Sympathetic Crystallization of Trapped Ions

P. Bowe, L. Hornekær, C. Brodersen, M. Drewsen, and J. S. Hangst
Institute of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C, Denmark

J. P. Schiffer
Argonne National Laboratory, Argonne, Illinois 60439
and University of Chicago, Chicago, Illinois 60637
(Received 5 November 1998)

We have created multispecies Coulomb crystals in a linear Paul trap containing up to a few hundred ions of which more than 50% were cooled only sympathetically through the Coulomb interaction with laser-cooled Mg^+ ions. In an extreme case, one laser-cooled ion maintained order in a 15 ion string. Ion species segregation was obtained by radiation pressure. Previous experiments and molecular dynamics simulations suggest the temperature is 10 mK or lower. These results indicate that a wide range of atomic and molecular ions can be cooled and localized in linear Paul traps which is important for improvements in spectroscopic studies of such ions. [S0031-9007(99)08637-8]

PACS numbers: 32.80.Pj, 42.50.Vk, 52.25.Wz

Trapped ions, when cooled sufficiently, form spatially ordered structures (Coulomb crystals). For smaller crystals (typically $\leq 10^5$ ions), where surface effects play a dominant role, shell and stringlike structures are the equilibrium states [1–3]. Molecular dynamics (MD) simulations of infinite one component plasmas predict a body centered cubic (bcc) structure [4] as the lowest energy state. By applying laser cooling, string and shell structures have been observed in both Paul and Penning traps by imaging the fluorescence from the ions [5–11], and recently bcc structures at the center of very large ion crystals in a Penning trap have been revealed by Bragg scattering techniques [12,13]. Since only ions with simple level schemes accessible to lasers can easily be laser cooled, most atomic ion species, and all molecular ions, due to their complex vibrational and rotational structure, are excluded from this type of cooling. Hence, to date only very few singly charged ion species have been laser cooled and crystallized. The equations of motion in both Penning and Paul traps allow, however, ions within a certain charge-to-mass ratio to be trapped simultaneously, which makes sympathetic cooling [14] through the Coulomb interaction possible.

Several authors have previously investigated sympathetic cooling, where directly laser cooled ions were used to cool ions of a different species through mutual Coulomb interaction [9,14–17]. In most of these experiments, the typically achieved temperatures of the sympathetically cooled ions were several hundred mK, which were too high for ordering of the whole plasma. In a ring-shaped [18] and a linear Paul trap [19] a few nonfluorescing sites in crystals consisting of laser cooled $^{24}Mg^+$ were attributed to indirectly cooled impurity ions. Recently, a crystal consisting entirely of Ca^+ was observed to stay crystallized when some of the constituent ions were decoupled from the cooling laser by being optically pumped into a metastable dark state [9].

In this Letter we report on formation of Coulomb crystals consisting of up to a few hundred ions in a linear Paul trap where the fraction of indirectly (sympathetically) cooled ions is greater than 50%. In one case particularly interesting for spectroscopy, 14 sympathetically cooled ions were maintained in an ordered string structure by one directly cooled $^{24}Mg^+$ ion. We have not measured the temperature of the sympathetically cooled ions in the crystals directly, but recent studies of the formation of $^{24}Mg^+$ ion crystals [10] and MD simulations of infinitely long cylindrical plasmas [20] indicate that the temperature is 10 mK or lower.

The radiation pressure of the cooling lasers has furthermore made it possible to segregate the ions according to species. Besides the potential interest of such mixed Coulomb crystals within plasma physics, the very cold and well-localized sympathetically cooled ions are very interesting in connection with high-resolution spectroscopy, where transit time broadening, Doppler shift and broadening, and laser-induced light shifts [15] can be problematic.

The experimental setup used is sketched in Fig. 1 and is practically identical to the one previously described in [10]. The ions are confined in a linear Paul trap [21] operating at an rf frequency of $\Omega = 2\pi \times 4.2$ MHz and an rf amplitude of $U_{\rm rf} = 20$–100 V. The Mathieu stability parameter q of the linear Paul trap is defined as

$$q = 2\,\frac{QU_{\rm rf}}{mr_0^2\Omega^2},$$

where r_0 is the distance from the trap axis to the electrodes and Q and m are the charge and mass of the ions, respectively. For $^{24}Mg^+$ ions the rf parameters employed give values of q within the range $[0.075, 0.38]$. As the trap is loaded by electron bombardment of an atomic magnesium beam at the trap center, ions can also be produced from the background gas, and these will be trapped if their charge-to-mass ratio gives values of q below the Mathieu stability

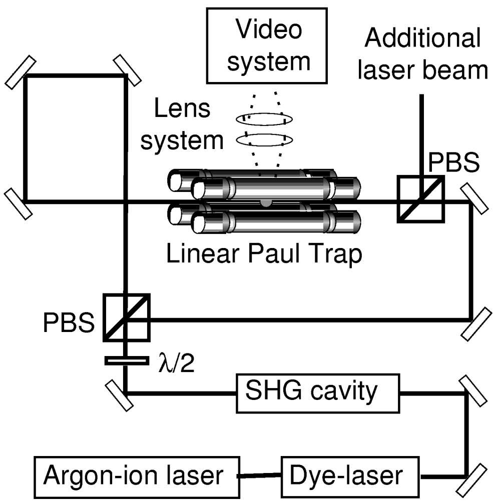

FIG. 1. Schematic overview of the experimental setup. The optional second UV laser beam of a separately adjustable frequency, produced by a similar laser and frequency doubling system, is also shown. PBS (polarizing beam splitter); $\lambda/2$ (half-wave plate).

limit of 0.9. By controlling the atomic beam flux, the fraction of background gas ions can be controlled to some extent. The $^{24}\mathrm{Mg}^+$, and in some cases also $^{26}\mathrm{Mg}^+$, ions are laser cooled axially by Doppler cooling on the $3s\,^2S_{1/2}$-$3p\,^2P_{3/2}$ transition. Sympathetic cooling of the transverse degrees of freedom of the laser-cooled ions as well as all degrees of freedom of the other trapped ions is accomplished through the Coulomb interaction between the ions. When cooling a single isotope, two counterpropagating laser beams at the same frequency and with adjustable intensity difference are sent into the trap region. When cooling two isotopes an extra laser beam at a separately tunable frequency is added. The transition frequency of $^{26}\mathrm{Mg}^+$ is 3 GHZ (equal to 70Γ, where Γ is the natural linewidth of the transition) above that of $^{24}\mathrm{Mg}^+$, and hence any heating of the $^{24}\mathrm{Mg}^+$ ions due to the laser resonant with $^{26}\mathrm{Mg}^+$ is very weak compared to the cooling by the laser resonant with $^{24}\mathrm{Mg}^+$. The positions of the ions are monitored by imaging the fluorescent light onto an image-intensified video camera (Proxitronic NCA), the time resolution of which is 20 ms. The camera system views the trap perpendicular to the trap axis.

In Fig. 2(a) we show an image enhanced single video frame of a single $^{24}\mathrm{Mg}^+$ ion in a string with 14 nonfluorescing ions. The dark ions are either atomic or molecular ions from the background gas [17,22] or other isotopes of magnesium. The evidence for the string comes about by monitoring the various positions the single $^{24}\mathrm{Mg}^+$ ion visits due to diffusion in a 2 min video sequence. In Fig. 2(b) a composite picture of the positions visited by the visible ion is shown together with black crosses indicating simulated positions of a string of 15 singly charged ions at zero temperature. Since each video frame always shows the visible ion in a single position, the rearrangement time of the string after a detectable disturbance must be faster than the frame integration time of 20 ms. This is in accordance with typical reordering times ($\sim$1 ms) found in the MD simulations discussed below. For the observed positional diffusion of the visible ion with a jump rate of 0.36 per sec, there are two probable processes: occasional collisions with atoms or molecules of the background gas and random walk of the ions induced by scattering of photons during the laser cooling process. For the experimental background pressure of approximately 10^{-10} Torr, we estimate 0.03 elastic collisions per ion per second [23]. If each such collision transfers enough momentum for a positional jump, this can explain the observed jump rate. We have initiated MD simulations where the rf field of the trap, collisions with the background gas atoms, and the random momentum kicks given to the ions in the photon scattering processes are included. Our preliminary simulations indicate that nearly every collision will lead to a positional jump without loss of trapped ions (the trap potential depth is typically more than an order of magnitude larger than the kinetic energy of a room temperature atom). However, future experiments conducted at various pressures and MD simulations should reveal more clearly the importance of the two mechanisms for positional jumps.

With an electron beam energy of 1 keV, one would also expect to produce and trap doubly charged ions in an observable amount [24]. The good agreement between the positions of the ions in the experiment and in the simulation presented in Fig. 2(b) suggests, however, that only singly charged ions are present in the string (the presence of doubly charged ions would lead to different ion spacings). The reason no doubly charged ions are present is probably

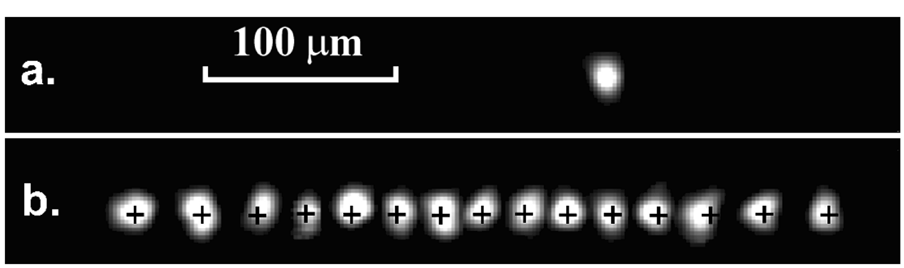

FIG. 2. (a) A single video frame showing the lone $^{24}\mathrm{Mg}^+$ ion in one of the fifteen sites. (b) The overall structure of the crystal visualized by making a superposition of 15 images in each of which a different site is occupied. The superimposed crosses indicate the positions predicted by molecular dynamics simulations for a string containing 15 singly charged ions.

that the string in Fig. 2 is the product of a larger crystal decaying over one hour or so, in which time electron transfer collisions with background gas atoms are highly possible. A collision between a doubly charged ion and an atom can lead to the production of two singly charged ions, while collisions between a singly charged ion and an atom will not lead to the formation of a doubly charged ion.

We have also observed formation of crystals containing some 200 ions where 60% of the ions were cooled sympathetically. Figure 3 shows a sequence where the ions are loaded into the trap in the presence of two counterpropagating lasers resonant with $^{24}Mg^+$ and $^{26}Mg^+$, respectively. Because of the net radiation pressure force exerted on the cooled ions in the direction of the laser beams, the crystal is spatially segregated according to species. The apparent void between the two magnesium isotopes cannot be attributed to the radiation pressure forces. These forces can at most displace the ions some 50 μm along the axial trap potential, while in Fig. 3(c) the space between the visible ions is 500 μm wide or 10 times larger. We conclude therefore that this dark region must be filled with other ionic species. The total number of ions present is estimated from measuring the aspect ratio of the crystals and using the model introduced by Dubin in [25]. The visible magnesium ions are clearly crystallized at the boundary with the dark region, indicating that the ions in the dark region are also crystallized, since the Coulomb interaction is long range and the otherwise chaotic motion at the center of the crystal would lead to heating of the entire ion plasma. By using two counterpropagating laser beams resonant with only one of the magnesium isotopes and temporarily changing their power balance, we can cause the visible ions to move back and forth through the center. The movement takes the form of discrete hops from one well-defined site to another. Such ordered migration is seen clearly in the video frames shown in Fig. 4 where in a separate experiment only 14 $^{24}Mg^+$ ions out of a total of about 60 ions were cooled directly.

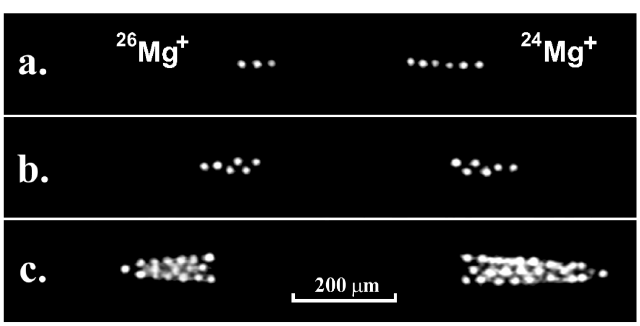

FIG. 3. Three successive images taken during a loading sequence. The $^{24}Mg^+$ ions are pushed to the right and $^{26}Mg^+$ ions to the left by their respective near resonant cooling lasers. The central region contains sympathetically cooled ions. Note the nearest neighbor separation between ions is ca. 20 μm while the space between the two magnesium isotopes is ca. 500 μm wide.

For judging the spectroscopic prospects of these sympathetically crystallized ions, it is important to know the kinetic energy of the ions. Previous studies of larger $^{24}Mg^+$ crystals of similar shapes [10] show that the onset of spatial ordering happens at a plasma coupling parameter comparable to the value predicted for infinite plasmas [20]. The plasma coupling parameter is defined as

$$\Gamma = \frac{E_{\mathrm{Coul}}}{k_B T},$$

where T is the temperature and E_{Coul} is the nearest neighbor Coulomb energy. From the simulation presented in [20], ordered shell structures are expected when $\Gamma \geq 100$. With the trap parameters used in the present experiment this lower limit of Γ sets an upper temperature limit of about 10 mK. Because of the strong coherent transverse micromotion of the ions, the maximum transverse kinetic energy of the outer shell ions in Fig. 3(c) corresponds, however, to a temperature of a few kelvin. The coupling of transverse micromotion into the axial motion is found (MD simulations) only to be a few mK, leading to a small increase in the first order Doppler shifts along the trap axis. The second order Doppler shifts will, however, generally be larger than implied by the estimated axial temperature due to the transverse micromotion. Exceptions are the ion string case presented in Fig. 2, as well as the case we have observed where a shell of directly cooled ions surrounded a string of indirectly cooled ions. Here the indirectly cooled species lie on the axis where there is essentially no micromotion. In the latter case, the spatial separation of the species is due to the q/M dependence of the effective transverse trap potential. Ions with high q/M values will experience a steeper potential than ions with lower values and hence tend to accumulate along the trap center axis.

In our present trap, we have observed stable crystals for the Mathieu stability parameter q ranging from $q = 0.075-0.38$. Observed instabilities at low q are due to dc field imperfections and fast ion loss due to collisions with background gas atoms, while the instabilities in the high q range are probably due to excessive rf heating [26]. Since $q \propto Q/m$, this indicates that singly charged ions with a mass ratio of up to about 10 could be trapped and cooled simultaneously. With the rf parameters employed,

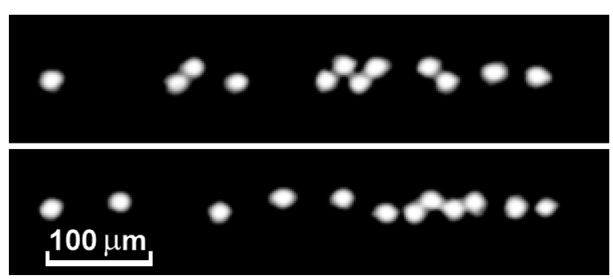

FIG. 4. A sequence of images recorded using only one cooling laser tuned to $^{24}Mg^+$. The images are taken while the ions are migrating due to power imbalance between the two counterpropagating laser beams.

the estimated mass range of sympathetically cooled, singly charged ions is 4–162 amu when cooled by $^{24}Mg^+$ ions. The sympathetic cooling rate is, however, highly dependent on the mass and charge of the directly and sympathetically cooled ions, both through the possible energy transfer in collisions and through the fact that the various ion species tend to separate spatially due to differences in effective potentials. The constraints on sympathetic crystallization due to these effects remain to be studied. However, since singly charged ions that can be laser cooled are scattered throughout the whole periodic table, any atomic and most small molecular ions should be able to be effectively cooled and localized.

In conclusion, we have demonstrated that in a linear Paul trap it is possible to make Coulomb crystals consisting of up to a few hundred ions where more than 50% of the ions are cooled sympathetically. We have proven it is possible to create and keep a string of 15 cold ions where only a single ion is directly cooled. Our previous studies of the formation of $^{24}Mg^+$ ion crystals and MD simulations indicate that the axial temperature of the sympathetically cooled ions is probably around 10 mK or lower, which is an order of magnitude lower than previously reported temperatures of sympathetically cooled ions. Our findings indicate that a wide range of atomic and molecular ions, in substantial amounts, can be effectively cooled and localized in linear Paul traps. This points towards improvements in spectroscopic studies of such ions.

This work was supported by the Danish National Research Foundation through the Aarhus Center of Advanced Physics, the Danish Research Council, the U.S. Department of Energy, Nuclear Physics Division, under Contract No. W-31-109-Eng-38, as well as the European Union under Contract No. ERBFMBICT972323.

[1] A. Rahman and J. P. Schiffer, Phys. Rev. Lett. **57**, 1133 (1986).
[2] D. H. E. Dubin and T. M. O'Neil, Phys. Rev. Lett. **60**, 511 (1988).
[3] R. W. Hasse and J. P. Schiffer, Ann. Phys. (N.Y.) **203**, 419 (1990).
[4] D. H. E. Dubin, Phys. Rev. A **42**, 4972 (1990).
[5] F. Diedrich, E. Peik, J. M. Chen, W. Quint, and H. Walther, Phys. Rev. Lett. **59**, 2931 (1987).
[6] D. J. Wineland, J. C. Bergquist, W. M. Itano, J. J. Bollinger, and C. H. Manney, Phys. Rev. Lett. **59**, 2935 (1987).
[7] S. L. Gilbert, J. J. Bollinger, and D. J. Wineland, Phys. Rev. Lett. **60**, 2022 (1988).
[8] G. Birkl, S. Kassner, and H. Walther, Nature (London) **357**, 310 (1992).
[9] W. Alt, M. Block, P. Seibert, and G. Werth, Phys. Rev. A **58**, R23 (1998).
[10] M. Drewsen, C. Brodersen, L. Hornekaer, J. S. Hangst, and J. P. Schiffer, Phys. Rev. Lett. **81**, 2878 (1998).
[11] T. B. Mitchell, J. J. Bollinger, H. E. Dubin, X.-P. Huang, W. M. Itano, and R. H. Baughman (to be published).
[12] J. N. Tan, J. J. Bollinger, B. Jelenkovic, and D. J. Wineland, Phys. Rev. Lett. **75**, 4198 (1995).
[13] W. M. Itano, J. J. Bollinger, J. N. Tan, B. Jelenković, and D. J. Wineland, Science **279**, 686 (1998).
[14] R. E. Drullinger, D. J. Wineland, and J. C. Bergquist, Appl. Phys. **22**, 365 (1980).
[15] D. J. Larson, J. C. Bergquist, J. J. Bollinger, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 70 (1986).
[16] H. Imajo, K. Hayasaka, R. Ohmukai, U. Tanaka, M. Watanabe, and S. Urabe, Phys. Rev. A **53**, 122 (1996).
[17] T. Baba and I. Waki, Jpn. J. Appl. Phys. **35**, 1134 (1996).
[18] I. Waki, S. Kassner, G. Birkl, and H. Walther, Phys. Rev. Lett. **68**, 2007 (1992).
[19] M. G. Raizen, J. M. Gillian, J. C. Bergquist, W. M. Itano, and D. J. Wineland, J. Mod. Opt. **39**, 233 (1992), and references therein.
[20] J. P. Schiffer in Proceedings of the Workshop on Crystalline Ion Beams, Wertheim, Gemany, 1988, edited by R. W. Hasse, I. Hofmann, and D. Liesen (GSI Report No. GSI-89-10, 1989).
[21] J. D. Prestage, G. J. Dick, and L. Maleki, J. Appl. Phys. **66**, 1013 (1989).
[22] G. Birkl, Dissertation, Ludwig-Maximilians-Universität München, München, 1993.
[23] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics—Non-Relativistic Theory* (Pergamon, Oxford, 1965).
[24] R. F. Boivin and S. K. Srivastava, J. Phys. B **31**, 2381 (1998).
[25] D. H. E. Dubin, Phys. Rev. Lett. **71**, 2753 (1993).
[26] R. Blümel, C. Kappler, W. Quint, and H. Walther, Phys. Rev. A **40**, 808 (1989).