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Relaxation of strongly coupled Coulomb systems after rapid changes of the interaction potential

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Abstract

The relaxation of charged particle systems after sudden changes of the pair interaction strength is investigated. As examples, we show the results for plasmas after ionization and after a rapid change of screening. Comparisons are made between molecular dynamics simulation and a kinetic description based on the Kadanoff–Baym equations. We found the latter very sensitive to the way the scattering cross section is treated. We also predict the new equilibrium state requiring only conservation of energy. In this case, the correlation energy is computed using the hypernetted chain approximation.

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1. Introduction

Strongly coupled laboratory plasmas are created by a variety of methods which in turn produce a wide range of system parameters. Examples are laser or particle beam interactions with gases and solids, shocks, explosions and laser excitation of semiconductors. Typically these plasmas are created in a state far from equilibrium. Therefore, it is of interest to understand the subsequent relaxation process which establishes equilibrium momentum distributions as well as the equilibrium balance between kinetic and potential energies. The latter is particularly important for strongly coupled systems, where the potential energy exceeds the kinetic energy.

Most strongly coupled plasmas are multicomponent systems, where the energy of the external driver, that is creating or subsequently disturbing the plasma, is almost entirely absorbed by one species. The heating of one species also changes the effective interactions of the other species. According to these new forces, the 'unaffected' species will now evolve on a new energy landscape to seek a new equilibrium state. Examples for rapid changes of the interactions are the ionization of ultracold gases [1], the creation of electron–hole pairs in semiconductors and non-thermal melting of solids [2].

In this paper, we focus on the question of how changing the interaction potential affects the properties, in particular the temperature, of the 'undisturbed' subsystem. Theoretically, it

is difficult to predict the development of a strongly coupled nonequilibrium system. Here, we compare two methods: classical molecular dynamics (MD) simulations and quantum kinetic theory (QKT). For strongly coupled Coulomb systems, it is especially important to have kinetic equations that conserve total energy (in contrast to standard kinetic approaches such as the usual Boltzmann equation). There have been several attempts to derive and apply kinetic equations that include potential energy contributions [3–7]. Here, we apply the Kadanoff–Baym equation approach of [5], which allows for a description of short times and conserves total energy. Estimates for the final temperature are also found by solving the hypernetted chain (HNC) equations.

While the MD and HNC calculations are limited to classical systems, they describe strong coupling effects. On the other hand, the QKT includes degeneracy effects but practical computations are limited to weak coupling since the Born approximation for the scattering cross section is usually used. To allow for calculations in the moderately coupled regime, we also employ a pseudopotential, which models close collisions and, therefore, partially overcomes the limitations of the Born approximation.

As an example, we will study the relaxation of a one-component plasma with a neutralizing, polarizable background (Yukawa model). Therefore, no energy exchange between the species is included. The interaction between the particles is given by

$$V(r) = \frac{q^2 e^2}{r} \exp(-\kappa r) \tag{1}$$

where κ is the inverse screening length. We can already demonstrate the qualitative behaviour with this simple model, while it considerably reduces the numerical effort compared to a full multicomponent calculation. Furthermore, we restrict ourselves to classical systems to allow for a comparison between MD and QKT.

2. Energy balance of an isolated subsystem

We consider a plasma after an excitation by an external energy source. Due to the large mass ratio of electrons and ions, most of the energy is typically absorbed by the electrons; the kinetic energy of the ions is only negligibly changed. However, the ion–ion pair potential is changed during the excitation since the electron properties determine the screening of the bare Coulomb potential. In this paper, we investigate the short-time dynamics of the ionic subsystem after the excitation. If the external excitation occurs rapidly, the relaxation of the ions to a new quasi-equilibrium state is usually fast enough that the ions can be treated as an isolated subsystem; i.e., we can neglect the energy transfer between ions and electrons and between the system and the environment. Under these circumstances, total energy is conserved during the relaxation[3]. Then the following holds:

$$E_{\mathrm{tot}}(0) \equiv K(0) + U(0) = K(t) + U(t) \equiv E_{\mathrm{tot}}(t) = E_{\mathrm{tot}}(\infty) \tag{2}$$

where $E_{\mathrm{tot}}(0)$ is the total energy just after the change of potential and/or kinetic energy, and $E_{\mathrm{tot}}(\infty)$ refers to the new quasi-equilibrium state of the ionic subsystem. The mean kinetic and potential energies per particle of a system with a momentum distribution $f(p, t)$ and pair distribution $g(r, t)$ are given by

$$\langle K \rangle = \frac{1}{n} \int \frac{\mathrm{d}\mathbf{p}}{(2\pi\hbar)^3} \frac{p^2}{2m} f(p, t) \qquad \text{and} \qquad \langle U \rangle = \frac{n}{2} \int \mathrm{d}\mathbf{r}\, V(r)[g(r, t) - 1]. \tag{3}$$

[3] Note that total energy is changed during the external excitation at $t < 0$, when the energy is provided by the driver. Only after the external driver is switched off (at $t \geqslant 0$), total energy is conserved.

There are two possibilities for the initial nonequilibrium state after the energy landscape has been modified:

(i) The system is initially undercorrelated, i.e. $|U(0)| < |U(\infty)|$. This includes also the totally uncorrelated initial state.
(ii) The system is initially overcorrelated, i.e. $|U(0)| > |U(\infty)|$.

The stability of the ionic quasi-equilibrium state requires that the correlation energy $U(\infty)$ is a negative quantity. Therefore, the system evolves in the first case to a state with a more negative potential energy. Accordingly, kinetic energy increases, i.e. the system heats. The opposite behaviour can be found in case (ii), where the system cools [5, 7].

3. Theoretical description of the relaxation

3.1. Quantum kinetic theory

To describe the relaxation after changes in the interaction, we must apply a kinetic approach which accurately describes: (i) conservation of total energy, i.e. the conversion of kinetic into potential energy and vice versa; (ii) ultrashort times, i.e. the initial stage of the evolution; (iii) initial correlations in the system; and finally, equilibrium solutions of correlated particles. These requirements are satisfied by the generalized Kadanoff–Baym equations (KBE) for the two-time correlation functions $g^{\gtrless}$ which include initial correlations (see [5] for the notation)

$$\begin{aligned}\left(\mathrm{i}\hbar\frac{\partial}{\partial t_1}-\frac{p_1^2}{2m_1}-\Sigma^{\mathrm{HF}}(\mathbf{p}_1,t_1)\right)g^{\gtrless}(\mathbf{p}_1;t_1,t_1')-I^{\mathrm{IC}}(\mathbf{p}_1;t_1,t_1')\\ =\int_{t_0}^{t_1}\mathrm{d}\bar{t}_1[\Sigma^{>}(\mathbf{p}_1;t_1,\bar{t}_1)-\Sigma^{<}(\mathbf{p}_1;t_1,\bar{t}_1)]g^{\gtrless}(\mathbf{p}_1;\bar{t}_1,t_1')\\ +\int_{t_0}^{t_1'}\mathrm{d}\bar{t}_1\Sigma^{\gtrless}(\mathbf{p}_1;t_1,\bar{t}_1)[g^{<}(\mathbf{p}_1;\bar{t}_1,t_1')-g^{>}(\mathbf{p}_1;\bar{t}_1,t_1')]\end{aligned}\tag{4}$$

together with the adjoint equations. The initial correlation term in equations (4) is given by

$$\begin{aligned}I^{\mathrm{IC}}(\mathbf{p}_1;t_1,t_1')=\pm\mathrm{i}\hbar^5\mathcal{V}\int\frac{\mathrm{d}\mathbf{p}_2\,\mathrm{d}\mathbf{q}}{(2\pi\hbar)^6}V(\mathbf{q})g^{\mathrm{R}}(\mathbf{p}_1+\mathbf{q};t_1,t_0)g^{\mathrm{R}}(\mathbf{p}_2-\mathbf{q};t_1,t_0)\\ \times c'(\mathbf{p}_1+\mathbf{q},\mathbf{p}_2-\mathbf{q},\mathbf{p}_1,\mathbf{p}_2;t_0)g^{\mathrm{A}}(\mathbf{p}_2;t_0,t_1)g^{\mathrm{A}}(\mathbf{p}_1;t_0,t_1')\end{aligned}\tag{5}$$

where $c'=(\mathrm{i}\hbar)^{-2}\left[F_{12}-F_{12}^0\right]$ describes initial pair correlations (correlated part of the pair density matrix). In our calculations, we compute the self-energy $\Sigma^{\gtrless}$ in the second Born approximation and with a pseudopotential approach. The latter has been obtained by requiring that the Born approximation reproduces the full T-matrix result for the total scattering cross section which was calculated by a phase shift analysis (see e.g. [9]).

3.2. Molecular dynamics simulations

We performed MD simulations using the Verlet integration algorithm with 3000–6000 particles and periodic boundary conditions [10]. The system is first equilibrated with a velocity scaling procedure before the microcanonical evolution is computed using the modified pair interaction. Finally, we average over different initial configurations to smooth the results and to yield error estimates.

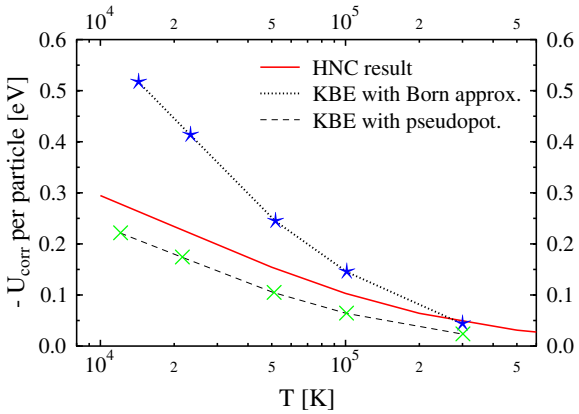

Figure 1. Correlation energy per particle for a system of singly charged particles with a density of $n = 10^{21}$ cm^{-3} interacting via a Yukawa potential with an inverse screening length of $\kappa = 0.2a_B^{-1}$. The applied approaches are HNC calculations and the Kadanoff–Baym equations using the Born approximation (stars) and a pseudopotential (crosses).

3.3. Equilibrium estimates with HNC calculations

If we are only interested in the final temperature the system evolves to, we simply have to match the total energy of the equilibrium system with the initial total energy. Since the total energy monotonically increases with temperature, the solution is unique. To calculate the correlation energy, we use the definition (3), where the binary distribution $g(r)$ is calculated from the well-known HNC equations

$$\begin{aligned} g(r) &= \exp[-V(r)/k_BT + h(r) - c(r)] \\ h(r) &= c(r) + n\int \mathrm{d}r'\, h(r-r')c(r') \\ g(r) &= 1 + h(r). \end{aligned} \tag{6}$$

This approach also describes strong coupling and is known to match Monte Carlo and MD simulations very well for equilibrium conditions and moderate coupling [11].

4. Numerical results and discussion

4.1. Equilibrium correlation energy

We first consider equilibrium predictions for the correlation energy. This will allow us to understand deviations of the nonequilibrium results and to propose improvements. Furthermore, these data are very useful for preparing a pre-perturbed, correlated initial state in the simulation from a random state. The equation of state data then provide the connection between the random state and the desired initial state.

In figure 1, the correlation energy per particle is shown for a system of singly charged particles. For the moderate coupling strengths shown in the figure, the HNC results agree very well with MD data (not shown). The results also show a good agreement between the HNC and QKT calculations for temperatures higher than $T = 2 \times 10^5$ K. For lower temperatures or larger coupling, one can observe the known fact that the Born approximation overestimates the correlation energy. It is quite interesting that the QKT can be considerably improved by a

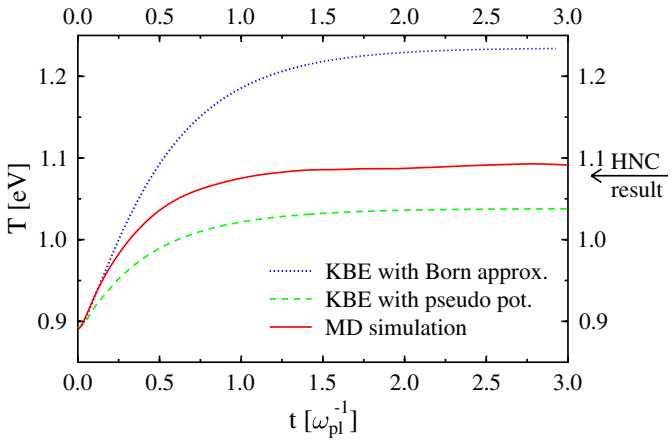

Figure 2. Time evolution of the temperature for an initially uncorrelated system with a particle density of $n = 10^{21}$ cm^{-3}. The interaction is given by a Yukawa potential with a screening parameter of $\kappa = 0.2a_B^{-1}$. The curves correspond to MD simulations and solutions of the Kadanoff–Baym equations. In the latter approach the Born approximation and the pseudopotential scheme have been used.

pseudopotential which models strong collisions. In this case, we obtain the correlation energy within 20% of the HNC result, where the QKT now underestimates the correlations for the special case shown. Since the QKT with a pseudopotential gives these improved results for the equation of state, we can hope that it also gives improved results in nonequilibrium situations.

4.2. Relaxation of nonequilibrium systems

Now we turn to the time evolution of the correlation energy. Again, we consider a model system with Yukawa-type interactions. For numerical convenience, we use particles with electron masses, but the results can be scaled to heavier particles.

First we show the relaxation of an initially uncorrelated system, which is the typical case after laser ionization of gases (see figure 2). The rapid build-up of (negative) correlation energy increases the temperature of the system during the first few plasma periods of the relaxation. All the schemes we applied show reasonable agreement for the time scale on which this process occurs, although they predict a different amount of heating. As we learned from the equilibrium calculations the Born approximation overestimates the potential energy and, therefore, the corresponding QKT approaches a much higher final temperature. Applying the pseudopotential, we find temperatures which are closer to the MD data for all times. This behaviour demonstrates that the T-matrix corrections are appropriate for nonequilibrium situations, too.

Figure 3 shows the results for the relaxation after a sudden increase of screening. That is, the system was in equilibrium for times $t < 0$, where the interaction was defined by the screening parameter κ_0. At $t = 0$ this parameter was increased by a factor of 2. Since the interaction is weaker now, the system is in an overcorrelated state. Accordingly, it relaxes to a state with less (negative) correlation energy and, therefore, a lower temperature. This behaviour is predicted by all applied schemes, and we also find similar time scales again. The amount of cooling predicted is quite different due to the different values for the correlation energy in the initial and final equilibrium states (see figure 1). The T-matrix corrected QKT yields again results closer to the MD data than the Born approximation. However, many-body

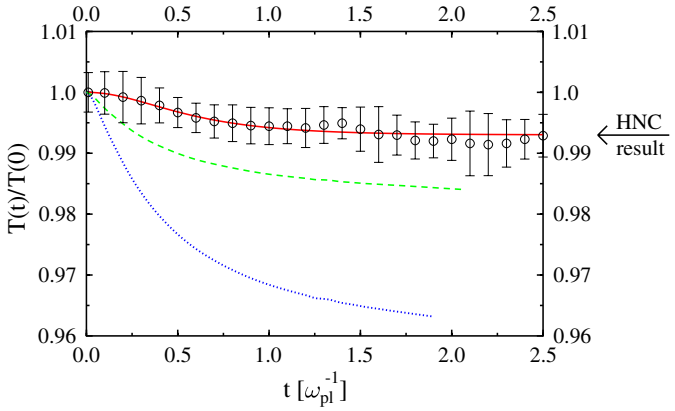

Figure 3. Temporal slope of the temperature after the inverse screening length κ was doubled at $t = 0$. Initially the system was in equilibrium with a screening parameter of $\kappa_0 = 0.319 a_B^{-1}$ and a temperature of $T_0 = 1.19 \times 10^4$ K. The same theoretical models as in figure 2 have been applied. The particle density is again $n = 10^{21}$ cm^{-3}.

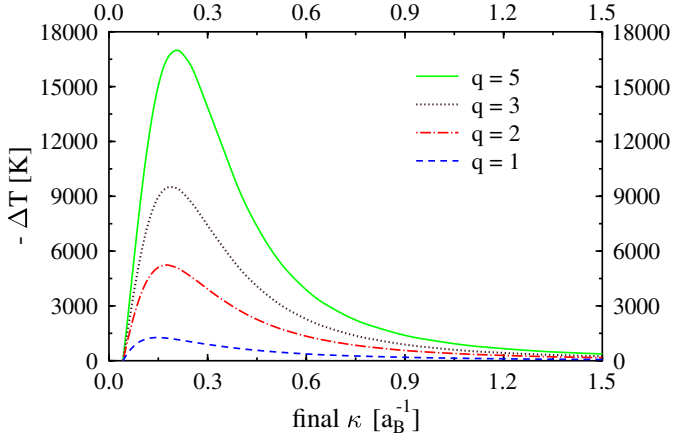

Figure 4. Temperature decrease after a sudden change of screening versus final screening parameter κ for different charges of the plasma particles q. The initial screening parameter and temperature are $\kappa_0 = 4.26 \times 10^{-2} a_B^{-1}$ and $T_0 = 1.13 \times 10^5$ K, respectively. The density is the same as in figure 3. Predictions were obtained by means of a HNC scheme which matches the total energy directly after the screening has been changed with the total energy of an equilibrium system.

effects other than strong binary collisions remain still neglected. For the screening length used in figure 3, this gives rise to an overestimation of the cooling.

The relaxation from an overcorrelated state driven by a sudden increase of screening is an interesting problem with possible applications. Therefore, the question of whether parameters exist for which the cooling is particularly strong arises. Figure 4 shows the temperature decrease for particles with different charges, which is obtained by the HNC scheme. There is clearly a region where the cooling is strongest. This behaviour is explained by the limiting cases: if screening is changed only slightly, the change in correlation energy is small; the same is true if the system is very strongly screened since the correlation energy is initially and finally very small in this case. The change in potential energy is also increased for particles

with higher charges. Therefore, moderate changes of screening and high charge states are preferable if a large amount of cooling should be achieved.

5. Conclusion

Rapid changes of the interactions in strongly coupled charged particle systems can trigger heating and cooling. A description of the relaxation process with classical MD simulations and a generalized quantum kinetic theory shows the same qualitative behaviour. Furthermore, both approaches agree on the time scale of the relaxation for classical plasmas. However, the QKT predicts larger temperature changes if the Born approximation is applied. This overestimation of the build-up or reduction of correlation energy can be considerably reduced by applying a pseudopotential that is based on an exact T-matrix treatment for strong binary scattering.

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