Ultrafast Quantum Kinetics of Time-Dependent RPA-Screened Coulomb Scattering

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We study the Coulomb quantum kinetics of relaxation and dephasing of an electron-hole gas in a semiconductor excited by a coherent femtosecond laser pulse. Our theory employs the full two-time-dependent RPA-screened Coulomb potential. The self-consistently calculated potential evolves from a bare potential to the well-known dynamically screened RPA potential for long times. The time dependence of the particle distributions, the interband polarization, and the potential are calculated. As a first application we study Rabi flopping with resonant femtosecond pulses. [S0031-9007(98)06707-6]

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Ever since the pioneering resonant photon echo experiments on bulk [1] and quantum well [2] GaAs, the problem of calculating the dephasing and relaxation kinetics in a dense electron-hole (e-h) plasma by Coulomb scattering has become one of the most challenging problems in nonequilibrium many-body theory. The temporal resolution of femtosecond four-wave mixing (FWM) experiments [3,4] allows one to study the nonequilibrium plasma on time scales smaller or comparable to the typical period of a plasma oscillation in this system. On these time scales the semiclassical Boltzmann kinetics has to be replaced by a quantum kinetics with memory structure [5]. The Coulomb scattering problem is complicated by the kinetics of the buildup of screening itself [6]. For this reason the quantum kinetics of Coulomb scattering [7,8] is far less developed than that of LO-phonon scattering [4,9]. The studies of the non-Markovian Coulomb quantum kinetics have so far been restricted to time scales considerably smaller than a plasma period, where the Coulomb potential is still bare [10,11]. While the $q \to 0$ divergence of the bare Coulomb potential makes a Markovian-Boltzmann scattering description inconsistent, the delayed quantum kinetic scattering integrals are well behaved and have been shown to explain the early-time development of a femtosecond-pulse excited plasma at least qualitatively [12]. In the framework of nonequilibrium Green functions, the screened Coulomb potential becomes a propagator which depends on two Keldysh contour time arguments [5] and obeys the equation

$$V_q(t_1, t_2) = V_q \delta(t_1 - t_2) + V_q L_q(t_1, t_3) V_q(t_3, t_2), \quad (1)$$

where integrations over repeated time arguments are implied in the matrix notation. V_q is the Fourier transformed bare Coulomb potential. The polarization function $L_q(t_1, t_2)$ will be evaluated here in the time-dependent random-phase approximation (RPA). For a two-band semiconductor excited with coherent light pulses, the particle propagators are two-by-two matrices in the band indices. Depending on the ordering of the time arguments on the contour, $V_q(t_1, t_2)$ will yield either the scattering potentials $V_q^{(<)_>}(t, t')$ or the retarded (advanced) potentials $V_q^{(r)_a}(t, t')$. The spectral potential functions can be represented as

$$\begin{Bmatrix} V_q^r(t, t') \\ V_q^a(t, t') \end{Bmatrix} = V_q \delta(t - t') + \begin{Bmatrix} \theta(t - t')[V_q^>(t, t') - V_q^<(t, t')] \\ \theta(t' - t)[V_q^<(t, t') - V_q^>(t, t')] \end{Bmatrix}. \quad (2)$$

Further symmetry relations like $V_q^{(<)_>}(t, t')^* = -V_q^{(<)_>}(t', t)$ and $V_q^>(t, t') = V_q^<(t', t)$ allow one to calculate only the integral equation for one of the four components of the nonequilibrium potential. From (1) one obtains, e.g., for $V_q^>$ the closed equation

$$V_q^>(t, t') = V_q \Big\{ L_q^>(t, t') V_q + \int_{-\infty}^{t} dt'' \, L_q^r(t, t'') V_q^>(t'', t') + \int_{-\infty}^{t'} dt'' \, L_q^>(t, t'') \times [V_q^>(t', t'') - V_q^>(t'', t')] \Big\}. \quad (3)$$

Note that for given polarization functions L_q only one integration from $-\infty$ to t or t' is necessary, while the use of nonlinear identities like $V_q^< = V_q^r L_q^< V_q^a$ would include two successive time integrations which requires much more computer memory for long-time integrations than the previously described strategy. For a two-band semiconductor the two-by-two density matrices $\rho_{\vec{k}}(t)$ (describing the carrier distributions in both bands and the interband polarization components) obey in the presence of coherent laser pulses the well-known semiconductor Bloch equations [5,13,14] with the following quantum kinetic collision rates:

$$\frac{\partial \rho_{\vec{k}}(t)}{\partial t} = -\int_{-\infty}^{t} dt' \times [\Sigma_{\vec{k}}^>(t, t') G_{\vec{k}}^<(t', t) - \Sigma_{\vec{k}}^<(t, t') G_{\vec{k}}^>(t', t) - G_{\vec{k}}^>(t, t') \Sigma_{\vec{k}}^<(t', t) + G_{\vec{k}}^<(t, t') \Sigma_{\vec{k}}^>(t', t)]. \quad (4)$$

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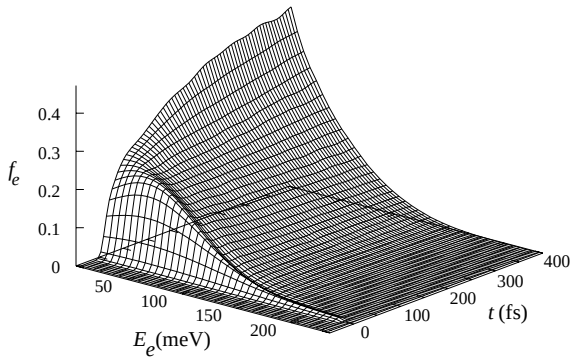

FIG. 1. Relaxation kinetics of the *electron* distribution in the conduction band $f_k^e(t)$ for a 15 fs pulse with 50 meV excess energy.

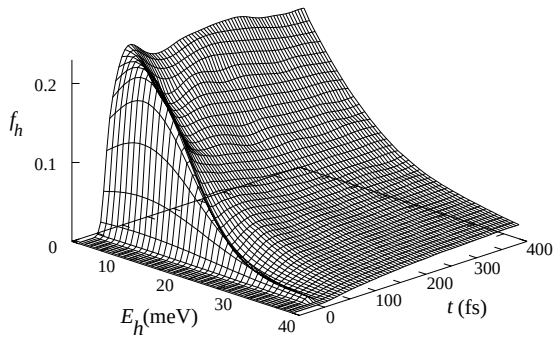

FIG. 2. Relaxation kinetics of the *hole* distribution in the valence band $f_k^h(t)$ for a 15 fs pulse with 50 meV excess energy.

In RPA, but without vertex correction, the self-energy matrix is given in terms of the scattering Coulomb potentials

$$\Sigma_k^{(>)_<}(t,t') = i\sum_{\vec{q}} G_{\vec{k}-\vec{q}}^{(>)_<}(t,t')V_q^{(>)_<}(t,t')\,.$$

The matrix of the propagators $G^{(>)_<}$ is expressed in terms of the density matrix by the generalized Kadanoff-Baym ansatz [5,15]

$$\begin{Bmatrix} G_{\vec{k}}^{<}(t,t') \\ G_{\vec{k}}^{>}(t,t') \end{Bmatrix} = \begin{Bmatrix} -G_{\vec{k}}^{r}(t,t')\rho_{\vec{k}}(t') + \rho_{\vec{k}}(t)G_{\vec{k}}^{a}(t,t') \\ G_{\vec{k}}^{r}(t,t')[1-\rho_{\vec{k}}(t')] + [1-\rho_{\vec{k}}(t)]G_{\vec{k}}^{a}(t,t') \end{Bmatrix}. \quad (5)$$

For simplicity the spectral functions will be taken in the diagonal free-particle approximation $G_{i,j,\vec{k}}^{r}(t,t') = G_{j,i,\vec{k}}^{a}(t',t)^{*} = -i\theta(t-t')\delta_{i,j}\exp[-ie_{i,k}(t-t')]$, where $e_{i,k}$ are the free-particle energies in band i. The polarization functions of Eq. (3) are in RPA given by $L_q^r(t,t') = \theta(t-t')[L_q^>(t,t') - L_q^<(t,t')]$ and $L_q^{(<)_>}(t,t') = -2i \times \sum_{\vec{p}} G_{\vec{p}+\vec{q}}^{(<)_>}(t,t')G_{\vec{p}}^{(<)_>}(t',t)$. Now the system of integro-differential equations for the density matrices and the screened Coulomb potential is closed. At no point of the solution does the $q=0$ divergence of V_q cause mathematical problems. We can show analytically that our quantum kinetic scattering integrals approach in the long-time limit the Boltzmann scattering rates calculated with a dynamically screened Coulomb potential with the Lindhard dielectric function. The frequency corresponds to the energy transfer in a collision.

In the following we present for bulk GaAs some numerical results for the relaxation kinetics of the carrier distributions and the interband polarization dephasing kinetics following a Gaussian excitation pulse with a half-width of 15 fs and a strength which corresponds to a 0.25π pulse. With the previously described strategy we were able to perform these calculation typically in less than 8 h on a standard RISC workstation. The excess energy with respect to the unrenormalized band edges was 50 meV. In Figs. 1 and 2 we show the evolution of the pulse excited e and h distributions. One sees that a few hundred femtoseconds after the exciting pulse the carriers approach thermal equilibrium distributions whose temperature is determined by the excess energy of the photoexcited carriers. Figure 3 shows the absolute amount of the optically induced interband polarization. After a fast decay in the first 50 fs, oscillations are seen up to about 200 fs. These oscillations have already been found in the bare Coulomb quantum kinetics [11] and have been shown to result in a density-dependent sideband in the spectrum of the four-wave mixing signal.

Repeatedly some skepticism has been expressed whether Coulomb quantum kinetics and, in particular, various screening models produce any measurable characteristic new features. Therefore we compare in Fig. 4 the absolute amount of the interband polarization, i.e., the polarization amplitude $|p_k(t)|$ for a given wave number $ka_B = 1.2$ (corresponding to a pair energy of 6 meV), where a_B is the exciton Bohr radius, calculated within various approximations for the scattering potential. Static screening, which rigorously speaking has no justification in the short-time quantum kinetics, but which has been used

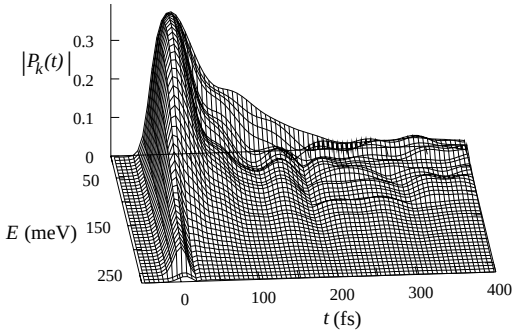

FIG. 3. Interband polarization amplitudes $|p_k(t)|$ versus time and pair energy for a 15 fs pulse with 50 meV excess energy.

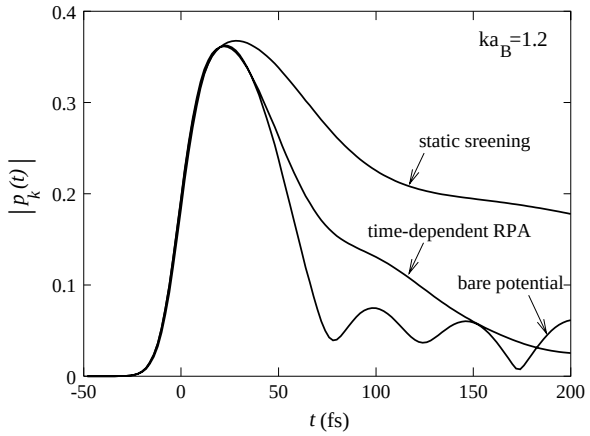

FIG. 4. Interband polarization amplitude $|p_{k=1.2a_B^{-1}}(t)|$ for a 15 fs pulse with 50 meV excess energy calculated in various approximations.

often [16–18], also with a self-consistent time-dependent screening length [17,18] results in a slow structureless decay of the polarization component as shown by the upper curve in Fig. 4. The bare Coulomb potential which holds for very short-time intervals after the carrier generation [6,8,11,12] results in a too strong dephasing with too pronounced coherent oscillations, while the full two-time-dependent RPA screened Coulomb potential is intermediate to the two simplified models and shows a weak shoulder at the time where the first oscillation of the bare Coulomb scattering peaks. Figure 4 demonstrates clearly that a phase sensitive variable, here the interband polarization amplitude, is influenced strongly by different screening models, while the resulting excited carrier distributions differ only quantitatively for the various potential approximations.

In order to analyze the resulting self-consistently calculated screened Coulomb potential we introduce the incomplete Fourier transform of the retarded Coulomb potential [6]

$$V_q(\omega, t) = \int_0^\infty d\tau\, e^{i\omega\tau} V_q^r(t, t-\tau) = \frac{V_q}{\epsilon(\omega, t)}. \quad (6)$$

In Fig. 5 the spectra of the imaginary and real parts of the inverse dielectric function are shown for various times for $qa_0 = 1$. The imaginary part shows that the plasmon pole is well developed after about 200 fs. The plasma frequency is about 31 meV, which corresponds to a plasma period of about 100 fs. This illustrates that the characteristic buildup time of screening is indeed the plasma oscillation period.

As a first application, we will study the currently disputed question under which conditions Rabi flopping by intense resonant pulse excitation is possible. Figure 6 shows the total excited plasma density for 3π pulses with various widths δt, again for a detuning of 50 meV. Note that the renormalized Rabi frequency $\omega_{R,k} = dE_0(t) + \sum_{\vec{q}} V_q p_{\vec{k}-\vec{q}}(t)$ corresponds to an effective pulse strength larger than a 4π pulse [here d is the dipole matrix element, $E_0(t)$ the pulse amplitude]. Figure 5 shows that two well-resolved Rabi flops occur as long as the pulse width is smaller than the time of the Coulomb dephasing kinetics and vanish under the present conditions at about $\delta t \simeq$ 200 fs. Such double Rabi flops have indeed been observed recently [19]. Rabi flopping in quantum wells has been observed already earlier for nonresonant excitation (9 meV below the exciton resonance) with about 100 fs pulses and explained in the framework of Maxwell-Bloch equations with phenomenological relaxation times [20].

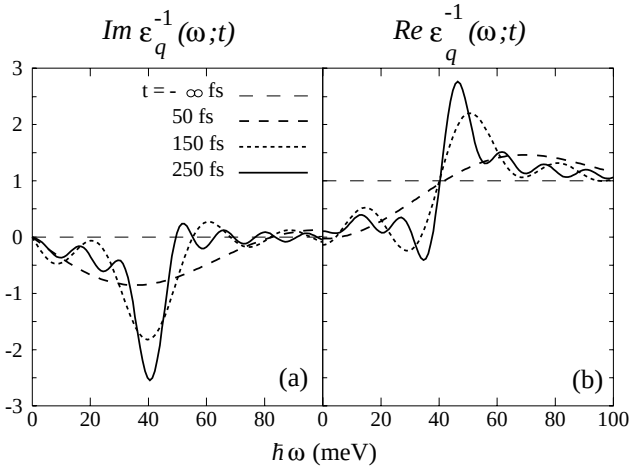

FIG. 5. Spectra of the imaginary (a) and real (b) part of the inverse dielectric function for $qa_0 = 1$ and various times after the 15 fs pulse at $t = 0$.

In conclusion, we presented the first complete Coulomb quantum kinetic calculations of the relaxation and dephasing kinetics of femtosecond-pulse excited carriers. The resulting self-consistently calculated two-time-dependent scattering potential is shown to develop from an initially bare potential into a stationary RPA potential. As a first example we investigated under which conditions Rabi

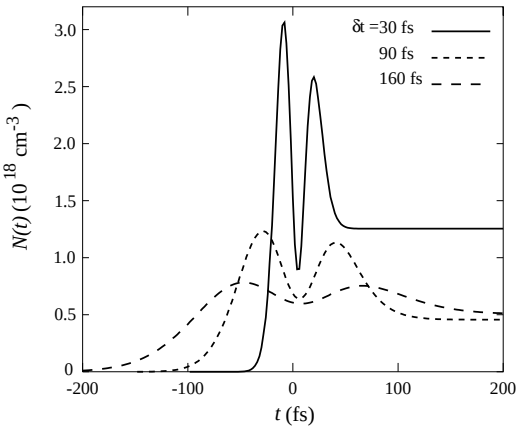

FIG. 6. Total excited plasma density $N(t)$ for 3π pulses and various pulse widths δt.

flopping is possible for intense resonant femtosecond-pulse excitation.

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