Acceleration of a fast atomic beam by laser radiation pressure

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We have observed the acceleration of 100-keV Ne atoms by the radiation pressure of a laser field, tuned into resonance with the fast-moving atoms. The acceleration was 0.12 V/μsec in agreement with an analytic expression of the light-pressure-induced modification of the atomic phase-space distribution function. Representing the first stage of laser cooling of a fast beam, the observed acceleration shows the potential for laser cooling in heavy-ion storage rings.

I. INTRODUCTION

The finite velocity distribution of atoms has been the limiting factor for the resolution obtainable in classical spectroscopy. With the advent of lasers and particularly narrow-band tunable dye lasers, a major step forward was possible through the application of nonlinear spectroscopic techniques. The first-order Doppler broadening was substantially reduced through saturation spectroscopy, two-photon absorption, or Raman processes. However, problems associated with the finite velocity distribution, e.g., the second-order Doppler broadening and transit-time broadening, limit the optical resolution and can only be reduced by cooling and confining the atomic sample. Cooling methods known from low-temperature physics are not easily applied in the spectroscopy of free atoms or ions; however, the recent development of laser cooling has offered several new possibilities.[1] This technique utilizes the velocity-dependent radiation pressure from a near-resonant laser field to obtain spectroscopic samples with submillikelvin temperatures.

There are two qualitatively different types of optical forces: the dissipative spontaneous force[2,3] due to the resonant scattering of photons on atoms and the conservative dipole force[4] due to the spatial variation of the ac Stark effect in the presence of an electric field gradient. Working through successive absorption–spontaneous-emission cycles, laser cooling based on the spontaneous force takes place at a rate bounded by the excited-state lifetime. The fundamental cooling limit (the recoil limit) corresponds to the homogeneously broadened linewidth of the cooling transition. However, substantially higher initial cooling rates can be obtained via stimulated processes.[5] By taking advantage of the fact that the dipole force does not saturate at high laser intensities, extremely large viscous forces can be obtained, but at the expense of a higher cooling limit.

These cooling methods have been used to deflect[6–8] and focus[9–11] atomic beams, and atoms have been laser cooled[12,13] and stopped.[14] Recoil-limited temperatures have been obtained in optical molasses,[15] making it possible to load atoms into a shallow, all-optical trap.[16] Cold, stopped atoms have also been loaded into magnetic traps.[17,18]

Combined with the well-established and highly developed field of electromagnetic trapping of ions, laser cooling has made possible the production of clouds of ultracold confined ions.[19] These are of importance in the development of an optical frequency standard[20] and a new interesting aspect is the production of ionic liquids and crystals,[21] both new forms of diluted matter.

As pointed out recently,[22,23] these condensed plasmas might also be observed in an ion beam "trapped" in a storage ring provided a strong and efficient cooling can be realized to overcome the beam-heating mechanisms.

In a storage ring the fast ions are confined transversely by magnetic focusing, while the longitudinal confinement is ensured by the closed-orbit motion. Besides the possibility of observing ionic crystals in this geometry, a stored ion beam with a very narrow velocity distribution and a well-defined trajectory is also of interest from a spectroscopic point of view. It will combine the ion trap's ultrahigh resolution with the wide range of experimental possibilities that the continuing development of fast-beam laser spectroscopy has offered. Studies of atomic and nuclear structures and molecular dynamics have thus been performed using a variety of spectroscopic techniques, known from thermal atom spectroscopy but taking advantage of the Doppler-tuning capability of the isotropic pure sample in an essentially collision-free environment. These unique possibilities have thus allowed a new generation of highly sensitive tests of the special theory of relativity.[24]

Two cooling schemes have so far been introduced successfully in storage rings, used in elementary particle physics: stochastic cooling[25] and electron cooling.[26] They have both been developed to increase the phase-space density of stored antiprotons before injection into large collider rings. Both these cooling methods work through the charge of the stored particles in contrast to laser cooling, which requires an excitation of the particle for the laser field to deliver its momentum. This limits the universality of laser cooling as compared to stochastic and electron cooling.

In stochastic cooling fluctuations in the average beam velocity is detected and an electronic correction is applied. The cooling rate scales inversely with the number of particles, making stochastic cooling a slow process for large samples.

In electron cooling, electrons with a narrow velocity distribution are merged collinearly with the stored ions. By matching the average velocities of these "cold" electrons and the ions, the Coulomb interaction will cause a reduction of the velocity spread of the "warm" ions. Electron cooling is a fast process and momentum spreads as low as $\Delta p_{\parallel}/p \sim 10^{-6}$ have been obtained in an electron-cooled proton beam.[27]

II. FAST-BEAM LASER COOLING

The mechanical effects of light on atoms is small in collinear fast-beam spectroscopy.[28] Only a few photons are scattered due to the high speed of the particles, limiting the interaction time. However, by combining a long straight beamline with an atom with a rapid spontaneous decay, enough photons may be scattered to study the onset of laser cooling of a fast beam. The closed transition $3s[\frac{3}{2}]_2^\circ$-$3p[\frac{5}{2}]_3$ in Ne I, with an upper-state lifetime of 19.4 ns, allows the scattering of ~ 100 photons in our experimental setup at a beam energy of 100 keV. On the average, each absorbed photon transfers its momentum $h\nu/c$ to the atom. This corresponds to a change of 6 meV in beam energy or 40 kHz in first-order Doppler shift. Thus, the total effect of the transfer of momentum from the laser beam to the atomic beam will be a displacement of the laser absorption on the order of a few MHz, i.e., less than the homogeneous linewidth of the cooling transition. Therefore, it is not necessary for instance to chirp the frequency of the cooling laser during the interaction period to compensate for the change in Doppler shift due to the photon recoils. If the homogeneous broadening is comparable to the inhomogeneous one, as in our case, the net result is an acceleration of all the particles. Actual cooling of the fast beam sample requires the light-pressure force to Doppler-shift the cooling transition at least corresponding to the homogeneous linewidth.

The fixed cooling frequency scheme has been analyzed theoretically for the case of a slow atomic beam.[29] More recently, the implementation of laser cooling in a fast, stored ion beam has been discussed.[30]

A. Experiment

The experimental setup is shown schematically in Fig. 1. Ne^+ ions were accelerated to 100 keV in an electrostatic accelerator and mass separated in a high-resolution magnet. At the entrance of a long, straight beamline the ions ($^{20}Ne^+$) were charge exchanged in a Na cell. A well-collimated dye-laser beam overlapped the fast atomic beam collinearly, providing a 4-meter-long interaction region for cooling. The laser-induced fluorescence from the beam was monitored with a spectrometer and a photomultiplier. In order to avoid losing the atoms to other fine-structure levels by optical pumping, the cooling laser was, as shown in Fig. 2, tuned to the closed transition from the metastable $3s[\frac{3}{2}]_2^\circ$ level to the $3p[\frac{5}{2}]_3$ level.

The change in the velocity distribution due to the interaction with the cooling laser was probed at the end of the interaction region by scanning a second dye laser across the $3s[\frac{3}{2}]_2^\circ$-$3p'[\frac{1}{2}]_1$ probe transition, which was

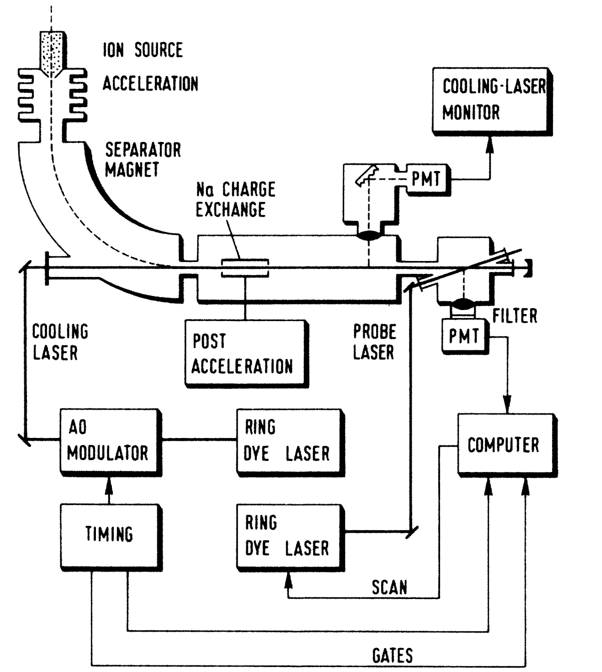

FIG. 1. In the experiment the ions are accelerated in an electrostatic accelerator and charge exchanged in Na vapor. Fluorescence from the beam is detected with photomultiplier tubes (PMT) both in the cooling section and in the probe region. The gating electronics and the computer control the timing and the frequency scan of the cw ring dye lasers, as well as the data acquisition.

monitored by observing the fluorescence on the $3p'[\frac{1}{2}]_1$-$3s'[\frac{1}{2}]_0^\circ$ branch, using a photomultiplier with a narrow-band interference filter. To avoid optical pumping to other fine-structure levels, the probe beam crossed the atomic beam at an angle of 5°, limiting the interaction region to approximately 2.5 cm, corresponding to an in-

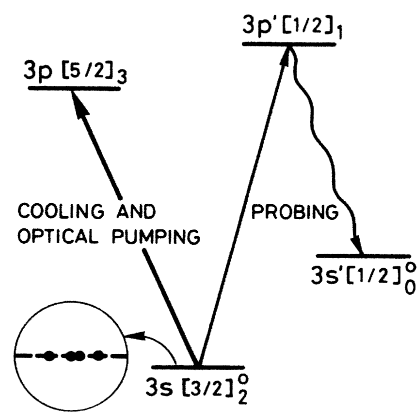

FIG. 2. The cooling laser was tuned in resonance with the closed $3s[\frac{3}{2}]_2^\circ$-$3p[\frac{5}{2}]_3$ transition. Optical pumping on this transition leaves 47.6% of the population in the $M=0$ and $M=\pm 1$ levels, respectively, and 4.8% in the $M=\pm 2$ levels. The velocity distribution was probed by scanning another dye laser across the $3s[\frac{3}{2}]_2^\circ$-$3p'[\frac{1}{2}]_1$ transition and observing the fluorescence from the decay to the $3s'[\frac{1}{2}]_0^\circ$ level.

teraction time of 25 nsec.

To further isolate the small changes in the Doppler profile due to the radiation pressure the cooling laser was chopped with an acousto-optic modulator, and the probe detection electronics was gated to detect cooled and noncooled atoms in two different spectra. This scheme also allowed us to rule out two systematic effects in the experiment: possible nonlinear laser interactions in the probe region and effects of the optical pumping between the magnetic sublevels in the cooling section. The former was avoided by recording the cooled velocity distribution as well as the noncooled reference while the cooling laser was off. The latter was eliminated by also optically pumping the reference atoms with a short cooling laser pulse. The repetition rate of this cooling and detection cycle was limited by the time of flight of the atoms through the apparatus (4 μsec).

The gate pulses and the cooling laser intensity, as detected with a photo diode, are shown in Fig. 3. After the cooling laser had been on for 5 μsec, the velocity distribution was recorded for 2 μsec in gate 1. After the "cooled" particles had passed the probe region, a 200-nsec pulse from the cooling laser optically pumped the new "warm" particles prior to opening of gate 2 for 2 μsec. Negligible momentum would be transferred in this short time interval, and a noncooled reference distribution was obtained.

Figure 4 shows the two Doppler profiles recorded in this way. The light-pressure force has clearly blue shifted (accelerated) the "cooled" velocity distribution (solid circles) relative to the reference distribution represented by the open circles. Due to a strong saturation of the cooling transition, the whole distribution, and not only the part closest to resonance, has been accelerated. The frequency shift is determined to be 4.0±0.4 MHz by fitting Gaussian line shapes to the two distributions. The similarity of the two distributions due to the rather uniform acceleration ensures an accurate determination of the actual shift despite a slight asymmetry towards lower velocities. The fitted lines are indicated in Fig. 4 by the dashed and the solid curves.

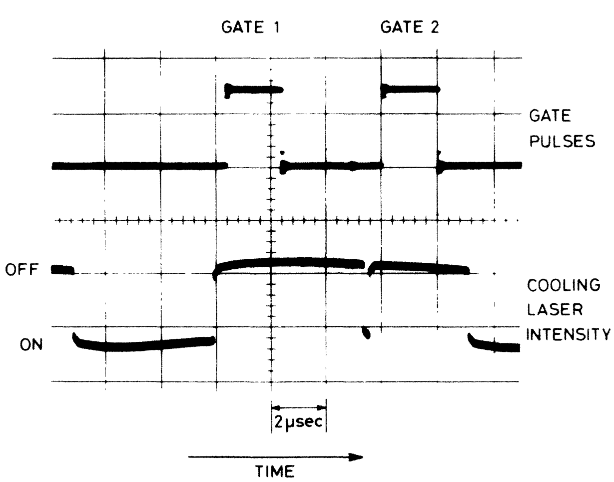

FIG. 3. Timing of the cooling laser and the detection gates. The upper trace shows the two gates while the lower one shows the intensity of the cooling laser.

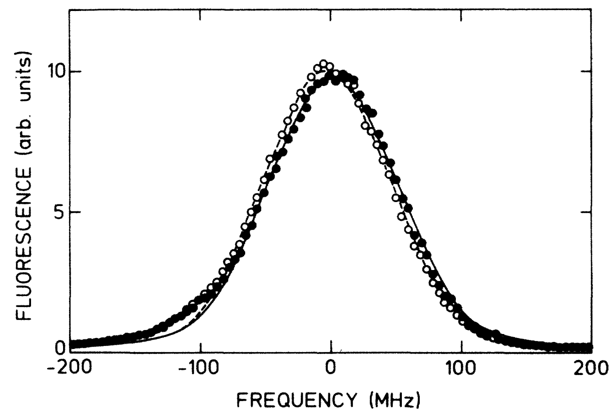

FIG. 4. Experimentally detected Doppler profile for "cooled" atoms (solid circles) and the noncooled reference (open circles). For clarity, points are averaged two by two. The dashed and the solid curves are Gaussian line shapes fitted to the two distributions, respectively.

The 4-MHz displacement of the Doppler profile corresponds to a velocity change of 2.4 m/s or a 0.5-eV increase in energy. Taking into account the interaction time of 4 μsec, we have achieved a laser-induced acceleration of 6×10^5 ms^{-2} or 120 keV/s. By tuning a counterpropagating laser field in resonance with the cooling transition the atoms were decelerated at the same rate.

B. Theoretical model

A simple theoretical model for the modification of the velocity distribution due to the photon recoil can be obtained if we neglect the heating caused by the random direction of motion of the spontaneously emitted photons. This approximation will be appropriate in the present experiment due to the short interaction time and therefore small number of scattered photons.

The equation of motion of a particle moving with longitudinal velocity v, which is accelerated by the light pressure of a copropagating laser beam, is[30]

$$\frac{dv}{dt}=a(v)=\frac{\Gamma\varkappa^2 v_r}{(\Omega_L-\omega-kv)^2+\gamma^2}\;, \tag{1}$$

where $v_r=\hbar k/M$ is the recoil velocity. $\varkappa$ is the Rabi frequency of the cooling transition, Γ the spontaneous decay rate of the upper level, and γ the power-broadened linewidth. Ω_L is the frequency of the cooling laser, while ω is the atomic transition frequency.

Given an initial distribution f_0 the phase-space distribution function f of particles in the beam direction can now be determined as a function of v and the interaction time τ. The particles that initially were in a velocity range δv_0 around v_0 will after the time τ occupy the range δv around v,

$$\delta v_0 f_0(v_0)=\delta v f(v,\tau)\;. \tag{2}$$

Thus:

$$f(v,\tau)=\frac{a(v_0(v,\tau))}{a(v)}f_0(v_0(v,\tau)) , \quad (3)$$

where $v_0(v,\tau)$, the velocity that in time τ will be accelerated to v, is obtained analytically by integrating Eq. (1).

The distribution function $f(v,\tau)$ is experimentally investigated by the second laser, crossing the fast beam at a small angle. The interaction time in the detection region is $\tau_{int}\approx 25$ nsec, thus transit time limiting the observed line shape. The fluorescence is proportional to

$$S(v)=\int_{-\infty}^{\infty} f(v',\tau)\mathcal{L}(v-v',\tau_{int})dv' , \quad (4)$$

where $\mathcal{L}$ is the transit-time-limited lineshape given by Stenholm.[31]

The numerically evaluated function $S(v)$ for the noncooled distribution is shown together with the experimental data in Fig. 5(a). For simplicity the initial longitudinal velocity profile is taken to be a 10-V-wide Gaussian distribution. Figure 5(b) shows the difference between the cooled and the noncooled Doppler profiles both for the experimental data and the numerically evaluated distributions. The only adjustable parameters are the amplitude of the signal and the cooling laser detuning (15 MHz).

This analysis has neglected the dipole force. It merely causes a slight focusing (defocusing) of the atoms for the cooling laser tuned below (above) resonance, respectively, amounting to a transverse displacement of less than 0.1 μm in the present experiment.[4]

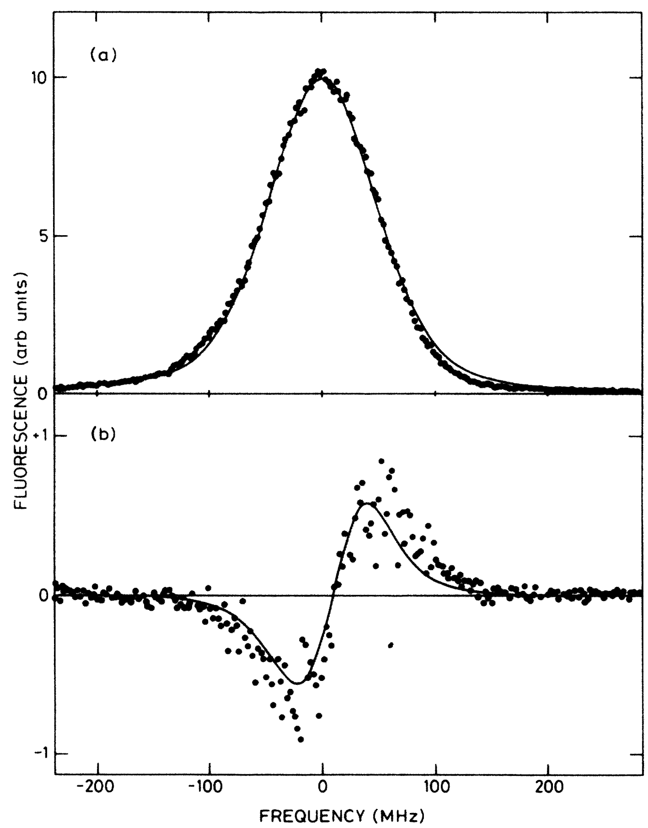

FIG. 5. (a) Experimentally detected (points) and numerically evaluated (solid curve) velocity distribution of noncooled atoms. (b) The difference between the cooled and noncooled velocity distributions. The cooling laser is detuned 15 MHz above exact resonance.

C. Optimization of laser cooling

In order to optimize the observable effect of the light-pressure force in this experiment, the cooling transition was strongly saturated and the laser tuned close to line center. Clearly this is not desirable in a real cooling experiment since cooling does not occur until the faster atoms are pushed away from resonance and the photon scattering rate is thereby reduced. Based on the good qualitative as well as quantitative agreement between the experiment and the theoretical model we can predict the evolution of the longitudinal velocity distribution for longer interaction times. Figure 6 shows that already after 100 μsec a substantial compression has taken place. However, the figure also indicates that after several tens of μsec the acceleration rate drops because the atoms are pushed too far out of resonance. Thus, in order to optimize the light-pressure cooling process not only a sufficiently long interaction time will be needed, but the atoms must also be kept close to resonance either by chirping the laser or by applying an external acceleration. This leads to the definition of the cooling time τ_c as the time required for the radiation pressure to accelerate the atoms corresponding to the width of the initial velocity distribution.

However, on a time scale comparable to the cooling time our simple theoretical model is no longer adequate. The phase-space density is instead obtained as the solution to a Fokker-Planck equation, where the random emission of spontaneous photons is accounted for by a diffusion term.[30]

Once the particles have been cooled enough to have a

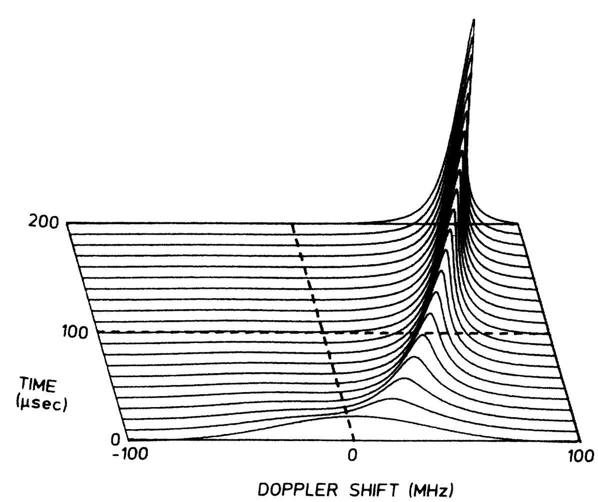

FIG. 6. The velocity profile of the beam for cooling times from 0 to 200 μsec as predicted by the theoretical model. The frequency of the saturating cooling laser is kept fixed at the center of the non-cooled Doppler profile, and no external acceleration is applied.

velocity distribution comparable to the power-broadened linewidth, the time scale of interest is the time τ_e to reach the diffusion limit, where the diffusive heating is exactly balanced by the light-pressure cooling. This equilibration time is also the time scale on which the system dissipates internal longitudinal energy (heat). It is typically on the order of the inverse of the recoil energy[30] (in cyclic frequency units), and is generally much shorter than any known heating process. Intrabeam scattering[32] (Coulomb scattering), for instance, mixes the transverse and longitudinal motions with a time constant of tens of milliseconds.

III. DISCUSSION

A possible candidate for fast-ion-beam laser cooling is $^{24}Mg^+$ which has two closed hyperfine-free transitions around 280 nm suitable for cooling. For optimum laser power and detuning we find $\tau_e = 3.0$ μsec and a final velocity distribution with a width of only about 1.0 m/s. This corresponds to a temperature of 0.7 mK or an energy spread at 100 keV of 220 meV. For an initially 10-eV-wide energy distribution we find $\tau_c = 5.7$ μsec.

Long-lived cooling transitions offer an even lower cooling limit. The closed $0_{13/2} - 18463_{15/2}$ transition in Er^+, for instance, has an upper-state lifetime of about 1 μsec. This leads to a final temperature in the diffusion limit of less than 3 μK or a velocity spread of about 2.5 cm/s. This low cooling limit will, of course, only be obtained at the expense of a considerably lower cooling rate. We thus find $\tau_e \approx 80$ μsec and $\tau_c \approx 1.4$ ms, again for a 10-eV-wide initial distribution.

An interesting aspect for future fast-beam laser cooling is the utilization of the recently proposed "stimulated molasses."[5] In connection with fast-beam cooling it requires the establishing of a strong-standing laser wave in the atomic beam rest frame. This is easily done by tuning two collinear and counterpropagating laser beams into resonance with the fast beam.

IV. CONCLUSION

Laser cooling of fast stored ions represents a subfield of atomic and laser physics. The aim is to produce highly specialized spectroscopic sources and utilize them in a variety of experiments ranging from studies of crystalline beams to tests of fundamental principles of physics, using "monovelocity" probes. These applications do not require a universal cooling scheme, but rather that a small number of elements can be cooled effectively.

This development awaits the completion of heavy-ion storage rings presently under construction in several laboratories. The prospects for successful laser cooling can, however, be evaluated using existing linear accelerators as has been done in the present experiment.

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