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Emission of fast atoms from a cold Rydberg gas

B. Knuffman and G. Raithel*
FOCUS Center, Department of Physics, University of Michigan, Ann Arbor, Michigan 48109-1120, USA
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Rydberg-Rydberg collisions in a cold, dense Rydberg gas can lead to the conversion of internal energy into center-of-mass energy of the colliding atoms, resulting in Rydberg-atom velocities much larger than the initial velocities in the gas. We prepare cold Rydberg-atom gases by laser excitation of laser-cooled atom clouds and use time-of-flight measurements to demonstrate the production of fast Rydberg atoms. The velocity distributions of Rydberg atoms emerging from the Rydberg-atom gases are obtained. State-selective field ionization spectra indicate a correlation between the production of fast Rydberg atoms and the presence of Penning ionization signatures. The dependence of the yield of fast Rydberg atoms on Rydberg atom density is measured.

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In several recent experiments, gases of cold Rydberg atoms have been created by photoexcitation of atoms laser-cooled in magneto-optical traps (MOTs) into Rydberg states. The internal states of atoms in cold Rydberg gases evolve through state-mixing and ionizing collisions with electrons and other Rydberg atoms [1–4]. In addition to the changes in their internal states, the velocities of the Rydberg atoms will increase from the initial thermal velocities of ~10 cm/s, as internal-state energy is converted to center-of-mass energy. This can result in the production of relatively fast Rydberg atoms having velocities of ~10 m/s [5]. Until now, there has been no experimental study of how the interactions in a cold Rydberg gas alter the center-of-mass energy of the Rydberg atoms. At high densities, cold Rydberg-atom gases have also been found to evolve into cold, neutral plasmas [6,7]. Such plasmas have also been created directly by excitation beyond the photoionization threshold, and have been studied extensively from both a theoretical and experimental perspective [8–12]. Presumably, fast Rydberg atoms with velocities in the range of 50 m/s are formed in cold plasmas via three-body recombination [13,14].

In this paper, we report on the detection of Rydberg atoms with velocities up to ~10 m/s emitted from cold Rydberg-atom gases. The velocity distributions of these fast atoms have been characterized using a time-of-flight (TOF) measurement. Furthermore, we have studied the production of fast Rydberg atoms as a function of the initial Rydberg-atom density. Using state-selective field ionization (FI) spectra to obtain the probability distribution of ionization electric fields, we are able to characterize the Rydberg-gas conditions that lead to the production of fast Rydberg atoms, and have found a correlation between the signature of Penning ionization and the production of fast Rydberg atoms.

A collision between two Rydberg atoms is likely to result in Penning ionization,

$$\mathrm{Ry}(n_0) + \mathrm{Ry}(n_0) \rightarrow \mathrm{Ry}(n) + \mathrm{ion} + e^-, \qquad (1)$$

with n_0 and n denoting the initial and final principal quantum numbers, respectively. In a cold Rydberg gas, two atoms are initially accelerated toward each other by attractive forces, increasing the kinetic energy of the atoms during the first leg of the collision. For atoms in high-angular-momentum states, the attraction is due to electric-dipole forces, which depend on the interatomic separation as R^{-4}, and on the mutual orientation of the permanent dipole moments of the atoms. When R is small enough that the Rydberg-atom interaction energy is comparable to the binding energy of the Rydberg state, one of the atoms can be ionized, and the other atom deexcited into a more deeply bound state. The free electron escapes within a fraction of a ns, leaving the ion and the Rydberg atom in close proximity to each other. A net repelling force between the ion and the positive core of the Rydberg atom then causes the atom and the ion to separate, further increasing the kinetic energy of the ion and the Rydberg atom [5]. In Eq. (1), the final state of the bound atom is constrained by $n \leqslant n_0/\sqrt{2}$, in order to provide at least enough energy to ionize one atom. Under the approximation that the initial kinetic energies of the atoms are negligible, the increase in binding energy of the atom that remains bound equals the sum of the initial binding energy of the ionized atom and the kinetic energies of the three product particles. While most of the energy is transferred to the electron, the kinetic-energy transfer to the product ion and Rydberg atom is sufficiently large that their velocities should exceed the initial velocity by a couple of orders of magnitude [5].

In our experiment, ^{85}Rb atoms are loaded into a MOT from a low-velocity intense source (LVIS [15]). The MOT light is used to excite atoms from the $5S_{1/2}$ to the $5P_{3/2}$ state. Atoms are then excited from the $5P_{3/2}$ state to the Rydberg state by a tunable pulsed dye laser ($\lambda \approx 480$ nm, pulse width ≈ 10 ns, bandwidth ≈ 15 GHz, pulse energy $\lesssim 1$ mJ, fluence $\Phi \approx 3 \times 10^{15}$ photons/cm^2). Figure 1 shows the apparatus used to measure time-of-flight distributions of the Rydberg atoms emitted from the cold Rydberg gas. Fast Rydberg atoms created in the gas travel 2 cm from the MOT location to a remote FI region located between a semicircular electrode and the first extraction electrode. Using relaxation calculations, we estimate the electric field in this region to be ≈ 400 V/cm, which is strong enough to ionize low-angular-momentum Rydberg atoms down to $n=30$ and high-angular-momentum atoms down to $n=42$. Electrons resulting from

*Email address: graithel@umich.edu

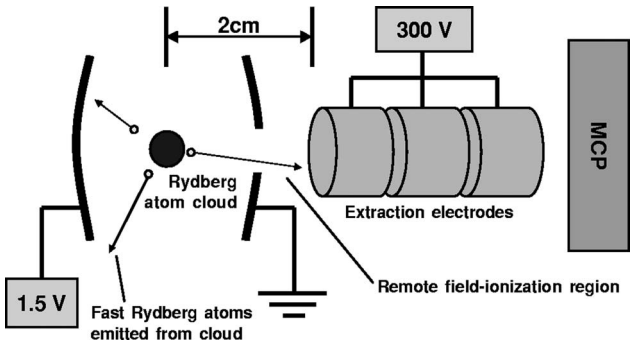

FIG. 1. Experimental setup for time-of-flight measurement. A fraction of the fast Rydberg atoms emitted from the cloud travels into the remote field ionization region between the semicircular electrode and the first extraction electrode. Rydberg atoms in this region are field-ionized, and the resulting electrons are counted by the microchannel plate detector.

Rydberg atoms ionized in this region are then counted by an MCP located approximately 10 cm away. A photon counter then records the MCP pulses over a 0.5–1 ms window with variable delay.

From earlier experiments [1] it is known that thermal ionization of high-angular-momentum Rydberg atoms produces time-delayed electrons up to tens of milliseconds after the excitation of the Rydberg gas. To ensure that such time-delayed electrons could not migrate from the atom cloud location into the remote FI region and produce a signal that could be mistaken for a FI signal of fast Rydberg atoms, a bias voltage of 1.5 V is applied to the left semicircular electrode in Fig. 1. The resultant electric field slightly overcompensates the fringe electric field produced by the extraction electrodes at the location of the atom cloud, leading to a weak net field of ≈ 0.2 V/cm (pointing from left to right in Fig. 1). This field deflects electrons generated by time-delayed thermal ionization of slow Rydberg atoms to the left semicircular electrode so that the signal measured in the remote FI region is solely due to fast Rydberg atoms.

Figure 2 shows typical time-of-flight data for $n_0=90$. Since the distance traveled by the atoms from the MOT to the detection region is $L=2$ cm, the velocity distribution $P_V(v)$ of the detected fast Rydberg atoms, shown in Fig. 2(b), can be calculated from the time-of-flight data $P_T(t)$, shown in Fig. 2(a), using the relation $P_V(v)=P_T(L/v)L/v^2$. The most probable velocity of the detected fast Rydberg atoms is found to be slightly less than 5 m/s, and the width of $P_V(v)$ about 5 m/s. The corresponding kinetic-energy range exceeds that of Rb atoms in a MOT (velocity ~ 0.15 m/s) by a factor of about 1000. The most probable velocity from Fig. 2 agrees within 20% with classical calculations by Robicheaux [5], and the shapes of simulated and measured velocity distributions are very similar. Based on these results, we believe that the fast Rydberg atoms are a product of Penning ionization. This assessment is supported by state-selective FI data (see Fig. 4 below).

The most probable time of flight in Fig. 2 is 4 ms, which exceeds the natural lifetime of most Rydberg states. In the following, we consider the effect of radiative decay and thermal ionization of fast Rydberg atoms prior to detection. For

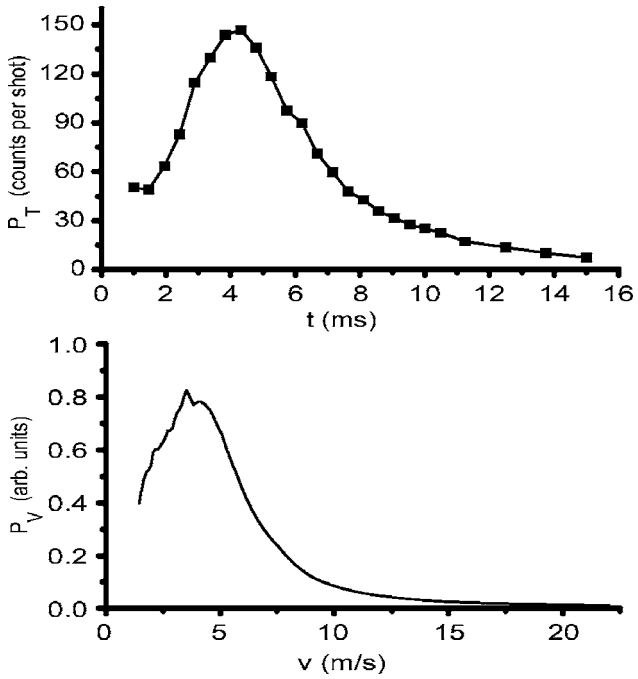

FIG. 2. Time-of-flight (top) and velocity distribution (bottom) of Rydberg atoms emitted from a cold Rydberg-atom gas with initial principal quantum number $n_0=90$, central Rydberg atom density 1.4×10^8 cm^{-3}, and Rydberg atom number 3×10^5. The time-of-flight t is the elapsed time between the Rydberg excitation pulse and the detection of the Rydberg electron by the MCP detector.

$n_0=90$, the principal quantum numbers of the fast Rydberg atoms are expected to be in the range $45\lesssim n\lesssim 65$, peaking around $n=60$ [5]. Also, based on the FI data presented in Fig. 4 below, we assume that the Rydberg-atom products of Penning-ionizing collisions typically are in high-angular-momentum states. Dipole selection rules allow atoms in high-angular-momentum states to only undergo transitions to other high-angular-momentum states, so these atoms remain in Rydberg states for much longer than the lifetime of the initial high-angular-momentum state. To obtain a quantitative estimate of the fraction of detectable atoms that reach the remote FI zone, we have simulated the evolution of an ensemble of $n=60$ Rydberg atoms with initially random angular momenta, weighted $\propto(2l+1)$ and prepared in a 300 K radiation field, using a rate equation calculation developed previously [16]. We find that 4 ms after excitation, about 80% of the atoms still reside in detectable states ($n\gtrsim 35$ in our case). We are therefore certain that under the conditions of Fig. 2 most of the fast Rydberg atoms survive until detection, and that our TOF method yields good representations of the velocity distribution of fast Rydberg atoms produced in Rydberg-Rydberg collisions.

The product Rydberg states of Penning ionization are distributed over a certain n range. A subtle consequence of radiative decay or ionization is that we preferentially measure the velocities of the longer-lived, higher-n portion of the distribution and lose more of the shorter-lived, lower-n portion. Since it is likely that atoms produced with below-average n have above-average velocities (because of generally larger energy exchanges in the Penning collision process), prefer-

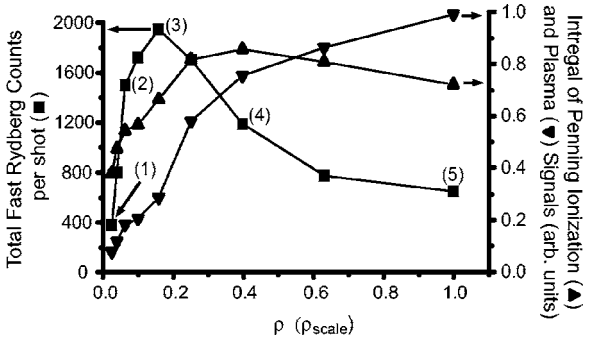

FIG. 3. Total production of fast Rydberg atoms (■) vs initial Rydberg atom density ρ at $n_0=90$, where $\rho_{\text{scale}}=9.1\times10^8$ cm^{-3} is a convenient density unit. The production of fast Rydberg atoms is compared with integrated Penning ionization (▲) and plasma (▼) signals obtained from state-selective FI spectra. For the numbered data points, the FI spectra are shown in Fig. 4.

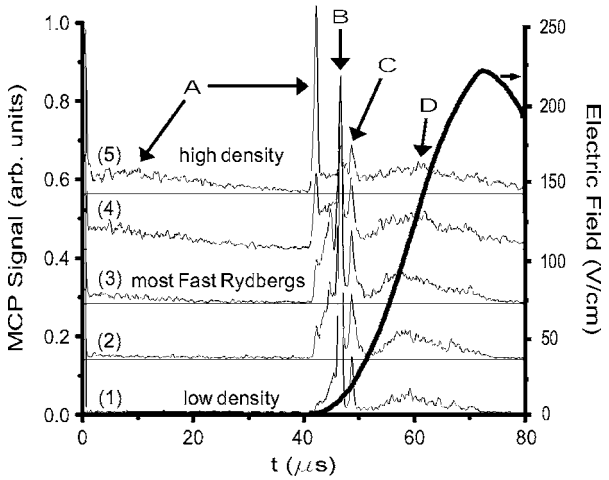

FIG. 4. Field ionization (FI) spectra for $n_0=90$ and Rydberg atom densities identified by the labels on the left and matching labels in Fig. 4. The Rydberg gas is excited at $t=0$, and the FI pulse (thick line) is applied between 40 μs$<t\lesssim$80 μs. Characteristic FI signatures: (A) plasma electrons, (B) initial state, (C) ℓ-mixing, (D) Penning ionization.

ential detection of high-n atoms distorts the measured velocity distribution. For the case $n_0=90$, we expect $45\lesssim n\lesssim 65$. Using the simulation discussed in the previous paragraph, we find that this n-range corresponds to survival probabilities ranging from about 66% at $n=45$ to 86% at $n=65$. Since the width of this range is quite small, for $n_0=90$ the difference between measured and actual velocity distributions of the fast Rydberg atoms is deemed minimal. However, we found that the estimated spread in survival probabilities of product atoms increases considerably with decreasing n_0. Therefore, we have limited most studies in this paper to large values of n_0 (namely 90 and 75).

We have studied the dependence of fast Rydberg atom production on the initial Rydberg atom density (■ in Fig. 3). Fast Rydberg atom production is determined by summing the MCP counts for a time-of-flight scan at a particular density over all times. At low densities, we observe a rapid increase of fast Rydberg atom production, while at the highest densities the production of fast Rydberg atoms is suppressed. Optimal production is observed at some intermediate density.

In order to understand the density dependence of fast-Rydberg-atom production, we have examined state-selective FI spectra (see Fig. 4) of cold Rydberg gases for various densities at which time-of-flight measurements were taken (data points in Fig. 3 labeled with numbers). During a Penning ionizing collision, one of the Rydberg atoms undergoes a transition into a more deeply bound state with $n\lesssim n_0/\sqrt{2}$ and presumably high angular momentum. The deexcited atoms should produce a broad FI signal at electric fields of order ten times the ionization electric field of the initial state, because the ionization electric field scales as n^{-4} and because high-angular-momentum atoms ionize at fields up to four times higher than those for low-angular-momentum atoms. We believe that the broad FI signal labeled "D" in Fig. 4 is due to the deexcited product atoms of Penning ionization. The net Penning ionization signal (▲ in Fig. 3), obtained by integrating over the "D"-signatures in Fig. 4, can be compared with the production of fast Rydberg atoms (■ in Fig. 3). Both quantities rapidly increase with density at low densities, exhibit broad maxima at intermediate density values, and decrease with density at high densities. These similarities indicate that Penning-ionizing collisions lead to the production of fast Rydberg atoms. At low densities, the Penning-ionization and fast-Rydberg signal steadily increase, because with increasing density it becomes increasingly likely that two atoms are sufficiently close to each other to "snap together" and undergo the reaction in Eq. (1). As the density significantly exceeds the density of optimal production of fast Rydberg atoms, the Rydberg gas increasingly evolves into a plasma, as evidenced by the free-electron signatures "A" in Fig. 4. Electron-Rydberg-atom collisions in the plasma cause the Rydberg atoms to ionize or to undergo transitions to lower Rydberg states that ionize at higher electric fields than are present in the remote FI region [2]. Both of these effects contribute to the detection of weaker Penning-ionization signatures in FI spectra and fewer fast Rydberg atoms at high densities.

The Penning-ionization signature is correlated with FI signatures of l-mixing (signal "C" in Fig. 4). Therefore, it seems likely that many Rydberg atoms first acquire large permanent electric dipole moments due to l-mixing collisions before undergoing Penning-ionizing collisions [2]. This sequence of processes appears reasonable because polar (high-l) atoms interact via dipole-dipole forces that are stronger than the van-der-Waals forces acting between the (initially) nonpolar low-l atoms.

By examining the production of fast Rydberg atoms at low densities, the number of fast Rydberg atoms produced should possess a simple power-law dependence on density. Figure 5 shows a plot of total fast Rydberg counts vs density for very low densities. The experimentally determined exponent for a power-law fit is 1.91. This exponent is close to 2, indicating that the production of fast Rydberg atoms results from binary collisions between Rydberg atoms, such as Penning ionization.

TOF data were taken for initial principal quantum numbers $n_0=90$, 75, 60, and 45. As n_0 was decreased from 90 to

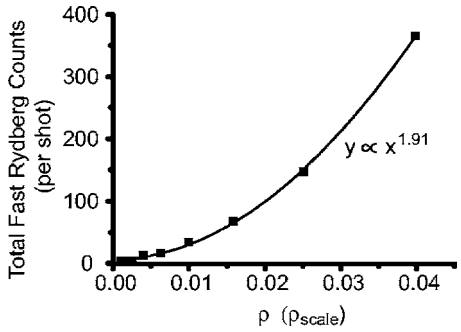

FIG. 5. Total production of fast Rydberg atoms vs initial Rydberg atom density ρ in the low-density limit at $n_0=75$, where $\rho_{\text{scale}}=7.7\times10^8$ cm^{-3}. A constant background of 17 counts was subtracted from the data set to compensate for dark counts on the MCP. The uncertainty for these counting measurements arises mainly from statistical noise, which is $\approx\sqrt{N}$. The data are fitted with the indicated power law.

45, the optimal yield of fast Rydberg atoms steadily decreased by about a factor 5, while the velocity distributions remained approximately the same. The signal decrease can be qualitatively explained as a consequence of radiative decay and thermal ionization [16], which cause a decrease of the survival probability of the Rydberg-atom products of Eq. (1) from about 85% at $n=60$ to less than 10% at $n=20$. The fact that we observe essentially n_0-independent velocity distributions contradicts the simulations in Ref. [5], where it is found that the velocities of the fast Rydberg atoms should scale as $1/n_0$. This discrepancy can partially be explained by the n-dependence of the survival probabilities of the product states in our time-of-flight experiment. An interesting possibility to further explain the discrepancy is that at lower n_0 we may increasingly observe "van de Graaf type" collisions that result in product velocities that are actually lower than those calculated for Penning ionization [5]. In these predicted collisions, the Rydberg atoms initially approach each other on an attractive dipole potential. During the collision, the Runge-Lenz vectors of the Rydberg atoms precess in such a way that the dipole potential becomes repulsive. In this process, both atoms remain bound and gain kinetic energy from the collision because the internal states of the atoms change during the collision. Future experiments will be required to distinguish between Penning ionization and van de Graaf type collisions of cold Rydberg atoms.

Fast Rydberg atoms could also form by three-body recombination in dense, expanding plasmas; however, we believe that this mechanism does not significantly contribute to our results. First, Rydberg atoms formed by three-body recombination are expected to have velocities $\sim$50 m/s [13], while our data indicate an average velocity of only $\sim$5 m/s and a rapid drop-off at velocities above $\sim$10 m/s (see Fig. 2). Second, the signal we observe already appears at low densities, where it scales like a binary collision effect (see Fig. 5) and where the Rydberg gas does not evolve into a plasma. In contrast, Rydberg-atom formation via three-body recombination presumably will exhibit a threshold behavior, because the evolution of the Rydberg gas into a plasma proceeds via an ionization avalanche [6,7].

In summary, we have performed TOF measurements on Rydberg atoms emitted from a cold Rydberg gas. We have characterized the velocity distribution of these atoms, and found that the atoms have acquired a factor of order 1000 in kinetic energy. Furthermore, we have correlated the production of these fast atoms with Penning ionization. While large parts of our results agree with simulations [5], it remains to be explained in future research why for small n_0 the experiments show lower atom velocities than the simulations. Also, in future experiments at higher atom densities we intend to study fast Rydberg atoms formed via three-body recombination.

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[1] S. K. Dutta *et al.*, Phys. Rev. Lett. **86**, 3993 (2001).
[2] A. Walz-Flannigan *et al.*, Phys. Rev. A **69**, 063405 (2004).
[3] W. Li, P. J. Tanner, and T. F. Gallagher, Phys. Rev. Lett. **94**, 173001 (2005).
[4] W. R. Anderson, J. R. Veale, and T. F. Gallagher, Phys. Rev. Lett. **80**, 249 (1998).
[5] F. Robicheaux, J. Phys. B **38**, S333 (2005).
[6] M. P. Robinson *et al.*, Phys. Rev. Lett. **85**, 4466 (2000).
[7] N. Vanhaecke *et al.*, Phys. Rev. A **71**, 013416 (2005).
[8] T. C. Killian *et al.*, Phys. Rev. Lett. **83**, 4776 (1999).
[9] S. Kulin, T. C. Killian, S. D. Bergeson, and S. L. Rolston, Phys. Rev. Lett. **85**, 318 (2000).
[10] D. Feldbaum *et al.*, Phys. Rev. Lett. **89**, 173004 (2002).
[11] F. Robicheaux and J. D. Hanson, Phys. Plasmas **10**, 2217 (2003).
[12] S. Mazevet, L. A. Collins, and J. D. Kress, Phys. Rev. Lett. **88**, 055001 (2002).
[13] T. C. Killian *et al.*, Phys. Rev. Lett. **86**, 3759 (2001).
[14] Y. Hahn, Phys. Lett. A **231**, 82 (1997).
[15] Z. T. Lu *et al.*, Phys. Rev. Lett. **77**, 3331 (1996).
[16] A. Walz-Flannigan *et al.*, in *Photonic, Electronic and Atomic Collisions (XXII ICPEAC Proceedings)*, edited by J. Burgdoerfer *et al.* (Rinton Press, Princeton, 2002).