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Author(s): Seiler, Christian; Hogan, S.D.; Schmutz, H.; Agner, J.A.; Merkt, Frédéric

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Collisional and radiative processes in adiabatic deceleration, deflection and off-axis trapping of a Rydberg atom beam

Ch. Seiler, S. D. Hogan, H. Schmutz, J. A. Agner and F. Merkt
Laboratorium für Physikalische Chemie, ETH Zürich, CH-8093, Switzerland
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A supersonic beam of Rydberg hydrogen atoms has been adiabatically deflected by 90°, decelerated to zero velocity in less than 25 μs and loaded into an electric trap. The deflection has allowed the suppression of collisions of the Rydberg atoms with atoms in the trailing part of the gas pulse. The processes leading to trap losses, i.e., fluorescence to the ground state, and transitions and ionization induced by blackbody radiation have been monitored over several milliseconds and quantitatively analyzed.

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The use of cold atoms and molecules in spectroscopic experiments and in studies of bimolecular collisions at low temperature has recently stimulated considerable efforts to devise experimental tools capable of producing state-selected cold molecular samples, either in electromagnetic traps or in velocity-controlled gas beams [1–4]. The use of supersonic beams in combination with deceleration represents one of the main approaches to generate cold gas-phase samples [3]. Deceleration methods enable one to produce cold samples of polar molecules by multistage Stark deceleration [5], of paramagnetic atoms and molecules by multistage Zeeman deceleration [6–8] and, at least in principle, of any atom or molecule by Rydberg-Stark [9–11] and optical deceleration [12]. These methods and variants based on travelling electric traps have been developed recently [10, 13], and further approaches, including the use of microfabricated devices [14] and continuous trap-loading schemes [15], are promising for many applications in chemical physics, high-resolution spectroscopy, metrology, quantum information processing, and in particular also antihydrogen studies.

Using these methods, the atoms or molecules to be decelerated only represent a small fraction of the gas in the supersonic expansion, the largest fraction being the carrier gas (usually a rare gas), precursor molecules (e.g., NH3 for NH [15]) and molecules in quantum states not suitable for deceleration. As the deceleration proceeds, the increasing-velocity difference between decelerated and undecelerated particles renders collisions unavoidable, potentially leading to loss and heating of the decelerated particles. These undesirable processes, referred to as the carrier-gas problem below, are difficult to quantify and rarely mentioned in the context of deceleration experiments. The carrier-gas problem becomes acute when (1) the deceleration is strong and a large velocity slip builds up before the trailing part of the gas pulse has overtaken the decelerated molecules, (2) the decelerated molecules are directed toward an on-axis target, such as a surface or another gas sample, because the impact of the carrier gas on the target may alter its properties or mask the effect one desires to study, and (3) cumulative trap loading from successive gas pulses is attempted, because the cold molecules trapped in a given experimental cycle are ejected by the carrier gas in subsequent pulses. The carrier-gas problem represents a limitation in our Rydberg-Stark deceleration and trapping experiments on atomic and molecular hydrogen [11, 16], and has so far prevented us from disentangling the effects of collisional and radiative trap loss processes.

We present here a Rydberg-Stark deceleration scheme which enables the rapid adiabatic 90° deflection of the Rydberg sample from the gas beam followed by the loading of an off-axis trap. By comparing the results of on-axis and off-axis trapping experiments at room temperature and at 125 K in the same experimental setup, we have (1) quantified important aspects of the carrier-gas problem in Rydberg-Stark deceleration, (2) studied the loss of Rydberg atoms from the trap under conditions where it is dominated by radiative processes, (3) clarified the role of fluorescence and blackbody-radiation-induced transitions in the trap, and (4) made first measurements of trap losses resulting from the zero-field region near the trap center.

A schematic diagram of the Rydberg-Stark decelerator and off-axis trap is depicted in Fig. 1(a). A supersonic beam of hydrogen atoms, indicated by the dashed arrow in Fig. 1(a), is produced by laser photolysis of NH3 seeded in krypton as described in Ref. [17]. The beam passes through a skimmer and enters the electrode setup which forms the Rydberg-Stark decelerator and trap. Photoexcitation to selected Rydberg-Stark states with principal quantum number n around 30 is achieved by resonant two-photon excitation via the 2S0P state. The two laser beams at 121 nm and ~ 365 nm are counterpropagating, enter the excitation region through a set of 1.6-mm-diameter holes in electrodes 1, 2 and 7, and cross the H atom beam at right angles. The excited hydrogen atoms move initially in the positive z direction. Following photoexcitation, a series of pulsed potentials are applied to the electrodes to decelerate and load the Rydberg atoms into the off-axis trap located 6 mm away from the axis of the undeflected supersonic beam.
trodes can be designed so as to either trap the atoms on-
34, 7, 8 (off-axis trap). Electrodes 5 and 9 (6 and 10) are separated by 10 mm in the x direction and are operated at potentials of −22 V (+22 V) to achieve confinement of the Rydberg atom cloud in the x direction and generate a field of 10 V/cm at the trap minimum. The lowest saddle point in the electric field distribution is 46 V/cm which corresponds to a trap depth of 1.3 cm⁻¹ (or $E/k_B = 1.9$ K) for the $n = 30, k = 19$ Rydberg atoms used in the experiments.

To study the effects of blackbody radiation, the electrode setup was enclosed on five sides by a 90 mm × 80 mm × 60 mm copper heat shield and cooled to $\sim 125$ K by connection to a liquid-nitrogen thermal bath. The Rydberg atoms loaded into the on-axis (or off-axis) trap are detected by pulsed field ionization (PFI) by applying a potential of +1.25 kV to electrodes 1 and 2 (or 2 and 7) which extracts the $H^+$ ions toward a microchannel-plate detector connected to a phosphor screen. Images of the $H^+$ ions are recorded with a CCD camera. The ion optics were designed so that atoms that are field ionized in the on- and off-axis traps are easily distinguished, as illustrated in Fig. 1(b) and (c). Numerical particle-trajectory simulations were carried out as described in Ref. [18] to predict and analyze the experimental results.

Figs. 1(b) and (c) display typical images of the $H^+$ ions recorded following deceleration of a 600 m/s beam of $n = 30, k = 19$ atoms. To obtain the image in panel (c), the atoms were extracted from the position of the on-axis trap, whereas to obtain the image in panel (b) the atoms were first deflected by 90° before being loaded into the off-axis trap, as described in more detail below. In the former (latter) case, the entire trapping process took less than 10 µs (25 µs). The asymmetric shape of the $H^+$ image in panel (b) results from the details of the electrode and ion optics configuration. The removal of the Rydberg atoms from the beam axis in only a few microseconds occurred after off-axis trapping opens up the possibility, exploited in the experiments described below, of studying the trap decay dynamics without interference from collisional processes.

The sequence of pulsed voltages applied to the ten electrodes can be designed so as to either trap the atoms on-axis between electrodes 1–4 as already demonstrated in Ref. [16] or off-axis between electrodes 2, 4, 7 and 8 (see Fig. 2(a) for the sequence used in this work). In the photoexcitation and trapping configuration (Fig. 2(b)), potentials of +12 V (−12 V) are applied to the electrodes 1, 4 and 7 (2, 3 and 8) forming two electric quadrupole traps in the yz-plane, with minima located at the midpoint of the four electrodes 1–4 (on-axis trap) and 2, 4, 7, 8 (off-axis trap). Electrodes 5 and 9 (6 and 10) are separated by 10 mm in the x direction and are operated at potentials of −22 V (+22 V) to achieve confinement of the Rydberg atom cloud in the x direction and generate a field of 10 V/cm at the trap minimum. The lowest saddle point in the electric field distribution is 46 V/cm which corresponds to a trap depth of 1.3 cm⁻¹ (or $E/k_B = 1.9$ K) for the $n = 30, k = 19$ Rydberg atoms used in the experiments.

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The operational principle of the trap and its main characteristics are described in Figs. 2 and 3. The electric potentials applied to the relevant electrodes during deceleration and trap loading are presented in Fig. 2(a). The pulse sequence consists of five phases: photoexcitation in the trap configuration ($t = 0$ µs), initial deceleration and deflection ($0 < t < 5$ µs), turning of the electric field minimum around electrode 2 ($5 < t < 15$ µs), final deceleration in the y direction ($15 < t < 25$ µs) and off-axis trapping ($t \geq 25$ µs). The relevant parts of the electric-
field distributions in the yz plane at the beginning of each of these five phases are displayed in Figs. 2(b) to 2(e). The corresponding times are indicated by dashed lines in Fig. 2(a).

![Simulated phase-space acceptance](image)

FIG. 3: (a, b, c) Simulated phase-space acceptance (grey areas) of the electrostatic deceleration and on-axis (top panels) and off-axis (bottom panels) trapping procedures for \( n = 30, k = 19 \) Rydberg states of H in the (a) x, (b) y and (c) z directions. The photoexcited Rydberg atoms are represented as black dots.

The grey areas in Fig. 3(a), (b) and (c) represent the calculated phase-space acceptance of the deceleration and trapping process outlined in Fig. 2 for the x, y and z dimensions, respectively. The top (bottom) panels correspond to on-axis (off-axis) trapping. For the simulations, we adopted the mean longitudinal velocity (600 m/s) and the velocity and position distributions defined by the geometric constraints of the experiment (see Ref. [18]). In the phase-space diagrams, the photoexcited atoms are represented as black dots. The figure demonstrates that approximately 90% of the initially prepared Rydberg atoms end up in the trap. Most of the loss occurs in the y dimension.

In all experiments described below the trap decay rates were measured by monitoring the H\(^+\) ion signal as a function of the delay time between photoexcitation and PFI. Fig. 4(a) shows measurements of the trap decay at room temperature following on-axis trapping (open circles) and off-axis trapping (full circles) of low-field-seeking \( n = 30 \) Rydberg states of H. The decay rates are identical beyond 170 \( \mu \)s, and the difference at early times is evidence for collisional losses of \( \approx 40\% \) induced by the trailing part of the gas pulse and of their suppression by the off-axis trapping procedure. The fluorescence lifetimes \( \tau_\text{fl} \) of a \( n = 30 \) Rydberg-Stark state can be estimated using Table 15 of Bethe and Salpeter and applying the expected \( n^{3.36} \) scaling-law [19] to be 220 \( \mu \)s and would correspond to the decay described by the thick dashed line. Decay channels other than fluorescence therefore play a role in the experiment. Their time constant can be determined to be \( \tau = 350 \mu \)s using \( 1/\tau_\text{obs} = 1/\tau_\text{fl} + 1/\tau \) with \( \tau_\text{obs} = 135 \mu \)s. The additional decay is attributed to blackbody radiation effects, but contributions from Rydberg-Rydberg collisions cannot be excluded at early times. Transitions between neighboring Rydberg states induced by blackbody radiation are such that they do not change the dipole moment significantly, and do not lead to trap losses but to a gradual redistribution of the initial population to neighboring Rydberg states. At room temperature, this redistribution rendered the observed trapping times largely independent of the \( n \)-value of the initially prepared Rydberg states. The observed trap loss can be attributed to direct ionization by blackbody radiation, if it is assumed that approximately 8% of the expected blackbody-radiation-induced depopulation of the initial state leads to ionization, as expected on the basis of the results presented in Refs. [20–22]. A further indication of the importance of blackbody-radiation-induced ionization was the observation that \( \tau \) was reduced by a factor of \( \approx 3 \) to \( \approx 110 \mu \)s after introduction of the copper heat shield under otherwise identical conditions (i.e., \( T = 300 \) K). The low-frequency radiation field is indeed strongly influenced by the mode structure resulting from metallic surfaces surrounding the experimental volume.

\( n \)-dependent trap decay becomes observable after cooling the device to \( \approx 125 \) K, as illustrated by the measurements presented in Fig. 4(b) which were carried out following excitation to \( n = 32, k = 17 \) and \( n = 36, k = 15 \) Rydberg states. The trap-loss curves in Fig. 4(b) show a nonexponential decay that was modeled by a Monte-Carlo simulation based on the following assumptions: (1) Trap loss is caused either by fluorescence to the ground state (\( \tau_\text{fl} \), see above), or by ionization induced by low-frequency radiation (\( \tau_\text{fl} \)), or by multiple transitions to Rydberg states with \( n \) values outside the range \( (n = 24 – 60) \) that can be trapped. (2) Transition rates \( K_{nk,n'k'} \) to neighboring Rydberg states with \( |n – n'| \leq 5 \) and \( nk = n'k' \) are dominant and can be calculated from the Einstein A coefficient and the photon occupation number \( \bar{n} \) with (in s\(^{-1}\))

\[
K_{nk,n'k'} = \bar{n} \frac{64\pi^4 \omega_0 a_{nk,n'k'} \Delta \omega}{3 \hbar c^2} \left| \langle nkm = 0 | \mu_z | n'k'm' = 0 \rangle \right|^2
\]

using the Stark wavefunctions in parabolic coordinates from Ref. [23]. (3) The ionization rate \( 1/\tau_\text{fl} \) corresponds to \( 8\% (4\%) \) of the total blackbody-radiation-induced depopulation rate \( 1/\tau_\text{BB} \) at 300 K (125 K) as expected from Ref. [24]. (4) This rate can be estimated from the sum \( 1/\tau_\text{BB} \) of the rates of all transitions with \( |\Delta n| \leq 5 \) using \( 1/\tau_\text{BB} = 1/\tau_\text{BB}^0 + 1/\tau_\text{BB}^\Delta \). This model accounts well for the experimental observations, however, only if the occupation number \( \bar{n} \) is raised by a factor of \( \approx 3 \) which we believe is also caused by the copper heat shield surrounding the device. Our calculations (solid lines in Fig. 4(b)) predict that fluorescence to the ground state, ionization, and multiple \( \Delta n \) transitions to untrappable Rydberg states account for 87%, 12% and 1%, respectively, of the observed trap loss during the first 2 ms at 125 K. At 300 K,
these percentages are 57%, 40%, and 3%, respectively. This prediction implies that continuous loading of ground state H atoms into a magnetic trap following Rydberg-Stark deceleration should be possible and become more efficient as the temperature is reduced.

Fig. 4(c) compares two measurements of trap decay following off-axis trapping of $n = 30$, $k = 19$ Rydberg atoms. The first was carried out with the trap minimum at 0 V/cm (electrodes 9 and 10 both at +22 V, open circles), the second with the trap minimum at 10 V/cm (electrodes 9 and 10 at ±22 V, full circles). In the former (latter) case, the decay up to 700 µs is fitted by an exponential function with a time constant of 300 µs (350 µs). This measurement reveals a further source of decay with a rate of $\approx 500 s^{-1}$ when the trap has a zero field minimum. We attribute this decay to transitions caused by interactions between neighboring Rydberg atoms and by fluctuating electric fields in the trap.

The results presented herein demonstrate that Stark deceleration and off-axis trapping of Rydberg atoms enable one to suppress collisional losses, to study trap-loss mechanisms over extended periods of time and open an attractive route toward continuous trap loading schemes. This work is supported by the Swiss National Science Foundation under Project 200020-132688 and by the European Research Council advanced grant program under Project 228286.


FIG. 4: (color online). (a) On-axis (open circles) and off-axis (full circles) trap loss of initially prepared $n = 30$, $k = 19$ Stark states. The dashed line corresponds to the $n = 30$, $k = 19$ radiative lifetime. The solid lines are single-exponential functions, fitted to the experimental data beyond 200 µs. (b) Measurement of the decay of atoms from the off-axis trap for $n = 32$ and $n = 36$ hydrogen Rydberg states at an electrode temperature of 125 K up to 2 ms with the corresponding Monte-Carlo simulations. (c) H$^+$ ion signal detected from the off-axis electrostatic trap as a function of the delay between photoexcitation and off-axis trapping of Rydberg atoms. The first was carried out with the trap minimum at 10 V/cm (electrodes 9 and 10 at −22 V, open circles), the second with the trap minimum at 0 V/cm (electrodes 9 and 10 both at +22 V, full circles). In the former (latter) case, the decay up to 700 µs is fitted by an exponential function with a time constant of 300 µs (350 µs). This measurement reveals a further source of decay with a rate of $\approx 500 s^{-1}$ when the trap has a zero field minimum. We attribute this decay to transitions caused by interactions between neighboring Rydberg atoms and by fluctuating electric fields in the trap.

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