Evaporative Cooling and Coherent Axial Oscillations of Highly Charged Ions in a Penning Trap

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Externally, in an electron beam ion trap, generated Ar^{16+} ions were retrapped in a Penning trap and evaporatively cooled in their axial motion. The cooling was observed by a novel extraction technique based on the excitation of a coherent axial oscillation which yields short ion bunches of well-defined energies. The initial temperature of the ion cloud was decreased by a factor of more than 140 within 1 s, while the phase-space density of the coldest extracted ion pulses was increased by a factor of up to about 9.

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Highly charged ions (HCIs) play an important role in many fields of physics, such as nuclear, astro, plasma, atomic, and surface physics as well as in metrology. In order to achieve ultimate sensitivity and accuracy in experiments with HCIs, cooling is a key prerequisite. Using HCIs one can test quantum electrodynamics (QED) in the strong field regime [1], achieve the highest precision in mass measurements [2], or accurately determine the nuclear-size effect [3]. Recently it was proposed [4] to use laser spectroscopy on cold HCIs to investigate a time variation of the fine structure constant at a much higher sensitivity than with atoms or lowly charged ions. The high sensitivity would be due to the mutually enhancing contributions from three factors: high nuclear charge Z, high ionization degree, and a significant difference in the configuration composition of the states involved.

For preparation, observation, and excitation of HCIs, Penning traps are unique tools [5]. However, most standard ion cooling techniques for Penning traps were developed for singly charged ions and are applicable to HCIs only to a limited extent. Resistive cooling has been demonstrated for HCIs [6], but generally suffers from long cooling times. Buffer gas cooling with a neutral coolant is very efficient for singly charged ions [7] but for HCIs, charge exchange poses a severe limitation. With laser cooling, temperatures much below 1 K have been obtained [8], but this is only applicable to singly charged ions and only to a few species. Laser cooling cannot be applied directly to HCIs due to a lack of suitable transitions. A complex alternative is sympathetic cooling of HCIs by Coulomb collisions with laser-cooled singly charged ions, e.g., Be^+ [9]. Electron and positron cooling of HCIs in Penning traps was proposed [10]. It has not been demonstrated so far, but calculations were performed [11,12]. Positron cooling requires a complex source and electron cooling might be applicable only at energies of the HCIs above $100q$ eV, where q is the charge state of the ion, as at lower energies the recombination rate could become too high.

Evaporative cooling is a technique which has proven extremely efficient to reach very low temperatures of magnetically trapped neutral atoms, achieving Bose-Einstein condensation [13]. Recently, evaporative cooling of antiprotons was reported [14], and for HCIs it was demonstrated in electron beam ion traps (EBITs) with the use of simultaneously injected light-mass ions [15]. Evaporative cooling requires the selective removal of particles with the highest energy. The remaining particles rethermalize by elastic collisions, reaching a lower temperature. In the case of charged particles, evaporation and rethermalization are driven by Coulomb interaction and its long-range nature results in much higher elastic collision rates compared to neutrals. Evaporative cooling should be especially favorable for HCIs as the collision strength is $\propto q^2$. While the technique is simple and applicable over a wide range of temperatures, it requires high initial ion numbers as the cooling is achieved through a substantial loss of particles.

Here we report the first observation of evaporative cooling of HCIs in a Penning trap. In such a trap the ions are effectively bound radially, but can be evaporated via the electrostatic axial trapping potential. In order to monitor the cooling process we have developed an extraction technique which is based on coherent axial oscillations of the confined ion cloud. In contrast to previous studies of oscillations of ion clouds in traps (see, e.g., [16–18]), we observe the oscillation by extracting a fraction of the trapped ions once each oscillation period, which yields a time-of-arrival (TOA) spectrum that directly images the oscillation.

The results reported in this Letter have been achieved with the cooling trap of the new high-precision mass spectrometer SMILETRAP II at AlbaNova, Stockholm University. Highly charged ^{40}Ar ions were produced by electron impact ionization in the Stockholm EBIT (S-EBIT) [19], then extracted with $4.5q$ keV energy, and transported via a 90° bending magnet for mass-over-charge (m/q) separation to the beam line of the

SMILETRAP II experiment. Ar^{16+} was selected because of the high intensity. Typically, the m/q-separated ion pulse was about 100 ns long and contained about 10^5 ions. Prior to injection into the cooling trap, the Ar^{16+} ions were decelerated to a few eV per q.

The cooling trap [Fig. 1] is an open end-cap Penning trap, composed of seven cylindrical electrodes (center ring electrode, two end-caps, and four compensation electrodes) in a 1.1 T conventional electromagnet. In the trap the ions are radially confined by the magnetic field. In the axial direction the confinement is achieved by a potential well, created by applying positive voltages up to 10 V to the lower end cap (LEC) and the upper end cap (UEC) relative to the center electrode which floats at the same potential as the S-EBIT trap. Fractions of the end-cap voltages are applied to the corresponding compensation electrodes in order to reach a close-to-harmonic axial potential in the center of the trap. An ion in a harmonic potential oscillates with frequency $\omega_z = (qeU/md^2)^{1/2}$, where U is the axial trap potential, e the elementary charge, and d the characteristic trap dimension. For the present trap $d = 15$ mm.

When ions entered the 4 mm trap opening, LEC was set to 0 V, while 10 V were applied to UEC. Some of the HCIs were turned around by the UEC potential and then trapped by biasing also LEC with 10 V. Because of the short and intense ion pulse from S-EBIT, typically about 10^4 Ar^{16+} ions were trapped. A lower limit for the initial ion density n_0 can be estimated from the maximum volume the ion cloud can occupy in the trap, given by the geometry, which yields $n_0 = 2 \times 10^{10}$ m^{-3}. However, a higher peak density is expected as the ion cloud is not uniform over the trap volume. The Brillouin density, which gives the maximum number of ions that can be confined in the trap, is $n_{\max} = 1 \times 10^{12}$ m^{-3}.

The Ar^{16+} ions were trapped for a preset storage time t_s, ranging from a few microseconds to a few seconds, during which evaporative cooling took place. The pressure in the trap region was about 1×10^{-9} mbar which made charge exchange and inelastic collisions with the background gas negligible for these storage times.

After storage, the ions were extracted from the trap as follows [Fig. 2(a)]: First, a coherent axial oscillation was excited by switching the UEC potential, referred to as "trapping potential" U_t, from 10 to 1 V in about 1 μs. The excitation occurred through suitable Fourier components of the time-dependent electric field by the fast potential switching. The LEC potential was kept constant at 10 V during excitation and extraction [see inset in Fig. 1]. Subsequently, the ions were extracted by slowly ramping the trapping potential linearly from 1 to 0 V in 200 μs. Reaching zero, U_t was reset to 10 V to prepare for trapping a new ion pulse.

While ramping, only those ions could leave the trap which had an axial energy $E > U_t q$. In addition, as a result of the excitation, these ions could leave the trap only once per oscillation, when the ion cloud reached UEC. Thus, the hottest ions were gradually extracted from the trap in bunches. After leaving the trap, the ions were smoothly accelerated to ground potential, and finally detected with a channeltron about 90 cm away from the trap. We recorded the ions' TOA in a multichannel scaler which started with the excitation pulse applied to UEC.

A typical TOA spectrum, obtained by accumulation of 200 cycles (injection, storage, excitation, extraction), is

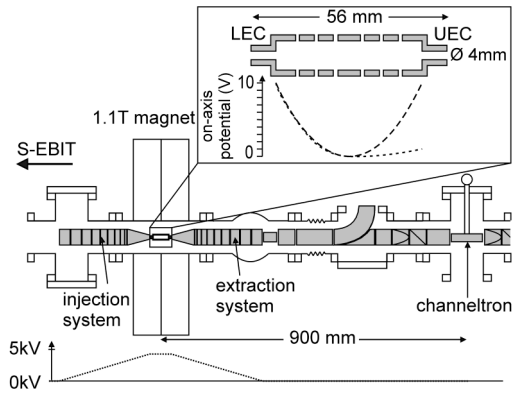

FIG. 1. Schematics of the cooling trap. The on-axis potential well during storage (dashed line) and after excitation (dotted line) is shown in the inset.

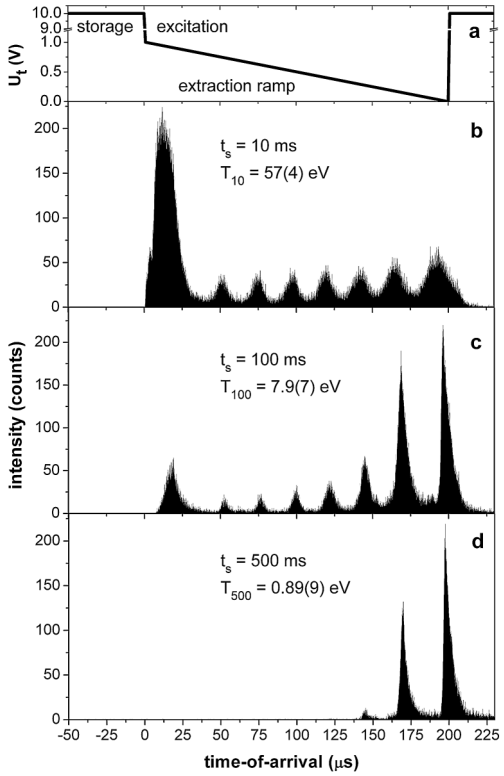

FIG. 2. (a) Switching of the trapping potential U_t and (b)–(d) TOA spectra of Ar^{16+} ions for different storage times t_s.

shown in Fig. 2(b). Each extracted ion bunch appears as a well-separated peak in the spectrum. The first strong peak at around 15 μs contains ions which left the trap with the excitation pulse. These ions had axial energies 1–10q eV. This peak is followed by a sequence of peaks as the trapping potential was slowly and linearly ramped down from 1 to 0 V. The colder the ions, the later they appear in the TOA spectrum. Since the extraction is fast compared to the time scale of the cooling process of milliseconds, the relative intensity of the peaks depicts the momentary energy distribution of the ions after the corresponding storage time.

From the time difference between subsequent peaks the axial oscillation frequency ω_z' of the ion cloud can be derived. In Fig. 3 ω_z' is shown as a function of the average trapping potential the ions experienced during the corresponding oscillation. In the case of a center-of-charge oscillation, which is the lowest oscillation mode of a group of ions, ω_z' should be identical to the single-ion frequency. For the part of the extraction ramp where $U_t < 0.35$ V, which is towards the end of the ramp, the observed frequency trend follows closely the trend expected for a single ion in a harmonic trap. Further experiments with other ions have shown that the trend in the first part of the ramp ($U_t > 0.35$ V) is due to a transient motion. Waiting for a time t_w between excitation and extraction, the initial frequency increases with increasing waiting time, and the frequency trend approaches the square-root behavior. This is shown for H_2^+ in the inset of Fig. 3.

The observed frequencies are well below the frequencies expected for higher oscillation modes. Plasma behavior is not expected as the Debye length is large (few 10 mm) compared to the dimensions of the ion cloud, and no dependence of the frequency on the ion density was observed. Finally, it should be noted that the anharmonicity of the trap potential due to asymmetry during extraction, and space-charge effects result in a shift of the axial frequency compared to ω_z.

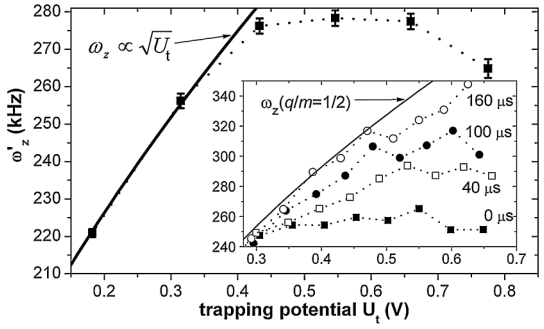

FIG. 3. Oscillation frequencies of the Ar^{16+} ion cloud. The trapping potential is decreased in time from right to left. Inset: Frequency trend of H_2^+ ($q/m = 1/2$) ions for different waiting times t_w indicated on the right side. Both axes have the same units as those of the main figure.

The evaporative cooling of the trapped Ar^{16+} cloud can be seen from Figs. 2(b)–2(d) where TOA spectra for 10, 100, and 500 ms storage times are shown. The cooling results in an intensity shift towards later TOA with increasing storage time. Evaporation of the hottest ions and an increase in the number of cold ions can be clearly seen as an intensity decrease of the first peak and increase of the last peaks, respectively. In addition, a narrowing of the last TOA peaks can be observed.

As an axial energy can be assigned to each peak, the axial temperature T of the ion cloud can be determined. Assuming that the ions are Boltzmann distributed, the axial energy E follows the chi-square distribution $\chi^2(x, 1)$ with $x = 2E/kT$, where k is the Boltzmann constant. By fitting the cumulative distribution function of the latter, i.e., the Gauss error function $\mathrm{erf}[(x/2)^{1/2}]$, to the cumulated peak intensities, we obtain T. A plot of the axial temperature as a function of storage time [Fig. 4] reveals the time evolution of the cooling process. For the shortest storage time of 5 ms the temperature $T_5 = 115(11)$ eV, while for the longest storage time $T_{1000} = 0.79(1)$ eV. This constitutes a temperature decrease by a factor of more than 140 in about 1 s. A fit with the exponential evaporation function $T(t) = T_0 \exp(t_s/\tau) + \mathrm{const}$ yields a cooling time constant of $\tau = 41(8)$ ms.

The characteristic time constant for relaxation to thermal equilibrium for charged particles is the self-collision time defined by Spitzer [20]. For a one-component plasma the collision relaxation time t_c in seconds is given by $t_c = 11.4A^{1/2}T^{3/2}/(nq^4 \ln\Lambda)$, where A is the atomic mass number of the ions, T the temperature in kelvin, n the ion density in cm^{-3}, and $\ln\Lambda$ the Coulomb logarithm which takes a numerical value between 10 and 30 for fully ionized gases. For $\ln\Lambda = 30$, $T = T_5$ and $n = n_0$ we find $t_c = 32$ ms as an upper limit for the relaxation time which is in good agreement with the observed time constant of the cooling process.

The Spitzer formula also illustrates the advantage of HCIs over singly charged ions for evaporative cooling: as $t_c \propto A^{1/2}q^{-4}$, the collision relaxation time for Ar^+, assuming same temperature and density as the Ar^{16+} cloud,

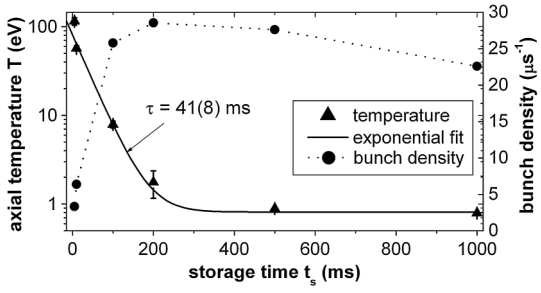

FIG. 4. Axial temperature T of the trapped Ar^{16+} ion cloud with exponential fit, and bunch density of the last TOA peak as a function of storage time.

would be higher by more than a factor of 10^5. Furthermore, the dependence of t_c on the mass is only weak, making evaporative cooling favorable for highly charged high-Z ions. Nevertheless, if high densities and long storage times without significant exchange losses can be realized, it can also provide a powerful technique for lowly charged ions.

The presented extraction technique also allows observing the change in phase-space density of the extracted ions. Because of the coherent oscillation all ions that can overcome the potential barrier leave the trap almost simultaneously. Thus, their time-of-flight spread at the detector is a good measure of their axial momentum spread. We can define the bunch density as the total number N_p of ions in a TOA peak divided by the peak's full-width-at-half-maximum. The observed narrowing of the peaks corresponds to an increase in the bunch density.

In Fig. 4 the bunch density of the last peak is shown as function of the storage time. While N_p increases by only a factor of 2.4 from 5 to 200 ms storage time, the corresponding bunch density increases by a factor of 8.6 to 27 ions/μs. For $t_s > 200$ ms the density decreases again. However, it has to be noted that the coldest ions ($E < 0.1q$ eV) are not extracted with the chosen scheme, restoring $U_t = 10$ V at the end of the extraction ramp. The bunch density of the coldest ions might still increase for $t_s > 200$ ms.

Despite the strong decrease of the axial temperature and the increase of the bunch density, the cooling of the Ar^{16+} ions was still quite inefficient in our experiment, reflected by a loss of about 85% of the ions after 1 s. The speed of evaporation depends on the ratio of the trapping potential to the ions' thermal energy, given by the dimensionless truncation parameter $\eta = U/\mathrm{kT}$ [21]. For small η a large fraction of the ions can overcome the potential barrier which results in a fast evaporation but inefficient cooling as the temperature reduction per escaping ion is small. In contrast, a high η means efficient cooling as each escaping ion removes much energy, but the cooling process is slow as fast particles are rarely produced. With U constant during the evaporation process, η varied between about 1 and 200. Especially for $t_s < 20$ ms, the cooling was inefficient. Controlling η by adjusting U during the storage time, it will be possible to optimize the cooling process in terms of cooling time and efficiency. In order to further reduce the ion temperature, a higher initial number of ions and ion density are necessary. This should be possible by optimizing the injection.

When reaching temperatures below about 500 meV, heating due to the excitation of the coherent oscillation will limit the observable temperature. In order to determine temperatures below that level other detection methods, such as the leaky mode [22], have to be used. The final limitation of the present setup is the room temperature of the trap. However, the evaporative cooling technique is capable of reaching lower temperatures [13,14]. For such low temperatures a cryogenic trap, ultrastable power supplies, and precooling using, e.g., resistive cooling is required.

In conclusion, we present clear evidence for evaporative cooling of HCIs in a Penning trap. This technique is particularly of advantage for high-Z HCIs. We also developed a novel extraction technique which allows us to monitor the cooling process and to extract HCI bunches of high density and low momentum spread. The presented results open the door for new experiments on preparation and manipulation of cold HCIs.

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