Stark Acceleration of Rydberg Atoms in Inhomogeneous Electric Fields

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A method is proposed to decelerate neutral atoms in a thermal beam for purposes of precision spectroscopy, atomic clocks, or loading into shallow traps. Rydberg atoms exhibit large electric dipole moments suggesting that inhomogenous fields can exert forces on them. The resultant change in kinetic energy is equal to the Stark shift of the Rydberg state. It has been found that a series of tandem electrode pairs can reduce atomic velocities by a factor of 10. Various limitations and constraints are discussed.

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There has recently been great interest in optical techniques for exerting controlled forces on free, neutral, single atoms.[1-6] Many of these have been motivated by the opportunity to perform precision spectroscopy on cooled or even trapped samples of free atoms. Wing[6] has suggested an electrostatic trap based on the "permanent" dipole moment that arises from the very nearly exact degeneracy of Rydberg states with different angular momenta. In the present paper we propose a method for electrostatic Rydberg cooling based on the Stark effect.

Outer electrons of highly excited (Rydberg) atoms are found at very large distances ($r \sim n^2$) and therefore exhibit large electric dipole moments in weak and moderate fields. This suggests that a nonuniform electric field can be used to decelerate such atoms from thermal velocity to much slower speeds (cooling). We propose a scheme that exploits this property to decelerate a thermal atomic beam and produce a sample of slow atoms in a vacuum for precision spectroscopy, atomic clocks, or loading into shallow atomic traps.

The classical picture given above has a straightforward quantum-mechanical counterpart: Rydberg atoms exhibit very large Stark shifts that cause their attraction into (or repulsion from) regions of strong electric fields. Atoms entering or leaving a region of strong field could be significantly slowed. Their kinetic energy would be reduced by the Stark energy.

These Stark shifts ΔE_s can become comparable to the separation between adjacent Rydberg levels, $\Delta E_n = 1/n^3$ (atomic units).[7,8] To avoid complications from Stark mixing into states from other n manifolds we exclude fields large enough to produce shifts larger than one-half this energy difference as well as states with large quantum defects. For $n \geq 5$ this also avoids destruction of the sample atoms by field ionization. The first-order Stark shift in l-degenerate states has a maximum $\Delta E_s = (3/2)n^2F$ (F = electric field in atomic units, 5×10^9 V/cm), and setting this equal to $\Delta E_n/2 = 1/2n^3$ gives $F = 1/3n^5$. The magnitude of moderate laboratory fields ($F = 10^{-6}$) suggests that n should be at least 12–15 in order to achieve a maximum Stark shift $\Delta E_s = 2\times 10^{-4} \approx 40$ cm^{-1}. Since thermal energy is about 10^{-3} a.u. $\simeq 200$ cm^{-1}, it is clear that the scheme proposed here, called Stark acceleration, can only produce significant changes in thermal velocity in some tandem arrangement.

We propose a series of decelerating stages, each consisting of an inhomogeneous dipole electric field produced by a pair of oppositely charged cylindrical electrodes along the y direction as shown in Fig. 1. An atom traveling along the x direction (transverse to the field) is excited in one or two steps by a y-directed laser beam(s) to a state with parabolic quantum number $n_2 \cong n \cong 12$–15 in the region between the electrodes where F_z is largest. (Separation of the Schrö-

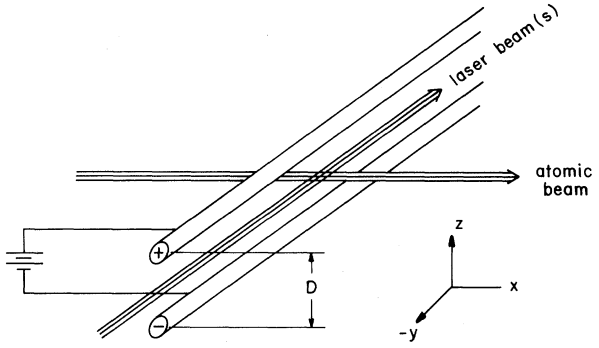

FIG. 1. Diagram showing coordinate system and a single pair of wire electrodes for one cooling scheme. The cylinder lens to focus the laser beam into a sheet of light in the yz plane is not shown. The electrodes may be a few millimeters in diameter, about 1 cm apart, and a few centimeters long.

dinger equation in parabolic coordinates, and identification of states by corresponding quantum numbers, is appropriate[7,8] for this domain of energies.) The atom would have to give up kinetic energy $\Delta E_s = (3/2)Fn(n_1 - n_2) \approx (3/2)Fn^2$ to escape to the low-field region further downstream. We choose states that radiatively decay to much lower n (much smaller Stark shift) before the atom encounters the next electrode pair downstream so that it is not accelerated into the next high-field region. In this way the atom is excited between the electrodes, is slowed as it escapes from the high-field region, decays to a state of much lower n and Stark coefficient, enters the next high-field region subject to little or no acceleration, and is again excited between the electrodes for further deceleration. A series of five such sets of electrodes crossed by laser beams could reduce atomic velocities significantly below thermal velocities, and might even cool a beam of atoms to a few kelvins.

We have carefully considered a large number of problems associated with Stark acceleration that might compromise its effectiveness. Each of these can be dealt with by appropriate choice of experimental arrangements, none of which are mutually exclusive. These problems include Doppler shifts, laser heating, defocusing, lifetimes, velocity distribution, optical pumping, Stark anticrossings with other states, and excitation probability. These problems and their solutions are discussed below.

Atoms moving at thermal velocities experience Doppler-shifted optical frequencies $\Delta\nu_D = \nu v/c \cong 1$–$2$ GHz. In order to eliminate these shifts from the excitation process, the laser beams are to be perpendicular to the atomic beam, crossing it between the electrodes as shown in Fig. 1.

The light beams of each frequency are all derived from the same laser by a series of mirrors, and cross the atomic beam alternately from the + and − y directions to minimize the transverse momentum imparted to the atoms. Since the average number of stages and therefore of excitations experienced by each atom is of order 10, the random motion imparted by spontaneous emission is a few times $h\nu/Mc \cong 10$ cm/sec (microkelvin heating). This additional transverse velocity is very small compared with the residual thermal component, about 10^3 cm/sec for 100:1 collimation of the atomic beam.

Atoms can be defocused from the beam if they begin slightly off axis (by amount z) or if there are small misalignments of the electrode structures. The magnitude of this effect can be estimated most easily from the classical point of view. A careful analysis of the force on a microscopic dipole in the (nondipole) field of charged rods separated by D shows that the z-directed force is $F_z \simeq 4pPz/R^4$ where p is the atomic dipole moment, $P = (\text{charge per unit length}) \times D$, and R is the average of the distances in the xz plane between atom and rods. The x-directed force is $F_x = 2pPx/R^4$. By calculating the impulse $= \int F\,dt$, we estimate that an atom will be deflected from the x axis through $\theta \simeq (z/2D)(\Delta E/E)$ where ΔE is its loss of kinetic energy E. In the early stages of our proposed decelerator $\Delta E/E \simeq \frac{1}{5}$, $D \simeq 1$ cm, and for $z \simeq 1$ mm we find $\theta \approx 0.01$ rad so that the beam would not be defocused by much more than its collimation ratio. In later stages $\Delta E/E$ is larger, and the effect of earlier stages could cause loss of half the atoms.

In order for Stark acceleration to be effective, the atoms must remain in the Rydberg state long enough to experience most of the field change. For a distance $s = (\frac{1}{2}\sqrt{7})D$ from the midpoint of the electrode pair, the field is reduced to $\frac{1}{8}$ of its maximum value and the energy loss for that stage is $\frac{7}{8}$ complete. Therefore, the lifetime of the Rydberg state must be at least $\tau = s/v \cong 8$ μsec, a typical[9] value for $n = 15$ atoms. As the atoms begin to slow down, they will experience less deceleration in each stage simply because they will decay before completely escaping the high-field regions. Fast atoms will therefore be decelerated more than slower ones, resulting in a narrowing of the velocity distribution. Even though the Stark acceleration technique proposed here is most effective for a particular velocity subgroup, that subgroup population is enhanced by the addition of a few more stages to take advantage of the "bunching" in velocity space. The price for this narrowing is the requirement of more stages and some slight additional defocusing.

Optical pumping can occur when an atom decays to a hyperfine sublevel of the ground state not in resonance with the exciting light. For alkali atoms there are two hfs ground-state components, typically separated by a few gigahertz. Atoms in either hfs level can be excited by the same laser if its amplitude is modulated to produce sidebands separated by the hfs so that both levels could be excited, but this precludes π-pulse excitation (see below). Two lasers, one in resonance with each hfs component, could also reduce the loss of excitable atoms by optical pumping.

There are limits to ΔE_s imposed by the atomic structure, particularly by the quantum defects of the various states.[8] Adiabatic anticrossings therefore limit the energy lost per stage to about $\Delta E_n/2$. There are n^2 Stark sublevels in Rydberg atoms emanating from each zero-field n state. Part of one section of the Stark map for $m=1$ Na is reproduced in Fig. 2.[10] We propose the use of an electric field and light frequency corresponding to point A, or another one with the following two properties. First, the Stark energy is linear in field so that it is subject to a rather large change from the operating field to zero. Using higher fields would not significantly alter the energy loss per stage of deceleration. Second, at the anticrossing there is a small but easily attainable region where the optical transition frequency is relatively independent of field. Field inhomogeneities will then have relatively little effect on the linewidth or transition probability. One must be careful in choosing such an anticrossing to avoid cancellation of the transition amplitude of interest.[11]

There is no way to assure that atoms passing through the laser beam will always emerge in the excited state. If the laser light is very weak only a small fraction of the atoms will be excited to the Rydberg states. As the intensity approaches saturation, as many as $\frac{1}{2}$ the atoms may be excited. Therefore it might take an average of twice as many stages for deceleration as expected since the atom spends only half its time in the excited state.

This problem can be ameliorated by excitation with a π pulse to assure a much higher probability of reaching the Rydberg state, achieved by tailoring the intensity and spatial extent of each transverse laser beam so that the atom experiences a π pulse passing through it. The most severe difficulty is achieving the same Rabi frequency ω_R for each atom in the beam, and assuring that the detuning Δ from the resonance frequency is $\leq \omega_R$.[12] The principal contribution to Δ comes from the Stark detuning of the atoms in the inhomogeneous field. By working near an anticrossing where the frequency is only weakly dependent on field, it is possible to minimize such effects. The energy of a state near an anticrossing is $E(F)=[E_0^2+(3n^2\delta F/2)^2]^{1/2}$ where $2E_0$ is the minimum separation of the anticrossing states and δF is the field separation from the center of the anticrossing. If the field inhomogeneity over the region occupied by atoms in the beam is $\delta F/F \cong 0.1\%$ (0.5-mm beam diam, electrodes 1 cm apart), and we choose $2E_0 \cong 1\ \mathrm{cm}^{-1}$ as in Fig. 2, and $3n^2F/2 \cong 40\ \mathrm{cm}^{-1}$, the energy spread of the atoms corresponds to about 48 MHz. In order to have $\omega_R/2\pi \geq 48$ MHz, and also produce a π pulse, the interaction time must be about 10 nsec, during which a thermal atom travels about 0.01 mm. A laser beam focused by a cylinder lens into a sheet of thickness 0.01 mm perpendicular to the atomic beam is a relatively simple accomplishment. The beam waist can easily have a Rayleigh length of several millimeters and cover the area of interest. The intensity required for $\omega_R \cong 48$ MHz is less than 100 mW/cm^2, which means a few-hundred-microwatt laser beam for focusing by a cylinder lens (one dimensional). Reduced effectiveness of π excitation might excite $\frac{3}{4}$ of the atoms. Coupled with an average 25% loss from defocusing results in net loss of $\simeq$ half the atoms per stage.

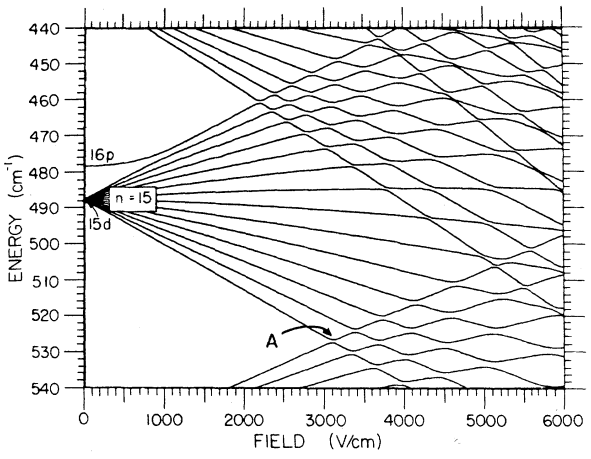

FIG. 2. The Stark levels of the $n=15$, $|m|=1$ levels of sodium.

Stark accelerators could have many variations. One particularly attractive alternative is electrodes composed of pairs (or triples) of plane parallel grids perpendicular to the atomic beam. The atoms would be excited in a high-field region between grids, pass through into a low-field region, and be decelerated. The electric field inhomogeneity would be considerably less in this case because the field between the mesh planes is much more uniform than that between charged wires. A value $\delta F/F \cong 0.01\%$ is readily achievable producing an energy spread of 0.48 MHz so that a π pulse requires an interaction time of 100 nsec, and a laser beam waist of about 0.1 mm. Another advantage of this design is a considerable reduction in the time an atom must spend in

the Rydberg state to be decelerated. The laser beams could be focused just a few millimeters upstream of the grid so that the travel time to escape the region between the grids is only a few microseconds. The defocusing would be slightly worse than for single pairs of wires because it is now two dimensional. The grids would not have to be aligned very carefully; as many as 10 pairs of 90% transmitting grids would still transmit $1/e$ of the atoms.

In another variation, atoms could be excited to appropriate angular momentum states in the low-field region upstream of dipole electrode pairs or parallel grid pairs, and be decelerated as they passed into the high-field region. (For dipole spheres the atoms would be subjected to x and y focusing.) Still another variation would use an rf field in some resonant structure that produced the appropriate inhomogeneity and a series of laser pulses, produced by optical delays of a single pulse, appropriately timed so that the Stark field was maximum when they excite the atoms. Then the Stark field would oscillate to its minimum, the atoms would be slowed, and they would decay or be π-pulsed to the ground state to avoid acceleration during the next rf cycle.

Other variations might use axial focusing by a laser tuned well below the atomic resonance frequency as described by Ashkin.[5] This laser could also provide the first of the usual two-step excitation to Rydberg states. It would be sufficiently intense and far enough off resonance to excite fully Doppler-shifted atoms as well as those that had been considerably slowed.

In all such schemes the fundamental principles are the same: Momentum is transferred to the electrodes by atomic dipoles moving in inhomogeneous electric fields. Work is done on the atoms by the circuits that maintain the voltages on the electrodes. Tandem operation is made possible by the excitation and decay of Rydberg states having large dipole moments even in very small fields.

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