Relaxation to Nonequilibrium 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 Γ_i increases considerably in later stages of the expansion, reaching the strongly coupled regime despite the well known initial drop of Γ_i to order unity due to disorder-induced heating. Furthermore, we formulate a suitable measure of correlation and show that Γ_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) [1] have attracted wide attention both experimentally [2–5] and theoretically [6–10]. 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 $\Gamma_i \approx 30\,000$, 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 [6,11,12] 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 nonequilibrium 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: first, we will formulate a consistent measure of correlation for expanding ultracold plasmas, and second, 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 Γ_i [12–17] 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 [9] 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 Γ_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 [9]. 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 [19,20]. The close agreement between experiment [20] and theory (Fig. 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 time scale t_{corr} of the initial buildup of ion-ion correlations is typically much smaller than the time scale 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 time scales become

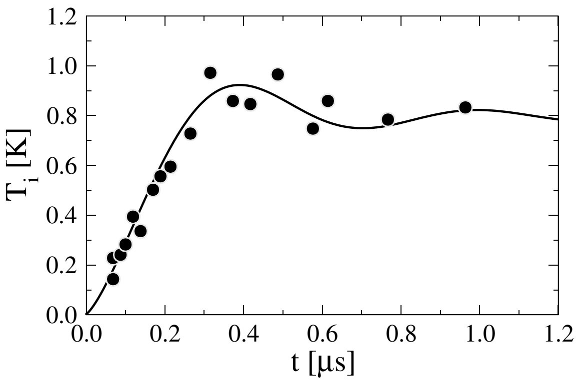

FIG. 1. Ion temperature from the H-MD simulation of a plasma of 10^6 Sr ions with initial peak density $\rho_0(0) = 2 \times 10^9$ cm^{-3} and electron temperature $T_e(0) = 38$ K (solid line), compared to experimental results (dots) [20]. The fact that the experimental ion number is about a factor of 10 larger than in our calculation does not affect the time evolution of the ionic temperature, since there is no significant adiabatic ion cooling on the time scale considered in Fig. 1.

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 time scale t_{corr}, the buildup 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 Γ_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 [7,12,19]. The present H-MD approach allows us to study also the long-time behavior of the ion coupling.

In Fig. 2, we show Γ_i (solid line) as a function of $\tau = \omega_{\text{p},0} t$ for a plasma with $N_i(0) = 5 \times 10^4$, $\bar{\rho}_i(0) = 1.1 \times 10^9$ cm^{-3}, and $T_e(0) = 50$ K, determined in a central sphere with a radius of twice the initial rms radius $\sigma(0)$ of the plasma. (In the following, dimensionless units are used where time is scaled with the initial plasma frequency $\omega_{\text{p},0} = \omega_{\text{p}}(t = 0)$ and $\omega_{\text{p}} = \sqrt{4\pi e^2 \bar{\rho}_i / m_i}$.) As can be seen in the inset, Γ_i quickly drops down to $\Gamma_i \approx 2$. After this initial stage, however, Γ_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 $T_i \bar{\rho}_i^{-2/3} =$ const. Hence, Γ_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 nonequilibrium systems. Initially, e.g., they are created in a completely uncorrelated state, so that the high value of Γ_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 nonequilibrium state at all times, and one must ask to what extent Γ_i really parametrizes the correlations present in the plasma.

To this end, we compare Γ_i as obtained above with an alternative value $\tilde{\Gamma}_i$ (dashed line in Fig. 2) parametrizing correlation properties of the plasma. As in [15], we have

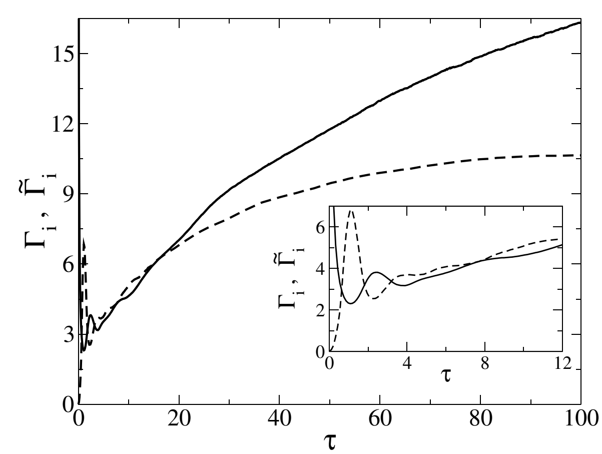

FIG. 2. Ionic Coulomb coupling parameter for a plasma with $N_i(0) = 5 \times 10^4$, $\bar{\rho}_i(0) = 1.1 \times 10^9$ cm^{-3}, and $T_e(0) = 50$ K. The solid line shows the CCP calculated from the average temperature and density; the dashed line marks the CCP extracted from pair correlation functions (see text). Inset: blowup of the short-time behavior.

calculated the distribution $P(r/a_{\mathrm{loc}})$ of interionic distances rescaled by the local Wigner radius. These distribution functions are fitted to the known pair correlation function $g(r/a, \tilde{\Gamma}_i)$ of an equilibrium plasma given in [25] (Fig. 3). From the fit, a value $\tilde{\Gamma}_i$ is extracted at several times. As can be seen in Fig. 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 Γ_i and $\tilde{\Gamma}_i$ in Fig. 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 Γ_i and $\tilde{\Gamma}_i$ since the plasma is too far away from equilibrium. As the plasma relaxes towards this equilibrium, Γ_i and $\tilde{\Gamma}_i$ rapidly approach each other, showing that during this stage Γ_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 Γ_i also appear in $\tilde{\Gamma}_i$, however, with a "phase shift" of π. This phase shift is due to the fact that a minimum in the temperature means a maximum in Γ_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 $\tilde{\Gamma}_i$.

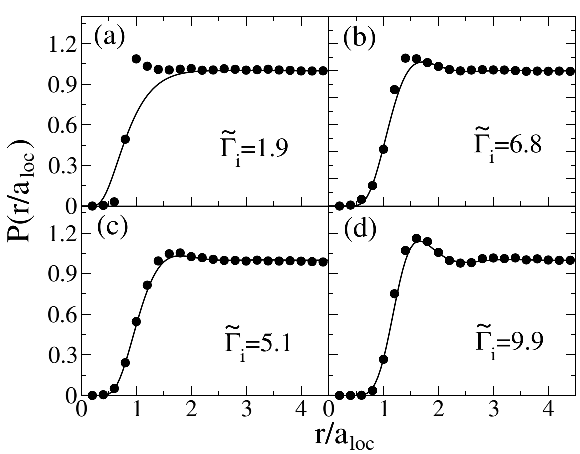

FIG. 3. "Pair correlation functions" of the plasma of Fig. 2 at four different times $\tau = 0.54$, $\tau = 1.1$, $\tau = 10$, and $\tau = 60.7$. The $\tilde{\Gamma}_i$ indicated in the figure is obtained by fitting the distribution of scaled interionic distances (dots) with pair correlation functions for a homogeneous plasma given in [25] (solid line).

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 Fig. 2 as being again evidence for the breakdown of the Bogoliubov assumption of a separation of time scales, in this case of the correlation time τ_{corr} and the hydrodynamical time scale τ_{hyd}, i.e., the characteristic time for the plasma expansion.

The time scale τ_{hyd} may be determined from the relative change of macroscopic plasma parameters, such as the ion temperature or density. Because of 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., $a \propto \bar{\rho}_i^{-1/3}$ lead to the same conclusions since they result in a simple constant proportionality factor $1/\alpha$ of order unity in the expression for τ_{hyd}). Then

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

where we have used the self-similar solution for the collisionless quasineutral plasma expansion [24] with $\tau_{\mathrm{exp}} = \sigma(0)\omega_{\mathrm{p},0}\sqrt{m_i/(k_B T_e)}$. On the other hand, binary correlations are known to relax on the time scale of the inverse of the plasma frequency in the strongly coupled regime [23] for an initially uncorrelated state, and somewhat slower if the initial state already exhibits spatial ion correlations [12], $\tau_{\mathrm{corr}} \gtrsim \omega_{\mathrm{p},0}/\omega_{\mathrm{p}}$. The self-similar plasma expansion then yields

$$\tau_{\mathrm{corr}} = (1 + \tau^2/\tau_{\mathrm{exp}}^2)^{3/4}. \tag{2}$$

Therefore, τ_{corr} is initially much smaller than τ_{hyd}, but ultimately exceeds τ_{hyd} as the plasma expands, leading to an inevitable breakdown of the Bogoliubov condition. Consequently, the buildup of correlations in the system cannot follow the changing equilibrium anymore, and correlations freeze out as indicated by the leveling off of $\tilde{\Gamma}_i$ towards a constant value $\tilde{\Gamma}_i \approx 10$.

Equating τ_{corr} and τ_{hyd} as given above yields

$$\tau_\rho^* = 2^{-1/2}\tau_{\mathrm{exp}}x^2\sqrt{1 + \sqrt{1 + 4x^{-4}}} \approx \tau_{\mathrm{exp}}x^2, \tag{3}$$

with $x \equiv \tau_{\mathrm{exp}}/(3\alpha)$, as the time when both time scales become equal. In Fig. 4, we show the time τ_Γ^* when correlations start to freeze out as a function of τ_{exp}^3, where τ_Γ^* is determined as the time when the relative deviation

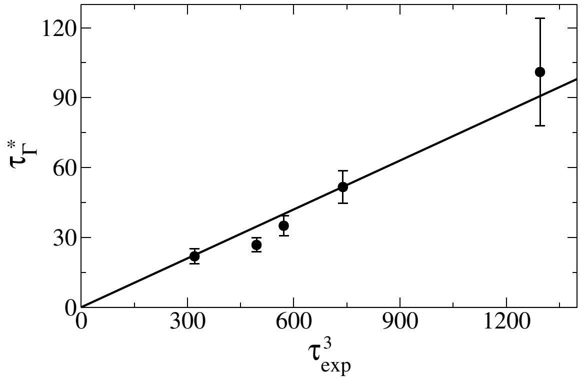

FIG. 4. τ_Γ^* as a function of $\tau_{\rm exp}^3$ for different initial conditions: $N_i = 5 \times 10^4$, $\bar{\rho}_i = 1.1 \times 10^9$ cm^{-3}, $T_e = 50$ K; $N_i = 4 \times 10^4$, $\bar{\rho}_i = 3 \times 10^9$ cm^{-3}, $T_e = 45$ K; $N_i = 5 \times 10^4$, $\bar{\rho}_i = 10^9$ cm^{-3}, $T_e = 33.3$ K; $N_i = 8 \times 10^4$, $\bar{\rho}_i = 10^9$ cm^{-3}, $T_e = 38$ K; $N_i = 10^5$, $\bar{\rho}_i = 1.3 \times 10^9$ cm^{-3}, $T_e = 33.3$ K (left to right). The solid line is a linear fit. Error bars show the range of 2% to 8% relative deviation between Γ_i and $\tilde{\Gamma}_i$ for determining τ_Γ^*.

between Γ_i and $\tilde{\Gamma}_i$ is less than 5% for the last time. The linear correlation visible in the figure strongly supports our reasoning that it is the crossover of time scales that is responsible for the freeze-out of correlations.

Thus, we may conclude that the system ultimately approaches a nonequilibrium 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 $\Gamma_i \gtrsim 10$. Ultimately, the time scale for equilibration becomes longer than the time scale on which the equilibrium changes; thus the system cannot equilibrate anymore and correlations freeze out. In all the calculations summarized in Fig. 4, $\tilde{\Gamma}_i$ approaches a value of $\tilde{\Gamma}_i(t \to \infty) \approx 10$, suggesting that this might be an approximate upper bound for the degree of correlations achievable in the current experimental setup. Clearly, ultracold neutral plasmas are unique systems that evolve through different thermodynamical stages of nonequilibrium and (near)-equilibrium behavior. Their further experimental and theoretical study thus should provide new stimulus for plasma physics as well as for nonequilibrium thermodynamics.

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