},\mathbf{k}-\mathbf{q}} := \langle c^\dagger_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}'\sigma'} c_{\mathbf{k}'+\mathbf{q}\sigma'} c_{\mathbf{k}-\mathbf{q}\sigma} \rangle. \tag{5}$$

In the spirit of the correlation expansion[8] it is useful to define the correlation function $\bar{K}$ by subtracting from the two-particle density matrix K its mean-field part. Here we will concentrate on spin-independent distributions $f_{\mathbf{k}}$ as generated, e.g., by unpolarized or linearly polarized light pulses which allows us to define

$$\bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} := \sum_{\sigma'} K^{\sigma,\sigma',\sigma',\sigma}_{\mathbf{k},\mathbf{k}',\mathbf{k}'+\mathbf{q},\mathbf{k}-\mathbf{q}} + f_{\mathbf{k}} f_{\mathbf{k}-\mathbf{q}} \delta_{\mathbf{k},\mathbf{k}'+\mathbf{q}}. \tag{6}$$

A description of Coulomb kinetics including exchange and screening in a fully dynamical way is then given by the equations[8,20,34–36]

$$i\hbar \frac{d}{dt} f_{\mathbf{k}} = 2i \sum_{\mathbf{k}',\mathbf{q}} V_{\mathbf{q}} \mathrm{Im}\{\bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}}\}, \tag{7}$$

$$\begin{aligned}
i\hbar \frac{d}{dt} \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} = {} & (\mathcal{E}_{\mathbf{k}-\mathbf{q}} + \mathcal{E}_{\mathbf{k}'+\mathbf{q}} - \mathcal{E}_{\mathbf{k}'} - \mathcal{E}_{\mathbf{k}}) \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} \\
& + 2V_{\mathbf{q}} [f_{\mathbf{k}} f_{\mathbf{k}'} (1 - f_{\mathbf{k}'+\mathbf{q}})(1 - f_{\mathbf{k}-\mathbf{q}}) - f_{\mathbf{k}-\mathbf{q}} f_{\mathbf{k}'+\mathbf{q}} (1 - f_{\mathbf{k}'})(1 - f_{\mathbf{k}})] \\
& - V_{\mathbf{k}'-\mathbf{k}+\mathbf{q}} [f_{\mathbf{k}} f_{\mathbf{k}'} (1 - f_{\mathbf{k}'+\mathbf{q}})(1 - f_{\mathbf{k}-\mathbf{q}}) - f_{\mathbf{k}-\mathbf{q}} f_{\mathbf{k}'+\mathbf{q}} (1 - f_{\mathbf{k}'})(1 - f_{\mathbf{k}})] \\
& + 2V_{\mathbf{q}} \left[(f_{\mathbf{k}} - f_{\mathbf{k}-\mathbf{q}}) \sum_{\mathbf{k}''} \bar{K}_{\mathbf{k}'',\mathbf{k}',\mathbf{q}} + (f_{\mathbf{k}'} - f_{\mathbf{k}'+\mathbf{q}}) \sum_{\mathbf{k}''} \bar{K}_{\mathbf{k},\mathbf{k}'',\mathbf{q}} \right] \\
& - V_{\mathbf{k}'-\mathbf{k}+\mathbf{q}} \left[(f_{\mathbf{k}} - f_{\mathbf{k}'+\mathbf{q}}) \sum_{\mathbf{k}''} \bar{K}_{\mathbf{k}'',\mathbf{k}',\mathbf{k}-\mathbf{k}'-\mathbf{q}} + (f_{\mathbf{k}'} - f_{\mathbf{k}-\mathbf{q}}) \sum_{\mathbf{k}''} \bar{K}_{\mathbf{k},\mathbf{k}'',\mathbf{k}-\mathbf{k}'-\mathbf{q}} \right] \\
& + (1 - f_{\mathbf{k}-\mathbf{q}} - f_{\mathbf{k}'+\mathbf{q}}) \sum_{\mathbf{q}'} V_{\mathbf{q}'} \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}+\mathbf{q}'} - (1 - f_{\mathbf{k}} - f_{\mathbf{k}'}) \sum_{\mathbf{q}'} V_{\mathbf{q}'} \bar{K}_{\mathbf{k}-\mathbf{q}',\mathbf{k}'+\mathbf{q}',\mathbf{q}-\mathbf{q}'} \\
& - (f_{\mathbf{k}} - f_{\mathbf{k}-\mathbf{q}}) \sum_{\mathbf{q}'} V_{\mathbf{q}'} \bar{K}_{\mathbf{k}-\mathbf{q}',\mathbf{k}',\mathbf{q}} - (f_{\mathbf{k}'} - f_{\mathbf{k}'+\mathbf{q}}) \sum_{\mathbf{q}'} V_{\mathbf{q}'} \bar{K}_{\mathbf{k},\mathbf{k}'+\mathbf{q}',\mathbf{q}} \\
& - (f_{\mathbf{k}} - f_{\mathbf{k}'+\mathbf{q}}) \sum_{\mathbf{q}'} V_{\mathbf{q}'} \bar{K}_{\mathbf{k}-\mathbf{q}',\mathbf{k}',\mathbf{q}-\mathbf{q}'} - (f_{\mathbf{k}'} - f_{\mathbf{k}-\mathbf{q}}) \sum_{\mathbf{q}'} V_{\mathbf{q}'} \bar{K}_{\mathbf{k},\mathbf{k}'+\mathbf{q}',\mathbf{q}-\mathbf{q}'} \\
& + \text{six-point correlations},
\end{aligned} \tag{8}$$

where we have introduced the energies renormalized by the exchange energy according to $\mathcal{E}_{\mathbf{k}} := \varepsilon_{\mathbf{k}} - \Sigma_{\mathbf{q}} V_{\mathbf{q}} f_{\mathbf{k}-\mathbf{q}}$. The dynamical variables are directly related to the system energies. By introducing the mean kinetic, exchange, and correlation energies per particle as

$$E_{\mathrm{kin}} := 2n^{-1} \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} f_{\mathbf{k}}, \tag{9a}$$

$$E_{\mathrm{ex}} := -n^{-1} \sum_{\mathbf{k},\mathbf{q}} V_{\mathbf{q}} f_{\mathbf{k}} f_{\mathbf{k}-\mathbf{q}}, \tag{9b}$$

$$E_{\mathrm{corr}} := n^{-1} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} V_{\mathbf{q}} \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}}, \tag{9c}$$

respectively, the total energy is given by $\langle H \rangle = n(E_{\mathrm{kin}} + E_{\mathrm{ex}} + E_{\mathrm{corr}})$ with the density $n = 2\Sigma_{\mathbf{k}} f_{\mathbf{k}}$.

The terms in Eq. (8) have been classified in Ref. 20 and their respective physical meanings have been identified. From Eq. (8) two well-known approximations can be derived as limiting cases: The Born approximation (BA) is obtained by retaining only the first two lines and the random-phase approximation (RPA) consists of keeping the terms in the first, second, and fourth lines of Eq. (8).[8,20] The BA is expected to be the relevant approximation level for not too strong short-range interactions. In the case of long-range Coulomb interaction studied here it serves mainly as a reference representing the limit of completely unscreened dynamics. The RPA, on the other hand, is the state-of-the-art level of theory for the analysis of a screened Coulomb dynamics. It is a distinguished level of theory because it comprises the contributions with the strongest singularities in the long-wavelength limit ($q \to 0$) on the right-hand side of Eq. (7). From the structure of the terms in the third and fifth lines of Eq. (8) it is apparent that they describe exchange corrections to the terms in the second and fourth lines, respectively, which are necessary to satisfy the correct antisymmetry of the two-particle density matrix upon exchange of the two final or initial states. Obviously this antisymmetry is not fulfilled in the two cases of the BA and RPA. In the following we discuss the influence of the exchange interaction on the dynamics by comparing BA and RPA results with calculations accounting for the respective exchange terms denoted by BAX [first–third lines of Eq. (8)] and RPAX [first–fifth lines of Eq. (8)]. The most advanced level of theory that will be discussed explicitly in this paper is RPAX. This level comprises all contributions that within density-matrix theory contribute to the carrier scattering in a dynamically screened potential including the corresponding exchange contributions.[42] However, as with any numerically tractable level of theory for an interacting many-body system, it does, of course, not represent the complete dynamics of the microscopic model. For example, the part leading to the ladder diagrams [sixth line of Eq. (8)] is not yet included. However, these terms contribute noticeably only when the density is either very low or when the system is almost inverted.[34] Otherwise, the prefactor $(1 - f_{\mathbf{k}} - f_{\mathbf{k}'})$ suppresses the ladder contributions. The densities studied in the present paper have been chosen such that the latter condition is met. The remaining terms correspond to vertex corrections [seventh and eigth lines of Eq. (8)] which are less singular then the scattering and screening contributions of the second–fifth lines of Eq. (8) because the Coulomb potential $V_{\mathbf{q}'}$ here occurs inside the sum over $\mathbf{q}'$. Finally, there is a coupling to six-point correlations which are of higher order according to the correlation expansion and are not specified explicitly here. Neglecting six-point correlations corresponds to a truncation of the quantum Bogolyubov-Born-Green-Kirkwood-Yvon hierarchy at the four-point level. Ladder terms, vertex corrections, as well as the six-point correlations are beyond the scope of the present paper.

According to Eqs. (7) and (8) relaxation can be identified as a two-step process: first, the correlation is built up from the distribution function and, second, the distribution function changes due to the feedback from the correlation. The screening terms, on the other hand, already require the existence of a correlation. This is the reason for the retarded buildup of screening. To clearly identify this buildup we compare the fully dynamical treatment according to Eqs. (7) and (8) with a model where the screening is built up instantaneously by invoking the Born approximation with a statically screened Coulomb potential where the screening is described by the Lindhard formula in its static limit,[43,44] i.e., by the dielectric function

$$\epsilon(\mathbf{q}, \omega = 0) = 1 - V_{\mathbf{q}} \sum_{\mathbf{k},\sigma} \frac{f^{\sigma}_{\mathbf{k}} - f^{\sigma}_{\mathbf{k}-\mathbf{q}}}{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{q}}}. \tag{10}$$

This level of the theory will be denoted as BAL. In the present case of a two-dimensional electron gas it is worth noting that in the long-wavelength limit the Lindhard screening only depends on the distribution function at the band minimum $f_{\mathbf{k}=0}$. The consequences of this point will be discussed below.

We apply our theory to a GaAs quantum well of width 10 nm. To see most clearly the effects of the various contributions we first consider the case of the relaxation of an uncorrelated initial distribution corresponding to a Gaussian in energy centered at 10 meV with a width of 15 meV and a maximum of $f_{\mathbf{k}} = 0.5$ corresponding to a density of about 5×10^{11} cm^{-2}. We have numerically integrated the above coupled equations of motion for f and $\bar{K}$ by using a time step of 1 fs. The k dependencies have been discretized resulting in a total number of complex valued dynamical variables of about 5×10^7. We have verified that our results do not critically depend on the width of our k mesh or the time step.

Figure 1 shows for the five levels of the theory introduced above the electron distribution function at the times 25, 75, 150, and 250 fs. As a general trend we clearly see in all cases a relaxation towards the shape of an equilibrium distribution. A close look reveals that at the earliest time (25 fs) the curves group into three sets (see inset): BA and RPA exhibit the fastest relaxation, BAX and RPAX are somewhat slower, and the BAL curve is still closest to the initial distribution. This shows that the Lindhard formula clearly overestimates the screening when initially a noticeable density $f_{\mathbf{k}}$ is present at $\mathbf{k} = 0$, which is the case under the present conditions. In contrast, the dynamically treated screening both with and

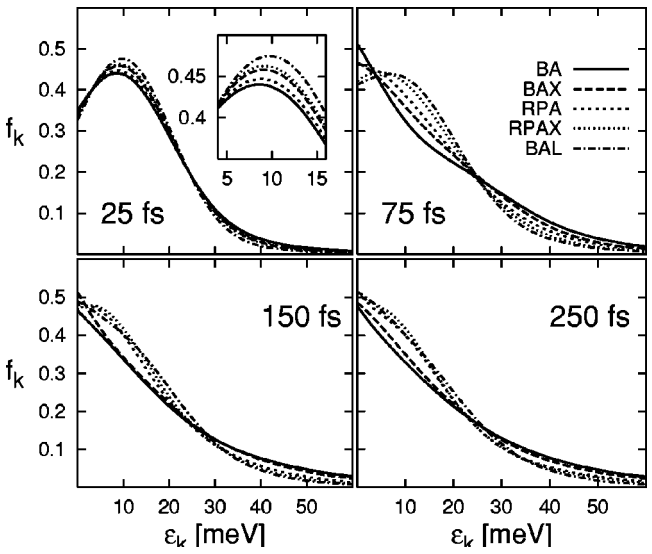

FIG. 1. Electron distribution function for the five cases discussed in the text at four different times. The inset shows an enlarged view of the region around the maximum.

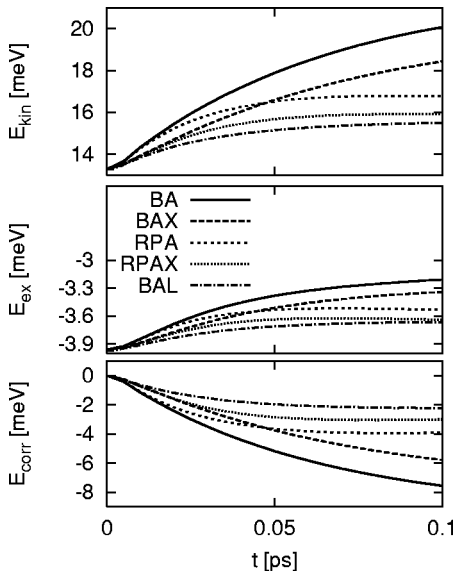

FIG. 3. Kinetic, exchange, and correlation energies per particle as functions of time obtained for the relaxation of an initial distribution.

without exchange contributions is not yet very efficient at this early time. At 75 fs no more grouping is observable, and all contributions to the dynamics are of the same order. At later times, and most pronounced at 250 fs, the curves regroup into two sets. Now the cases BA and BAX neglecting any kind of screening exhibit a significantly faster relaxation while all curves including the screening (RPA, RPAX, and BAL) are close to each other. In a different representation the same behavior can be seen in Fig. 2 where we plot the time evolution of the distribution function at two specific energies. Figure 2(a) refers to the band minimum ($\varepsilon_{\mathbf{k}}=0$), and Fig. 2(b) to $\varepsilon_{\mathbf{k}}=10$ meV, corresponding to the maximum of the initial distribution. At early times both curves, including dynamically the screening (RPA and RPAX) coincide with the respective cases without screening (BA and BAX) while

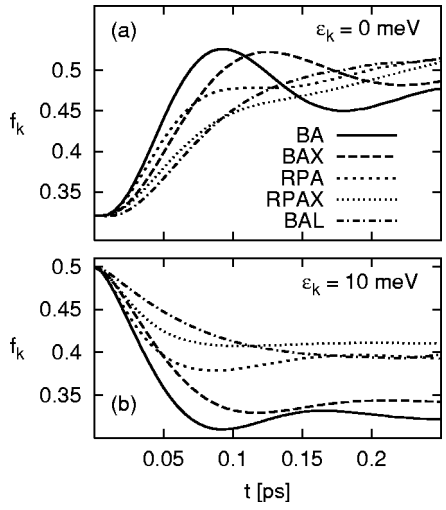

FIG. 2. Evolution of the electron distribution function at the minimum of the band ($\varepsilon_{\mathbf{k}}=0$) and at the maximum of the initial distribution ($\varepsilon_{\mathbf{k}}=10$ meV) for the five cases.

the statically screened case (BAL) relaxes much more slowly. The curves without screening exhibit the most pronounced relaxation dynamics showing even an oscillatory or overshoot behavior and at later times they are well separated from the cases with screening.

In Fig. 3 the energies as defined in Eq. (9) are plotted as functions of time. We find that the exchange energy does not depend very sensitively on the approximation level. The correlation and kinetic energies, however, exhibit remarkable differences. Without screening a large negative correlation energy builds up which in turn is complemented by a corresponding increase in the kinetic energy; the correlations still increase after 100 fs. In all cases with screening the correlation energies are reduced and saturate at about 50 fs. The early-time behavior again exhibits the grouping discussed above.

Let us now come to the physical interpretation of the results. A comparison of BA with BAX reveals that the exchange-scattering contribution [third line of Eq. (8)] leads to a pronounced reduction of the scattering efficiency especially at early times due to the opposite signs of direct and exchange terms [second and third lines of Eq. (8)]. Since scattering processes are effective just from the beginning the corresponding curves separate from each other immediately and already at 25 fs remarkable differences can be seen. The reduced scattering efficiency is a consequence of the less efficient buildup of correlations resulting in a slower rise of the absolute value of the correlation energy and, by energy conservation, of the kinetic energy.

At very early times RPA and RPAX coincide with BA and BAX, respectively. Then, however, the screening builds up and the relaxation of the distribution function dramatically decreases. After about 50–70 fs this reduction starts to dominate over the reduction by the exchange-scattering term [third part of Eq. (8)] as can be seen from the crossing of the curves BAX and RPA in Fig. 2. This is just the time scale

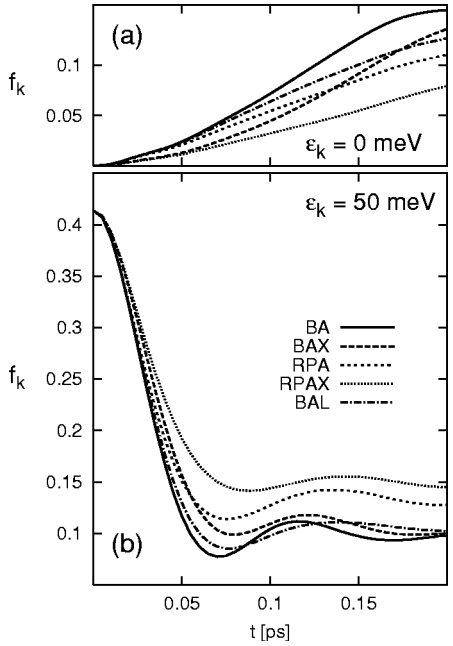

FIG. 4. Evolution of the electron distribution function for the case of an excess energy of 50 meV at the minimum of the band ($\varepsilon_{\mathbf{k}}=0$) and at the maximum of the initial distribution ($\varepsilon_{\mathbf{k}}=50$ meV) for the five cases.

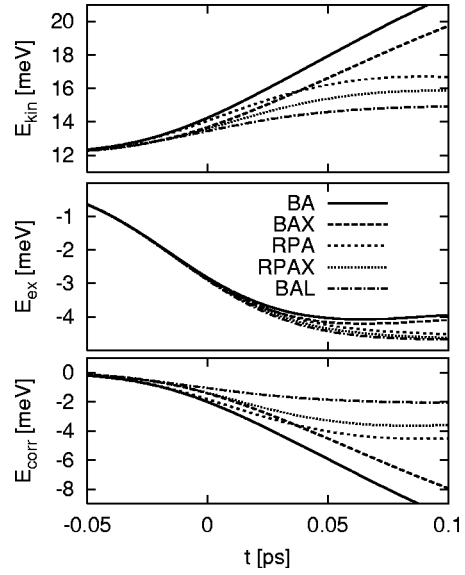

FIG. 5. Kinetic, exchange, and correlation energies per particle as functions of time including a semiclassical carrier generation with an excess energy of 10 meV.

where in Fig. 1 we have found that all terms are of equal importance. At about 150 fs the time evolutions of the distribution functions obtained by RPA and RPAX become more or less parallel. This can be interpreted as a mutual cancellation of the exchange contributions [third and fifth lines of Eq. (8)]. Thus, at later times the dynamics is well described by the commonly used RPA.

By completely neglecting the buildup of screening, the statically screened model BAL strongly overestimates the screening and consequently underestimates the relaxation at short times. At times longer than about 230 fs BAL leads to results similar to the RPA/RPAX levels of theory which means that the dynamics can now be well described by a static screening model.

However, a quite different scenario is obtained when the initial distribution is centered at a higher excess energy such that initially there is almost no density at the band minimum $\varepsilon_{\mathbf{k}}=0$. This is clearly seen in Fig. 4 where the temporal evolution of the electron distribution function at the bottom of the band and at the maximum of the initial distribution is plotted for an excess energy of 50 meV. The general features discussed above for the cases BA/BAX and RPA/RPAX remain unchanged. Again, the RPA initially coincides with BA while RPAX coincides with BAX demonstrating the retarded buildup of screening. As for an excess energy of 10 meV and also for the higher excess energy of 50 meV the relaxation is reduced by the exchange contributions. The static Lindhard screening BAL, however, now strongly underestimates the screening in contrast to the previous case where an overestimation of the screening has been found. The reason for this different behavior is the fact that, as mentioned above, the long-wavelength limit of the static screening, which is the most important part, in a two-dimensional electron gas only depends on the distribution function at the band minimum. In the present case of an excess energy of 50 meV and a width of 15 meV the occupation at $\mathbf{k}=0$ is negligible at early times. Therefore, initially there is no screening also in the BAL case. Indeed, up to about 50 fs the BAL curves coincide with the BA curves. They start to separate only after a noticeable occupation at the band minimum has built up and consequently screening reduces the relaxation in the BAL case. This overestimation of the scattering efficiency in a statically screened model is in qualitative agreement with previous findings for a three-dimensional electron gas studied in the Boltzmann limit, where scattering rates including static and dynamical screenings have been calculated for a situation characterized by strongly athermal electron distributions.[45]

The assumption of an uncorrelated initial distribution function is typically not realized in an actual experiment where carriers first have to be created. However, it has allowed us to clearly separate the various physical phenomena entering the relaxation dynamics. In order to demonstrate that assuming an uncorrelated initial state does not crucially affect our results and that the effects discussed above are not masked by the generation process we have repeated the calculations accounting for a semiclassical generation rate in Eq. (7) corresponding to a 75-fs laser pulse with an excess energy of 10 meV which, in the absence of relaxation, would result in the same distribution function used above. Scattering processes, however, are present already during the carrier generation and also the screening starts to build up before the generation has been completed. Therefore, due to the carrier-carrier interaction the distribution after the end of the laser pulse differs from the previously assumed initial distribution. In particular, already during the pulse the occupation of the low-momentum states will increase which will affect the screening dynamics.

In Fig. 5 we display the temporal evolution of the energies

for this approach and obviously by comparing with Fig. 3 we find that all three energies behave quite similarly, such as in the case of the initial distribution. In particular, all the features discussed above are also present here. At longer times the separation between screened and unscreened cases is even more pronounced. Clearly also this generation model is a simplification. The main point, however, responsible for the characteristic behavior of the different contributions is the fact that the light field only couples to the single-particle density matrices and these density matrices then act as source terms for the two-particle correlations. This two-step behavior also holds for the case of a two-band model which is necessary for a fully coherent treatment of the generation process. Therefore we expect that even if there will be some quantitative changes in the shapes of the distribution functions, the role of the exchange contributions for scattering and screening in the relaxation dynamics is not affected by the details of the generation process.

In conclusion, we have shown that on an ultrashort-time scale the inclusion of quantum-mechanical exchange terms results in a significant reduction of the scattering efficiency when compared to the case of direct interaction only. With increasing time the screening of the bare Coulomb interaction builds up, but also the screening is reduced by exchange contributions. When the screening has built up, the exchange contributions to scattering and screening essentially compensate each other and the subsequent relaxation dynamics is in good agreement with the standard RPA case. Finally we have shown that at even longer times the relaxation dynamics can satisfactorily be described in terms of a static screening model. Interestingly, particularly in a two-dimensional electron gas a static screening model may initially either over- or underestimate the screening efficiency, depending on whether or not there is initially a noticeable occupation at the band minimum.

This work has been partially supported by the Schwerpunktprogramm Quantenkohärenz in Halbleitern of the Deutsche Forschungsgemeinschaft and the TMR network Ultrafast Quantum Optoelectronics of the European Commission.

APPENDIX: NONDIVERGENT SCATTERING WITH AN UNSCREENED POTENTIAL

It is well known that the scattering rates in the Boltzmann treatment of Coulomb scattering diverge when an unscreened potential is used.[44] However, it is essential for our present discussion that initially the potential is unscreened and that screening is subsequently built up. The purpose of this Appendix is to show explicitly that within our quantum-kinetic density-matrix approach the scattering contributions yield a finite nondivergent contribution to the dynamics of our two-dimensional system. Similar considerations have been presented earlier for a three-dimensional system within the Green's-function formulation of the theory.[1] To facilitate the subsequent discussion we will use the following abbreviations:

$$I_{\mathbf{k}}(t,t') := \frac{1}{\hbar}\int_{t'}^{t} dt'' \mathcal{E}_{\mathbf{k}}(t''), \tag{A1}$$

$$e_{\mathbf{k},\mathbf{q}}(t,t') := \exp\{i[I_{\mathbf{k}}(t,t') - I_{\mathbf{k}-\mathbf{q}}(t,t')]\}, \tag{A2}$$

$$c_{\mathbf{k},\mathbf{q}}(t,t') := \cos\{I_{\mathbf{k}}(t,t') - I_{\mathbf{k}-\mathbf{q}}(t,t')\}, \tag{A3}$$

$$s_{\mathbf{k},\mathbf{q}}(t,t') := \sin\{I_{\mathbf{k}}(t,t') - I_{\mathbf{k}-\mathbf{q}}(t,t')\}, \tag{A4}$$

$$g_q(t,t') := \sum_{\mathbf{k}} e_{\mathbf{k},\mathbf{q}}(t,t') f_{\mathbf{k}}(t')[1 - f_{\mathbf{k}-\mathbf{q}}(t')]. \tag{A5}$$

The most critical case is obviously the BA level of theory. However, in this case it is easy to invert the equation of motion for $\overline{K}$ [Eq. (8)] and insert the result on the right-hand side of the equation of motion for f [Eq. (7)]. This procedure results in

$$\frac{d}{dt} f_{\mathbf{k}} = \frac{4}{\hbar^2} \frac{L^2}{(2\pi)^2} \int_0^t dt' \int_0^\infty dq\, F_{\mathbf{k},\mathbf{q}}(t,t'), \tag{A6}$$

where

$$\begin{aligned} F_{\mathbf{k},\mathbf{q}}(t,t') := \mathrm{Re}\Bigg(q \int_0^{2\pi} d\varphi V_{\mathbf{q}}^2 e_{\mathbf{k},\mathbf{q}}(t,t')\{ f_{\mathbf{k}}(t')[1 \\ - f_{\mathbf{k}-\mathbf{q}}(t')] g_q(t,t') - f_{\mathbf{k}-\mathbf{q}}(t')[1 \\ - f_{\mathbf{k}}(t')] g_q^*(t,t')\} \Bigg). \end{aligned} \tag{A7}$$

Here, φ is the angle between the vectors $\mathbf{k}$ and $\mathbf{q}$ and $V_{\mathbf{q}}$ is the Coulomb potential projected on the lowest quantum well sublevel. In order to study the singular point at $\mathbf{q}\to 0$ we use the asymptotic form of the Coulomb potential valid for a quasi-two-dimensional system

$$V_{\mathbf{q}} \propto \frac{1}{q}. \tag{A8}$$

Therefore, we can write the integrand $F_{\mathbf{k},\mathbf{q}}$ [Eq. (A7)] in the limit of small $|\mathbf{q}|$ values in the following form:

$$\begin{aligned} F_{\mathbf{k},\mathbf{q}}(t,t') \propto \frac{1}{q} \int_0^{2\pi} d\varphi (c_{\mathbf{k},\mathbf{q}}(t,t') \mathrm{Re}[g_q(t,t')][f_{\mathbf{k}}(t') \\ - f_{\mathbf{k}-\mathbf{q}}(t')] + s_{\mathbf{k},\mathbf{q}}(t,t') \mathrm{Im}[g_q(t,t')]\{ f_{\mathbf{k}}(t')[1 \\ - f_{\mathbf{k}-\mathbf{q}}(t')] + f_{\mathbf{k}-\mathbf{q}}(t')[1 - f_{\mathbf{k}}(t')]\}). \end{aligned} \tag{A9}$$

The terms in the integral in Eq. (A9) with the prefactor $s_{\mathbf{k},\mathbf{q}}(t,t')$ can be expanded as follows:

$$s_{\mathbf{k},\mathbf{q}}(t,t') \approx q\mathbf{e}_{\mathbf{q}} \cdot \nabla_{\mathbf{k}} I_{\mathbf{k}}(t,t') + \ldots, \tag{A10}$$

$$\mathrm{Im}[g_q(t,t')] \approx q \sum_{\mathbf{k}} \mathbf{e}_{\mathbf{q}} \cdot \nabla_{\mathbf{k}} I_{\mathbf{k}}(t,t') + \ldots, \tag{A11}$$

$$f_{\mathbf{k}}(t')[1-f_{\mathbf{k}-\mathbf{q}}(t')]+f_{\mathbf{k}-\mathbf{q}}(t')[1-f_{\mathbf{k}}(t')] \approx 2f_{\mathbf{k}}(t')[1-f_{\mathbf{k}}(t')]+q[1-2f_{\mathbf{k}}(t')]\mathbf{e}_{\mathbf{q}}\cdot\nabla_{\mathbf{k}}f_{\mathbf{k}}(t') +\cdots, \tag{A12}$$

where $\mathbf{e}_{\mathbf{q}}$ denotes the unit vector in the $\mathbf{q}$ direction. Thus, the product of the terms Eqs. (A10)–(A12) which occurs in Eq. (A9) asymptotically scales as q^2 in the limit of small q. Therefore, the $1/q$ singularity is canceled for this term.

The contributions to the integral in Eq. (A9) with the prefactor $c_{\mathbf{k},\mathbf{q}}(t,t')$ can be expanded analogously resulting in

$$c_{\mathbf{k},\mathbf{q}}(t,t')\approx 1-\frac{1}{2}q^2[\mathbf{e}_{\mathbf{q}}\cdot\nabla_{\mathbf{k}}I_{\mathbf{k}}(t,t')]^2+\ldots, \tag{A13}$$

$$\mathrm{Re}[g_q(t,t')]\approx\sum_{\mathbf{k}}\{f_{\mathbf{k}}(t')[1-f_{\mathbf{k}}(t')] -qf_{\mathbf{k}}(t')\mathbf{e}_{\mathbf{q}}\cdot\nabla_{\mathbf{k}}I_{\mathbf{k}}(t,t')\}+\ldots, \tag{A14}$$

$$f_{\mathbf{k}}(t')-f_{\mathbf{k}-\mathbf{q}}(t')\approx -q\mathbf{e}_{\mathbf{q}}\nabla_{\mathbf{k}}f_{\mathbf{k}}(t')+\ldots. \tag{A15}$$

The corresponding product is therefore proportional to q for small q values. Consequently, the total contribution to scattering does not diverge in the limit $q\to 0$ even when the scattering due to an unscreened potential is considered.

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