Many-Body Ionization in a Frozen Rydberg Gas

Paul J. Tanner, Jianing Han, E. S. Shuman, and T. F. Gallagher
Department of Physics, University of Virginia, Charlottesville, Virginia 22904-0714, USA
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In a dense gas of 300 μK ^{85}Rb atoms of $n \sim 50$ ionization occurs on a 100 ns time scale, far too fast to be explained by the motion of the atoms or photoionization by 300 K blackbody radiation. Rapid ionization is accompanied by spectral broadening, with the spectrum becoming continuous at $n = 88$ at a density of 5×10^{10} cm^{-3}. The atomic transitions broaden both smoothly and by the emergence of new features, which we attribute to multiple atom absorptions. We attribute the rapid ionization to a sequence of near resonant dipole-dipole transitions through virtual states in this intrinsically many-body system, culminating in the ionization of some of the atoms.

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A low density, 10^{10} cm^{-3}, 300 μK gas of Rydberg atoms, those in states of high principal quantum number n, resembles an amorphous solid, and for this reason it is often termed a frozen Rydberg gas [1]. It is effectively frozen because on the experimentally interesting 1 μs time scale the atoms move at most a few percent of the average interatomic spacing. In spite of the low density the atoms interact with each other, primarily by means of the dipole-dipole interaction, which scales as n^4, and the frozen Rydberg gas can exhibit many-body effects typically seen in solids, such as excitonlike diffusion of excitation [1–5]. Because of the exaggerated properties of Rydberg atoms the frozen Rydberg gas is an easily manipulated artificial solid and has been proposed as the basis of quantum gates [6,7]. To be useful the frozen Rydberg gas must be stable on at least a microsecond time scale. The obvious criterion for stability is that the orbits of the Rydberg atoms not overlap, for then Auger ionization occurs [8,9]. Since the orbital radius r is given by $r = 2n^2$, overlap occurs when atoms are spaced by $R = 4n^2$, corresponding to a density N of

$$N = \frac{3}{256\pi n^6}. \quad (1)$$

We use atomic units unless stated otherwise. Classical Monte Carlo simulations have verified that ionization occurs when pairs of atoms are separated by $R \simeq 4n^2$ in agreement with Eq. (1). This suggests that $n = 50$ atoms are stable for densities $N \leq 1.6 \times 10^{12}$ cm^{-3} [10], a density far above that specified above. The importance of other ionization mechanisms such as photoionization by 300 K blackbody radiation and motion of initially stationary atoms along attractive potentials, resulting in ionizing collisions has clearly been demonstrated [10–14]. However, for $n = 50$ and $N = 10^{10}$ cm^{-3} these processes occur on a time scale of many microseconds. Another possible mechanism for the initial ionization is molecular autoionization, a diatomic dipole-dipole energy transfer process in which one atom is ionized and the other loses the corresponding energy in one step, but it is even slower [8,9]. It would seem that a frozen Rydberg gas of $n = 50$ should be stable on a microsecond time scale.

Here we report rapid ionization of a frozen Rydberg gas of ^{85}Rb. In this experiment atoms are excited to nd Rydberg states, then ionize on a time scale of 100 ns. For $n \approx 90$ the ionization is consistent with the physical overlap of atomic orbits as described by Eq. (1), but for $n \sim 50$ the ionization is far too rapid to be attributed to any of the above processes. We attribute the rapid ionization to a many-body form of molecular autoionization roughly analogous to intramolecular vibrational energy redistribution (IVR) in molecules [15]. Stated another way, the atoms are never isolated, but always interacting with each other. As a result, a many-atom state in which all the atoms are in single-atom nd states is not an eigenstate of the many-atom system, although it is the initial state by virtue of the laser excitation. The many-atom system rapidly evolves from its initial state through a series of near resonant, dipole-dipole coupled states, which results in the diffusion of the atomic population over a band of energies, including the ionization continuum. In the sections that follow we describe our experimental method and observations, present a simple model to explain them, and summarize the implications of this work.

The experiment is conducted using a vapor loaded ^{85}Rb magneto-optical trap (MOT) [16]. The trapped atoms are at the center of a four rod structure which allows us to field ionize the Rydberg atoms and drive all ions to a microchannel plate detector (MCP) 10 cm from the trap volume. The typical parameters of the MOT are a Rb $5p_{3/2}$ density of 5×10^{10} cm^{-3} and a volume of 1 mm^3. We excite atoms to a chosen Rydberg nd state with a 480 nm blue laser pulse 10 ns long at a 20 or 30 Hz repetition rate. At a chosen time after the laser pulse we apply the field ionization pulse of variable amplitude and 1 μs rise time to two of the rods, driving free ions and those resulting from field ionization to the MCP. The signal from the MCP is recorded with gated integrators or a digitizing oscilloscope, and the resulting data are stored in a computer.

We have used two sources of blue light, a 480 nm dye laser with a 10 GHz bandwidth, and a frequency doubled, pulse amplified, single mode Sacher Tiger series 960 nm diode laser, which produces 480 nm light with a 100 MHz bandwidth. The blue laser beam is focused to a diameter of 0.1 mm so we can produce a high density of Rydberg atoms without significantly depleting the trap on each laser shot.

A surprisingly large number of free ions are present immediately after laser excitation, as shown by Fig. 1, which shows the time resolved ion signals obtained when atoms are excited to the Rb $40d$ state with the dye laser and exposed to the field ionization pulse 40 ns later. The ionization pulse amplitude is set to ionize the $40d$ state. At low atom density the signal at $t = 1.1$ μs is due to field ionization of the $40d$ state. At high density the signal is observed from $t = 0.4$ to 1.7 μs, due to transfer to higher and lower energy states. The sharp signal observed at 0.4 μs is due to free ions, and it comprises 5% of the total signal.

Insight into the origin of the ionization can be obtained from the excitation spectra of the frozen Rydberg gas. In Fig. 2 we show the spectra for the excitation of the $32d$, $45d$, and $88d$, states obtained by scanning the diode laser through the atomic $5p_{3/2} - nd$ transitions. In all cases the laser is operated at maximum intensity, with 480 nm pulse energies of $\sim$100 μJ. The laser frequency ν is measured by monitoring the diode laser transmission through a 1.5 GHz free spectral range etalon, and the scale of Fig. 2 is relative to the center of gravity of the atomic Rb $5p_{3/2} - nd$ transition frequency ν_0. The field ionization pulse is set to ionize the nd state excited by the laser. For the $32d$ and $45d$ states we can separate the time resolved signals due to free ions and the nd atoms, but for the $88d$ state we cannot resolve the two signals. The field ionization pulse begins 10 ns after the laser pulse, but the results are almost identical if it begins 250 ns later. In Fig. 2 we show both the free ion and nd state signals for the $32d$ and $45d$ states and the total signal for the $88d$ state. For the $32d$ state we see free ions only at the frequencies of the atomic transitions to the resolved $32d_{3/2}$ and $32d_{5/2}$ states, which are 370 MHz apart [17], with no observable broadening. Even at the maximum trap density the free ion signal is only 5% of the atom signal. At a trap density of 5.0×10^{10} cm^{-3} the $45d$ free ion signal is 800 MHz wide with a satellite feature 1 GHz to the blue. The $45d$ signal is near the atomic frequency. For $88d$ the combined atom and free ion signal shows two clearly resolved peaks due to the $88d$ and $90s$ states when the trap density is lowest ($\approx 2.3 \times 10^9$ cm^{-3}). Even in this case the 227 MHz width of the observed spectral feature at the location of the $88d$ state exceeds the laser linewidth. When the trap density is increased to 1.6×10^{10} cm^{-3} the $88d$ feature broadens to 490 MHz, and when the density is increased to the maximum value, the spectral features corresponding to the $88d$

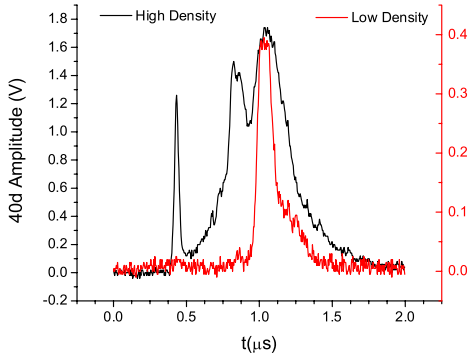

FIG. 1 (color online). Oscilloscope trace of signals obtained with a field ionization pulse applied 80 ns after dye laser excitation of the Rb $40d$ state. With low density we observe the $40d$ signal at $t = 1.1$ μs. At high density we observe signals from a range of final states: predominantly free ions at $t = 0.46$ μs and the $42p$ state at $t = 0.8$ μs.

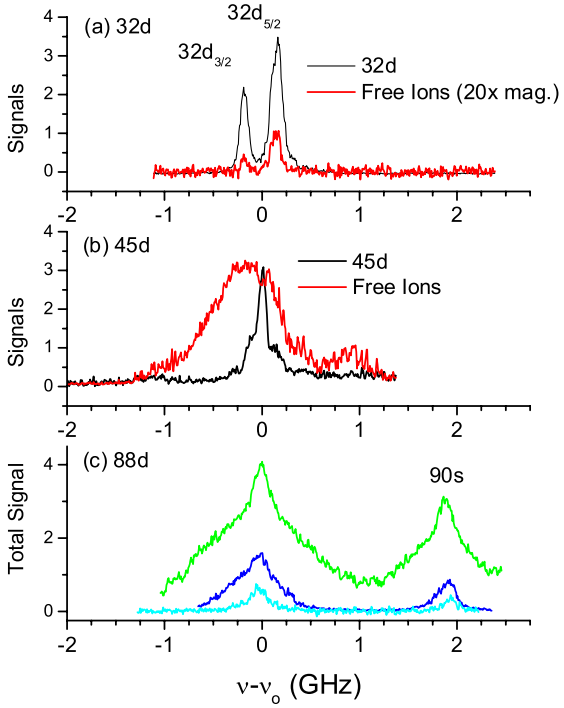

FIG. 2 (color online). Excitation spectra of the Rb $32d$, $45d$, and $88d$ states. The frequency is relative to the center of gravity of the atomic $5p_{3/2} - nd$ transition frequency. (a) The $32d$ state at density 5.0×10^{10} cm^{-3}. Neither the $32d$ signal nor the free ion signal exhibits broadening beyond the laser linewidth. (b) The $45d$ state at a density of 5×10^{10} cm^{-3}. The central free ion signal feature is 800 MHz broad, and is several hundred megahertz broader than the $45d$ signal. Also a new feature appears at $\nu - \nu_0 = 1$. The hole at relative frequency zero is an artifact due to trap depletion. Here the peak atom and free ion signals are equal. (c) The $88d$ and $90s$ states total (atom plus free ion) signals. At 2.3×10^9 cm^{-3} (light blue or light gray) the $88d$ and $90s$ are well resolved features. At 1.6×10^{10} cm^{-3} (blue or dark gray) the two states are still well resolved, but at 5.0×10^{10} cm^{-3} (green or gray) the states are completely overlapped; the spectrum has become a continuum.

and 90s states overlap. The spectrum has become continuous. At a density of 5×10^{10} cm^{-3} the condition given by Eq. (1) is met, and ionization results from the orbital overlap of the Rydberg atoms.

In Fig. 3 we show the free ion signals from the 51d state at several intensities of the 480 nm light. The field ionization pulse begins 10 ns after the laser pulse and is set to ionize the 51d state. The 51d signal appears only at the frequency of the atomic transition, and it is easily saturated. We interpret it as arising mainly from the edge of the excitation volume, where there is reduced interaction among the Rydberg atoms. In contrast, the spectral width of the free ion signal increases with laser power. However, it does not simply broaden, but it develops new features as well, such as the ones at $\nu - \nu_0 = -1$ GHz and 600 MHz. Near the atomic transition, at $|\nu - \nu_0| < 400$ MHz for example, the free ion signal depends quadratically on the blue light intensity at low intensity and saturates at high intensity. Presumably the excitation occurs by two photon excitation of pairs of atoms, or multiple atoms, via a virtual intermediate state, as described by Raimond *et al.* [18]. However, the signals at -1 GHz and 600 MHz exhibit a thresholdlike, i.e., higher than quadratic, behavior as a function of blue laser intensity, presumably due to the simultaneous excitation of more than two atoms. With a broadband, 10 GHz linewidth, laser these excitations can occur via a real intermediate state and are thus more efficient. Since we do not observe any prompt ions when we excite nd states of $n \leq 29$, and the photoionization cross section of Rydberg states decreases as n^{-3} we infer that laser photoionization plays a negligible role.

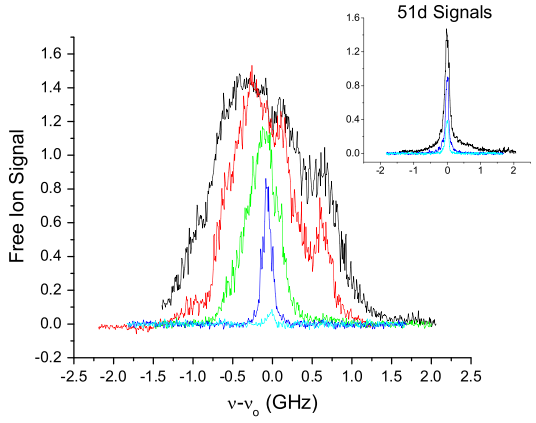

FIG. 3 (color online). Excitation spectra of the 51d state at different relative laser intensities, 0.01 (cyan or lightest gray), 0.03 (blue or dark gray), 0.2 (green or light gray), 0.65 (red or gray), and 1.0 (black). The frequency is relative to the atomic $5p_{3/2} - 51d$ transition frequency. At low intensity the free ion signal is only visible at the atomic transition frequency, but as the intensity is raised the free ion signal broadens and develops new features, for example, at $\nu - \nu_0 = -1.0$ GHz and 0.6 GHz. Inset, signals from the field ionization of the 51d state at the same laser intensities. The signals are essentially confined to the atomic line except at the highest density.

We attribute the ionization to the fact that the frozen Rydberg gas is an inherently many-body system in which the optically accessible state is not an eigenstate, but simply the entry into a band of levels [19]. The specific mechanism in this case is similar to superexchange in IVR in a polyatomic molecule. The essential idea is shown in Fig. 4, which is simplified in that we only consider two atoms. In the molecular energy level diagram of Fig. 4(a) the initial excitation of the atoms is to the $ndnd$ state which is coupled by the dipole-dipole interaction to the energetically nearby $(n+2)p(n-2)f$ state, which is in turn coupled to other states. In terms of the atomic energy levels, shown in Fig. 4(b), this leads to the possibility of a sequence of near resonant dipole-dipole transitions through virtual states in which one atom loses energy passing through n_d states separated in energy by ω, while the other gains energy, passing through n_u states energetically separated by $\omega + \Delta$. This sequence ends when one atom has reached the ionization limit and the other has reached a state of principal quantum number $n_d \leq n/\sqrt{2}$. The process is a multistep variant of molecular autoionization, and a reasonable criterion for its occurring is the one used in a similar process, microwave ionization at high scaled frequency, i.e., requiring that in the intermediate steps the dipole-dipole coupling equals the detuning from resonance, Δ [20,21]. Explicitly,

$$\frac{\langle n'_u|\mu|n''_u\rangle\langle n'_d|\mu|n''_d\rangle}{R^3} = \Delta. \qquad (2)$$

Here R is the distance between atoms. For simplicity we assume that for the downward transitions $n''_d - n'_d = -1$, and for the upward transitions $n''_u - n'_u > 1$. In this case ω is the energy separating the n'_d and $n'_d - 1$ states, and Δ is a fraction of the spacing between adjacent n_u levels. For

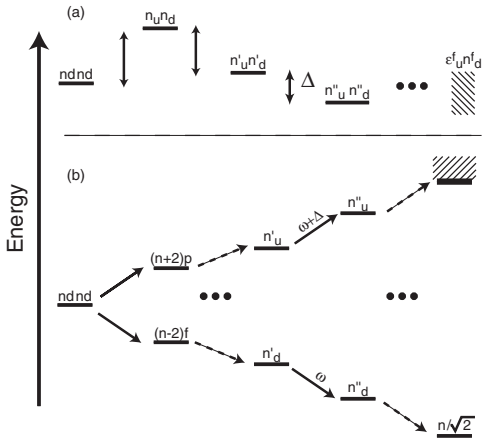

FIG. 4. Schematic of ionization in a simple binary model in terms of (a) molecular two-atom energy levels and (b) the single-atom levels. (a) Pairs of atoms initially excited to the $ndnd$ state are coupled to other nearly degenerate pairs of states composed of atomic states of higher and lower energy. (b) The atomic population rapidly diffuses over a band of levels, which includes the ionization continuum.

large n the dipole matrix element $\langle n'_u|\mu|n''_u\rangle \approx 0.41(n'_u n''_u)^{-3/2}\omega^{-5/3} \approx 0.41 n'^{-3}_u \omega^{-5/3}$, and $\langle n'_d|\mu|n''_d\rangle \approx 0.3 n'_d n''_d \approx 0.3 n'^2_d$. Using $\omega = n'^{-3}_d$ and $\Delta = \delta n'^{-3}_u$, with $\delta < 1/2$, we can rewrite Eq. (2) as

$$\frac{0.12 n'^7_d}{R^3} = \delta. \tag{3}$$

We take n'_d to be its average value over the sequence, which leads to an ionization requirement of

$$\frac{0.04 n^7}{R^3} = \delta, \tag{4}$$

which can be written as a density requirement for ionization. Explicitly,

$$N_i = \frac{3}{4\pi R^3} = \frac{18\delta}{\pi n^7}. \tag{5}$$

This density exhibits the same n scaling as that which produces spectral overlap of adjacent n levels due to the dipole-dipole interaction.

To evaluate Eq. (5) we need to assign δ a value. In a two-atom hydrogenic system the average detuning, $\Delta = 1/4 n'^3_u$ ($\delta = 1/4$). In the real frozen Rydberg gas each atom interacts with, not one, but roughly six other nearby atoms, substantially raising the density of states [19] thus lowering the detuning and introducing the possibility that many atoms lose a relatively smaller amount of energy in order that a few may be ionized. There are 15 possible pairs of atoms which can have a dipole-dipole interaction, and each of these pairs has 8 possible coupled states yielding 120 accessible states. Accordingly, we reduce δ from $1/4$ to $1/480$, in which case Eq. (5) predicts ionization to occur for $n = 50$ at $N = 1.0 \times 10^{11}$ cm^{-3}. This density is higher than the experimentally observed density, but we have ignored all other ionization processes, such as ionization pathways in which $\Delta n_d > 1$, and other effects which may be important. Examples are stray fields and high angular momentum states which develop permanent dipole moments [22]—both of which blunt the dipole selection rule. Finally, it is useful to compare the strength of the dipole-dipole interaction, given by $0.1 n^4/R^3$ to the observed time scale of 100 ns. For $n = 50$ and $N = 3 \times 10^{10}$ cm^{-3} the dipole-dipole interaction strength is 78 MHz, or 5×10^8 s^{-1}, compatible with population transfer from the initial state in 100 ns.

The picture of Fig. 4 requires that some atoms lose energy in order that others be ionized. Accordingly, we have examined the atomic n state distributions subsequent to laser excitation. Specifically, we have measured the time dependence of the total signals due to free ions and field ionization for different field pulse amplitudes. When we set the pulse amplitude to 55 V/cm, adequate to ionize atoms in the $51d$ state [23], the signal increases from its initial value with increasing delay of the field pulse, reaching a peak at 10 μs delay, then slowly declining. When we set the pulse amplitude to 400 V/cm, a field strong enough to ionize adiabatically atoms in states as low as $n = 30$, we observe a larger signal with a slow decline in time. These observations are consistent with the very rapid formation of a distribution of atomic states, of n as low as 30, followed by a relaxation of this distribution, with most of the atoms being ionized and a small number going to states of n too low to be ionized.

These measurements show that a wide distribution of states, including free ions, is present 100 ns after laser excitation of the frozen Rydberg gas. Ionization occurs because the frozen Rydberg gas is not composed of isolated atoms but is an interactive many-body system which exhibits a form of molecular autoionization roughly analogous to IVR in a polyatomic molecule. Similar processes may be operative in rapid and nearly complete population transfer reported by Reinhard *et al.* [24]. These measurements confirm that blockades to excitation disappear when the laser intensity is raised [25], and they demonstrate that excitation of high n states with a broadband dye laser can easily produce free ions as well as atoms. This rapid ionization process is clearly important in the transformation of the frozen Rydberg gas into a plasma [10,11], and more generally, it renders the frozen Rydberg gas less stable than previously thought.

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