# High-Angular-Momentum States in Cold Rydberg Gases

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Cold, dense Rydberg gases produced in a cold-atom trap are investigated using spectroscopic methods and time-resolved electron counting. Optical excitation on the discrete Rydberg resonances reveals long-lasting electron emission from the Rydberg gas (>20 ms). Our observations are explained by $lm$-mixing collisions between Rydberg atoms and slow electrons that lead to the population of long-lived high-angular-momentum Rydberg states. These atoms thermally ionize slowly and with large probabilities.

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Laser-cooled atoms can be excited to Rydberg states in order to study Rydberg gases at both high densities and low atomic velocities [1,2]. Dense clouds of cold Rydberg atoms have been found to undergo virtually complete ionization if the density of the excited Rydberg atom population exceeds a critical value dependent on the principal quantum number $n$ [3]. The ionization dynamics has been explained by ionizing collisions among Rydberg atoms and between Rydberg atoms and electrons, and appears to be similar to the dynamics observed earlier in hot Rydberg gases excited in an atomic beam [4]. Because of the difference in atomic velocities, the rates of ionizing collisions between the atoms in cold Rydberg gases should be reduced compared to the case of a hot Rydberg gas. Therefore, new density-dependent interaction phenomena may become observable.

In this paper, we report the spontaneous evolution of cold Rydberg atoms into long-lived states. These states reveal themselves via a long-lasting electron emission signal, the time scale of which exceeds the natural lifetime of the initially excited Rydberg levels by approximately 2 orders of magnitude. We identify the long-lived states as high-$l$ Rydberg states, and explain their appearance by $l$-changing collisions between slow, free electrons and the initially excited low-$l$ Rydberg atoms. A similar process is at work in molecular zero kinetic energy (ZEKE) spectroscopy [5].

In our experiment, a vapor cell (pressure $<5 \times 10^{-9}$ Torr) magneto-optic trap (MOT) is operated in a 10 Hz cycle. $^{87}$Rb atoms are cooled to $\sim 50$ $\mu$K and trapped for 75 ms, after which the MOT light and magnetic fields are turned off for 25 ms. 1 ms after these fields are turned off, a 5 $\mu$s long diode laser pulse ($\lambda = 780$ nm) resonant with the $5S_{1/2}, F = 2 \rightarrow 5P_{3/2}, F = 3$ transition is applied, exciting a fraction of the ground state atoms to the $5P_{3/2}$ level. While the 780 nm pulse is on, a tunable blue dye laser pulse ($\lambda \approx 480$ nm, pulse width $\approx$10 ns, bandwidth $\approx$15 GHz, pulse energy $<5$ mJ) illuminates the entire atom cloud and excites $5P_{3/2}$ atoms to $ns$- and $nd$-Rydberg states. The dye laser, pumped by a tripled Nd:YAG (355 nm), consists of an oscillator and two optional stages of amplification. A microchannel-plate (MCP) detector placed 10 cm from the atom cloud is used to detect electrons emitted from the Rydberg gas. No electric field ionization pulse is applied. Based on the highest principle quantum number $n$ that we can spectroscopically resolve ($\sim$85), the electric field due to the MCP at the location of the atoms is estimated to be $\lesssim$0.5 V/cm ([6], p. 75). A photon counter records MCP pulses over a window with variable delay and a 1 ms width.

When the dye laser is tuned to a discrete Rydberg resonance, a long-lasting electron emission signal is observed. Operating the dye laser at a fixed wavelength and with the oscillator only, we obtain the signal shown in Fig. 1a for the $20d$ and $50d$ Rydberg states. The Rydberg atom densities are of the order of $10^9$ and $10^8$ cm$^{-3}$, respectively; the cloud volume is 1 mm$^3$. Conversely, by fixing the gate delay and scanning the dye laser wavelength, spectra such as shown in Fig. 1b are obtained. The spectra show that a delayed electron signal is present only when the dye laser is tuned to a Rydberg resonance, and that the signal does not appear either when the dye laser is tuned off of a resonance or tuned above the ionization limit ($\lambda_{\text{ion}} = 479.1$ nm). In addition, the $d$ peaks are significantly larger than the $s$ peaks, which are nearly absent for $n < 30$. The source of the detected electrons and the reasons for the long time scales associated with their emission are the main subject of this paper.

The key element in our explanation is that Rydberg atoms are transferred from their initial low angular momentum states ($l = 0$ or 2) into an approximately statistical mixture of $lm$ states. The natural lifetime of low-$l$ states scales as $n^3$, while for a statistical mixture of $lm$ states it scales as $n^{4.5}$ [6]. The long-lived high-$l$ states in the mixture decay over tens of milliseconds, due in large part to ionization by blackbody radiation. The thereby released thermal electrons are detected as the "delayed" electrons shown in Fig. 1.

One possible mechanism for transforming low-$l$ into high-$l$ Rydberg states is by rapid ionization [3] of the initial Rydberg atoms followed by recombination into high-$l$ states. If the recombination were significant, then the electron signal for $\lambda < \lambda_{\text{ion}}$, where a plasma is directly created, would be comparable to the average signal level right below the ionization threshold, $\lambda > \lambda_{\text{ion}}$ (because the

  

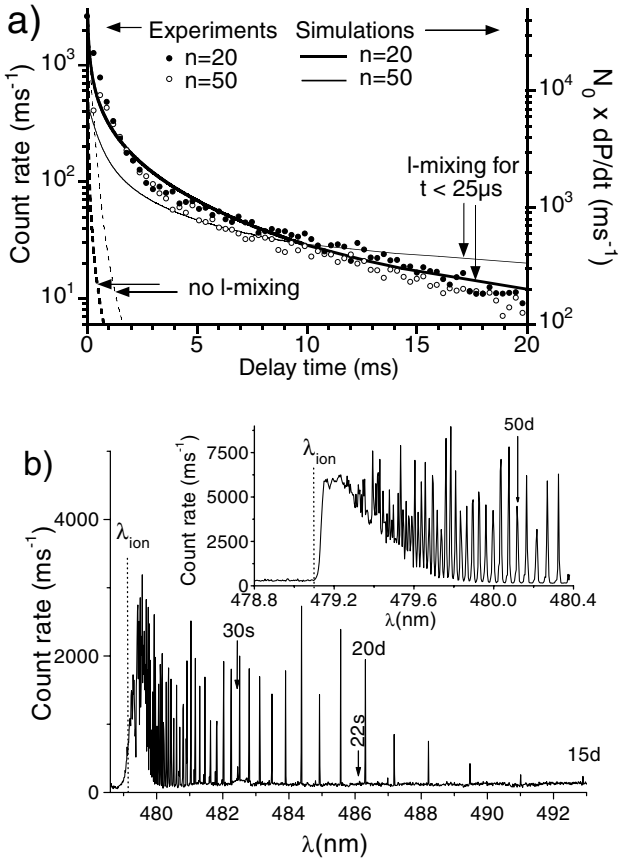


FIG. 1. Long-lasting electron emission. (a) Experimental and simulated electron signals vs time for $50d$ and $20d$ excitation. The simulations show the thermally induced population flux $N_0 * dP/dt$ into the continuum for the cases of no $lm$ mixing and efficient $lm$ mixing during the first 25 $\mu$s. The initial number of Rydberg atoms $N_0$ is $10^6$ and $10^5$ for $n = 20$ and $n = 50$, respectively. (b) Electron signal vs excitation wavelength, measured 10 ms after the Rydberg excitation pulse. The inset shows the count rate at 250 $\mu$s for $\lambda$ near the ionization threshold.

energy-averaged oscillator strength does not change at $\lambda_{\text{ion}}$). This is not the case, as shown in the inset of Fig. 1b.

Instead, we believe that the transfer of population into high-$l$ states originates from electric-field-induced mixing of $l$ states due to collisions of Rydberg atoms with bypassing "early" electrons. A single-atom simulation which statistically considers $l$-changing collisions reproduces the qualitative features in our data as seen in Fig. 1a for $n = 20$ and $n = 50$. In the simulation, dipole transitions due to thermal radiation and spontaneous decay drive the excited state population either toward ionization, thereby producing detectable electrons, or back to the ground state. Further, population can be redistributed within the hydrogenic manifolds and the nearby nonhydrogenic states using the statistical weights of the $l$ levels $(2l + 1)$. When the redistribution is applied over the first 25 $\mu$s (in accordance with the dynamics outlined below), the duration of the simulated electron signal is in qualitative agreement with our experimental results and is over an order of magnitude longer than if the redistribution is not applied (see Fig. 1a). The result of the simulation is insensitive to the exact duration of the redistribution phase. The simulations do not account for the efficiency of the electron counting and the free fall of the Rydberg atom cloud.

The early electrons which cause the $lm$ mixing are primarily due to short-time-scale thermal ionization of the Rydberg atoms. Using rate equation calculations of the thermal ionization in a 300 K radiation field and the experimental conditions of Fig. 1a, the $20d$ and $50d$ states are estimated to yield ~5000 electrons with an average kinetic energy of 8 meV. We have estimated that other sources of these early electrons, including cold-cold and cold-hot [3] Rydberg atom ionizing collisions, are less important under the conditions of Fig. 1a.

The velocities of the early thermal electrons ($10^4$ to $10^5$ m/s) are on the order of the Rydberg electron velocity ($\sim 5 \times 10^4$ m/s for $n = 40$), where the rates of $l$-changing collisions are the highest [6]. Although Rydberg atom-ion collisions are thought to cause $lm$ mixing in ZEKE spectroscopy [5], we discount the influence of ions because in our system the ions originate from laser-cooled atoms, the velocities of which are too low to result in appreciable rates of $lm$-mixing collisions.

The mechanism for the $l$-changing collisions is illustrated as step A in Fig. 2. Consider a Rydberg atom in the $21d$ state in the presence of the time-varying electric field produced by a bypassing electron. As the electron approaches and the electric field increases, the levels in the hydrogenic manifold of $n = 20$ intersect with the $21d$ energy level. The anticrossing between the initial $21d$ state and the hydrogenic manifold at electric fields of $\approx 1 \times 10^{-7}$ a.u. leads to mixing into the hydrogenic manifold. As the electron moves away and the electric field decreases, there exists a large probability that the atom remains in the hydrogenic manifold. We find that step A promotes the initial Rydberg atoms into high-$l$ states with an average value of $l \approx n/2$. Weak stray electric fields

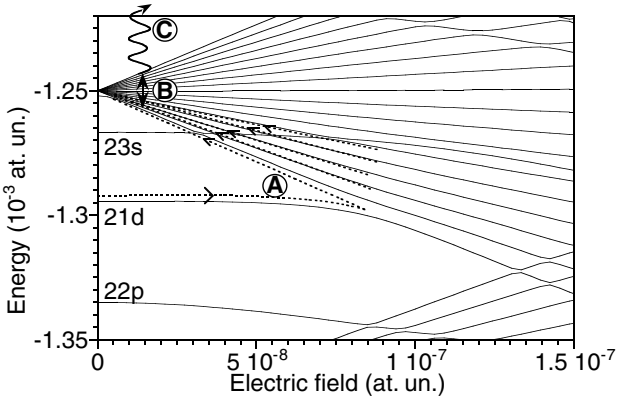


FIG. 2. Stark map of Rb near $n = 20$ illustrating $l$-mixing collisions. Step A shows mixing from the $21d$ state into the $n = 20$ hydrogenic manifold. In step B, the population is further randomized among the levels in the manifold. In step C, thermal radiation ionizes atoms on a slow time scale.

are likely to further randomize the population distribution within the hydrogenic manifolds, as indicated by step B in Fig. 2.

With the overall mechanism outlined, we present the results of calculations and experiment that support our model. We have numerically solved the time-dependent Schrödinger equation for Rydberg atom–thermal electron collisions. For an electron energy of 5 meV, the calculated $lm$-mixing cross section for the $d$ states has the form $\sigma \approx 2.2 \times 10^{-8} n_{\text{eff}}^5$ $\mu\text{m}^2$ (where $n_{\text{eff}} = n - \delta_l$ is the effective quantum number and $\delta_l$ is the quantum defect). Using this cross section and assuming that the electrons escape the Rydberg atom cloud ballistically, we calculate that for the estimated number of thermal electrons in Fig. 1a only 0.02% of $n = 20$ and 2% of $n = 50$ Rydberg atoms would be transferred to high $l$ states. This is not enough to account for our experimental observations.

This deficit is remedied by an electron trapping mechanism which has been identified in studies of cold plasmas [7]. In our system, an electron trap is formed by thermal ionization of a fraction of the Rydberg atom cloud. Electrons escape until the cloud is sufficiently positively charged to prevent further electrons from leaving. The electrons trapped in this space charge cloud make many oscillations through the cloud before the trapping potential diminishes due to Coulomb expansion. While the electrons oscillate, they collide frequently with the Rydberg atoms that are embedded in the space charge cloud. The work in [7] and our own observations show that the electron trapping time is of the order of 10 to 100 $\mu$s, allowing us to estimate that the electron-trapping mechanism enhances the probability of $lm$-mixing collisions by a factor of order 1000. This enhancement is critical in our explanation of the observed number of delayed electrons.

The number of early electrons that must leave the atom cloud in order to form an electron trap deep enough to retain electrons depends on the kinetic energy of the electrons as well as the size of the atom cloud [7]. For our conditions, this number is calculated to be ~2000. As estimated above, ~5000 early electrons are produced on the $d$ peaks for the conditions of Fig. 1a, ensuring the formation of an electron trap filled with ~3000 electrons. We believe that the strong delayed electron signal observed on the $d$ peaks in Fig. 1 hinges on the formation of this electron trap.

The critical role of the electron trap is made further evident by explaining the near-absence of the $s$ peaks compared to the neighboring $d$ peaks in Fig. 1b. The number of early electrons depends on the $5P \rightarrow ns$ photoexcitation cross sections and the thermal ionization probability of the Rydberg states. The photoexcitation cross section for the $s$ states is about six times smaller than for the neighboring $d$ states. The thermal ionization probability of the $s$ states was calculated using rate equations to be one half of that of the neighboring $d$ states. Therefore, if the Rydberg transitions are not saturated, the yield of early electrons from the $s$ states is ~1/10 that of the $d$ states. We hypothesize that on the $s$ states, under the conditions of Fig. 1a, the number of early electrons created is not sufficient to produce a filled electron trap, and thus there is not efficient $lm$ mixing and, consequently, no long-lasting, delayed electron signal.

To test this hypothesis, we modify the experimental conditions such that the formation of a (filled) electron trap can be guaranteed. The spectrum in Fig. 3a is taken under the same conditions as Fig. 1b (except for the delay time); no significant $s$ peaks are present, implying that for the excitation of $s$-Rydberg states no electron trap has formed. The spectrum in Fig. 3b is taken with the same pulse energy as in Fig. 3a, but with ≈10% of the pulse energy contained in amplified spontaneous emission (ASE). The ASE has been added to the pulse spectrum by activating one dye amplifier. The ASE spectrum is about 5 nm wide and centered at $\lambda = 478$ nm; therefore, a large fraction of the ASE is above the ionization threshold and creates free electrons and ions regardless of the wavelength of the lasing component. We estimate that in Fig. 3b ~10 000 free electrons with an average kinetic energy of 10 meV are produced by the ASE, i.e., enough to form a filled electron trap. As a result, the $s$ peaks emerge. The constant background count rate in Fig. 3b occurs because the fraction of the ASE with $\lambda < \lambda_{\text{ion}}$ excites a range of Rydberg states, which, after experiencing $lm$-mixing collisions in the electron trap, contribute to the long-lasting signal.

A second method of guaranteeing the formation of a (filled) electron trap is to preionize the atom cloud using a pulse of 355 nm ultraviolet (UV) light from the Nd:YAG pump laser that arrives 10 ns before the dye laser pulse. The UV photons generate photoelectrons with a kinetic energy of 0.9 eV. When the dye laser pulse arrives 10 ns later, the UV-excited electrons have moved 3 cm away from the atom cloud and thus have no effect on the Rydberg atoms. However, the potential well associated with the UV-generated positive space-charge cloud is deep enough

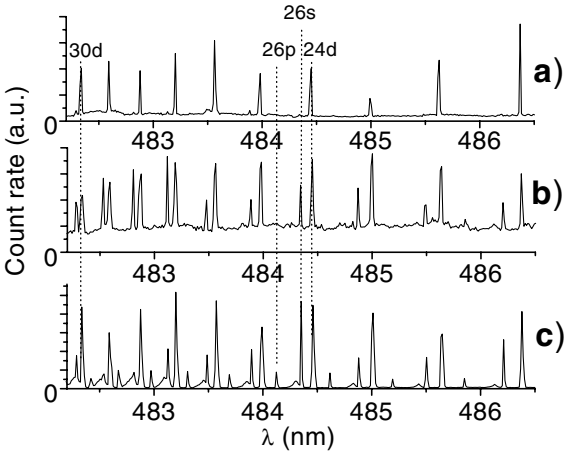


FIG. 3. Effect of electron trap for $n < 30$. (a) No electron trap present for $s$ peaks. (b) Electron trap formed by ASE for $s$ and $d$ peaks. (c) Electron trap formed by UV prepulse for $s$ and $d$ peaks. The electron counts are measured at a delay of 5 ms.

to prevent any of the early ~10 meV thermal electrons released from Rydberg atoms from escaping. Figure 3c shows a spectrum taken under the same conditions as in Fig. 3a with the addition of the UV prepulse. The emergence of the *s* peaks demonstrates that efficient *lm* mixing can be forced by providing an electron trap that is already in place at the time of the Rydberg atom excitation. The dipole-forbidden transitions seen in Fig. 3c ($p$- and $l > 2$ peaks) are due to the substantial electric fields caused by the ionic space charge cloud and will be discussed in a future paper.

Using a system similar to ours, recent investigations have shown that when an electron trap forms, Rydberg atom-electron ionizing collisions may lead to nearly complete ionization of the Rydberg cloud within ~20 $\mu$s of the Rydberg excitation [3]. We have observed indications of such large-scale ionization when the dye laser pulse energy is increased by operating the dye laser with the oscillator and both amplifiers (pulse energy ~2 mJ, ASE-contribution ≈10%). As a result, we saturate all Rydberg transitions and obtain an $n$-independent Rydberg atom density $>10^9$ cm$^{-3}$. The loss of atom trap population caused by direct optical photoionization, by collisional ionization of Rydberg atoms, or by *lm* mixing and subsequent thermal ionization of Rydberg atoms can be measured by detecting the absorption of a low-power probe pulse resonant on the $5S_{1/2} \rightarrow 5P_{3/2}$ transition. From the absorption, the effective area density of the cloud is computed and plotted as a function of the Rydberg excitation wavelength in Fig. 4a. The minima of the area density occurs on the Rydberg transition wavelengths and for $\lambda < \lambda_{\text{ion}}$. Using the same pulse energy, we obtain the electron signal at 10 ms as shown in Fig. 4b. For $n < 35$, *lm* mixing produces trap loss and a long-lasting signal on the Rydberg peaks, as discussed above. However, for $n > 40$ there are no peaks in the electron signal, even though Fig. 4a clearly shows that Rydberg atoms are being excited and lost from the trap in this range. The absence of a long-lasting electron signal for $n > 40$ suggests that collisional ionization of Rydberg atoms dominates the trap loss in this regime. To explain the observed behavior, we note that even though the cross sections of ionizing Rydberg atom-electron collisions are smaller than the ones for *lm* mixing by a factor of ~$n$ [6], a single ionization collision overrides the effect of many earlier *lm*-mixing collisions. This is the case for $n > 40$ in Figs. 4a and 4b, where the ionization probability saturates as a result of both high density and large $n$. The data from Fig. 1b are reproduced for comparison in Fig. 4c to reiterate that at lower densities the long-lasting electron signal persists to $n \gg 40$.

In conclusion, cold Rydberg gases in a room-temperature thermal radiation field can exhibit production of high-$l$ Rydberg states and subsequent slow thermal ionization. The effect hinges on the temporary existence of a space-charge electron trap. From our results and the work of other groups we conclude that in a regime of moderate Rydberg atom density (~$10^8$ cm$^{-3}$ at $n \sim 40$) the dynamics of cold Rydberg gases is dominated by *lm* mixing and thermal ionization, while at higher densities rapid collisional ionization prevails. Since the radiation temperature is an important parameter in our system, we intend to perform future studies in a cryogenic enclosure with a variable wall temperature.

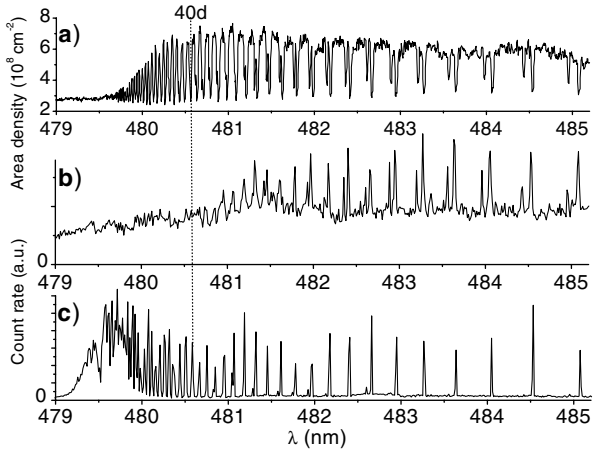


FIG. 4. Comparison of high and low Rydberg atom density. (a) Trap loss spectra for high density. (b) Electron signal at 10 ms for high density. (c) Electron signal at 10 ms for low density.

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*Note added.*—After acceptance of our Letter, we became aware of a related paper by Killian *et al.* [8]. We believe the recombination mechanism described in their paper does not play a role in our studies largely because our peak ion densities are lower than the ones used by Killian *et al.*

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