Observation of Ordered Structures of Laser-Cooled Ions in a Quadrupole Storage Ring

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We observed phase transitions and ordered structures of laser-cooled Mg^+ ions stored in a radio-frequency quadrupole storage ring with a diameter of 115 mm. The ions are confined two dimensionally by a quadrupole field and can move freely along the ring circumference. By cooling the ions with tangentially intersecting laser beams, we observed a linear string of ions aligned along the center line of the quadrupole field. For higher ion densities the linear structure changes into a helical structure. The dynamics of the stored ions was studied.

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Laser cooling of ions stored in Paul traps has made it possible to investigate single ions in an unperturbed environment [1] and study phenomena of the radiation-matter interaction [2,3]. It has also been demonstrated that a single ion can be cooled to its vibrational ground state in the trap [4] and can thus be used as a basis for optical frequency standards and precision spectroscopy [5]. Furthermore, it is possible to investigate phase transitions from a disordered to an ordered state with a few ions [6] and to study spectroscopically the oscillation spectrum of these ion clusters [7,8]. In the disordered phase the ions show nonlinear dynamics [8,9]. A phase-space reduction for ions in high-energy storage rings may also lead to ordered structures [10]. Laser cooling of high-energy Li^+ ions has already been demonstrated [11] and numerical simulations have been performed to reveal the expected ion configurations [12]. These ion structures are much easier to realize in low-energy storage rings such as radio-frequency (RF) quadrupole rings or race-track traps [13]. Quadrupole ring traps offer an important advantage over Paul traps with respect to micromotion of the stored ions. The amplitude of the micromotion is proportional to the distance from the saddle point of the field. Thus, in a Paul trap the micromotion is zero only for a single cooled ion at the trap center. This is different in a quadrupole storage ring. The field leads to a harmonic pseudopotential in the plane transversal to the electrodes, whereas the position of the ions along the field axis is determined by their Coulomb repulsion. Ions aligned along the center line of the quadrupole ring show no micromotion and can thus be laser cooled to very low temperatures. In this Letter we describe experiments where phase transitions and ordered structures of ions were investigated in a storage ring for the first time.

Figure 1 shows the quadrupole storage ring. The setup is contained in a UHV chamber with a pressure below 10^{-8} Pa. The storage ring consists of pairs of outer and inner rings held together by ceramic supports. The electrodes define a toroidal trap region with a diameter of $2R = 115$ mm; the distance between opposite electrodes is $2r_0 = 5$ mm. They are made of stainless steel and are gold coated. The part of the electrodes facing the trap region has a circular cross section with a radius of 2.9 mm. There is a gap of 2 mm between the electrodes, affording space for atomic, electron, and laser beams to enter the trap region. The ions are produced by electron bombardment of the atomic beam. The trapping is accomplished by applying an RF voltage of 7.2 MHz and an amplitude of a few hundred volts, resulting in a potential depth ψ_0 of several eV in the plane transversal to the electrodes.

The experiments described in this paper were performed with magnesium ions; the first results have been described in [14]. An order-of-magnitude estimate for the total number N of stored ions is obtained by switching off the trapping voltage and accelerating part of the ions towards a secondary electron multiplier. The laser frequency is tuned to the $3^2S_{1/2}$-$3^2P_{3/2}$ (280 nm) resonance transition of $^{24}Mg^+$ which has a natural linewidth of 43 MHz. Using a frequency-doubled ring dye laser, we obtain 0.5 mW of UV light [3]. The laser beam is focused tangentially into the storage volume and has a beam waist of roughly 100 μm. The overlap between the laser

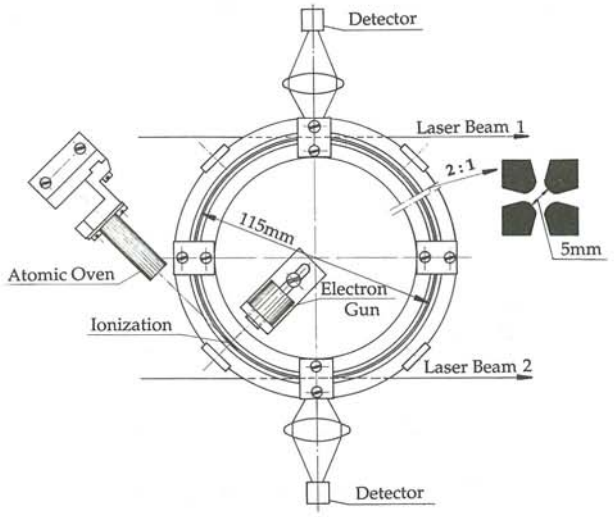

FIG. 1. Experimental setup of the quadrupole storage ring. Inset: An enlarged cross section of the ring electrodes. The electron multiplier for counting the stored ions is not shown.

beam and the trap region is about 2% of the total ring circumference. The resonantly scattered light is imaged onto a photomultiplier tube or an imaging photon detector system through magnifying optics.

In a Paul trap, stored ions can be cooled by using a single laser beam with a frequency tuned below resonance. In the ring trap, the ions can move freely around the ring. If the laser frequency is tuned into resonance from below, the velocity distribution of the ions is changed so that they finally end up with a bunched (cooled) velocity distribution, moving in the direction of the laser beam. Two laser beams in opposite directions are therefore necessary to cool all stored ions to rest. In our experiment, we achieve efficient cooling with a single beam since a contact potential is produced in the region where the atomic beam enters the storage volume by deposition of stray magnesium atoms on the electrodes. The potential of the coated trap section is roughly 1 eV above that of the rest of the ring. As a consequence, laser-cooled ions with a kinetic energy below that value cannot pass the potential barrier. They oscillate in the direction along the field axis and come periodically into resonance with the laser light like the ions in a Paul trap. Fast ions are first cooled sympathetically [5] via Coulomb collisions until they are unable to pass the potential barrier, which then compensates the radiation pressure.

A typical excitation spectrum of $^{24}Mg^+$ ions is shown in Fig. 2, where the total number N of stored ions is 10^4. The drop in the fluorescence signal at resonance is due to laser heating for positive detuning, whereas the sharp decrease in intensity at a detuning of -270 MHz is characteristic of the phase transition of laser-cooled ions from a chaotic gaseous state to an ordered crystalline state as also observed in a Paul trap [6,8]. The intensity decrease results from the sudden contraction of the velocity distribution of the ions. We also find similar behavior when one of the other control parameters of the trap such as the laser power or RF voltage is changed. Though the ions could not be resolved individually with the imaging

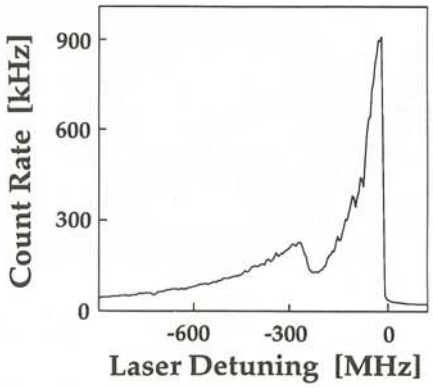

FIG. 2. Fluorescence intensity as a function of laser detuning. The spectrum shows a phase transition ($\psi_0 = 3$ eV, laser intensity about 0.3 W/cm^2).

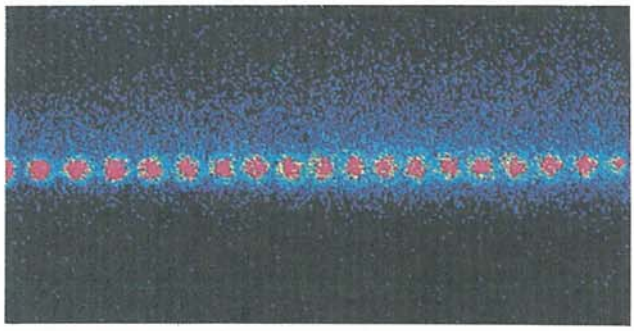

FIG. 3. Spatially resolved image of the fluorescence light emitted by an ordered structure of 19 ions forming a linear string. The distance between the ions is 33 ± 1 μm. The image is color coded, with red indicating high and blue indicating low fluorescence intensity (vacuum pressure 10^{-8} Pa).

system in this measurement, a sudden contraction in the transversal distribution of the ions is observed at the point of the phase transition. Laser cooling of the transversal motion would not be expected to be very effective owing to its small component in the direction of the laser beam. However, there is strong Coulomb coupling between the ions, and therefore cooling in the direction along the field axis is always combined with cooling in the transversal direction as well.

An improvement of the imaging optics allows one to resolve the ions individually. Figure 3 shows a string of ions aligned along the center line of the ring trap for a total number N of stored ions of about 10^4. The signal counts were integrated for several seconds. Under the conditions used for this measurement it is possible to observe also the phase transition from a disordered state to an ordered structure.

The temporal behavior of an ion string at increased vacuum pressure and lower laser intensity, observed at the maximum time resolution of the imaging system of 40 ms, is shown in Fig. 4. The images reveal two characteristics: (a) the ions appear at unequal distances, $^{24}Mg^+$ ions apparently being missing, and (b) the spatial

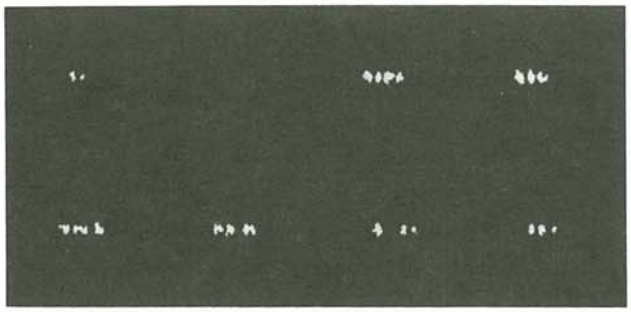

FIG. 4. Eight successive time-resolved images of a linear string of ions taken with an integration time of 40 ms for each frame (parameters as in Fig. 3, except magnification of the imaging system, vacuum pressure 10^{-7} Pa, and lower laser intensity).

distribution of the fluorescing ions changes randomly. We can obtain time-averaged string patterns for integration times as long as a minute, which indicates that the absolute ion positions stay fixed on that time scale. Since the Coulomb repulsion gives equal ion separations, there must be positively charged particles at the dark "lattice points" in the ion string. By monitoring the stored ion number when only the background gas was ionized, we could estimate that under the conditions of Fig. 4 the number of stored residual gas ions is comparable to that of $^{24}Mg^+$ ions. Thus, we deduce that the change in the string pattern is due to $^{24}Mg^+$ ions interchanging positions with other only sympathetically cooled species such as $^{25}Mg^+$, $^{26}Mg^+$, or ions of the residual gas included in the ion string. At $\psi_0 = 3$ eV and ion numbers N of 10^3 to 10^4 a residual thermal energy of about 100 mK is needed for two ions to exchange positions, i.e., to climb up the confining potential far enough so that a jump becomes possible. This effect can be inhibited by reducing the thermal energy of the ions or by increasing the trapping potential. We reach ion temperatures of below 10 mK, as can be deduced from the width of the excitation spectra. The string of ions is also stabilized by decreasing the number of residual gas ions, thus increasing the laser cooling efficiency. This can be achieved by operating the ring trap at a stability edge, where ions heavier than $^{24}Mg^+$ are expelled.

The ion storage ring allows us to investigate the transfer of laser cooling from one side of the ring to the other. For this purpose two laser beams are applied on opposite sides of the ring (see Fig. 1). Figure 5 shows two excitation spectra with roughly $N = 10^4$ ions stored in the ring: the upper one with both laser beams on and the lower one with only one beam on. The potential depth ψ_0 is considerably higher here than in Fig. 2, resulting in a stronger RF heating [8]. Therefore a phase transition is

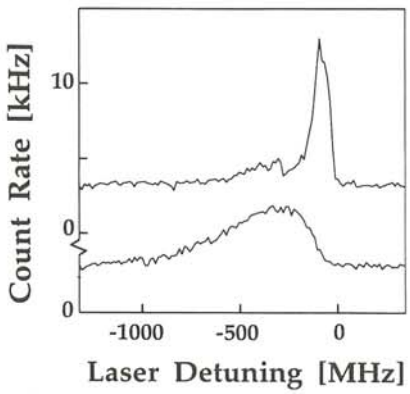

FIG. 5. Fluorescence intensity as a function of laser detuning. Lower spectrum: only the observation zone is illuminated. Upper spectrum: both interaction zones are illuminated. In the latter case a significant reduction in the linewidth is observed ($\psi_0 = 14$ eV, laser intensity about 1 W/cm^2 in each interaction zone).

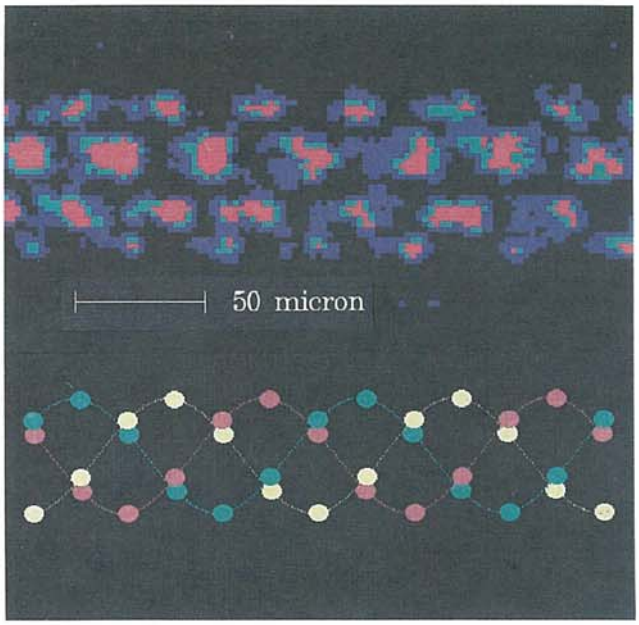

FIG. 6. Helical structure of $^{24}Mg^+$ ions with a diameter of 63 ± 2 μm. The experimental image (top) corresponds to three interwoven helices (shown in different colors, bottom). The closely appearing pairs of ions are sitting on opposite sites, resulting in twice the intensity at those positions ($\psi_0 = 1.1$ eV).

only observed when both laser beams are applied. A reduction in thermal energy is thus transferred from one side of the ring to the other (even though the ions stay essentially in their places). Another interesting effect can be observed when the two laser beams are very different in intensity and when in addition the ring contains a large fraction of residual gas ions: We find that $^{24}Mg^+$ ions are collected in the region with the higher laser intensity. This effect is directly caused by the redistribution of $^{24}Mg^+$ and residual gas ions between the different sections of the storage ring. The species interacting with the light field is collected in the stronger laser field, whereas the others are forced to move out and establish an equilibrium distribution of charges. When the storage ring is cleaned of residual gas ions, this effect is not observed. The concentration of the ions in the more intense zone is predominantly due to a temperature gradient generated by laser cooling, which is more effective in the stronger laser field.

Increasing the ion density changes the configuration of ions in the ordered structure. When about $N = 10^5$ ions are injected into the ring, laser cooling leads to the spectroscopic observation of a phase transition as in Fig. 2, resulting in the helical structure shown at the top of Fig. 6, which was recorded with the improved imaging optics. In numerical simulations [12] the normalized linear particle density $\lambda = (N/2\pi R)/(3q^2 r_0^2/16\pi\epsilon_0\psi_0)^{1/3}$, where q is the ion charge, is used to characterize the expected structures. A helical structure is predicted for values in the

range of $0.964 < \lambda < 3.10$. The experimental parameters of Fig. 6 yield $\lambda = 3.0 \pm 0.2$, in agreement with the theoretical predictions. Evaluation of the intensity distribution of the recorded image leads to the structure shown at the bottom of Fig. 6, consisting of three interwoven helices. The crystallization is in contrast to the observation of shell structures in Penning traps [15].

There is an important application of the storage ring we would like to mention: The linear ion structure allows us to apply the new Sisyphus cooling method [16] for which the lower temperature limit achievable is given by the recoil limit, corresponding to 6 μK for Mg^+ ions. At this temperature the harmonically bound ions reach their vibrational ground state, i.e., a Mössbauer situation is generated.

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