First Laser Cooling of Relativistic Ions in a Storage Ring

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The first successful laser cooling of ions at relativistic energies was observed at the Heidelberg TSR storage ring. A 7 Li⁺-ion beam of 13.3 MeV was overlapped with resonant copropagating and counterpropagating laser beams. The metastable ions were cooled from 260 K to a longitudinal temperature of below 3 K and decelerated by several keV. The longitudinal velocity distribution was determined by a fluorescence method. After laser cooling a strongly enhanced narrow peak appeared in the Schottky noise spectrum in addition to the uncooled ion distribution.

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The possibility of controlling the motion of charged and neutral particles by light has caused a rapid development of cooling and trapping techniques. In the field of laser cooling, all previous approaches, from the initial ones¹ up to the latest ones² have concentrated on velocity control close to or at rest in the laboratory frame. The observed temperatures either approach the "Doppler limit" for neutral atoms in optical molasses³ or even reach below.⁴ Cooled ions in a trap show clustering and crystallization.⁵ Light pressure influence on fast atomic beams has been observed in collinear geometry.⁶ In storage rings, so far, stochastic cooling^{$\overline{7}$} and electron cooling⁸ have been applied. These methods are limited to final ion temperatures of a few kelvin. In this paper we present the first experimental results on laser cooling of a relativistic ion beam in a storage ring leading to similar final temperatures. The theoretical limit for "Doppler cooling" depends on the lifetime of the cooling transition and is as low as 89 μ K in the case of ⁷Li⁺. Collinear geometry exhibits the maximum dynamical compression and a small momentum change Δp leads to a large energy change $\Delta E = \Delta p p / M$. Since the interaction time of the stored ions with the laser is long, their energy can be changed by several keV, even though the photon momentum is only 10^{-9} of the ion momentum.

As the first treatment of its kind the heavy-ion storage ring TSR in Heidelberg came into operation in 1988.⁹ It is connected to a tandem accelerator followed by a rf postaccelerator. Ions are injected into the storage ring by a multiturn injection method. The magnetic rigidity of the ring extends up to $B\rho = 1.7$ T m and its circumference is $l_{\text{ring}} = 55.4$ m. The ring has a fourfold symmetry as shown in Fig. 1(a). Laser cooling is performed in the experimental section opposite to the beam-injection area.

⁷Li⁺ ions are produced in the accelerator terminal by stripping of LiH⁻ ions in a gas target. A coasting beam of 10⁷-10⁸ particles is stored in the ring at an energy of 13.3 MeV corresponding to a velocity of $\beta = v/c = 0.064$. The storage lifetime τ is limited by collisions with residual gas molecules leading to a particle loss due to ionization or scattering. A mean value of $\tau \approx 2$ s is observed



FIG. 1. (a) Experimental setup and (b) partial ${}^{7}\text{Li}^{+}$ level scheme. Two counterpropagating laser beams (single-mode Ar⁺-ion laser and ring dye laser) are merged with the ${}^{7}\text{Li}^{+}$ -ion beam stored in the TSR at $\beta = 0.064$. The fluorescence signal is detected by a photomultiplier at 90°. The dynamical properties of the stored beam are monitored by a Schottky noise pickup in (a). A schematic ${}^{7}\text{Li}^{+}$ level scheme is shown in (b). The closed two-level system ${}^{3}S_{1}(F = \frac{5}{2}) \rightarrow {}^{3}P_{2}(F = \frac{7}{2})$ is used for cooling as well as for fluorescence detection.

in a vacuum of $\approx 10^{-8}$ Pa. Furthermore, ionization of residual gas molecules leads to a continuous energy loss of ≈ 100 eV/s at these conditions.

In the experimental section laser cooling is applied to the metastable $1s 2s^{3}S_{1}$ ions, present from the stripping reaction as a fraction of about 10% to 30% of the total beam. The ion beam is merged with a copropagating beam from a single-mode Ar⁺-ion laser and a counterpropagating single-mode dye-laser beam. The geometrical overlap of the ion beam with the two laser beams is achieved by the insertion of two diaphragms with 18-mm apertures. With careful adjustment, 25% of the total ion beam can pass through this collimator. The two laser beams of 10-mm diam are also centered through the apertures. The Doppler shift is determined by v_{\pm} $= v_0 \gamma (1 \pm \beta_z)$, where v_0 and v_{\pm} are the absorption frequencies in the center of mass and laboratory frame, β_z is the longitudinal ion velocity, and γ denotes the usual relativistic factor. The ion velocity is chosen to excite the ${}^{3}S_{1}(F = \frac{5}{2}) \rightarrow {}^{3}P_{2}(F = \frac{7}{2})$ transition in the ${}^{7}Li^{+}$ spectrum [Fig. 1(b)] with the 514.5-nm line of the Ar⁺-ion laser. The counterpropagating dye laser is set to 584.8 nm in order to scan the same transition, which forms a closed two-level system. The lower ${}^{3}S_{1}$ state has a radiative lifetime of 50 s, long compared to the storage lifetime of 2 s. The ${}^{3}P_{2}$ state has a mean radiative lifetime of $\tau_{sp} = 43$ ns with a less than 10^{-5} branching ratio to the ${}^{1}S_{0}$ ground state. Regarding the ${}^{3}S_{1}(F)$ $= \frac{5}{2} \longrightarrow {}^{3}P_{2}(F) = \frac{7}{2}$ transition as a two-level system, one obtains a saturation intensity $I_S = 8.8 \text{ mW/cm}^2$. The corresponding light-pressure acceleration a_{lp} on resonance is given by

$$a_{\rm lp} = \frac{\Gamma}{2} \frac{h}{\lambda M} \frac{S}{1+S} = 1.15 \times 10^6 \frac{S}{1+S} \frac{m}{s^2} \,, \tag{1}$$

where $\Gamma = 1/\tau_{sp}$, *h* is Planck's constant, *M* is the mass of the ⁷Li⁺ ion, λ is the wavelength, $S = I/I_S$ is the saturation parameter, and *I* is the laser intensity. However, the effective mean acceleration in the ring is reduced by the factor $I_{ex}/I_{ring} = 0.09$, since the ions are accelerated only in the laser-cooling section of length I_{ex} . A further reduction is caused by betatron oscillations of the ions. Because of these transverse oscillations, their Dopplershifted resonance frequencies and their geometrical positions in the interaction zone change randomly and thus they exhibit a reduced light interaction.

The laser cooling of the ion beam is performed in the following way. The frequency of the copropagating single-mode Ar^+ -ion laser beam is kept constant and is set in resonance with ions at the low-energy side of the velocity profile of the ion beam. The frequency of the counterpropagating tunable dye-laser beam is correspondingly set to the high-energy side. When sweeping the latter frequency towards higher values, ions getting into resonance are decelerated and accumulated in a narrow peak.

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The initial and compressed velocity distributions are measured by fluorescence detection at 90° in the experimental section. This signal is shown in Fig. 2 as a function of the laser frequency detuning Δv (from right to left) and the corresponding energy change ΔE of resonant ions. The original distribution seen in Fig. 2, trace a, has been obtained by reverse scanning (decreasing laser frequency) thus avoiding the cooling compression. The absolute energy of the ion beam may be obtained via the relativistic Doppler formula. Normal scanning leads to Doppler cooling and velocity compression as shown in traces b and c of Fig. 2. The position of the peak in the fluorescence spectrum is determined by the Ar⁺-ion laser frequency. While scanning over the initial distribution, ions in the metastable ${}^{3}S_{1}(F = \frac{5}{2})$ state are collected. Continuing the scan results in a further acceleration and in an almost constant fluorescence intensity until finally the cooled ions are brought to bright fluorescence in the combined fields of both lasers. The signal in Fig. 2, trace c, has been obtained at high laser intensities $(S \ge 50)$ and a scanning speed of 4 GHz/s. This cooling speed (close to the maximum) is below the speed in atomic-beam-cooling experiments and estimates from Eq. (1) by a factor of $\approx 10^3$ because of the reduction of the mean light-pressure acceleration \bar{a}_{1p} in the storage ring.



FIG. 2. Laser fluorescence signals. The fluorescence is shown as a function of the dye-laser frequency detuning and the equivalent energy change. Trace a shows the original energy distribution of the ion beam, obtained by a dye-laser scan from left to right. Trace b is obtained under identical conditions in the TSR. Scanning the dye-laser frequency from right to left (higher to lower ion energies) results in a dramatic compression of the velocity distribution. Simultaneous irradiation with a single-mode Ar⁺-ion laser at fixed frequency leads to a strong resonance, when the cooled ions interact with both lasers. In the high-resolution measurement shown as trace c, the frequency position of the Ar⁺-ion laser is different. The dashed lines represent the background signal without ions in the TSR.

The original distribution shows a frequency shift of 7.5 GHz, which corresponds to an energy change in the laboratory frame of more than 5.5 keV. The original signal width of 1.9 GHz due to kinematic compression is equivalent to a temperature of the longitudinal motion of T = 260 K. By laser cooling it is compressed by almost a factor of 10, but due to the high intensity of both lasers, the width of the compressed peak only allows one to quote an upper limit $T \le 2.9$ K according to

$$T = \frac{Mc^2}{4k_B} \beta^2 \left(\frac{\delta p}{p}\right)_{\text{FWHM}}^2 \le \frac{Mc^2}{4k_B} \left(\frac{\delta v}{v_-}\right)_{\text{FWHM}}^2, \quad (2)$$

where δv is the full width of the fluorescence peak, $v_$ the redshifted resonance frequency of the cooled distribution, and k_B is Boltzmann's constant. Observing the fluorescence with a weak probe beam, independently of the cooling process, would result in the determination of a presumably lower temperature.

An independent measurement of the beam properties of all ions (${}^{1}S_{0}$ ground state and all ${}^{3}S_{1}$ metastable) is achieved with the electronic Schottky noise signal from the longitudinal pickup shown in Fig. 1(a). The noise frequency distribution is obtained by a rf spectrum analyzer. The measured signal is proportional to the amplitude $|A_{n}(f)|$ of the beam density fluctuations¹⁰ at the *n*th harmonic of the revolution frequency. For hot, low-density dc beams the measured power spectrum $|A_{n}(f)|^{2}$ is proportional to the revolution frequency distribution of the particles. The integral $\int |A_{n}(f)|^{2} df$ = N, with N the number of particles in the ring. For cooled beams additional peaks may occur in the power spectrum at phase velocities, where collective fluctuations can propagate as waves.¹⁰

In the present experiment the noise spectra at n=18(Fig. 3) were analyzed with a delay of 2.5 s after the ion injection. Figure 3, trace a, shows the uncooled distribution without laser interaction. In Fig. 3, trace b, the delay time is used for a laser-cooling sweep, which stops \approx 500 MHz ahead of the maximum fluorescence signal of the compressed peak. The copropagating and counterpropagating lasers were then held at fixed frequencies in order to confine the ions in velocity space by the combined friction forces of both lasers. The broad distribution due to the uncooled ions yields similar width and particle number to those for trace a of Fig. 3. Besides a narrow intense peak at frequency f_{cool} occurs from the laser-cooled ions almost 250 MHz shifted from the maximum of a fluorescence spectrum similar to that shown in Fig. 2, trace c. The signal width is limited by the bandwidth of the spectrum analyzer. A high-resolution signal taken at n=36 and adapted to the frequency scale of the previous spectra is shown in Fig. 3, trace c. At an effective bandwidth of 14 Hz a signal width of 30 Hz is observed, which corresponds to a relative frequency width of 2.5×10^{-6} .

If one assumes that the narrow peak is caused by incoherent single-particle fluctuations, the ratio of cooled to uncooled ions is $N_{\text{cool}}/N_{\text{hot}} = 30$. This value is about 100 times larger than expected from the metastable beam fraction (0.1-0.3). Any collisional cooling of ground-state ions by the laser-cooled metastable ions during scanning is ruled out by low exchange rates. These are deduced from the long lifetime (100 ms) observed for Bennett holes,¹¹ produced by optical pumping. Therefore only a strong signal enhancement produced by a collective beam response can explain the observed spectrum. Also, the integral power spectrum is by a factor of 4 larger than the corresponding integral of the uncooled spectrum. This is in contrast to the situation after electron cooling, where the distortion of the shape of the Schottky spectrum due to collective effects is always accompanied by strong noise suppression.

This signal enhancement for a narrow phase-velocity region is observed for a large span of harmonic numbers $(6 \le n \le 36)$, different laser intensities $(1 \le S \le 100)$, and different energy shifts between cooled and uncooled distributions. The phenomenon is likely due to some instability, such as a two-stream instability¹² between cooled and uncooled ions, which at higher wave intensities is stabilized by the strong friction forces of the lasers. It is challenging to investigate this one-dimensional problem with known longitudinal velocity distributions and to compare theoretical and experimental Schottky noise spectra. Obviously, the width of the nar-



FIG. 3. Schottky noise signals. The signals are taken with a delay of 2.5 s after the injection of the ions into the TSR, the time required for the laser-cooling sweep. They are shown in a linear scale. Traces a and b are taken with identical sensitivity but different resolution bandwidths of 100 and 50 Hz, respectively, at the 18th harmonic of the revolution frequency at $f_{\rm cool}$ =6.210340 MHz. Without laser cooling sweep yields spectrum b. Finally trace c, plotted with the same frequency scale, is taken at the 36th harmonic resulting in a higher-frequency resolution.

row Schottky noise peak is not a direct measure of the ion-beam temperature.

We have demonstrated for the first time laser cooling of a relativistic ion beam in a storage ring and have obtained longitudinal temperatures at least as low as a few kelvin. This is still far above the theoretical limit of laser cooling, but we are close to the limit of present-day electron cooling. The absolute velocity of the ion sample can be determined with high accuracy, which is of considerable interest for precision experiments in atomic and nuclear physics. Schottky noise signals of the cooled-ion beam exhibit a strong enhancement of the total noise power, which is currently not well understood.

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