Atomic-Ion Coulomb Clusters in an Ion Trap

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We observe small numbers of laser-cooled Hg^+ ions, which are confined in a Paul radio-frequency ion trap, to crystallize into regular arrays or clusters. We investigate the structure of these clusters by direct imaging, optical spectroscopy, and numerical calculations. The spectroscopy of such "pseudomolecules" is unique in that we can probe individual atoms of the molecule separately. The ratio of Coulomb potential energy per ion to k_BT in these clusters is observed to be as high as 120.

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The success of various techniques for cooling of charged atomic and elementary particles in traps and storage rings has stimulated considerable interest in the observation of the expected spatial ordering of a collection of such particles. The study of ordering not only has a direct bearing on the dynamics of these stored particles but has applicability to the study of cluster formation, Wigner crystallization, strongly coupled plasmas, and spectroscopy.[1-8]

In this experiment, we investigated, both by direct imaging and by spectroscopic methods, the structure of small clusters or crystals of laser-cooled atomic ions which were confined in a Paul radio-frequency (rf) trap.[9] Photographic images of one, two, and three ions in a Paul trap have previously been reported,[10] but the resolution achieved did not allow investigation of possible ordering. Indirect evidence for ordered structures of trapped atomic ions was reported by Diedrich *et al.*[11] Imaging of ordered structures of macroscopic charged particles stored in a Paul trap was reported in the experiments of Wuerker, Shelton, and Langmuir.[12] The ordering observed here was similar but occurred under substantially different experimental conditions. One advantage of the present experiments is that the charge and mass of the particles were well defined. In addition, the structure of clusters of atomic ions can be spectroscopically probed. As a demonstration, we have measured the vibrational frequency of the diatomic pseudomolecule $(^{198}\mathrm{Hg}^+)_2$ by measuring its optical spectrum. This experiment is unique in that we were able to probe a specific atom by fixing the pseudomolecule in space.

Small numbers of $^{198}\mathrm{Hg}^+$ ions were stored in a miniature Paul trap,[13] which has properties equivalent to those of a hyperbolic trap with dimensions $r_0 \simeq 466$ μm and $z_0 \simeq 330$ μm.[14] An oscillating voltage V_{ac} with peak amplitude between 145 and 325 V at a frequency of 23.26 MHz was applied between the ring and end-cap electrodes for trapping. The ions were laser cooled by 1–2 μW of cw 194-nm radiation (bandwidth <2 MHz), whose beam waist was varied between 5 and 15 μm at the position of the ions. This radiation was tuned near the $5d^{10}6s\,^2S_{1/2} \rightarrow 5d^{10}6p\,^2P_{1/2}$ first resonance line, which has a natural linewidth of 70 MHz. Some of the 194-nm fluorescence from the ions was focused onto the photocathode of a resistive-anode photon-counting imaging tube as shown schematically in Fig. 1. The optics for this imaging was provided by a three-stage fused-quartz lens system with an aberration-compensated first stage. The first lens was apertured to give an f number of 4.5. The overall magnification of the lens system was 60×. The positions of the photons detected by the imaging tube could be displayed on an oscilloscope in real time or stored by a computer in order to make time exposures. The resolution was limited by the imaging tube to about a factor of 3 worse than the diffraction limit. In Fig.

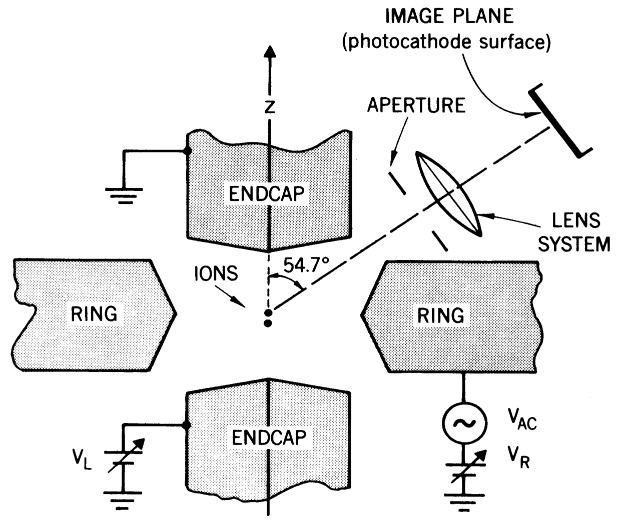

FIG. 1. Schematic drawing of the trap electrodes (to scale) and imaging system (not to scale). The end-cap to end-cap separation along the z axis is approximately 625 μm. The overall magnification of the lens system is 60×. The laser beam used to illuminate the ions also enters the trap at an angle of 54.7° with respect to the z axis and is perpendicular to the observation axis shown.

2(a) we show the trap coordinate system as viewed by the imaging tube. If we apply a positive static potential V_L to the lower end cap in Fig. 1 and hold the static ring voltage V_R at ground potential, a single ion[15] moves in the positive z direction by a known amount.[16] This determined the length scale in the images (after we accounted for viewing angle) and therefore also determined the magnification.

The harmonic pseudopotential well of the trap[9] is characterized by the single-ion resonance frequencies ν_z and ν_r for the motion in the axial and radial directions, respectively. For $V_L = 0$, these frequencies take the form[9]

$$\nu_r^2 = \nu_{r0}^2 + BV_R, \tag{1a}$$

$$\nu_z^2 = \nu_{z0}^2 - 2BV_R, \tag{1b}$$

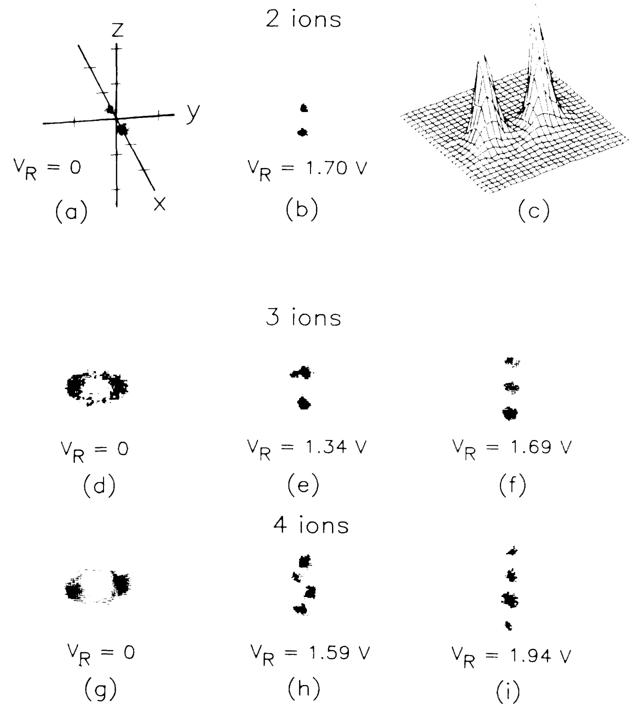

FIG. 2. Images of clusters of two, three, and four trapped and laser-cooled Hg$^+$ ions for the condition where $\nu_{z0} \simeq 2\nu_{r0} \simeq 500$ kHz. The coordinate system of (a) applies to the remaining pictures except for (c). The tick marks on the axes are separated by 10 μm. (c) A three-dimensional intensity plot for two ions pinned along the z axis ($V_R = 1.95$ V) and separated by 7.4 μm in the plane perpendicular to the direction of observation. The variation in the intensity for different ions is due, in part, to variations in intensity of the laser beam at the sites of the different ions. Peak counting rates were about 2000 s^{-1}/ion. Transitions between the observed configurations can be induced by the variation of the ring voltage V_R, and thereby the variation of the relative strengths of the axial and radial pseudopotential wells.

where ν_{z0} and ν_{r0} ($\nu_{z0} = 2\nu_{r0}$) are proportional to $V_{\rm ac}$. ν_z and ν_r were measured from the sideband spectra of an optical transition[15] and by direct electric field excitation of the motional resonances.[17] From these measurements we obtained $B = 0.0567(15)$ MHz^2/V. This agrees fairly well with the calculated value 0.0596 MHz^2/V based on the designed trap dimensions.

If two ions were loaded into the trap with $V_R = V_L = 0$, and the 194-nm source was slowly swept into resonance, the ions were observed to first "swarm" around the center of the trap with orbit radii up to a few tens of micrometers. As the laser detuning was further reduced, thereby increasing laser cooling, the ions oriented or "pinned" themselves along a preferred direction, separated by a distance d. Here the inward pseudopotential force on each ion is balanced by their mutual Coulomb repulsion. Since $V_R = 0$, the pseudopotential well in the axial direction is four times as strong as in the radial direction and the ions lie in the x-y plane. Although the trap was designed to have axial symmetry, the preferred spatial alignment of the cold ions indicates an asymmetric potential due to trap imperfections, contact-potential variations, or some other trap asymmetry. For convenience, we choose this preferred direction to be the x axis as indicated in Fig. 2(a).

If V_R is increased until $\nu_z < \nu_r$, we expect the ions to align along the z axis since this is the configuration which minimizes the potential energy. This configuration is shown in Fig. 2(b). For the conditions of Fig. 2 ($\nu_{r0} \simeq 250$ kHz), we expected this transition to occur at $V_R \simeq 1.11$ V and observed it to occur at $V_R \simeq 1.17$ V. The transition appeared to occur gradually over a range of about 30 mV rather than suddenly; this was probably related to the source of trap asymmetry in the x-y plane. For two ^{198}Hg$^+$ ions pinned along a certain direction, the interparticle spacing is theoretically given by $d = 3.29\nu_d^{-2/3}$ μm, where ν_d (in megahertz) is the single-ion oscillation frequency along the direction of separation. For the conditions of Fig. 2(b), we predicted $d = 8.5$ μm and measured 7.5(1.1) μm.

In Figs. 2(d)–2(i), we show various configurations for three and four ions. We also observed ordered structures for five or more ions. For three or four ions, when V_R was equal to zero, the ions appeared to lie the same distance from the z axis but were not pinned, as was the case for two ions. Instead, they appeared to circulate about the z axis [see Figs. 2(d) and 2(g)]. This interpretation is supported by the observation that the size of the circular image could be made to increase by more than a factor of 2 by appropriate positioning and tuning of the 194-nm laser beam. This is not surprising since the trap asymmetry in the x-y plane was relatively weak and since it was fairly easy to position the laser to apply a radiation-pressure torque about the z axis. Three stable configurations were observed for three and four ions. For large values of V_R, the ions were aligned along the z

axis as in Figs. 2(f) and 2(i). In the intermediate configuration for three ions [see Fig. 2(e)], one ion appeared to lie on the negative x axis and the others equidistant from the x-y plane and on a line parallel to the z axis which intersects the positive x axis. In the intermediate configuration for four ions [see Fig. 2(h)], two of the ions appeared to lie in the x-y plane pinned along the x axis, and two ions were positioned above and below the x-y plane along the z axis. We have verified the ordered structures in Figs. 2(d)–2(i) by calculating the configurations which minimize the potential energy for the specified trap parameters.

We also observed clusters where an ion apparently occupied a normal cluster position but did not fluoresce. This "phantom ion," which may have been $^{199}\mathrm{Hg}^+$, or perhaps an impurity ion like HgOH^+, switched positions with $^{198}\mathrm{Hg}^+$ ions of the cluster if the cooling was weak.

Such clusters of atomic ions can be viewed as pseudomolecules whose optical spectra would be expected to show rovibronic structure. As a demonstration, we have measured the vibrational structure of the diatomic pseudomolecule $(\mathrm{Hg}^+)_2$ by probing its optical structure. By pinning the molecule along the z axis as in Fig. 2(b), we could probe the optical structure of one of the atoms of the molecule by detecting the fluorescence of only this atom. In particular, we measured the structure of the $5d^{10}6s\,{}^2S_{1/2}(m_J=\frac{1}{2}) \rightarrow 5d^9 6s^2\,{}^2D_{5/2}(m_J=-\frac{1}{2})$ electric quadrupole transition ($\lambda \simeq 282$ nm) of one of these Hg^+ ions.

We first adjusted V_{ac} and V_R to make $\nu_r \simeq 2\nu_z \simeq 473$ kHz for a single ion in the trap. We than observed the quadrupole transition on this single trapped ion by the following method.[15] The 194-nm source was first tuned to give an easily detectable value of fluorescence. A radiation source near the quadrupole transition frequency was focused onto the ions and slowly swept through resonance. While being swept, the 282- and 194-nm sources were alternately chopped to avoid ac Stark shifts on the quadrupole transition from the 194-nm source. Each time the 282-nm source was shut off and the 194-nm radiation readmitted, the absence of 194 nm fluorescence from the ion indicated that it had made the transition to the ${}^2D_{5/2}$ level. The presence of 194-nm fluorescence indicated that the ion had remained in the ${}^2S_{1/2}$ state. The spectrum of the quadrupole transition could be taken by our plotting the probability of our observing 194-nm fluorescence as a function of the tuning of the 282-nm source. Figure 3(a) shows the central part of the spectrum for a single trapped $^{198}\mathrm{Hg}^+$ ion. The carrier is identified by the invariance of its frequency upon variation of V_{ac} and V_R. The resolution achieved was limited by the 282-nm source spectral width; the natural linewidth of the quadrupole transition is about 2 Hz. The sidebands are Doppler-effect–generated FM sidebands due to the harmonic oscillations of the trapped ion at frequencies ν_z and ν_r.[15] They can also be viewed as the vibration modes of the pseudomolecule composed of the ion plus the trap. For optimum laser cooling, the maximum amplitude of the ion's motion for the conditions of this experiment was about 0.18 μm.[15,16]

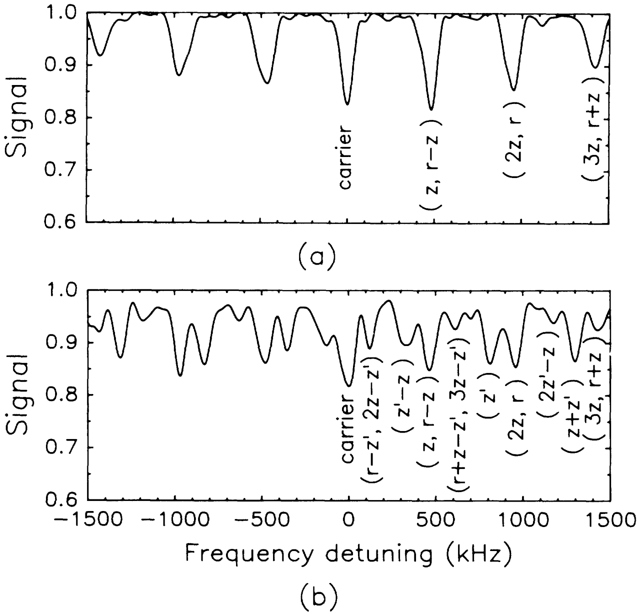

FIG. 3. Absorption spectra of the electric quadrupole transition ${}^2S_{1/2}(m_J=+\frac{1}{2}) \rightarrow {}^2D_{5/2}(m_J=\frac{1}{2})$ for individual $^{198}\mathrm{Hg}^+$ ions. (a) The central part of the spectrum for a single trapped ion. Adjacent to the carrier are Doppler-effect–generated sidebands due to the harmonic motion of the ion in the trap. Trapping conditions were adjusted to satisfy $\nu_r \simeq 2\nu_z \simeq 473$ kHz. The identifications below the lines show the order of the sideband. For example, the identification $(z, r-z)$ denotes absorption at the frequencies $\nu_0+\nu_z$ and $\nu_0+\nu_r-\nu_z$, where $\nu_0 \simeq 1.07\times10^{15}$ Hz is the carrier frequency. (b) The quadrupole spectrum for one of two ions, trapped in the same orientation as in Fig. 2(b). The trapping conditions were the same as for (a). The new lines are due to the stretch vibration mode of the ions at frequency $\nu_z' = \sqrt{3}\nu_z$.

When two ions are confined in the trap and aligned along the z axis, an additional vibrational mode is allowed; this is the stretch mode between the two ions. For small amplitudes of motion, this mode is expected to have a frequency $\nu_z' = \sqrt{3}\nu_z$. By using the double resonance method described above, we have observed the quadrupole spectrum of one of the trapped Hg^+ ions by detecting the fluorescence of only that ion. This spectrum is shown in Fig. 3(b). It is possible to attribute all of the new lines to the internal vibration mode at frequency ν_z' and its combinations with ν_z and ν_r. By fitting the observed spectra (assuming $\nu_r = 2\nu_z$), we derive the frequency of this mode to be $\nu_z' = 1.725(25)\nu_z$, which is in agreement with the theory. It should also be possible to observe this internal vibrational mode by direct excitation with spatially nonuniform electric fields.[18] Finally, by measuring the width of the Doppler envelope of the quadrupole resonance,[15] we determined that the temperature of the ions was less than 8 mK. This gave a ratio of Coulomb potential energy per ion to k_BT greater than 120.

The imaging and spectroscopic techniques described here should prove useful in the study of spatial ordering under various experimental conditions.[1-8] With careful control of the trap parameters, such as static field offsets, it should be possible to distinguish different cluster patterns whose minimum energies differ by small amounts[5,7] and to study the transitions between these patterns for different experimental conditions. Careful control of the balance between heating and cooling,[11] and spectroscopic measurements of the temperature, should allow the study of the transition from ordered to disordered structures. These studies may be aided by observation of the mobility of ions "tagged" by optical pumping or ions of different species in substitution as observed here. In a Paul trap, the amplitude of an ion's micromotion[9] due to the oscillating electric fields is proportional to the distance that the ion is from the center of the trap. Therefore rf heating is expected to be more of a problem for large-number clusters. This problem does not occur for particles trapped in static electric and magnetic fields.[1]

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