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Plasma formation from ultracold Rydberg gases

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Recent experiments have demonstrated the spontaneous evolution of a gas of ultracold Rydberg atoms into an expanding ultracold plasma, as well as the reverse process of plasma recombination into highly excited atomic states. Treating the evolution of the plasma on the basis of kinetic equations, while ionization/excitation and recombination are incorporated using rate equations, we have investigated theoretically the Rydberg-to-plasma transition. Including the influence of spatial correlations on the plasma dynamics in an approximate way, we find that ionic correlations change the results quantitatively but not qualitatively.

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Advances in cooling and trapping of neutral gases have opened up a new branch of atomic physics, namely, dynamics in ultracold ($T \ll 1$ K) systems. Ultracold neutral plasmas have been produced from a small cloud of laser-cooled atoms confined in a magneto-optical trap [1–5]. At NIST [1–3], a plasma was produced by photoionizing laser-cooled Xe atoms with an initial ion temperature of about 10 μK. Tuning the frequency of the ionizing laser, the initial electron energy E_e was varied corresponding to a temperature range 1 K$<E_e/k_B<$1000 K, and the subsequent expansion of the plasma into the surrounding vacuum was studied systematically. A significant amount of recombination was observed as the plasma expands, leading to the formation of Rydberg atoms from the plasma. In a complementary type of experiment [4,5], ultracold atoms were laser excited into high Rydberg states rather than directly ionized. In these experiments, also the reverse process has been observed, namely, the spontaneous evolution of the Rydberg gas into a plasma.

An issue in the theoretical considerations [6–9] stimulated by the experiments has been the question whether the evolving plasma is strongly coupled or not. Due to the very low initial temperature, the electron Coulomb coupling parameter $\Gamma_e(t=0)$ is found to be significantly greater than 1 [$\Gamma_e = e^2/(a k_B T_e)$, where a is the Wigner-Seitz radius]. On the other hand, it has been pointed out in Ref. [6] that, for the initial conditions of the NIST experiments [1–3], the development of equilibrium electron-electron correlations leads to a rapid heating of the electron gas, bringing Γ_e down to order unity on a very short time scale. The molecular-dynamics calculation [6] was limited to a short time interval ($\approx$100 ns) in the initial stage of the plasma evolution due to the large numerical effort required. For the first quantitative comparison with experiment, the plasma dynamics has been modeled within a kinetic approach, while ionization, excitation, and recombination have been treated by a separate set of rate equations [7]. The electron dynamics was described in an adiabatic approximation with the atom and ion temperatures set to zero. Since this model does only account for the mean-field potential created by the charges, possible correlation heating could not be described. Nevertheless, due to heating by three-body recombination, Γ_e does not exceed a value of $\approx$0.2 during the plasma expansion. Hence, the influence of electronic correlations on the plasma dynamics could be neglected. With zero ionic temperature, the *ionic* Coulomb coupling parameter, however, is infinite in the framework of this model. Consequently, the role of ion-ion correlations could not be estimated.

Our description is similar to that of Ref. [7] with small differences, e.g., inclusion of black-body radiation as a source for photoionization. This is necessary to describe the initial ionization of the Rydberg gas in the experiments [4,5] since the ionization rate through cold atom-atom collisions is much lower at these densities. More importantly, in addition to Ref. [7], we allow for spatial correlation effects. Briefly, we use a set of kinetic equations for the plasma expansion, combined with rate equations for the description of ionization/excitation and recombination. The kinetic equations are derived from the first equation of the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy, which yields the standard Vlasov equation for the evolution of a collisionless plasma, augmented by additional terms accounting for spatial correlations. Since the electronic Coulomb coupling parameter is well below unity under all experimental conditions realized so far (as we have convinced ourselves including electronic correlations on the basis of two-component Debye-Hückel theory; see also Ref. [7]), we neglect them and treat the ions as a one-component plasma embedded in a neutralizing background.

Assuming that the correlation function vanishes at distances larger than the correlation length a_c and that the spatial density varies slowly over a distance a_c, we find that the force induced by ionic correlations can be expressed in terms of the local ion-ion correlation energy per particle,

$$u_{ii}(\mathbf{r}) = \frac{e^2}{2}\rho_i(\mathbf{r}) \int d\mathbf{y} \frac{g_{ii}(y;\mathbf{r})}{y}, \tag{1}$$

with the correlation function $g(y)$ and the density ρ_i related to the one-particle distribution function f_i by $\rho_i = \int d\mathbf{v} f_i$. The correlation energy for a one-component plasma in thermal equilibrium is a function of the Coulomb coupling parameter only [10], and simple analytical formulas have been given for a wide range of Γ. We use

$$u_{ii}^{(\mathrm{eq})} = -\frac{9}{10} k_B T_i \Gamma_i = -\frac{9e^2}{10}\left(\frac{4}{3}\pi\rho_i\right)^{1/3}, \tag{2}$$

derived from the ion sphere model, which is a good approximation for $\Gamma > 1$ [11].

The time scale for relaxation towards the individual equilibrium of electrons and ions, respectively, is determined by the corresponding inverse plasma frequency $\omega_{\mathrm{p}}^{-1} = \sqrt{m/4\pi\rho e^2}$ [12]. Under typical conditions of the experiments [1–4], it is of the order of 10^{-9} s for electrons, while equilibration of the ions takes between 10^{-7} and 10^{-5} s depending on ρ. These time scales for equilibration have to be seen relative to the expansion time of the plasma, which is of the order of several 10^{-6} s. Hence, an adiabatic approximation for the electron distribution function can be safely applied. The kinetic equation for the electronic component of the plasma then yields a relation allowing one to express the mean-field potential in terms of the density ρ_e, which can be inserted into the corresponding equation for the ions.

The much slower equilibration of the ions renders an adiabatic approximation *a priori* difficult. However, even in situations where static parameters, e.g., relevant masses, periods, or rates, speak against an adiabatic treatment, it is sometimes made possible through dynamical adiabaticity (see, e.g., Ref. [13]). Of course, this can only be verified *a posteriori* by more elaborate calculations without adiabatic approximation. For now, we simply assume an adiabatic time evolution of the plasma, i.e., we estimate the ionic correlation energy from its equilibrium value [Eq. (2)] and use a homogeneous ion temperature at all times. This is the opposite limit to a zero ionic temperature which would never lead to equilibration. Hence, our results are expected to clarify whether (and to what extent) correlation effects can have influence on the dynamics at all.

For typical temperatures and densities, the Debye length is considerably smaller than the size of the plasma. Moreover, the overall charge of the plasma is small compared to the number of free charges. (In the experiment [3], typically more than 80% of the electrons are trapped. For the cases starting from a Rydberg gas [5], we estimate the fraction of trapped electrons to be roughly 95%.) Together, this justifies the quasineutral approximation $\rho_e = \rho_i = \rho$ [14]. Hence, a closed equation for the ion distribution function is obtained, and the resulting equation *without the correlation term* permits self-similar analytical solutions [14]. One of them is the Gaussian profile describing the initial state in the experiments under consideration. Exact self-similar solutions exist even in the case of an additional linear force [15]. This condition is satisfied by the correlation force to a good approximation save for the outer periphery of the plasma [16]. Hence, we can write

$$f_i \propto \exp\left(-\frac{r^2}{2\sigma^2} - \frac{m_i[\mathbf{v}-\mathbf{w}(\mathbf{r})]^2}{2k_{\mathrm{B}}T_i} \right), \tag{3}$$

where σ is the width of the spatial density distribution and $\mathbf{w}(\mathbf{r}) = \gamma\mathbf{r}$ is the hydrodynamic velocity (see also Ref. [7]). Substitution of Eq. (3) into the ionic kinetic equation finally leads to the set of equations

$$\frac{d\sigma^2}{dt} = 2\gamma\sigma^2,$$

$$\frac{d\gamma}{dt} + \gamma^2 = \frac{N_e}{M\sigma^2}[k_{\mathrm{B}}(T_e + T_i) + W_c/N_e], \tag{4}$$

where $W_c = 1/3\int d\mathbf{r}\, \rho\partial(u_{ii}\rho)/\partial\rho$ arises from the correlation pressure and M is the total mass of the plasma. The thermal energy is determined from the total energy of the system,

$$E_{\mathrm{tot}} = \frac{3}{2}N_e k_{\mathrm{B}}(T_e + T_i) + \frac{3}{2}M\gamma^2\sigma^2 + U_c, \tag{5}$$

i.e., the sum of the kinetic energy and the correlation energy $U_c = \int d\mathbf{r} u_{ii}\rho$. The set of equations (4) reduces to the one used in Ref. [7] if the correlation term is dropped and $T_i \equiv 0$. Very recently, it has been shown that this simple ansatz provides a good description of the plasma dynamics even if some of the assumptions, such as the quasineutrality of the plasma and the Gaussian distribution of the densities, only hold approximately [17].

The low-temperature enhancement of the expansion velocity of the ultracold plasma produced in Ref. [2] can be well modeled by combining a hydrodynamic description of the expansion dynamics with conventional rate equations accounting for inelastic collisions between the plasma particles and Rydberg atoms [7]. In our calculations, we include bound-bound transitions by electron-impact excitation and deexcitation, and bound-free transitions by three-body recombination, electron-impact ionization, and black-body radiation. We use the collision rate coefficients from Ref. [18] and black-body photoionization is described by first-order perturbation theory [19], using an asymptotic expression for the atomic oscillator strengths [20]. We have estimated the rate of ionization by quasistatic dipolar atom-atom interaction including threshold lowering on the basis of Ref. [21], and found it to be more than two orders of magnitude smaller than the black-body ionization rate.

Finally, the question of the expansion velocity of the Rydberg atoms has to be addressed, since they are not driven by the Coulomb drag of the expanding electrons. Under typical conditions ($\rho_e = 10^9$ cm^{-3}, $T_e = 20$ K), the time scale for electron-atom collisions is of the order of 10^{-8} s. Therefore, in the course of the expansion of the plasma (10^{-5} s), any given atom with binding energy of several $k_{\mathrm{B}}T$ will recombine and reionize many times. In addition, collisions between ions and atoms significantly equilibrate the hydrodynamical velocities even for lower Rydberg states [22]. Following this reasoning, we use the approximation of assuming equal hydrodynamical velocities and density profiles for the ions and atoms. This implies that the expansion of the neutral Rydberg atoms can be simply taken into account by changing the mass of the ions to an effective mass M in Eq. (4), which is the total mass of the *total* system (plasma and atoms).

As a test, we have simulated the expansion of an initially fully ionized plasma as in Refs. [2,3] (the atoms not ionized by the laser pulse are bound so deeply that they can be neglected for the current purpose). In accordance with previous

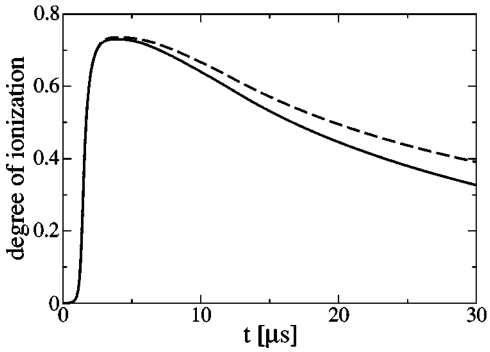

FIG. 1. Time evolution of the degree of ionization for the following initial conditions: atom density $\rho=8\times10^{9}$ cm^{-3}, atom temperature $T_a=140$ μK, plasma width $\sigma=60$ μm, and initial principal quantum number of the Rydberg atoms, $n_0=70$. Results are shown with ionic correlations (solid) and without ionic correlations (dashed).

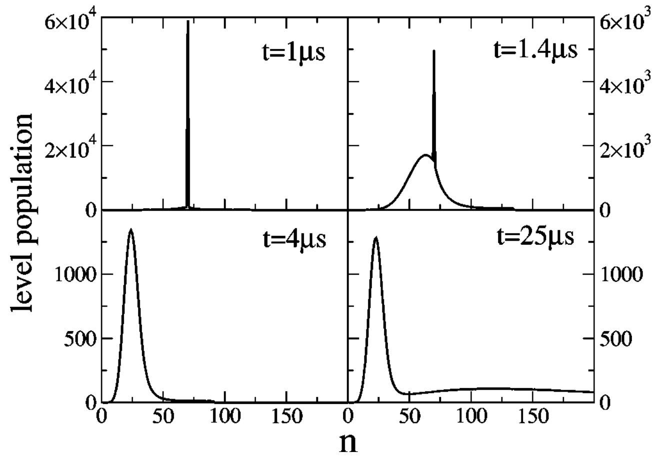

FIG. 3. Level distribution of Rydberg atoms after 1 μs, 1.4 μs, 4 μs, and 25 μs.

calculations [7], we find quantitative agreement for the electron energy dependence of the asymptotic expansion velocity. Furthermore, the calculations qualitatively reproduce the nonmonotonic time dependence of the number of *detected* atoms observed in Ref. [3].

The results of our calculations for the case starting from Rydberg atoms are summarized in Figs. 1–3. Figure 1 shows the degree of ionization of the Rydberg gas, i.e., the plasma fraction of the system, as a function of time during the expansion of the system. For comparison, the result without correlation [$U_c=T_i\equiv0$ in Eqs. (4) and (5)] is also shown. As has been observed experimentally, it takes of the order of two to three microseconds before significant plasma formation occurs, which sets in with an ionization avalanche. Ionization mechanisms other than black-body ionization, such as atom-atom collisions, etc., may affect the preavalanche behavior quantitatively. However, we do not expect a significant influence on the ionization avalanche itself, which is due to the rapid increase in the number of free electrons produced by electron-impact ionization at this stage, nor on the later stages of the plasma evolution. Note that the early time development including the ionization avalanche is obviously not influenced by the ionic correlations. Their development requires a significant amount of charged plasma particles which are only available after the ionization avalanche.

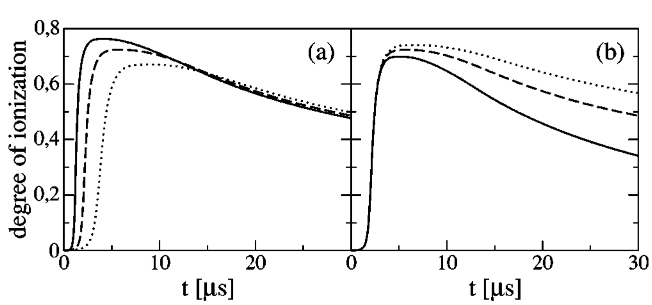

FIG. 2. Degree of ionization as a function of time for various initial conditions ($n_0=70$). (a) $\sigma=80$ μm fixed, $\rho=2.5\times10^{9}$ (dotted), 5×10^{9} (dashed), 1×10^{10} cm^{-3} (solid); (b) $\rho=5\times10^{9}$ cm^{-3} fixed, $\sigma=60$ (solid), 80 (dashed), and 100 μm (dotted).

The production of initial "seed charges" by black-body radiation does not depend on ionic correlations. The ionization avalanche appears somewhat (≈1 μs) earlier than in the experiment since in reality the first electrons produced by the black-body radiation will leave the atom cloud until a sufficiently strong positive space charge builds up to trap the electrons. With increasing number of free charges, the plasma forms quickly, followed by a partial back evolution into a Rydberg gas. This last stage of the expansion is qualitatively similar to the expansion of a directly produced plasma [1–3] under similar conditions. Figure 1 shows that this part of the gas dynamics may indeed be affected by ion-ion correlation, which leads to a more efficient recombination in the final stage of the plasma evolution. One sees from Eqs. (4) and (5) that W_c and U_c lead to an acceleration [$2/9U_c$ if Eq. (2) is used for u] in addition to the ideal thermal electron pressure $k_{\mathrm{B}}T_e$. Hence, adiabatic electron cooling during the expansion is faster than without correlations. In turn, recombination is accelerated, since the corresponding rate strongly increases with decreasing temperature. Corresponding effects of the same order of magnitude are also found for the experiments [2,3], where they do not significantly affect the expansion dynamics of the plasma part because the correlations mainly influence the recombination into high-lying states at later stages of the evolution. Since these are very weakly bound, they hardly influence the kinetic energy of the system which determines the expansion velocity. Hence, good agreement is found between calculations with and without inclusion of correlation effects as long as quantities related to the plasma expansion are compared.

Figure 2 illustrates the dependence of the plasma evolution on the initial conditions. For fixed initial width of the Rydberg atom cloud [Fig. 2(a)], the plasma formation occurs earlier with increasing density, due to the fact that ionization is faster and more efficient at higher densities. The later stages of the evolution, on the other hand, depend only weakly on the density. This may be attributed to the fact that in the evolution equation (4), ρ enters only in the form N/M, i.e., essentially as the degree of ionization. For fixed initial density ρ, the ionization avalanche occurs at the same time, in accordance with the argument made previously [Fig. 2(b)]. However, for smaller width, the plasma expands faster, leading ultimately to lower electron temperatures and increased recombination.

Figure 3 shows the population of Rydberg levels in the initial and final stages of the expansion, as well as at the time of the maximum degree of ionization. Initially, all atoms are prepared with a principal quantum number $n_0 = 70$. At later times, the interplay between ionization, recombination, and exciting and deexciting collisions leads to a decrease of the average excitation. This decrease in excitation is the main source of energy that triggers the ionization and the plasma expansion, and the energy absorbed from the black-body radiation field is negligible in comparison. Finally, recombination repopulates higher levels (which ''freeze out'' due to the rapid decrease of electron temperature), but the peak of the distribution is still found at relatively low excitations. This redistribution scenario is in qualitative agreement with Ref. [5], but in contrast to Ref. [4] where no redistribution has been observed. This discrepancy between the findings of the experiments [4], carried out at much lower initial excitation $n_0 = 40$, and the experiment [5] and our calculations, hints at the possible importance of other effects at these lower excitations. So far, this discrepancy remains unexplained and certainly has to be addressed in the future. In general, we have found the distribution of Rydberg atoms to be much more sensitive to the initial conditions (density, initial excitation, and Rydberg gas vs directly produced plasma) than ''macroscopic'' plasma parameters such as expansion velocity, etc. In order to shed more light on this issue, further experimental studies of the final distribution of Rydberg atoms (as a function of initial excitation n_0) would be very interesting and should provide a stringent test for theoretical models.

In summary, we have described the formation of a cold plasma from a highly excited Rydberg gas, using a simple model based on kinetic equations for the plasma evolution. Microscopic molecular-dynamics simulations are limited to much shorter time scales due to the large numerical effort required. We have estimated the maximum effect of ionic correlations by including them into the model assuming instant equilibration of the ions. In this respect the calculations without equilibration (no correlation, $T_i = 0$) and with instant equilibration should bracket the actual ionization yield of the Rydberg gas. Given the difference in the ionization yields with and without ionic correlation (Fig. 1), more elaborate calculations are worthwhile, although at present it is unclear whether the plasma evolution including correlation can be followed over sufficiently long times, e.g., with P^3M codes.

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