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Non-Lorentzian spectral functions for Coulomb quantum kinetics

M. Bonitz[1,a], D. Semkat[1], and H. Haug[2]

[1] Universität Rostock, FB Physik, Universitätsplatz 3, 18051 Rostock, Germany
[2] Institut für Theoretische Physik, Universität Frankfurt, Robert-Mayer Strasse 8-10, 60054 Frankfurt, Germany

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Abstract. Numerical solutions of the Kadanoff-Baym equations with self-energies in second Born approximation are presented for electrons in a bulk semiconductor. The results are used to analyze in detail the two-time spectral function $A(\mathbf{p}, t_1, t_2)$. In particular, (I) the damping of the spectral function is investigated over a broad density range both, for low and high temperatures; (II), $A(\mathbf{p}, t_1, t_2)$ is compared to analytical expressions – an exponential and a recently proposed inverse hyperbolic cosine decay law; (III), the two analytical spectral functions are studied with respect to conservation of total energy, where for the inverse hyperbolic cosine an improved behavior is demonstrated.

PACS. 05.20.Dd Kinetic theory – 05.30.-d Quantum statistical mechanics

1 Introduction

In the relaxation behavior of Coulomb systems, being excited by transverse electromagnetic (*e.g.* femtosecond laser pulses) or longitudinal electric fields, a variety of short-time phenomena is observed. The latter include collisional energy broadening, the buildup of correlations and screening and, instead of kinetic energy conservation, the conservation of total energy (kinetic plus interaction energy), see *e.g.* [1,2] for a detailed overview. These effects cannot be described by conventional Boltzmann-type (semiclassical) kinetic equations, but require generalized quantum kinetic equations. Among them, the most general are the Kadanoff-Baym equations (KBE) for the two-time correlation functions. Alternatively, one can consider quantum kinetic equations for single-time functions (Wigner distributions and interband polarizations) which are essentially simpler but, the price for this is some arbitrariness in the choice of the spectral functions A (or, retarded Green's functions G^{R}) appearing in the scattering kernels. In the weak-field limit and within a one-band model, the simplest ansatz for the single-particle spectral function $[G^{\mathrm{R}}(\mathbf{p}, t_1, t_2) = \Theta(t_1 - t_2)A(\mathbf{p}, t_1, t_2)]$ is the local approximation,

$$A(\mathbf{p}, t_1, t_2) \approx A(\mathbf{p}, \tau) = \mathrm{e}^{-\mathrm{i}\frac{E(\mathbf{p})}{\hbar}\tau}\mathrm{e}^{-\gamma\tau}; \quad \tau \equiv t_1 - t_2, \quad (1)$$

where $E(\mathbf{p})$ is the single-particle energy. Often, for the damping a phenomenological constant γ is taken (Wigner-Weisskopf approximation), but also improved self-consistent momentum- and time-dependent expressions have been derived, see *e.g.* [2]. In all cases, however, the ansatz (1) leads to serious problems – it violates energy conservation and causes an artificial heating and electron runaway in the long-time limit. The reason is the Lorentzian form of the Fourier transform of equation (1), which exhibits a too slow decay ($\sim 1/\omega^2$) for large energies. In the time representation, this is traced back to the behavior in the vicinity of the time diagonal $\tau = 0$, where the ansatz (1) has a finite slope $-\gamma$. This is in contrast to known analytical results and, as we will see below, also the Kadanoff-Baym equations give rise to a zero slope of the real part of the spectral function on the diagonal.

Therefore, recently an improved inverse hyperbolic cosine damping model has been proposed [3] which is of the form

$$A(\mathbf{p}, \tau) = \mathrm{e}^{-\mathrm{i}\frac{E(\mathbf{p})}{\hbar}\tau}\frac{1}{\cosh^{\alpha}(\omega_0\tau)}, \quad (2)$$

and reproduces the correct limits: (i) zero slope of $\mathrm{Re}A$ at $\tau = 0$ and (ii) exponential decay for large τ, where the damping constant is $\gamma = \alpha\omega_0$. This ansatz leaves open the two parameters α and ω_0. For the case of electron-LO-phonon scattering, the natural choice for α is the dimensionless Fröhlich constant and for ω_0 the LO-phonon frequency [3].

While the electron-phonon case has been analyzed in detail [4], where essentially improved properties of the ansatz (2) were confirmed, its critical test for the more complex carrier-carrier scattering is still missing. It is the aim of the current paper to present an in-depth study of this case. To this end, we solve numerically the Kadanoff-Baym equations for electron-electron scattering which provides us with the "exact" two-time spectral function $A(\mathbf{p}, t_1, t_2)$. This function is analyzed in detail with respect to its momentum dependence as well as its behavior for different densities and temperatures.

[a] e-mail: micha@elde.mpg.uni-rostock.de

Furthermore, we compare the two-time spectral functions to the Lorentzian and non-Lorentzian forms, equations (1, 2), respectively. A reasonable choice for ω_0 for Coulomb scattering is given by the plasma frequency ω_{pl}, whereas the parameter α is determined as the best fit of (2) to the KB result. Finally, we test the spectral functions (1, 2) in quantum kinetic relaxation calculations, using the obtained before values for α and ω_0. Our results show that the $1/\cosh$ form reduces the total energy increase, observed in the Lorentzian case, by about 50%.

2 Survey of the used quantum kinetics results

We recall the Kadanoff-Baym equations [5] for the two-time correlation functions $G^{\gtrless}(1,1')$, for a spatially homogeneous one-band system $(1=\mathbf{p}_1,t_1)$,

$$\Big(\mathrm{i}\hbar\frac{\partial}{\partial t_1}-\frac{p_1^2}{2m}\Big)G^{\gtrless}(1,2)-\int \mathrm{d}r_3\,\Sigma^{\mathrm{HF}}(1,3)G^{\gtrless}(3,2)$$
$$=\int_{t_0}^{t}\mathrm{d}t_3\left\{\Sigma^{>}(1,3)-\Sigma^{<}(1,3)\right\}G^{\gtrless}(3,2)$$
$$-\int_{t_0}^{t'}\mathrm{d}t_3\Sigma^{\gtrless}(1,3)\left\{G^{>}(3,2)-G^{<}(3,2)\right\}, \quad (3)$$

where $G^{\gtrless}(1,2)$ simultaneously obey equation (3) and its adjoint, and initial correlations are neglected, as we limit ourselves to not preexcited semiconductors, (for the account of initial correlations in the KBE, see [2,6]). From the solutions $G^{\gtrless}(1,2)$ of the KBE, all relevant quantities may be computed, in particular, the Wigner function f, the spectral function, and the kinetic and correlation energy density $\langle T\rangle$ and $\langle V\rangle$,

$$f(\mathbf{p},t)=-\mathrm{i}\hbar\,G^{<}(\mathbf{p},t,t) \quad (4)$$

$$A(\mathbf{p},t_1,t_2)=\mathrm{i}\hbar[G^{>}(\mathbf{p},t_1,t_2)-G^{<}(\mathbf{p},t_1,t_2)] \quad (5)$$

$$\langle T\rangle(t)=\int\frac{\mathrm{d}\mathbf{p}}{(2\pi\hbar)^3}\frac{p^2}{2m}(-\mathrm{i}\hbar)G^{<}(\mathbf{p},t,t) \quad (6)$$

$$\langle V\rangle(t)=\frac{1}{4}\int\frac{\mathrm{d}\mathbf{p}}{(2\pi\hbar)^3}\left\{\left(\mathrm{i}\hbar\frac{\partial}{\partial t_1}-\mathrm{i}\hbar\frac{\partial}{\partial t_2}\right)-\frac{p^2}{m}\right\}$$
$$\times(-\mathrm{i}\hbar)\,G^{<}(\mathbf{p},t_1,t_2)\big|_{t_1=t_2=t}, \quad (7)$$

from which the total energy follows according to $\langle H\rangle=\langle T\rangle+\langle V\rangle$. The Hartree-Fock contribution is not important for our considerations and will, therefore, be neglected. It is well known that the KBE conserve total energy for a broad class of self-energy functions $\Sigma^{\gtrless}$ [5]. Among them, the static second Born approximation is the simplest approximation for carrier-carrier scattering, involving the Debye potential V_s,

$$\Sigma^{\gtrless}(\mathbf{p},t_1,t_2)=$$
$$\mathrm{i}\hbar\int\frac{\mathrm{d}\mathbf{q}}{(2\pi\hbar)^3}V_s^2(\mathbf{q})\Pi^{\gtrless}(\mathbf{q},t_1,t_2)\,G^{\gtrless}(\mathbf{p}+\mathbf{q},t_1,t_2), \quad (8)$$

$$\Pi^{\gtrless}(\mathbf{q},t_1,t_2)=$$
$$-\mathrm{i}\hbar\int\frac{\mathrm{d}\mathbf{p}'}{(2\pi\hbar)^3}G^{\gtrless}(\mathbf{p}'-\mathbf{q},t_1,t_2)\,G^{\lessgtr}(\mathbf{p}',t_2,t_1). \quad (9)$$

and will be used in the following. In the framework of quantum kinetics, the screened Coulomb potential should be treated also as a two-time-dependent quantity [7,8], but for our present studies we use the time-independent Debye potential as a simple model potential.

The Kadanoff-Baym equations fully include retardation effects and are, thus, well suited for the investigation of short-time phenomena. Most importantly, with the self-energies being fixed, a fully self-consistent quantum kinetic model is given, and no further assumptions are necessary.

This is in contrast to the quantum kinetic equations for single-time functions which follow in well-known manner from the difference of equation (3) and its adjoint, but require additional approximations to eliminate the two-time functions in the collision integral I,

$$\frac{\mathrm{d}}{\mathrm{d}t}f(\mathbf{p},t)=I(\mathbf{p},t)$$
$$=2\mathrm{Re}\int_{t_0}^{t}\mathrm{d}\bar{t}\left\{\Sigma^{>}(\mathbf{p},t,\bar{t})G^{<}(\mathbf{p},\bar{t},t)\right.$$
$$\left.-G^{>}(\mathbf{p},t,\bar{t})\Sigma^{<}(\mathbf{p},\bar{t},t)\right\}, \quad (10)$$

which is achieved by means of the generalized Kadanoff-Baym ansatz (GKBA) [9],

$$(-\mathrm{i}\hbar)G^{\gtrless}(\mathbf{p},t_1,t_2)=A(\mathbf{p},t_1,t_2)\Big\{\Theta(t_1-t_2)f^{\gtrless}(t_2)$$
$$+\Theta(t_2-t_1)f^{\gtrless}(t_1)\Big\}, \quad (11)$$

where $f^{<}\equiv f$ and $f^{>}\equiv 1-f$. As a result, with the self-energies in Born approximation (8), the r.h.s. of equation (10) becomes

$$I(\mathbf{p},t)=\frac{2}{\hbar^2}\mathrm{Re}\int_{t_0}^{t}\mathrm{d}\bar{t}\int\frac{\mathrm{d}\mathbf{q}}{(2\pi\hbar)^3}V_s^2(\mathbf{q})$$
$$\times\int\frac{\mathrm{d}\mathbf{p}'}{(2\pi\hbar)^3}A(\mathbf{p},t,\bar{t})A(\mathbf{p}',t,\bar{t})A(\mathbf{p}+\mathbf{q},\bar{t},t)A(\mathbf{p}'-\mathbf{q},\bar{t},t)$$
$$\times\left\{f^{<}(\mathbf{p})f^{<}(\mathbf{p}')f^{>}(\mathbf{p}+\mathbf{q})f^{>}(\mathbf{p}'-\mathbf{q})-(f^{<}\leftrightarrow f^{>})\right\}\big|_{\bar{t}}, \quad (12)$$

which contains the two-time spectral functions. To eliminate this last relict from the KBE, additional approximations for $A(\mathbf{p},t_1,t_2)$ are necessary, two of which have been discussed in the introduction, *cf.* equations (1, 2).

3 Numerical Results

In this section, we present results from numerical solutions of quantum kinetic equations describing carrier relaxation due to electron-electron scattering in bulk semiconductors. We solve the KBE (3) and the equation for the Wigner distribution with the non-Markovian collision integral (12),

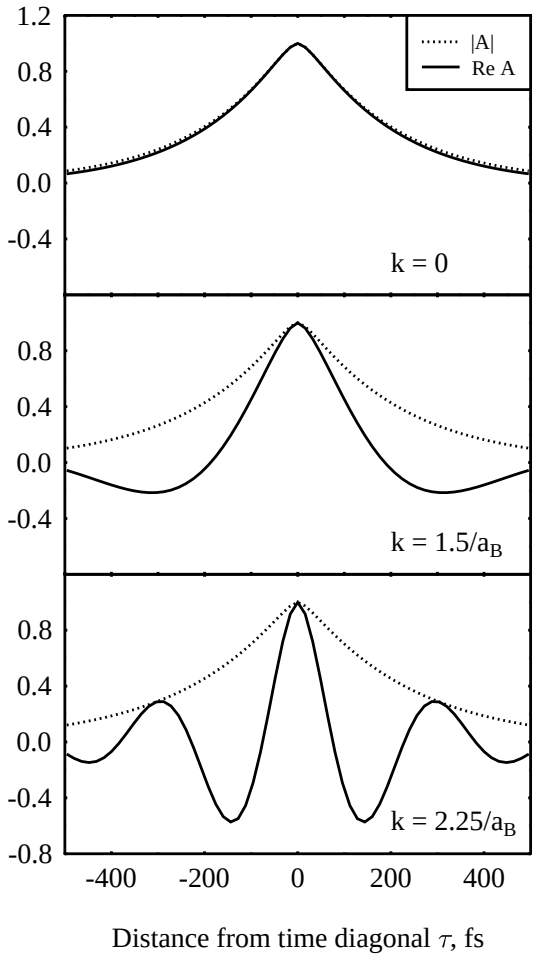

Fig. 1. Real part and absolute value of the spectral function A *versus* relative time $\tau = t - t'$ from a two-time (KB) calculation for three different wave numbers. The initial distribution was a Fermi function with $n = 5 \times 10^{17}$ cm^{-3} and $T = 290$ K. The plot corresponds to a macroscopic time of $T = 248$ fs after which a correlated equilibrium distribution has been built up.

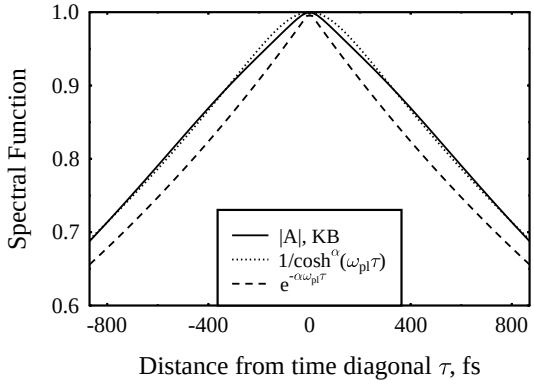

Fig. 2. Absolute value of the spectral function from a KB calculation (full line) compared to two one-time calculations: with Lorentzian damping (dashed line) and with the 1/cosh ansatz (dotted line). The damping increment is $\gamma = \alpha\omega_{\rm pl}$, where $\omega_{\rm pl}$ is the plasma frequency and α is obtained as the best fit of the non-Lorentzian curve to the KB-result. The same damping is used for the Lorentzian spectral function. The density is $n = 10^{16}$ cm^{-3} and temperature $T = 10$ K.

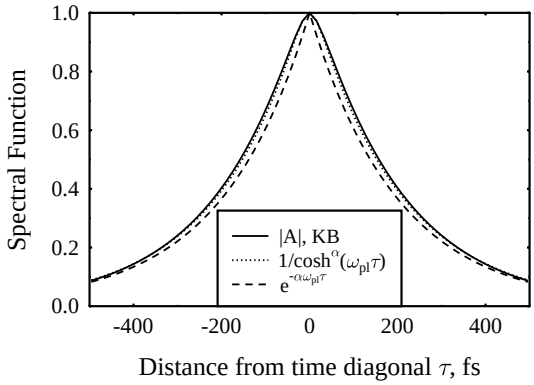

Fig. 3. Same as Figure 2, but for $n = 10^{17}$ cm^{-3} and $T = 290$ K.

using the spectral functions (1, 2), respectively, and also an undamped function (free GKBA), for numerical details, see references [2,10]. To be specific, we choose GaAs parameters for the background dielectric constant and the exciton Bohr radius, $\epsilon_{\rm B} = 13$ and $a_{\rm B} = 132$ Å.

We limit ourselves to a single-band relaxation and study various initial conditions. First we consider the equilibrium two-time spectral function, Figures 1–5. To this end, the KBE were solved starting with a Fermi distribution. For a correlated many-particle system, this distribution is a nonequilibrium one. The system relaxes towards a stationary correlated state and the damping builds up.

Figure 1 shows $A(\mathbf{p}, t, \tau)$ for a fixed macroscopic time $t = (t_1 + t_2)/2 = 248$ fs for three different wave numbers. The damping is strongest for zero momentum and decreases weakly with growing k. We, therefore, will concentrate in the following on the $k = 0$ case. Furthermore, one clearly sees the zero slope on the time diagonal, $\tau = 0$. Therefore, as expected, $A(t, \tau)$ can be approximated much better by the non-Lorentzian function (2), than by the form (1). Figures 2 and 3 show the results of the fits of approximations (1, 2) to $A(\mathbf{p}, t, \tau)$ for low and high temperatures, respectively. The agreement is generally better for high temperature where the damping is stronger. At low temperature, the decay of the two-time spectral function is more complex at intermediate times τ, so to improve the quantitative agreement would require to include further corrections into the function (2).

In Figures 4 and 5 we investigate the density dependence of the fit parameter α and of the damping increment $\gamma = \alpha\omega_{\rm pl}$ for a low and a high temperature. Notice the minimum in the curves $\gamma(n)$ which resembles the behavior of the scattering rates in static Born approximation. At high densities, where the role of correlations decreases, α approaches the value

$$\alpha(n) = \frac{1}{3^{2/3}\pi^{5/6}} \sqrt{\frac{me^2}{\hbar^2\epsilon_0}} n^{-1/6} \qquad (13)$$

which is independent of temperature. On the other hand, for low densities, the Born approximation scattering rates

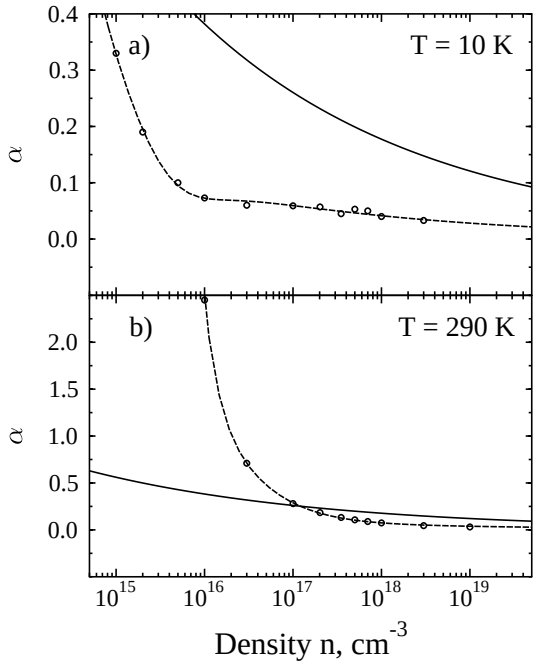

Fig. 4. Density dependence of the fit parameter α for $T = 10$ K (a) and 290 K (b). Symbols are extracted from the numerical solutions of the KB calculations by fitting to formula (2), *cf.* Figures 2 and 3, dashed line corresponds to the analytical fit formula (14), full line is the high density limit (13).

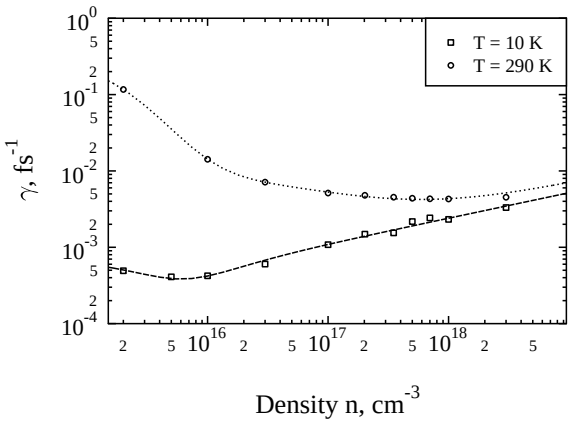

Fig. 5. Density dependence of the damping coefficient $\gamma = \alpha\omega_{\rm pl}$ (b) for two different temperatures. As in Figure 4, the circles and squares represent KB results, the lines to formula (14).

in a charged particle system approach a constant. Using these two limits, we construct a simple Padé formula to fit the density dependent two-time data,

$$\alpha(n) = a\frac{n^{-1/2} + b + c\,n^{1/2} + d\,n^{3/2}}{1 + e\,n^{5/3}}, \qquad (14)$$

where the parameter values are shown in Table 1.

Let us now consider nonequilibrium situations. The analysis of relaxation processes allows us to consider several further interesting questions including the influence of the initial distribution and the quality of the spectral functions (2) *versus* (1) in nonequilibrium. In order

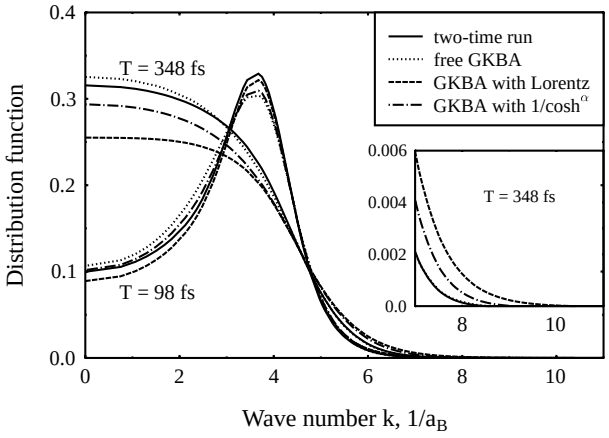

Fig. 6. Relaxation of a nonequilibrium Gaussian electron distribution function, shown for two different times and four different approximations for the spectral function, for explanation, see inset. The density is $n = 0.97a_{\rm B}^{-3}$.

to investigate these problems, we have performed a series of solutions of the quantum kinetic equation for the Wigner function with the collision term (12) using various nonequilibrium initial distributions, *e.g.* Gaussians, and zero initial correlations. For each case, first the KBE equations were solved. Then, as before, from the two-time spectral functions the parameter α was extracted as the best fit to the form (2). This is not trivial since α changes with time. However, the analysis shows that the damping of the spectral function grows during the initial time (correlation time), but afterwards approximately remains constant. Therefore, α was fitted to the spectral function for the final time.

Furthermore, the investigation showed that the spectral functions for initial "equilibrium" (Fermi) and nonequilibrium (Gaussian) distributions are quite similar. Provided, the two distributions lead to the same equilibrium state (same density and temperature), the time evolution of the spectral functions is not qualitatively different. There are, of course, quantitative differences in the transient relaxation behavior. In particular, at intermediate times our results show a stronger damping for Gauss distributions which is due to a higher correlation energy.

Figure 6 shows the relaxation of the distribution according to the four scattering models. Clearly, the Lorentzian spectral function slows down the relaxation compared to the two-time calculation. On the other hand, the free GKBA (*i.e.* Lorentzian spectral function with zero damping) turns out to be faster than the KBE, approaching the latter for large times, see also [11]. The distributions calculated with the $1/\cosh$ form lie approximately in the middle between the Lorentz and the free GKBA cases. Furthermore, the results confirm that for the Lorentzian case, the distribution decreases with increasing momentum significantly slower than the KB run. Again, the $1/\cosh$ form reduces this effect.

This behavior is more clearly seen in the evolution of macroscopic averages, in particular kinetic and potential energy. Consider first potential energy, Figure 7a.

Table 1. Fit parameters for the Padé formula (14) for two temperatures.

Temperature/K	a	b	c	d	e
10	0.022 6733	−3.579 64	−8.0729	2418.74	1115.7
290	11.6349	−8.7345	32.5059	26.9986	4864.86

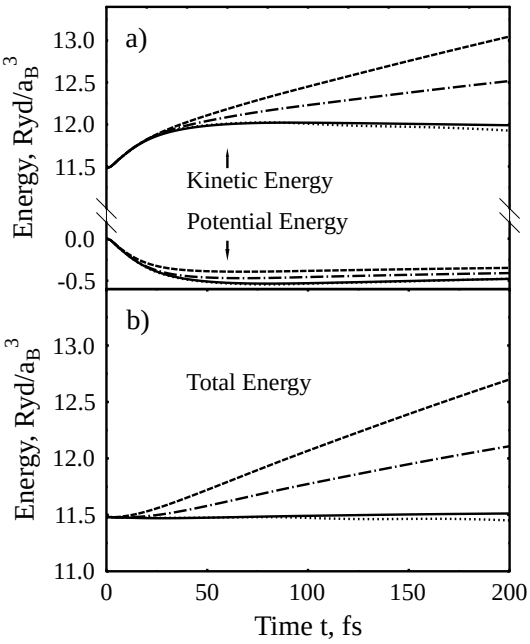

Fig. 7. Time evolution of kinetic and potential energy (a) and of total energy (b) for the relaxation conditions of Figure 6, where also the different approximations are explained.

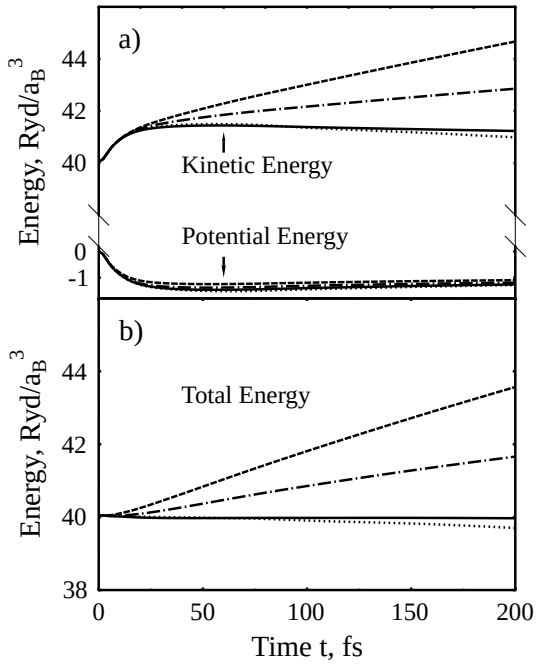

Fig. 8. Same as Figure 7, but for a higher density of $n = 2.77 a_{\mathrm{B}}^{-3}$.

Interestingly, there are only small differences between the four models, that is, correlation energy is quite insensitive of details of the scattering model: correlation energy decreases over a short time (correlation time τ_{cor}) which reflects the formation of (attractive) binary correlations. After this period, it remains nearly constant, as the system enters the regime of (Markovian) Boltzmann-type kinetics, *cf.* [11]. The same trend, but with the opposite sign, is observed for the kinetic energy, Figure 7a, in the case of the KBE and of the free GKBA, which is a consequence of exact total energy conservation in these models, see Figure 7b. In contrast, the Lorentzian spectral function leads to a much stronger growth of kinetic energy which, moreover, does not saturate after $t > \tau_{\mathrm{cor}}$. As one can see, the non-Lorentzian spectral function shows an improved behavior, reducing this heating effect approximately by 50%. This is a rather general result which was verified also for other situations, see for example Figure 8, where a higher density has been used.

4 Discussion

In this paper, a numerical study of the effect of different spectral functions for Coulomb quantum kinetics has been performed. While the Lorentzian spectral function shows the well-known problems, for the previously proposed 1/cosh function, improved behavior could be demonstrated. Nevertheless, this improvement turns out to be quantitative, and it does not overcome the qualitative problems. Also, the achieved effect is less than for the phonon case [8], which is, most likely, due to the more complex momentum structure of the Coulomb collision integrals containing summations (averaging) over two internal momenta. One may, of course, ask why not use at all the free GKB (zero damping) approximation, as it shows the same conservation behavior than the two-time result, and the stationary distributions reached by both are rather close to each other. There are two aspects to this question. The first is a technical: without damping, the memory duration in the collision integral (12) becomes unlimited leading to increasingly dense oscillations of the integrand. As a consequence, an accurate integration is possible only for a limited time (which is set by the chosen discretization). On the other hand, there is a more fundamental objection: in a correlated system, there do exist damping effects, as clearly shown by the KBE, *cf.* Figure 1, therefore, the choice of free spectral function would be inconsistent.

Of course, our one-band calculations are model studies which directly apply *e.g.* to a correlated electron gas. Nevertheless, our approach is expected to be of use also for the interband dynamics of optically excited semiconductors where one has to consider the intraband ($A_{\rm cc}, A_{\rm vv}$) and interband ($A_{\rm cv}$) spectral functions. Solutions of the interband KBE [10] allow us to conclude that the proposed 1/cosh approximation is directly applicable to the intraband spectral functions. On the other hand, the interband functions are more complex and require further studies.

Finally, let us mention that the ansatz (2) could be even more important for two-time calculations. One problem of the latter is that the generated data are depending in an extremely complex way on the two times and the momentum and are usually reduced in the calculation of macroscopic quantities. For example, a central quantity in quantum statistics is the damping of the spectral function, *i.e.* its τ-dependence. For this, the $1/\cosh$ form might be very useful, *e.g.* to extract the density and temperature dependencies, as was demonstrated in Figure 5.

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