Relaxation to non-equilibrium in expanding ultracold neutral plasmas

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We investigate the strongly correlated ion dynamics and the degree of coupling achievable in the evolution of freely expanding ultracold neutral plasmas. We demonstrate that the ionic Coulomb coupling parameter $\Gamma_i$ increases considerably in later stages of the expansion, reaching the strongly coupled regime despite the well-known initial drop of $\Gamma_i$ to order unity due to disorder-induced heating. Furthermore, we formulate a suitable measure of correlation and show that $\Gamma_i$ calculated from the ionic temperature and density reflects the degree of order in the system if it is sufficiently close to a quasisteady state. At later times, however, the expansion of the plasma cloud becomes faster than the relaxation of correlations, and the system does not reach thermodynamic equilibrium anymore.

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Freely expanding ultracold neutral plasmas (UNPs) have attracted wide attention both experimentally and theoretically. A main motivation of the early experiments was the creation of a strongly coupled plasma, with the Coulomb coupling parameter (CCP) $\Gamma = e^2/(ak_BT) \gg 1$ (where $T$ is temperature and $a$ is the Wigner-Seitz radius). From the experimental setup of [1], the CCPs of electrons and ions were estimated to be of the orders of $\Gamma_e \approx 30$ and $\Gamma_i \approx 30000$, respectively. By changing the frequency of the ionizing laser, the electronic temperature can be varied, offering the prospect of controlling the coupling strength of the electrons and creating UNPs where either one, namely the ionic, or both components could be strongly coupled.

However, due to unavoidable heating effects [4,11,12], these hopes have not materialized yet, and only $\Gamma_e \approx 0.2$ and $\Gamma_i \approx 2$ have been confirmed. Furthermore, the evolution of the expanding plasma turns out to be a rather intricate problem of non-equilibrium plasma physics for which a clear definition of the degree of correlation is not obvious to begin with.

The goal of this letter is twofold: Firstly, we will formulate a consistent measure of correlation for expanding ultracold plasmas, and secondly we demonstrate that the strongly correlated regime with $\Gamma_i \approx 10$ for the ionic plasma component can be reached by simply waiting until the plasma has (adiabatically) expanded long enough under already realized experimental conditions. This is remarkable in the light of alternatives proposed to increase $\Gamma_i$ [12,13,14,15,16,17] which are experimentally rather involved.

Substantiating both of our statements theoretically requires the ability to propagate the plasma numerically over a long time with full account of the ionic correlations. To this end, we have developed a hybrid molecular dynamics (H-MD) method [5] for the description of ultracold neutral plasmas. In our approach, ions and recombined atoms are propagated in the electronic mean-field potential with the full ion-ion interaction taken into account. The much faster and weakly coupled electrons, on the other hand, are treated on a hydrodynamical level. Elastic as well as inelastic collisions, such as three-body recombination and electron-impact ionization, are incorporated using a Monte-Carlo procedure [16,18]. The H-MD approach accurately describes the strongly coupled ionic dynamics and therefore allows us to realistically study the plasma relaxation behavior for long times.

Assigning $\Gamma_i$ for an expanding plasma by extracting a temperature from the kinetic energy of all ions is complicated by the fact that the radial expansion contributes considerably to this energy [19]. In our approach, we can determine a local temperature from the ion velocity components perpendicular to the (radial) plasma expansion [5]. Additionally, the distribution of thermal velocities of all plasma ions is found to be well described by a Maxwell-Boltzmann distribution corresponding to an average temperature $T_i$ even at relatively early times. Experimentally, the time evolution of the average ion temperature is determined from the corresponding Doppler broadening of optical transition linewidths [14,20]. The close agreement between experiment [20] and theory (figure 1) supports both the experimental scheme of extracting an ionic temperature as well as the assignment of a temperature to the transversal ion velocities in the H-MD approach.

Remarkably, the initial relaxation of the average ion temperature exhibits temporal oscillations, in contrast to the known behavior of weakly coupled plasmas. For the latter, the timescale $t_{corr}$ of the initial build-up of ion-ion correlations is typically much smaller than the timescale $t_{rel}$ for the relaxation of the one-particle distribution function. Based on this so-called Bogoliubov functional hypothesis, which is one of the fundamental concepts in kinetic theory [21], the different relaxation processes can be separated, resulting in a monotonic behavior of the correlation energy (and hence the ion temperature) [22]. Molecular dynamics simulations of the relaxation behavior of homogeneous one-component
plasmas show that the ion temperature starts to undergo damped oscillations around its equilibrium value if both of these timescales become equal, which happens for $\Gamma_i(0) \gtrsim 0.5$ 23. Therefore, the nonmonotonic ion relaxation observed in ultracold plasmas may be seen as a direct manifestation of the violation of Bogoliubov’s hypothesis.

Compared to the homogeneous plasmas considered in 23, the oscillations of the average ionic temperature damp out much quicker in the present case. This can be attributed to the fact that the Gaussian density profile of the UNPs created in current experiments leads to a spatial dependence of the correlation timescale $\tau_{corr}$, the build-up of correlations being fastest in the center of the plasma where the density is highest, and becoming slower towards the edge of the plasma cloud. As a consequence, the local ionic temperature shows not only temporal, but also pronounced spatial oscillations, which however tend to become averaged out if the spatial average over the whole plasma cloud is taken.

Having established the approximate validity of assigning a global temperature to the plasma ions, it becomes possible to define a corresponding CCP $\Gamma_i$. While the initial ion relaxation reveals some interesting strong-coupling effects as discussed above, disorder-induced heating 7,12 drives the ion component to the border of the strongly coupled fluid regime $\Gamma_i \approx 2$ and therefore limits the amount of correlations achievable in UNPs. However, so far this could be verified only for the early stage of the plasma evolution 7,12,14. The present H-MD approach allows us to study also the long-time behavior of the ion coupling.

In figure 2, we show $\Gamma_i$ (solid line) as a function of $\tau = \omega_p,0 t$ for a plasma with $N_i(0) = 5 \cdot 10^4$, $\bar{\rho}_i(0) = 1.1 \cdot 10^9$cm$^{-3}$ and $T_e(0) = 50$K, determined in a central sphere with a radius of twice the root-mean-square radius of the expanding plasma. (In the following, dimensionless units are used where time is scaled with the initial plasma frequency $\omega_p = \omega_p(t = 0)$ and $\omega_p = 4\pi e^2 \bar{\rho}_i/m_i$.) As can be seen in the inset, $\Gamma_i$ quickly drops down to $\Gamma_i \approx 2$. After this initial stage, however, $\Gamma_i$ starts to increase again due to the adiabatic cooling of the ions during the expansion. Indeed, CCPs of more than 10 are realized at later stages of the system evolution, showing that cold plasmas well within the strongly coupled regime are produced with the present type of experiments.

Neglecting the influence of the changing correlation energy as well as inelastic processes, the adiabatic law for the plasma expansion 24 yields $T_i \Gamma_i^{-2/3} = const.$. Hence, $\Gamma_i$ should increase $\propto \bar{\rho}_i^{-1/3}$ as the plasma expands, ultimately leading to coupling strengths of $10^2$ or even larger at very long times. For a classical plasma in thermodynamical equilibrium, the Coulomb coupling parameter is a direct measure of the amount of correlations, and properties such as pair correlation functions etc. can be parametrized by this single quantity. However, the UNPs created in the present type of experiments are non-equilibrium systems. Initially, e.g., they are created in a completely uncorrelated state, so that the high value of $\Gamma_i$ caused by the ultralow temperature of the ions has no relation at all with the correlation properties of the system. At later times, the system relaxes towards a local equilibrium. However, the plasma is freely expanding, and hence constantly changing its steady state. Thus, the plasma is in a non-equilibrium state at all times, and one must ask to what extent $\Gamma_i$ really parametrizes the correlations present in the plasma.
To this end, we compare $\Gamma_i$ as obtained above with an alternative value $\tilde{\Gamma}_i$ (dashed line in figure 2) parametrizing correlation properties of the plasma. As in [12], we have calculated the distribution $P(r/a_{\text{loc}})$ of interionic distances rescaled by the local Wigner radius. These distribution functions are fitted to the known pair correlation function $g(r/a, \Gamma_i)$ of an equilibrium plasma given in [25] (figure 3). From the fit, a value $\tilde{\Gamma}_i$ is extracted at several times. As can be seen in figure 3 at very early times the distribution of scaled interionic distances is not very well fitted to a pair correlation function of a homogeneous plasma in equilibrium. Again, this is due to the fact that the system is far away from its steady state, and a single parameter does not describe the correlation properties of the plasma in an adequate way. However, the interionic distances quickly relax, and they are well described by a pair correlation function of an equilibrium system at later times. Hence, we conclude that the value of $\tilde{\Gamma}_i$ is suitable for parametrizing the correlation properties of the plasma cloud once it came sufficiently close to equilibrium, and that it indeed reflects the degree of coupling in the plasma.

Comparing $\Gamma_i$ and $\tilde{\Gamma}_i$ in figure 2 several conclusions can be drawn. As discussed above, and has been well known before, in the very early phase of the system evolution there is no relation between $\Gamma_i$ and $\tilde{\Gamma}_i$ since the plasma is too far away from equilibrium. As the plasma relaxes towards this equilibrium, $\Gamma_i$ and $\tilde{\Gamma}_i$ rapidly approach each other, showing that during this stage $\Gamma_i$ is a good measure for the correlation properties of the ions.

In particular, the correlations building up in the system are indeed those of a strongly coupled plasma with a CCP well above unity. Moreover, the transient oscillations characteristic of the relaxation process which are apparent in $\Gamma_i$ also appear in $\tilde{\Gamma}_i$, however with a “phase shift” of $\pi$. This phase shift is due to the fact that a minimum in the temperature means a maximum in $\Gamma_i$ for a given density. Since total energy is conserved, a minimum in the thermal kinetic energy corresponds to a maximum in the potential energy, i.e. to an increased number of pairs of closely neighboring ions, and therefore to a pair correlation function with enhanced probability for small distances and consequently a minimum in $\Gamma_i$.

At later times, both curves diverge again and the plasma evolves back towards an undercorrelated state. At first sight, this seems very surprising since the plasma should relax towards equilibrium rather than away from it. However, as argued above, the plasma is freely expanding and the corresponding equilibrium properties are constantly changing. We interpret figure 2 as being again evidence for the break-down of the Bogoliubov assumption of a separation of timescales, in this case of the correlation time $\tau_{\text{corr}}$ and the hydrodynamical timescale $\tau_{\text{hyd}}$, i.e. the characteristic time for the plasma expansion.

The timescale $\tau_{\text{hyd}}$ may be determined from the relative change of macroscopic plasma parameters, such as the ion temperature or density. Due to the transient oscillations of the ion temperature we choose the ion density to characterize the change of the plasma properties (other choices such as, e.g., $a \propto \tilde{\rho}_i^{-1/3}$ lead to the same conclusions since they result in a simple constant proportionality factor $1/\alpha$ of order unity in the expression for $\tau_{\text{hyd}}$). Then

$$\tau_{\text{hyd}} \approx \frac{1}{\alpha} \tilde{\rho}_i = \frac{1}{\alpha} \left(1 + \frac{\tau^2}{\tau_{\text{exp}}^2}\right) \frac{\tau_{\text{exp}}}{3\tau},$$

where we have used the selfsimilar solution for the collisionless quasineutral plasma expansion [24] with $\tau_{\text{exp}} = \sigma(0) \omega_{p,0} \sqrt{m_i/(k_B T_e^*)}$. On the other hand, binary correlations are known to relax on the timescale of the inverse of the plasma frequency in the strongly coupled regime [25] for an initially uncorrelated state, and somewhat slower if the initial state already exhibits spatial ion correlations [12], $\tau_{\text{corr}} \gtrsim \omega_{p,0}/\omega_p$. The selfsimilar plasma expansion then yields

$$\tau_{\text{corr}} = \left(1 + \frac{\tau^2}{\tau_{\text{exp}}^2}\right)^{3/4}.$$  

Therefore, $\tau_{\text{corr}}$ is initially much smaller than $\tau_{\text{hyd}}$, but ultimately exceeds $\tau_{\text{hyd}}$ as the plasma expands, leading to an inevitable break-down of the Bogoliubov condition. Consequently, the build-up of correlations in the system cannot follow the changing equilibrium anymore, and correlations freeze out as indicated by the leveling-off of $\Gamma_i$ to a constant value.

Equating $\tau_{\text{corr}}$ and $\tau_{\text{hyd}}$ as given above yields

$$\tau_p^* = 2^{-1/2} \tau_{\text{exp}} x^2 \sqrt{1 + \sqrt{1 + 4x^{-4}}} \approx \tau_{\text{exp}} x^2$$

![Figure 3: “Pair correlation functions” of the plasma of fig. 2 at four different times $\tau = 0.54$, $\tau = 1.1$, $\tau = 10$ and $\tau = 60.7$. The $\Gamma_i$ indicated in the figure is obtained by fitting the distribution of scaled interionic distances (dots) with pair correlation functions for a homogeneous plasma given in [25] (solid line).]
percent relative deviation between \( \Gamma \) is a linear fit. The error bars show the range of two to eight \( \tau \) translations start to freeze out as a function of \( \tau \), with \( \Gamma \) plasma reaches a state well inside the strongly coupled regime, with \( \Gamma \sim 10 \). This is due to the correlation freeze-out described above. Still, the pair correlation functions can well be fitted to those of a CCP due to the correlation properties of an equilibrium plasma at this stage (fig. 3(d)), in contrast to the behavior at early times. This is due to the fact that the system went through a phase where equilibrium changes, thus the system cannot equilibrate anymore and correlations freeze out. Clearly, ultracold neutral plasmas are unique systems that evolve through different thermodynamical stages of non-equilibrium and (near)-equilibrium behavior. Their further experimental and theoretical study thus should provide new stimulus for plasma physics as well as for non-equilibrium thermodynamics.

FIG. 4: \( \tau^\star \) as a function of \( \tau^3_{\text{exp}} \) for different initial conditions:

- \( N_i = 5 \cdot 10^4, \rho_i = 1.1 \cdot 10^9 \text{cm}^{-3}, T_e = 50K; N_i = 4 \cdot 10^4, \rho_i = 3 \cdot 10^9 \text{cm}^{-3}, T_e = 45K; N_i = 5 \cdot 10^4, \rho_i = 10^9 \text{cm}^{-3}, T_e = 33.3K; N_i = 8 \cdot 10^4, \rho_i = 10^9 \text{cm}^{-3}, T_e = 38K; N_i = 10^4, \rho_i = 1.3 \cdot 10^9 \text{cm}^{-3}, T_e = 33.3K \) (left to right). The solid line is a linear fit. The error bars show the range of two to eight percent relative deviation between \( \Gamma_1 \) and \( \Gamma_1 \) for determining \( \tau^\star \).

with \( x \equiv \tau_{\text{exp}}/(3\alpha) \) as the time when both timescales become equal. In fig. 4 we show the time \( \tau^\star \) when correlations start to freeze out as a function of \( \tau^3_{\text{exp}} \), where \( \tau^\star \) is determined as the time when the relative deviation between \( \Gamma_1 \) and \( \Gamma_1 \) is less than five percent for the last time.

The linear correlation visible in the figure strongly supports our reasoning that it is the cross-over of timescales that is responsible for the freeze-out of correlations.

Thus, we may conclude that the system ultimately approaches a non-equilibrium undercorrelated state again due to the correlation freeze-out described above. Still, the pair correlation functions can well be fitted to those of an equilibrium plasma at this stage (fig. 3(d)), in contrast to the behavior at early times. This is due to the fact that the system went through a phase where equilibrium spatial correlations have developed which are preserved during the further evolution of the plasma. Hence, the system has the correlation properties of an equilibrium system, however “with the wrong temperature”.

In conclusion, we have simulated an expanding ultracold neutral plasma with special attention to the formation of ionic correlations. We have found that several phases can be distinguished in the evolution of the system. First, a quick relaxation to local equilibrium occurs, together with its characteristic transient oscillations of the ion temperature. After that, the system is close to a — changing — local equilibrium. In this stage, a CCP defined from temperature and density indeed is a measure for correlations in the plasma. Moreover, and this has, to our knowledge, not been pointed out so far, the plasma reaches a state well inside the strongly coupled regime, with \( \Gamma_1 \gtrsim 10 \). Ultimately, the timescale for equilibration becomes longer than the timescale on which the equilibrium changes, thus the system cannot equilibrate anymore and correlations freeze out. Clearly, ultracold neutral plasmas are unique systems that evolve through different thermodynamical stages of non-equilibrium and (near)-equilibrium behavior. Their further experimental and theoretical study thus should provide new stimulus for plasma physics as well as for non-equilibrium thermodynamics.

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