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Spontaneous Evolution of Rydberg Atoms into an Ultracold Plasma

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We have observed the spontaneous evolution of a dense sample of Rydberg atoms into an ultracold plasma, in spite of the fact that each of the atoms may initially be bound by up to 100 cm^{-1}. When the atoms are initially bound by 70 cm^{-1}, this evolution occurs when most of the atoms are translationally cold, <1 mK, but a small fraction, ~1%, is at room temperature. Ionizing collisions between hot and cold Rydberg atoms and blackbody photoionization produce an essentially stationary cloud of cold ions, which traps electrons produced later. The trapped electrons rapidly collisionally ionize the remaining cold Rydberg atoms to form a cold plasma.

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In a conventional plasma the lower bound to the electron temperature is set by the requirement that there be enough ionizing collisions to sustain the plasma. Since electron kinetic energies of 1–10 eV are required, it is difficult to make laboratory plasmas with temperatures less than 10^4 K. It would be quite interesting to do so, though, because cold plasmas can be strongly coupled; i.e., the Coulomb interaction between adjacent particles can exceed their kinetic energies, and such plasmas exhibit qualitatively new phenomena [1].

The first truly cold plasmas were one component plasmas composed of trapped laser cooled ions, which formed crystals at low temperatures [2,3], and the rigid body rotation of such ionic crystals has recently been reported [4]. In the past year a cold, nearly neutral plasma has been produced by photoionizing Xe atoms held in a magneto-optical trap (MOT) [5]. The Xe^+ ions have the temperature of the Xe atoms in the trap, 30 μK, and the electrons' kinetic energy is the excess of the laser photon energy over the binding energy, which can be quite small, less than 0.1 cm^{-1}, corresponding to a temperature of 100 mK. The Xe plasma is not quite neutral; the plasma electrons are bound to the macroscopic excess positive charge, and the plasma slowly expands.

Here we report the spontaneous evolution of Rydberg atoms, each initially bound by up to 100 cm^{-1}, into an ultracold plasma. Related observations of self-ionization of Rydberg atoms have been reported [6,7], and a mechanism with some similarities to that proposed here has been advanced [7]. In this paper we first describe the experimental approach and show that the Rydberg atoms evolve into a plasma. We then outline the mechanism of the evolution, describe experimental tests which support the proposed mechanism, and, finally, suggest some of the implications.

We have done experiments with both Rb and Cs, using essentially the same techniques. The Rb (Cs) is held in a vapor loaded MOT. The cloud of cold atoms in the MOT has a diameter of 1 (1.2) mm, contains 1.5×10^7 Rb $5p$ (1.2×10^7 Cs $6p$) atoms, and has a temperature $T = 300$ μK (140 μK). The cloud of cold atoms is midway between two parallel grids to which we apply voltage pulses to ionize Rydberg atoms and drive charged particles to a microchannel plate (MCP) detector behind one of the grids. We can detect either ions or electrons.

The Rb (Cs) atoms are excited from the $5p_{3/2}$ ($6p_{3/2}$) state to the Rb (Cs) Rydberg states with a pulsed 480 (520) nm dye laser running at a 20 (10) Hz repetition rate. The dye laser pulse is 7 ns long, has a bandwidth of 0.3 cm^{-1}, and has an energy of 100 μJ. The dye laser beam diameter is 0.2 mm, so we make a cylinder of up to 10^5 Rydberg atoms on each laser shot. Equally important, we also excite room temperature (hot) atoms to the Rydberg state so that under typical conditions 1% of the Rydberg atoms in the trap volume are hot. While we can measure relative populations to 10%, the absolute populations are uncertain by a factor of 3.

We analyze how the Rydberg population has evolved by applying a 1 μs rise time voltage pulse to the plates at delay times of 0–50 μs after the dye laser excitation. As the pulse rises it first frees electrons weakly bound to the plasma, then ionizes any bound Rydberg atoms, and, depending on its polarity, drives ions or electrons to the MCP detector. We detect the time resolved MCP signal which typically has two components, an early one due to the plasma ions or electrons and a later one due to the Rydberg atoms. Examples are shown for several delay times in the insets in Fig. 1. A very important point to note is that we see the plasma signal, which comes early in the ionization pulse, with both ion and electron detection, even with a delay time of 20 μs. The fact that the electrons are present and weakly bound to the macroscopic excess positive charge 20 μs after laser excitation demonstrates that the Rydberg atoms do indeed evolve into a plasma.

The insets in Fig. 1 show the major qualitative feature of our data. Shortly after the dye laser pulse, with a 2 μs delay, there are predominantly Rydberg atoms. Much later, with a 12 μs delay, there is predominantly a plasma, and at all delay times there are only two significant components of the signal. To see how the evolution occurs we must

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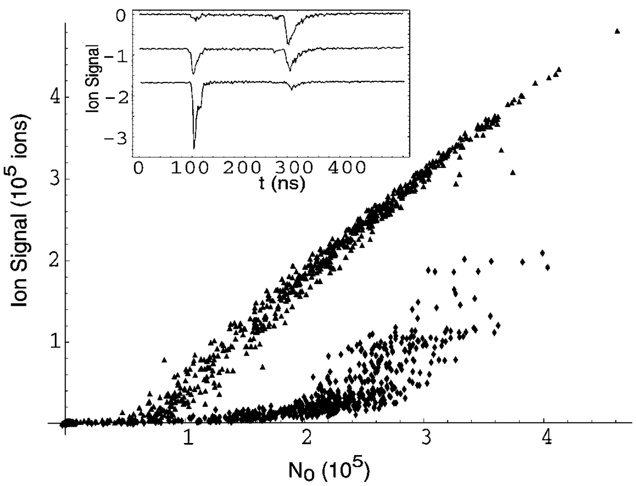

FIG. 1. Ion signals observed for different initial populations of the Rb 36*d* state. The two curves show the ion signals 2 μs (◆) and 12 μs (▲) delays after the dye laser excitation. The inset shows the time resolved signals obtained for $N_0 = 1.9 \times 10^5$ atoms at delays of 2 μs (upper trace), 5 μs (middle trace), and 12 μs (lower trace). In the upper trace there is no early ion signal and a large late atom signal while the reverse is true in the lower trace, indicating the formation of the plasma by 12 μs after laser excitation.

look at intermediate delay times, at which the plasma signal is nonlinear in the number of Rydberg atoms. The shot to shot dye laser frequency fluctuations lead to large fluctuations in the initial Rydberg population and 100% fluctuations in the plasma signal. To extract useful data we have binned the data by the total detected signal, which reflects the initial population in the Rydberg state, N_0 (the average total detected signal does not change with delay time for the first 8 μs). Explicitly, we fix the delay time and record the time resolved signal from each of 1000–3000 laser shots and plot the plasma signal vs the total signal. Examples are shown in Fig. 1 for the Rb 36*d* state at delays of 2 and 12 μs after the dye laser pulse. In addition to the shot to shot dye laser fluctuations neutral density (ND) filters as strong as ND 1.5 were used to obtain the data of Fig. 1. In Fig. 1 it is apparent that there are clear thresholds for the formation of the plasma at initial Rydberg state signals of $N_0 = 2.2 \times 10^5$ and 0.7×10^5 and that at higher initial numbers of atoms the number of plasma ions approaches the initial population in the 36*d* state. Similar data were obtained for Cs. For both Rb and Cs, for higher lying states the plasma forms at earlier times and with a smaller initial number of atoms.

To extract the time dependence of the evolution to the plasma we repeat measurements such as those shown in Fig. 1 at a series of delay times separated by 100 ns intervals. From these data we select those data points which have the same total signal, and, using them, construct the time dependence of the plasma signal as shown in Fig. 2 for the Rb 36*d* state with the total initial number of atoms $N_0 = 2.4 \times 10^5$ and 3.3×10^5. As shown by

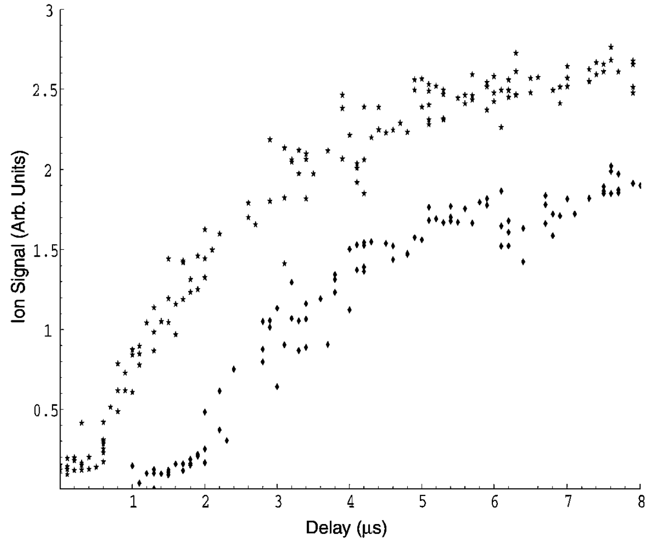

FIG. 2. Time resolved ion signals subsequent to the population of the Rb 40*d* state. The initial Rydberg populations are 3.3×10^5 (★) and 2.4×10^5 (◆). The data taken with the lower population are offset by 1 μs for clarity.

Fig. 2, for $N_0 = 3.3 \times 10^5$ the ion signal rises slowly for $0 < t < 0.6$ μs, after which the signal resembles the voltage on a charging capacitor. With a smaller initial number of atoms, $N_0 = 2.4 \times 10^5$, the exponential rise comes later, but the onset occurs at the same value of the ion signal. As noted above, the data for Cs are similar, and Fig. 3 shows the development of the plasma subsequent to excitation of the Cs 39*d* state, using electron detection, which demonstrates that the plasma lives for 30 μs.

While the data of Figs. 1–3 make it apparent that a plasma is formed, it is not obvious how it happens. Experiments which will be described shortly suggest the following sequence of events. At time $t = 0$ the dye laser pulse produces mostly cold, but some hot, 300 K, Rydberg

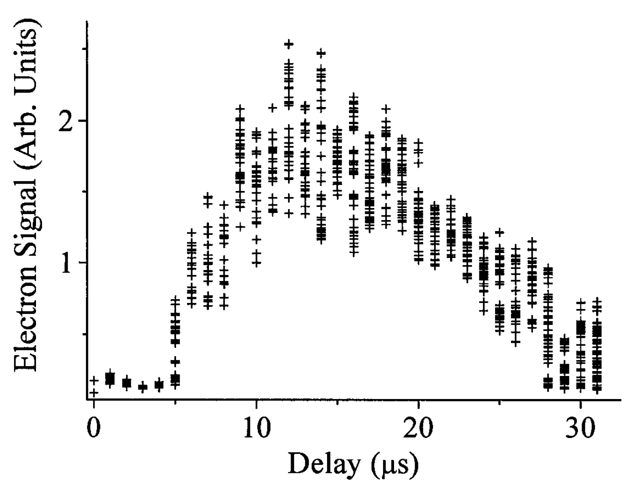

FIG. 3. Electron signal vs delay after excitation of the Cs 39*d* state, showing that the plasma lives for about 30 μs.

atoms. Photoionization by blackbody radiation and collisions between hot and cold Rydberg atoms produce ions and free electrons. These processes account for the linear rise in the ion signal of Fig. 2 at early time. The electrons produced leave, until enough positive charge of cold ions accumulates to trap the electrons subsequently produced in the MOT volume. Once electrons are trapped in the MOT volume they repeatedly collide with the remaining Rydberg atoms. Since electrons move rapidly and the cross sections for ionization of Rydberg atoms are large, roughly the Rydberg atom's geometric cross section, avalanche ionization ensues [8]. The onset of this process leads to the abrupt changes in the signals shown in Figs. 1–3.

Several additional observations led us to propose this mechanism. If we apply a small static field, 2 V/cm, the plasma never forms, presumably because the electrons cannot be trapped. With only hot Rydberg atoms the plasma never forms. If we turn off the diode lasers used for trapping and use a pulsed 780 nm laser to drive the Rb $5s_{1/2}$-$5p_{3/2}$ transition we can produce hot Rydberg atom samples, which have 40% of the peak Rydberg density obtained using the MOT. When Rb $40d$ atoms are excited at this density using the MOT we observe a plasma, but when only hot atoms are excited we see no evidence of the formation of the plasma; the cloud of hot ions formed expands too rapidly to trap the electrons.

We do not see plasma formation for initial states of $n \sim 40$ when there are no hot atoms although we do for $n > 50$. We have removed the hot atoms in two ways. After exciting the Cs $39d$ state we turned off the trapping light with a Pockels cell with a 200 ns switching time. If we switch off the trapping light 50–100 ns before the dye laser pulse, so the dye laser comes in the decaying tail of the trapping light, the plasma production practically disappears, although there is not a noticeable decrease in the number of Rydberg atoms excited. We presume that the total number of Rydberg atoms does not change because the cold Cs $6p$ population decreases slowly due to radiation trapping in the MOT [9]. Hot Cs $6p$ atoms, with much larger Doppler widths, do not experience radiation trapping. As a result, when we measure the hot Rydberg atom production, by blocking one of the trap laser beams, we see a rapid decrease as the delay of the dye laser pulse is increased from 50 to 100 ns after switching the Pockels cell. In other words, it is correlated with the decrease in the plasma production, pointing to the importance of hot Rydberg atoms for plasma formation.

As a clearer test of the importance of hot Rydberg atoms we have turned off the trapping diode lasers 20 μs before the dye laser pulse and turned on one diode laser beam a factor of 10 weaker than the trapping beams, colinear with the dye laser beam, and tuned exactly on resonance with the Cs $6s_{1/2}$ $F = 4 \rightarrow 6p_{3/2}$ $F = 4$ transition, 500 ns before the dye laser pulse. With this approach we remove 98% of the hot Rydberg atoms, and using this sample of atoms we never observe the production of a plasma for the Cs $39d$ state. For example, in Fig. 4 we show the plasma signals obtained with a delay of 3 μs after the dye laser pulse with only cold Rydberg atoms and with the normal composition of the MOT. The data are binned in the same way as those in Fig. 1. It is quite evident that no plasma is formed when there are only cold atoms. We have verified that hot $6p$ atoms have no effect.

To test the notion that the initial ionization is from blackbody photoionization and hot Rydberg atom-cold Rydberg atom collisions we have measured the ionization rate at early times. When only cold Cs atoms are excited to the Cs $39d$ state we find an ionization rate of 1×10^3 s^{-1}, roughly consistent with earlier measurements of blackbody photoionization rates [10]. For the normal MOT, with 1% hot Rydberg atoms, we find an ionization rate of 2×10^3 s^{-1}, consistent with the 1% hot Rydberg atoms ionizing the cold Rydberg atoms with a cross section 10 times the geometric cross section, in reasonable agreement with the theoretical value [11].

The abrupt changes in Figs. 1 and 2 should occur when enough cold ions have accumulated to trap electrons produced by ionization [5]. For an electron kinetic energy of 70 cm^{-1}, equal to the initial binding energy, and a spherical volume of radius 0.1 mm an excess charge of 10^3 ions is required for trapping, and for our cylindrical excitation volume approximately 5×10^3 ions are required, roughly 5% of the maximum of the Rb signals observed. Furthermore, neglecting expansion of the ion cloud the onset of trapping should occur for the same number of ions irrespective of the initial number of Rydberg atoms in the trap. As shown by Fig. 2, such is the case.

In conclusion, we have shown that a mixture of cold Rydberg atoms from a MOT with a small number of hot atoms spontaneously evolves to a cold plasma even though the atoms initially are rather deeply bound. There are still aspects of this evolution which remain to be

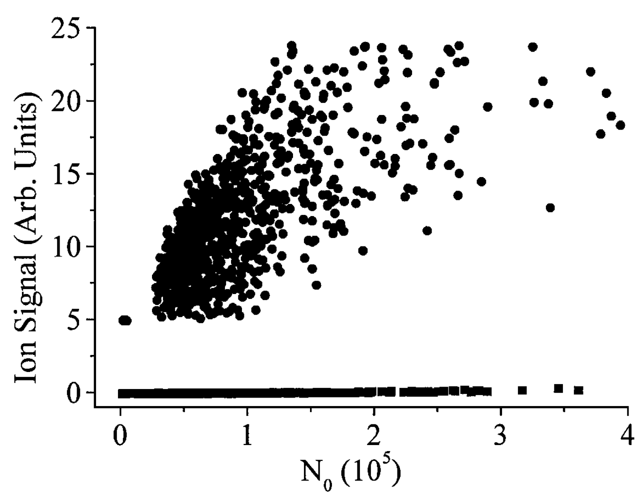

FIG. 4. Ion signals obtained with a delay of 3 μs after the excitation of the Cs $39d$ state with (●) and without (■) hot atoms. The signal hot atoms is offset by five units.

explored. For example, initial collisions with hot Rydberg atoms and blackbody photoionization seem to account for at most 10% of the energy required to convert the bound atoms to a plasma, although a possible source is superelastic scattering of the electrons from Rb5*p* (Cs6*p*) atoms [12]. Nonetheless, these observations suggest that cold, dense Rydberg atom samples may evolve into something like an amorphous metallic solid [8].

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