PHYSICAL REVIEW A **75**, 031402(R) (2007)

Multistage Zeeman deceleration of hydrogen atoms

Nicolas Vanhaecke,[1,2] Urban Meier,[1] Markus Andrist,[1] Beat H. Meier,[1] and Frédéric Merkt[1]
[1]*Physical Chemistry, ETH Zürich, CH-8093 Zürich, Switzerland*
[2]*Laboratoire Aimé Cotton, batiment 505, Campus d'Orsay, 91405 Orsay, France*
(Received 30 November 2006; published 14 March 2007)

The deceleration of beams of neutral particles possessing an electron spin with time-dependent inhomogeneous magnetic fields is demonstrated experimentally. Half the kinetic energy of a velocity-selected part of a pulsed supersonic beam of hydrogen atoms in the ground state is removed using six pulsed magnetic field stages.

DOI: 10.1103/PhysRevA.75.031402 PACS number(s): 33.55.Be, 33.80.Rv, 39.10.+j

Over the last few years there has been a growing interest in developing tools to manipulate the translational motion of cold and ultracold neutral atoms and molecules in the gas phase. This interest is stimulated by important applications of cold atoms and molecules in several fields of physics and chemistry such as (ultra)cold chemistry, metrology, or quantum-information technology [1,2]. Among the experimental techniques developed so far, the deceleration of supersonic beams of ground- or excited-state atoms or molecules stands out in terms of versatility. So far, only the interaction with electric fields has been exploited in the so-called Stark-deceleration technique [3], in the optical Stark-deceleration technique [4], and in the Rydberg-Stark-deceleration technique [5,6]. All these techniques rely on the forces which result from the interaction between inhomogeneous electric fields and particles with either a permanent or an induced electric-dipole moment. Although neutral atoms or molecules possessing a magnetic-dipole moment are numerous, to date no real experimental attempt has been made toward decelerating such species using inhomogeneous magnetic fields. Deceleration techniques based on the magnetic interaction would allow the deceleration of a wider range of species, in particular atoms and nonpolar free radicals to which the Stark-deceleration techniques cannot be applied. Free radicals are usually very reactive and radical-radical reactions are often barrierless and exothermic; i.e., they are expected to remain highly reactive even at very low temperature. General methods to generate (ultra)cold samples of free radicals are thus particularly relevant in the context of cold chemistry.

The spatial manipulation of neutral particles with external magnetic fields has a long tradition in physics and chemistry [7]. It relies on the force $\vec{F}$ acting on the particles, which amounts to

$$\vec{F} = \mu_{eff} \vec{\nabla} B, \tag{1}$$

where μ_{eff} is the quantum-state-specific effective magnetic-dipole moment of the species and B is the magnetic field strength. Such manipulations started with the celebrated demonstration of space quantization by Gerlach and Stern in their historic experiment splitting a beam of silver atoms by means of an inhomogeneous magnetic field [8]. The deflection of molecular beams in inhomogeneous magnetic fields has also been an important step in the development of nuclear magnetic resonance spectroscopy [9].

In this Rapid Communication, we report the successful deceleration of hydrogen atoms in their ground state using pulsed inhomogeneous magnetic fields. In a multistage setup of six magnetic stages, half the kinetic energy of ground-state H atoms is removed, resulting in a decrease in velocity from 313 m/s to 225 m/s.

The experimental setup consists of an atom beam machine, schematically depicted in Fig. 1. A pulsed supersonic beam of H atoms with a mean velocity of about 380 m/s is produced by 193 nm photolysis of NH_3 seeded in Xe (1:10) in a quartz capillary located at the orifice of a pulsed valve (General Valve Series 9, repetition rate 10 Hz, stagnation pressure 3 bar). The H atoms are exclusively in the $1\,^2S_{1/2}$ state, which is split at zero magnetic field into a singlet ($F=0$) and a triplet ($F=1$) component by the hyperfine interaction. The top-left inset of Fig. 1 shows the Zeeman effect of

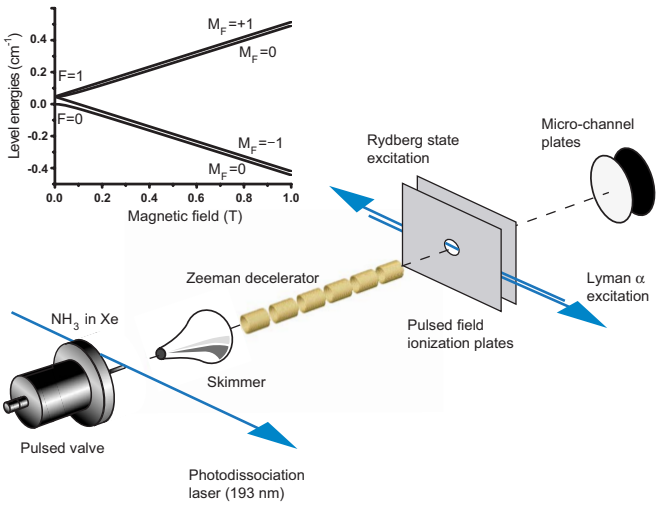

FIG. 1. (Color online) Schematical representation of the experimental setup. The skimmed supersonic beam of H atoms passes through the center of an array of coils and then intersects both vuv and uv laser beams between two metallic plates. The H^+ ions produced by pulsed field ionization are then detected on the microchannel plates. The top-left inset shows the details of the Zeeman effect of the H atom in the $1\,^2S_{1/2}$ state. The hyperfine structure dominates at low magnetic field strength whereas at high magnetic field strength the electron spin is strongly coupled to the field axis.

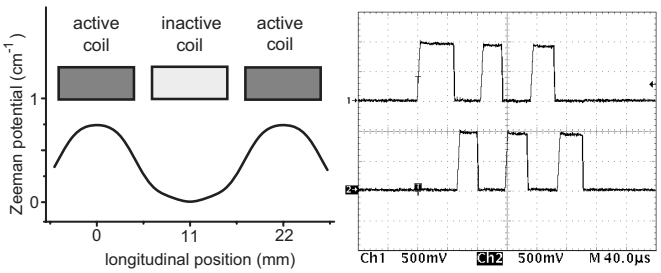

FIG. 2. Left panel: Zeeman potential energy experienced on the beam axis by a H atom in the upper low-field-seeking state ($M_F=+1$), when a current of 200 A flows through every second coil (referred to as "active"). Right panel: screen snapshot of a two-channel oscilloscope measuring currents flowing through both sets of three coils (see text). The current is about 200 A, measured through a small 10-mΩ shunt resistor with a 6-dB attenuator.

the M_F components in the presence of a magnetic field, where M_F is the projection of the total angular momentum F onto the magnetic field axis. The H atom beam passes through a 2-mm-diameter skimmer 1.7 cm downstream from the capillary exit and enters the deceleration chamber. In the current setup, the decelerator (called Zeeman decelerator hereafter) consists of six magnetic stages. The magnetic field in these stages is generated by 7.8-mm-long solenoids (5 mm inner diameter), which consist of two layers of each 21 windings of a 300-μm-diameter insulated copper wire. The cylindrical symmetry axis of the solenoids coincides with the beam axis, as shown in Fig 1. Pulsed currents up to 300 A can be driven through these solenoids, with rising and falling times as short as 5 μs. A pulse of current lasts typically a few tens of microseconds, and the maximum magnetic field at the center of each solenoid is about 2 T. In order to remove the power dissipated during operation, the solenoids are thermally connected to water-cooled ceramic pieces.

The H atoms in quantum states with a positive Zeeman effect (low-field-seeking states) lose kinetic energy when they enter a solenoid in which the current is flowing, because they experience an increasing magnetic field strength until they reach the center of the solenoid. The left panel of Fig. 2 shows the Zeeman potential energy of a H atom in the upper low-field seeking state ($M_F=+1$) on the beam axis, when a current of 200 A flows through every second coil. In the experiments decribed in this paper, the array of six coils is divided into two sets of three coils connected in series. The first (second) set consists of the first, third, and fifth (second, fourth, and sixth) coils. Homebuilt pulsed power supplies are used to drive currents up to 200 A through each set of coils with pulse sequences like the typical one displayed in the right panel of Fig. 2. When the H atoms approach the first coil the first set of coils is pulsed. It is switched off before the atoms reach the maximal field strength. The second set of coils is then turned on, and the procedure is repeated in the following coils. The time sequence to be applied to the decelerator is calculated in order to remove a fixed amount of kinetic energy per stage from the atoms in the $M_F=+1$ state, such that the bunch of decelerated atoms remains together stage after stage, according to the concept of longitudinal phase stability in synchrotronlike accelerators [10] and in Stark decelerators [11]. The field distribution is such that atoms in low-field seeking states are strongly transversally focused inside the solenoid, but slightly defocused outside. On average this leads to a net focusing force which ensures that the bunch of atoms in low-field-seeking states remains stable in the transverse dimensions throughout the Zeeman decelerator.

To detect the H atoms at the end of the decelerator, they are excited to a Rydberg state (typically with a principal quantum number around 37) using a two-color resonant excitation via the $2\,^2P$ level. The photoexcitation is carried out between two metal plates used to pulsed field ionize the Rydberg atoms; the H^+ ions are then accelerated towards microchannel plates and detected at the end of a short flight tube (see Fig. 1). The H atom beam intersects counterpropagating vuv and uv laser beams at right angles. The vuv radiation (300 μm beam waist, tuned to the Lyman-α transition) is generated by resonant four-wave mixing in a 10-mbar Kr cell where the 212-nm output of a frequency-tripled pulsed dye laser and a 845-nm dye laser beam are superimposed. The uv radiation (tuned to $\approx$368 nm) is provided by a frequency-doubled tunable pulsed dye laser. All three dye lasers are pumped by the same pulsed Nd:YAG laser so that vuv and uv beams arrive in the interaction zone at the same time, in order to avoid signal losses caused by the decay of the atoms in the $2\,^2P$ state, which has a radiative lifetime of only about 1.2 ns.

The intensity of the H^+ signal is displayed in Fig. 3(a) as a function of the time delay between firing the photodissociation laser and exciting the H atoms to the Rydberg state—i.e., as a function of the time of flight (TOF) of the neutral atoms between the source and the detection—for different operational conditions of the Zeeman decelerator. TOF profiles result from the contribution of all M_F components, and one expects therefore the TOF profiles to present complicated structures. Trace (i) shows the slowest part of the TOF distribution of the beam of H atoms when no current flows through the solenoids. From a measurement (not shown) of the entire TOF distribution, a mean velocity of 380 m/s and a velocity spread of 40% (full width at half maximum) of the beam of H atoms are deduced. Trace (ii) of Fig. 3(a) shows the TOF distribution of the H atoms when the decelerator is operated with a time sequence calculated to guide the atoms at a velocity of 313 m/s. A current of 140 A flows in the solenoids and the switching times—i.e., the timings when the currents are switched on and off—are calculated such that in each stage the H atoms in the $M_F=+1$ component initially moving at 313 m/s first gain kinetic energy, then lose the same amount of kinetic energy. This is equivalent to the operation of a Stark decelerator at the so-called phase angle of 0° [11]. The non-negligible effects of the rising and falling times of the magnetic fields are taken into account in the calculation of the switching times. The signature of the phase-space stability of the guiding process is clearly visible: the peak of H atoms that arrive around 655 μs after the photodissociation is sharp and corresponds to the atoms that are guided at 313 m/s. Moreover, the amount of atoms that make it through the decelerator is enhanced compared to the free-flight operation as a result of the transverse focusing properties of the decelerator. For the measurements presented

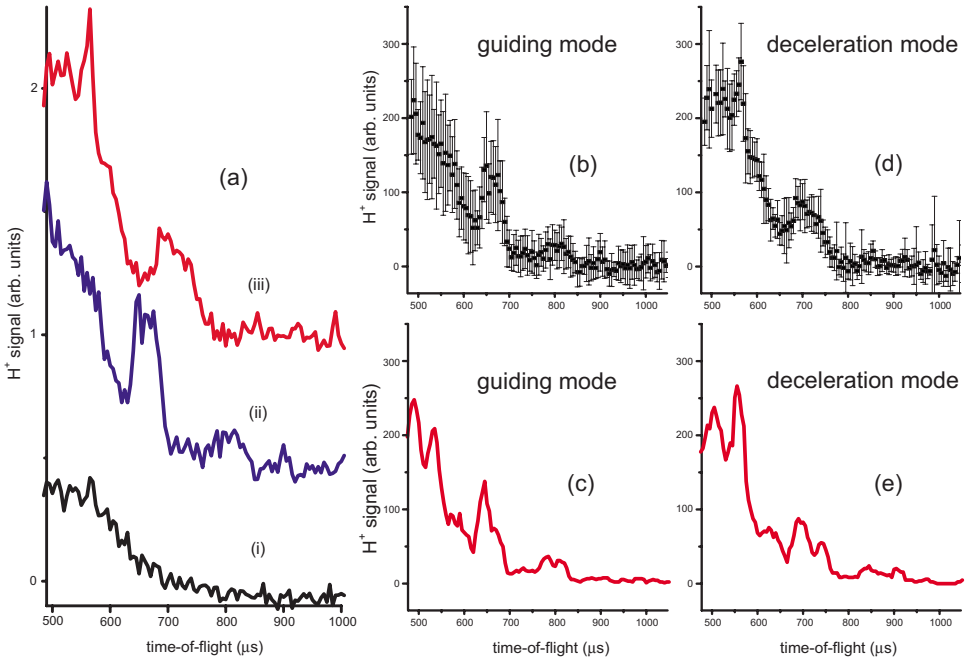

FIG. 3. (Color online) (a) Measured time-of-flight profiles of a beam of H atoms in the $^2S_{1/2}$ ground state passing through the Zeeman decelerator: (i) decelerator kept off, (ii) decelerator operating in a guiding mode at 313 m/s, and (iii) decelerator operating to slow down H atoms from 313 m/s down to 225 m/s. For the sake of clarity, measured profiles are given an offset, but are all on the same vertical scale. (b) [(d)] Measured time-of-flight profiles of H atoms after passing through the decelerator operating in the guiding mode at 313 m/s [decelerating from 313 m/s down to 225 m/s], including experimental error bars (one standard deviation). (c) [(e)] shows the output of a trajectory simulation using the same guiding [decelerating] time sequence.

in trace (iii), the switching times are set to decelerate atoms that enter the decelerator with a velocity of 313 m/s. With this time sequence, which corresponds to a phase angle of 55°, the atoms in the $M_F=+1$ component lose on average 0.33 cm^{-1} of kinetic energy per stage. The selected atoms are slowed down to a final velocity of 225 m/s, corresponding to a TOF of 740 μs. For such atoms, about half the initial kinetic energy is removed in the six magnetic stages.

The upper curves (b) and (d) in Fig. 3 show experimental data averaged over 250 shots in the guiding and the deceleration experiments, repectively, together with the experimental error bars (one standard deviation). The lower curves (c) and (e) show the results of three-dimensional Monte Carlo trajectory simulations of the guiding and the deceleration experiments, respectively. These calculations include the three-dimensional distribution of the magnetic field and the Zeeman effect of all M_F components of the $1\ ^2S_{1/2}$ state, as well as the time dependence of the magnetic field according to the actual time sequences applied to the decelerator. Special care has been taken to include the exact time dependence of the magnetic fields: the experimental rising and falling times of the magnetic field were taken as measured in the experimental setup. With the present electronic switching device, the applied magnetic field has to be switched off between two pulses, leaving the atoms in Earth's magnetic field for about 100 ns. This can induce changes of the quantum number M_F. To evaluate the probability that such a change takes place between two pulses, we solved the time-dependent Schrödinger equation for a four-M_F-component vector of the $1\ ^2S_{1/2}$ manifold during the switching off of the applied magnetic field, the 100-ns time interval in Earth's magnetic field, and the switching on of the applied magnetic field. Initially the applied magnetic field is on and the vector is an eigenstate of the Hamiltonian which includes at all times the hyperfine and the Zeeman interactions with the applied and Earth magnetic fields. Earth's magnetic field and the axis of the decelerator are assumed to be at right angles, which corresponds to the experimental configuration. Probabilities that a change of the quantum number M_F takes place within the $F=1$ manifold between two magnetic field pulses have been found to be on the order of 10%–20% and included in the trajectory simulations. As shown in panels (b)–(e) of Fig. 3, the overall structures of the experimental TOF profiles are quantitatively reproduced in the calculations. Agreement between calculated and measured TOF distributions is not only good for the arrival times of the atoms, but also for the intensity ratio between the bunch of atoms that are captured by the decelerator and the rest of the atoms that are not. The reliable modeling by Monte Carlo trajectory simulations of the experiment without any adjustable parameter validates the operational principle of the Zeeman decelerator and can be used to design the next generation of experiments aiming at slowing further H atoms and decelerating heavier species. The electronic switching device is currently being improved to control each coil independently, which will allow us to avoid the time intervals during which the magnetic field had to be switched off.

In conclusion, we have demonstrated the proof of principle of a multistage Zeeman decelerator and efficiently decelerated a supersonic beam of H atoms with time-dependent inhomogeneous magnetic fields. For this proof-of-principle

experiment the lightest atom was chosen, so that a substantial deceleration could be achieved with only a few magnetic stages. The deceleration technique presented here can be applied to a large variety of atoms and molecules with an unpaired electron, including nonpolar molecules such as molecular oxygen. All radicals can be decelerated with pulsed magnetic fields, as well as atoms and molecules in metastable states of nonzero electron spin, provided that the lifetime of these states is compatible with the deceleration time. The number of stages needed to decelerate a molecule in a Zeeman decelerator and possibly bring it to a standstill depends on the ratio between its effective magnetic moment and its mass. Calculations based on trajectory simulations show that molecular oxygen could be brought to a standstill in a Zeeman decelerator of about 100 stages. The Zeeman-deceleration technique presented here offers the possibility to slow molecular beams of a broad range of species that are of great interest in scattering and reaction experiments.

We thank Dr. M. Tomaselli for his contribution in the early phase of this project, Dr. S. Hogan and Dr. E. Vliegen for many fruitful discussions and their technical assistance, M. Vecellio for his technical support, and Dr. F. Jenni (PSI Villigen) for providing expertise concerning the current drivers. This work is supported by ETH Zürich including QSIT (Quantum Systems for Information Technology) and the Swiss National Science Foundation under Project No. 200021-113886.

[1] H. L. Bethlem and G. Meijer, Int. Rev. Phys. Chem. **22**, 73 (2003).
[2] J. Doyle, B. Friedrich, R. Krems, and F. Masnou-Seeuws, Eur. Phys. J. D **31**, 149 (2004).
[3] H. L. Bethlem, G. Berden, and G. Meijer, Phys. Rev. Lett. **83**, 1558 (1999).
[4] R. Fulton, A. I. Bishop, and P. F. Barker, Phys. Rev. Lett. **93**, 243004 (2004).
[5] S. R. Procter, Y. Yamakita, F. Merkt, and T. P. Softley, Chem. Phys. Lett. **374**, 667 (2003).
[6] E. Vliegen and F. Merkt, J. Phys. B **39**, L241 (2006).
[7] N. F. Ramsey, *Molecular Beams* (Oxford University Press, London, 1956).
[8] W. Gerlach and O. Stern, Ann. Phys. **16**, 673 (1924).
[9] I. I. Rabi, J. R. Zacharias, S. Millman, and P. Kusch, Phys. Rev. **53**, 318 (1938).
[10] E. M. McMillan, Phys. Rev. **68**, 143 (1945).
[11] H. L. Bethlem, F. M. H. Crompvoets, R. T. Jongma, S. Y. T. van de Meerakker, and G. Meijer, Phys. Rev. A **65**, 053416 (2002).