Large Ion Crystals in a Linear Paul Trap

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Plasmas of Mg^+ ions, containing more than 10^5 ions, have been observed to reach well-ordered (crystalline) states by applying laser cooling. The crystals are highly elongated with up to ten concentric cylindrical shells surrounding a central string. Such large structures have not previously been observed in a Paul trap. The amplitude of the micromotion of the ions can be larger than the shell spacings. As the diameter changes along the crystals, sharp transitions are observed when new shells form, in good agreement with molecular dynamics simulations. The predictions from simulations of how ordering develops with decreasing temperature are also confirmed. [S0031-9007(98)07188-9]

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Clouds of laser-cooled, trapped ions have previously been observed to condense and to exhibit quasicrystalline spatial order in Penning [1,2] and Paul (radiofrequency or rf) traps [3–5]. A classical, *infinite*, one-component plasma undergoes a transition from liquidlike behavior to a body-centered-cubic (bcc) lattice when the ratio Γ of the Coulomb energy between adjacent particles to the random thermal kinetic energy exceeds 175 [6]. In contrast, for *finite* plasmas molecular dynamics (MD) simulations predict formation of concentric shells with near-hexagonal ordering within the shell [7,8]. (Though these structures have no long-range periodic order, they are often referred to as crystals, a usage we follow here.)

In Paul traps the rf field drives micromotion, which is a modulation (at the rf frequency) of the secular harmonic motion in the effective trapping potential. The magnitude of the micromotion increases with an ion's distance from the trap central axis, and such motion can couple energy from the rf drive into the *random* motion of the ions through their mutual Coulomb interaction. This so-called rf heating (see, e.g., [9,10]) is widely assumed to limit attainable crystal sizes. The kinetic energy associated with the micromotion can be several orders of magnitude higher than the thermal energy at which spatial ordering occurs in static potentials, so the coupling of micromotion into thermal motion can be expected to be critical for crystal formation. The noninertial constraints and the continually changing shape of the cloud in the rf field can affect the ordering process and the resultant structure of the crystals in interesting ways that at present are poorly understood.

Crystals consisting of at most five shells have been attained earlier [5] in a ring rf trap, and simulations with up to 512 ions in a standard Paul trap, with the emphasis on the averaged position of ions in the crystal [11] have been reported previously. By contrast, the largest ordered systems so far were seen in Penning traps, where more than 10^5 ions have been crystallized and show evidence of central bcc structures [2].

In this Letter, we present evidence for Coulomb crystals of the largest transverse size observed in a linear Paul trap [12], as well as evidence on how the ordering develops gradually as the random motion or "temperature" of the ions is reduced. We have also performed simulations for up to 10^4 ions, taking into account the micromotion. These simulations predict the same detailed structures as are observed in the experiments.

The linear Paul trap [12] is a quadrupole composed of four parallel cylindrical electrodes (55 mm long), each sectioned longitudinally into three pieces (15, 25, and 15 mm), so that a positive dc potential can be added to the rf field on the eight end pieces to provide axial confinement [13]. The diameter of the electrodes is 4 mm, and they are at the closest 1.75 mm from the trap center axis in order to obtain the best approximation to a perfect quadrupole field [14]. An rf frequency $\Omega = 2\pi \times 4.2$ MHz and an amplitude $U_{\mathrm{rf}} = 30\text{–}50$ V are typically used. This gives rise to a value of the stability parameter q [15] in the range $[0.07\text{–}0.2]$ for Mg^+ ions and to a radial effective (pseudo) potential with a single-particle frequency $\omega_R = q\Omega/\sqrt{8}$. The very open trap configuration allows good access to laser beams along the central axis. The ions are directly cooled along this axis while the transverse degrees of freedom are cooled due to the coupling to the axial motion through Coulomb interactions.

The $^{24}Mg^+$ ions were produced by ionizing an effusive atomic Mg beam with a 1 keV electron beam at the center of the trap. In order to preferentially load the trap with the ^{24}Mg isotope, ions are laser cooled during loading using two counterpropagating laser beams which are continually frequency scanned through 8 GHz just

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below the $3s\ ^2S_{1/2}$-$3p\ ^2P_{3/2}$ transition for this isotope. The laser light at 280 nm needed for cooling is produced by frequency doubling of a dye laser system. The frequency doubled laser beam is spatially filtered and then power stabilized using an acousto-optical modulator. The resulting beam has a nearly Gaussian profile (waist of $\sim$0.5 mm) at the trap center with a power of 15 mW. The integrated fluorescence from the ions is monitored by a photomultiplier tube, and the shape and structure of the ion cloud is observed through a lens system with a 9X magnification, to image the fluorescence from the ion plasma onto a liquid nitrogen cooled, 1024×1024 pixel CCD camera having a pixel size of 24 μm.

Figure 1a shows a CCD picture (2 sec exposure) of the end of a very prolate ion crystal containing about 3500 ions. The well-separated points of high intensity indicate that some of the ions are relatively fixed in position with respect to the plane perpendicular to the line of sight. Since the ions (except for the ones on the trap axis) undergo the collective micromotion in the rf field with a period very short compared to the detection time, the sharpest image will be of those ions whose motion has the smallest amplitude in the plane perpendicular to the axis of viewing. These are the ions close to the midplane of the crystal, where the micromotion is perpendicular to the plane. In Fig. 1a the transitions between different shell structures along the axis are well defined. In order to test our understanding of these systems we have carried out MD simulations with 3500 ions using a radial rf field and assuming a harmonic potential along the trap axis. The result of such a simulation is presented in Fig. 1b. The calculated motion is integrated over an rf period to simulate the exposure time of the camera. As in the experiments, well-defined edge structures are observed. Transitions between different shell structures along the crystal are also seen in the simulation, but not as pronounced as in the experiment. The potential along the trap axis is known to be slightly anharmonic, so we do not expect the simulations to quantitatively match our measurements.

Since the crystal in Fig. 1a is very prolate, it is furthermore interesting to compare the experimental results with MD simulations of infinitely long cylindrical crystals in a static potential [16,17]. In this limit the shell structure is determined by the dimensionless linear density λ, defined as the number of ions per Wigner-Seitz radius a_{WS} [$a_{\mathrm{WS}} = (3/4\pi n_0)^{1/3}$, where n_0 is the asymptotic density for a cold system with many shells]; in our experiments a_{WS} is about 15 μm. The relative values of λ were extracted from the CCD data under the assumption that all ions contribute equally to the fluorescence signal. The absolute value of the linear density can be obtained by normalizing to the amount of light detected from the three single ions in the tip of the crystal or by assuming that the MD simulations for infinitely long crystals are valid in predicting the linear density for a particular

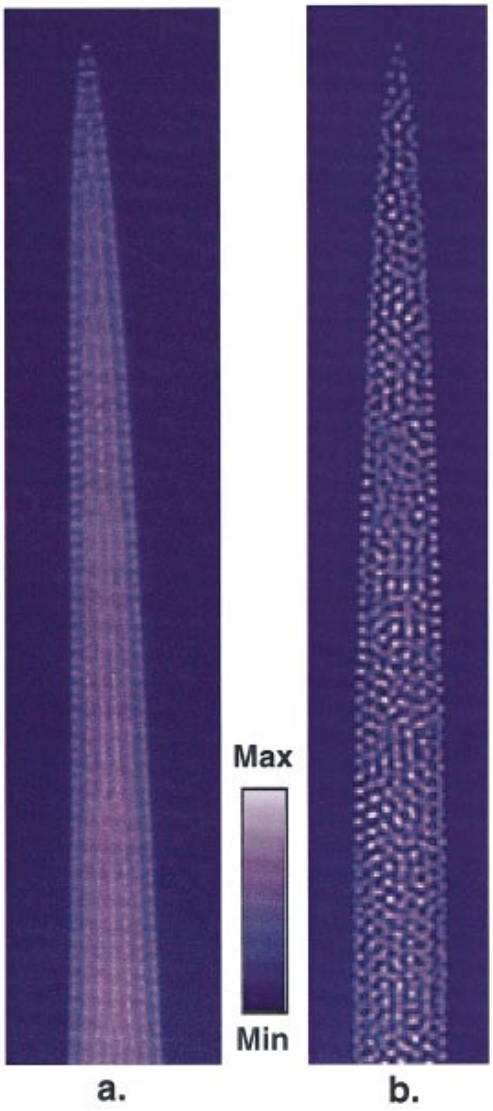

FIG. 1(color). (a) CCD picture integrated over 2 sec of a very prolate crystal of about 3500 ions. (b) Molecular dynamic simulation with 3500 ions in a crystal with roughly the same aspect ratio as the one in (a).

structural transition. For the crystal depicted in Fig. 1a, the two methods give nearly the same result. In Fig. 2, the dimensionless linear density λ, normalized to the MD simulation in the vicinity of the transition from a two-shells-plus-string to a three-shell configuration is plotted as a function of the position along the crystal (solid line). The dotted vertical bars indicate the uncertainty in the measured positions of the transitions while the dashed horizontal lines correspond to linear densities where transitions are expected to happen for infinitely long crystals. For the transitions for which theoretical values of λ have previously been calculated [17], the agreement is strikingly good. A similar analysis of the simulation presented in Fig. 1b shows the same correspondence with theory for infinite plasmas. Birkl *et al.* [5] had confirmed the

predictions of MD simulations in experiments where up to four-shell-plus-string ion crystals of constant linear charge densities were observed in a ring-shaped Paul trap.

In Figs. 3a and 3b, a CCD picture and associated transverse intensity profile of the crystal are presented for one of the largest crystals produced. This crystal consists of a central string surrounded by ten shells and holds more than 10^5 ions. The maximum linear density λ is about 5 times larger than that previously obtained in Paul traps [5]. MD simulations on infinitely long cylindrical crystals predict that the radial spacing between shells δr becomes constant at a value of around $1.48 a_{\mathrm{WS}}$ [17] once there are more than about three shells. From Fig. 3b we deduce that the spacing is indeed constant and has a value $\delta r = (1.35 \pm 0.15) a_{\mathrm{WS}}$. The uncertainty in δr comes partly from the uncertainty in measuring the radii from the pictures, and partly from the uncertainty in the measurement of the rf voltage (which is used in determining a_{WS}). In experiments with slightly smaller linear densities and in which the rf voltages were measured more precisely, we find an even better agreement with the MD results. For these larger crystals, the amplitude of the micromotion is, in fact, larger than δr and the corresponding kinetic energy about 4 orders of magnitude higher than the limited thermal energy needed for ordering. We have observed larger partially crystallized plasmas, in which the outer shells have been well defined, while structures in the central part were lacking. Estimated from the diameter of the outer shell, such crystals would consist of 13 shells if fully crystallized. Since the laser cooling rate saturates at a level specific to the ions used, while the rf heating grows with crystal radius, eventually there may be a limit on the size of obtainable crystals in Paul traps. Whether this limit is reached in the above-mentioned experiments is still unclear, but future experiments using stroboscopic observation as well as realistic MD simulations could reveal this.

The structural evolution of a strongly coupled one-component plasma with temperature has been studied through simulations [18], but, to our knowledge, has not been addressed experimentally in a systematic fashion. In Fig. 4, we present transverse profiles of an ion plasma in our trap during the cooling process. In these measurements, as the laser detuning, defined as the difference between the laser frequency and the atomic transition frequency, gets smaller, the laser cooling becomes more effective and the component of kinetic energy for motion along the laser is reduced. The concept of temperature in such a dynamic system is not clearly defined, but it seems to be a reasonable working assumption that with cooling, the coherent transverse motion in the rf field does not couple significantly into longitudinal velocities, and thus that the value of Γ relevant to ordering may be computed from this velocity component alone. This assumption is supported by the results of the MD simulations referred to above. The values of Γ listed in Fig. 4 are derived from the longitudinal laser detuning with the assumption that the random velocities have a Boltzmann distribution with a FWHM corresponding to the velocity that makes the ions Doppler shifted into resonance. For the largest detuning (smallest Γ) shown in this figure only a hint of an outer crust is seen, with the outer shell sharpening and interior structure developing to a fully ordered seven

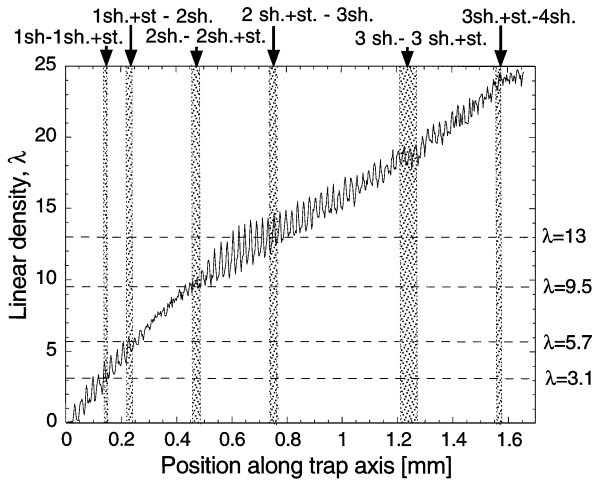

FIG. 2. The dimensionless linear density λ as a function of position along the crystal is shown in Fig. 1a (solid line). The shaded vertical areas indicate the uncertainties in the measured position of structural transitions, while the dashed horizontal lines correspond to linear densities where these transitions should happen according to MD simulation for infinite long crystals.

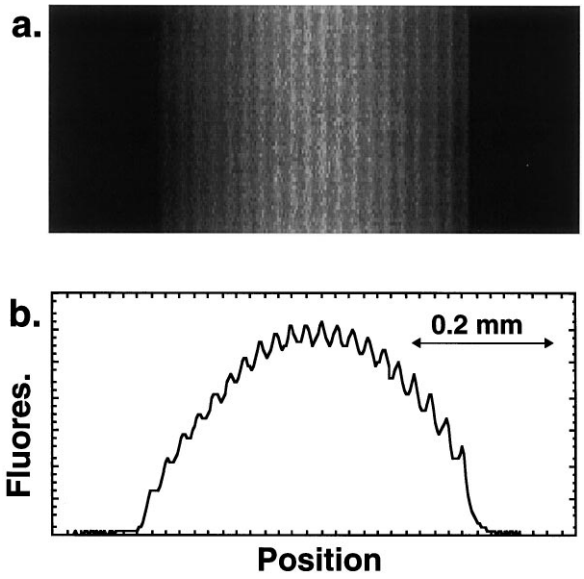

FIG. 3. (a) Part of a picture showing a crystal with ten shells surrounding a central string of ions. (b) Transverse intensity profile corresponding to the picture in (a). As expected from MD simulations the shells are nearly equally spaced.

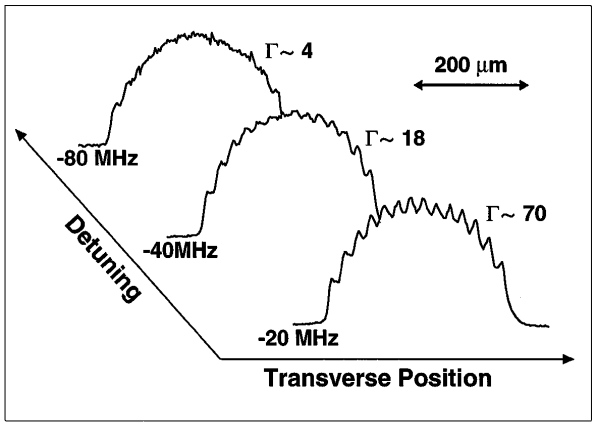

FIG. 4. Transverse intensity profiles of ion crystals laser cooled with various final detunings. The corresponding values of the coupling parameter Γ are estimated by the procedure described in the text.

shells for the coldest system. This pattern of ordering has also been found in simulations [18].

Our results also point to the possibility of using rf traps for sympathetic cooling [19] of large numbers of ions of species not directly addressed by the cooling laser. This technique, in particular, is interesting when one is dealing with very weak atomic or molecular ionic transitions. The Hamiltonian of ions in an rf trap is also quite similar to that of ions in a bunch in a storage ring [20]. The ability to cool and crystallize large numbers of ions in the trap may shed some light on certain aspects of the dynamics of cold and dense ion beams.

In conclusion, we have shown that crystals having up to ten shells and linear densities of as much as $\lambda = 150$ can be created in a linear Paul trap. Whether this represents a fundamental limit because of rf heating remains to be understood, but even if bulk structure turns out to be unobtainable in rf traps, our work suggests many new opportunities for studies of intermediate size structures. The observed structure in large crystals is found to be in agreement with the predictions of MD simulations.

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