Observation of a Phase Transition of Stored Laser-Cooled Ions

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(Received 8 July 1987)

Clouds of two to about fifty simultaneously stored, laser-cooled Mg^+ ions in a Paul trap were observed in two phases, which are clearly distinguishable by their excitation spectra. Transitions between these phases can be induced either by a variation of the power of the laser radiation used to cool the ions or by a change of the size of the radio-frequency voltage applied to the trap. Transitions between a "crystalline" phase and a "gaseous" phase can be repeatedly observed by variation of the appropriate parameters. The two phases and the transitions between them have also been recorded by a photon-counting image system.

PACS numbers: 42.50.Vk, 05.70.Fh, 32.50.+d, 64.70.−p

There is growing interest in the use of particle traps for high-resolution spectroscopy. The ultimate resolution limits which can be achieved in principle are beyond all other known experimental possibilities. It seems quite probable that particle traps will be the basis of future frequency standards.[1,2] The traps most extensively employed to date for ions are the static Penning trap and especially the dynamic Paul trap. Storage in the Penning trap is achieved by use of a strong magnetic field; the Paul trap uses an oscillating electric quadrupole field.[2,3]

In the Paul trap a single ion can, in principle, be kept at rest at a point of vanishing field amplitude in the center of the trap. By cooling of single Mg^+, Ba^+, and Hg^+ ions[4-7] in the millikelvin range, previous experiments have demonstrated that this ideal situation can be realized in very good approximation. In these experiments the kinetic energy of the ions (≈ 1 eV after the loading cycle) was reduced by radiation-pressure cooling. This was done by the excitation of a strong dipole transition with laser light whose frequency was detuned from resonance towards longer wavelengths. In this way a single Ba^+ ion may be localized in a region with dimensions small compared with the optical wavelength of the exciting light.[6] In this so-called Lamb-Dicke regime, the first-order Doppler effect is suppressed and, in addition, the second-order Doppler effect is negligible, so that single laser-cooled ions have been proposed as the basis for frequency standards.[8]

The Lamb-Dicke regime can easily be obtained for microwave transitions. Because of the longer wavelength, even clouds of 10^5 ions can be confined to a space-charge-limited spatial region which is inside the Lamb-Dicke regime.[2] In this connection the question arises of how a cloud of ions behaves when cooled to low temperatures. Crystallization is expected when the Coulomb coupling constant Γ, which corresponds to the ratio of the Coulomb energy to the kinetic energy of a particle, is much bigger than 1.[9] Experiments with laser-cooled Be^+ ions have shown that the observation of such a phase transition in a Penning trap should be possible.[10] The cooling of an ion crystal is predicted by theory to be as efficient as the cooling of independent particles.[11]

To our knowledge, no theoretical prediction exists for a phase transition of an ion cloud in a Paul trap. If the number of simultaneously stored ions is increased, the micro-oscillations become increasingly important as the average distance of the ions from the trap center is increased. It is possible that energy is transferred from the trapping field to the ions. This radio-frequency heating is assumed to cause the temperature of a laser-cooled ion cloud in the Paul trap to be much higher than that of a single laser-cooled ion.[5] Nevertheless, it has been shown that under the influence of strong cooling, a cloud of charged particles will arrange itself in a regular array. Such behavior has been observed by Wuerker, Shelton, and Langmuir,[12] who cooled a cloud of charged aluminum microparticles using a light buffer gas. Subsequent crystallization of the particles to this regular array and subsequent melting of it were achieved by variation of the storage potential and the cooling force.

In this Letter we report the observation of a phase transition of laser-cooled Mg^+ ions in a Paul trap.[13] Details of the apparatus used in this experiment have been reported elsewhere.[14] Mg^+ ions are stored in a Paul trap (diameter of ring electrode 5 mm, end-cap separation 3.54 mm, radio frequency 11 MHz) and excited by coherent ultraviolet (UV) radiation (focus 20×50 μm^2, laser linewidth about 1 MHz) on the transition $3^2S_{1/2} \rightarrow 3^2P_{3/2}$ (wavelength $\lambda = 280$ nm, natural linewidth 43 MHz). During loading of the trap the laser frequency is detuned by -700 MHz to the long-wavelength side from resonance. In this way the ions are cooled in a few seconds from their initial kinetic energy of ≈ 1 eV to the final temperature.

A typical excitation spectrum of a single Mg^+ ion is shown in Fig. 1, curve *b*. It is slightly Doppler and

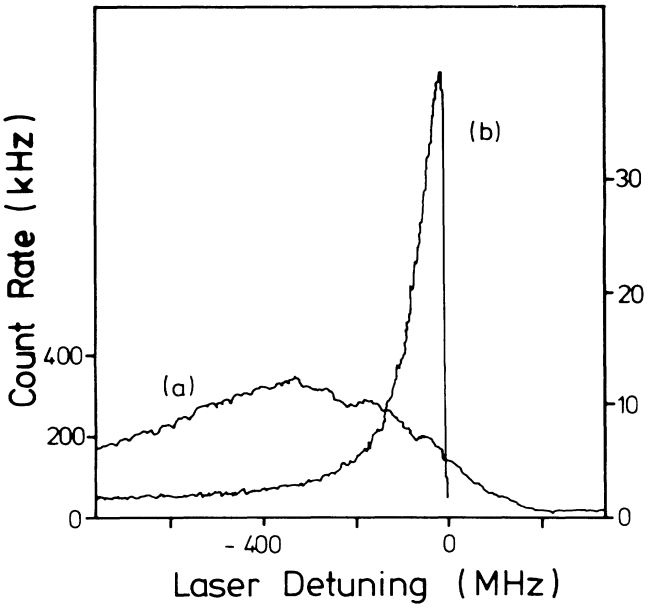

FIG. 1. Excitation spectra of stored laser-cooled ions: Curve *a*, cloud of ≈ 50 ions (left scale); curve *b*, single ion (right scale). The recording time for the spectra was 10 min in each case. The zero of the frequency scale marks the position of the resonance.

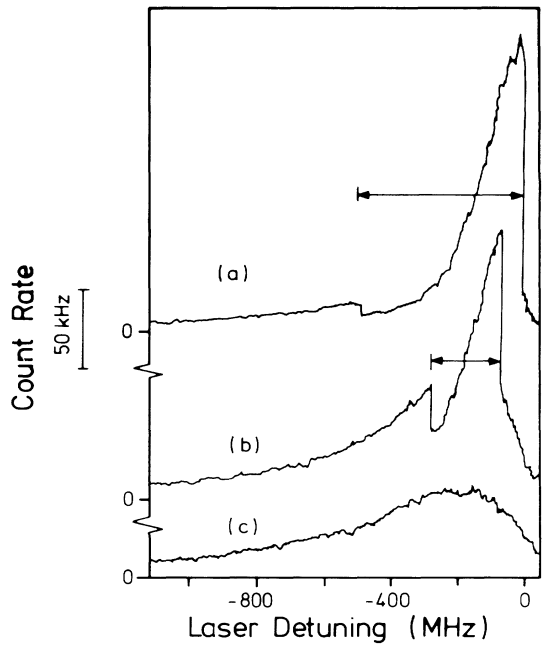

FIG. 2. Dependence of the excitation spectra of about five simultaneously stored ions upon the radio-frequency amplitude U_0 for fixed UV power $P_{UV}=500$ μW: (a) $U_0=360$ V, (b) $U_0=460$ V, and (c) $U_0=570$ V. The laser-frequency region where an ordered structure of the ions is observed is marked by arrows.

power broadened; the fluorescence intensity abruptly vanished on resonance. Then the ion is no longer cooled by the laser light but rather is heated and thus pushed out of the laser focus. The single-ion spectrum is totally different from that of the ion cloud containing ≈ 50 ions shown in Fig. 1, curve *a*. Here the Doppler width is much larger than the natural linewidth. Furthermore, the maximum of the signal occurs red-shifted from resonance. (Absolute frequency calibration of the spectra is achieved by comparison with a simultaneously recorded Doppler-free saturated-absorption signal of iodine vapor.) The signal profile results from frequency-dependent laser cooling on the one hand and radio-frequency heating on the other. The decrease of the fluorescence signal close to resonance occurs since there the oscillating amplitude of the ions becomes larger than the focal region of the laser light. This is due to the smaller laser cooling in this region and, in contrast to the single-ion spectrum, a little signal is also observed for positive detunings.

Under suitable conditions we observed that the spectrum of an ion cloud can abruptly change into a structure with a much smaller linewidth very much resembling a single-ion spectrum. This situation is shown for different experimental conditions in Figs. 2(a)–2(c). They show the excitation spectra of about five simultaneously stored ions. Jumps in the fluorescent signal are observed at certain values of the detuning. These cannot be explained by ion losses since they always show up in the same way when the scanning of the spectrum is repeated. The positions of the jumps strongly depend on the radio-frequency amplitude U_0.

Our observations suggest that the jumps in the spectra are caused by a phase transition of the stored ions between a state of uncorrelated motion (characterized by the cloudlike spectrum), where the radio-frequency heating is important, and an ordered state characterized by a sharper spectral structure. At large storage-field amplitudes no jumps occur and the spectrum resembles that of an ion cloud [Fig. 2(c)]. As was realized by direct observation of the ions by means of a photon-counting image system (model ARGUS 100, from Hamamatsu), the jump observed for large negative detuning when the laser frequency is increased [Fig. 2(b)] indeed corresponds to transition to a crystalline state and the jump at smaller negative detuning to transition from a crystalline state to an ion cloud. The jumps could be observed as well when the laser was tuned in the opposite direction. For direct observation of the crystalline structure, the cathode of the ARGUS system was placed in the picture plane of the lens system, which was attached to one of the end caps of the trap.[14] The crystalline structures observed mostly consisted of planar structures arranged in a plane perpendicular to the symmetry axis of the trap. The planar configuration is favored since no dc voltage was used

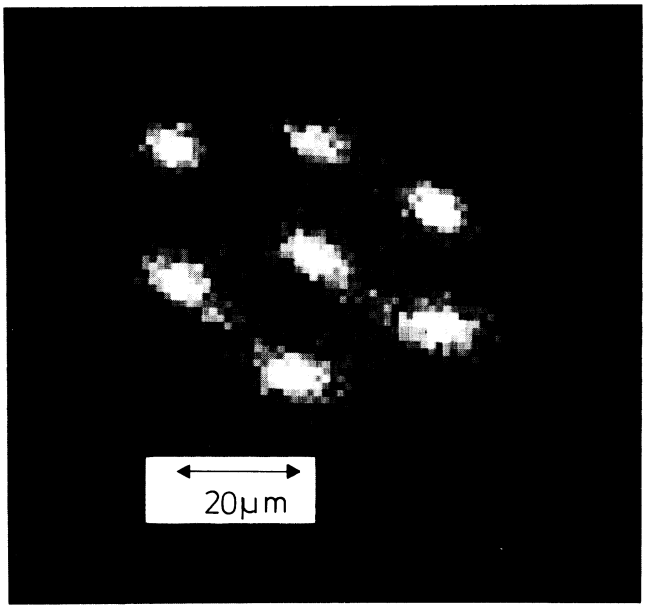

FIG. 3. Crystalline structure of seven $^{24}Mg^{+}$ ions observed with the ARGUS system. For the figure the fluorescent light was averaged for 40 s. The mean distance between the ions is about 23 μm. One picture element corresponds to about 1.3 μm in the plane of the ions. The magnification of the lens system is about 16×. If U_0 is increased, the structure abruptly changes into a diffuse cloud.

for the operation of the trap. Figure 3 gives an example of seven ions. The structure is slightly unsymmetric. This is caused by a small contact potential due to a thin Mg coating on the ring electrode opposite the Mg oven. This potential is also the reason that no rotation of the ion structure is observed. The transitions between the crystalline and the cloud state were observed by the image system and recorded by a video recorder. They occur between subsequent frames of the video, that is, faster than 0.04 s.

In Fig. 4 we show the fluorescence of about five ions as a function of the storage-field amplitude for fixed detuning at −120 MHz. At this particular detuning, the crystalline state shows higher fluorescence than the ion cloud [see Fig. 2(b)]. Transition from the high-fluorescence (crystalline) state to the low-fluorescence (cloudlike) state always takes place at higher radio-frequency amplitudes than transitions in the opposite direction. Such a hysteresis, which was not mentioned by Wuerker, Shelton, and Langmuir,[12] must be expected with laser-cooled ions because the cooling power is strongly dependent upon the motion of the ions as a result of the Doppler shift. At small detuning (approximately half a natural linewidth) in particular, the cooling power rapidly decreases when the kinetic energy increases. The changes of the switching points for different signal transients are caused by UV-power fluctuations. These occasionally occur because of the variations of the efficiency of the UV generation in the external cavity caused by mechanical vibrations.[14]

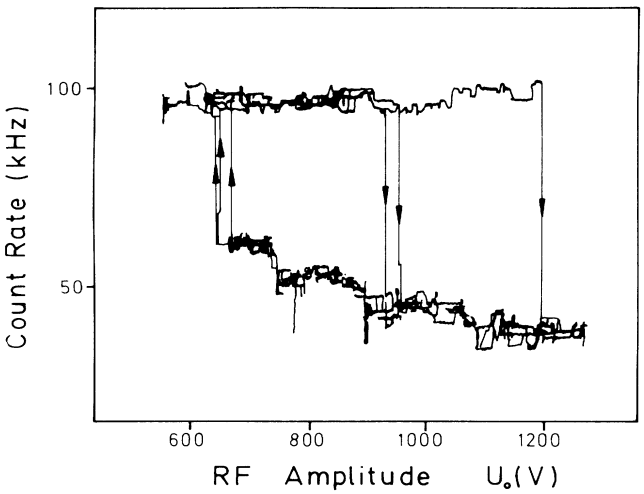

FIG. 4. Observation of the phase transitions as a function of radio-frequency heating. Shown is the dependence of the fluorescence intensity upon the amplitude of the radio-frequency voltage. The loops are scanned clockwise, and the arrows indicate the direction of the jumps. The time constant of the recording system was 0.1 s, and the time for one signal transient was about 10 s.

The dependence of the fluorescence upon the UV

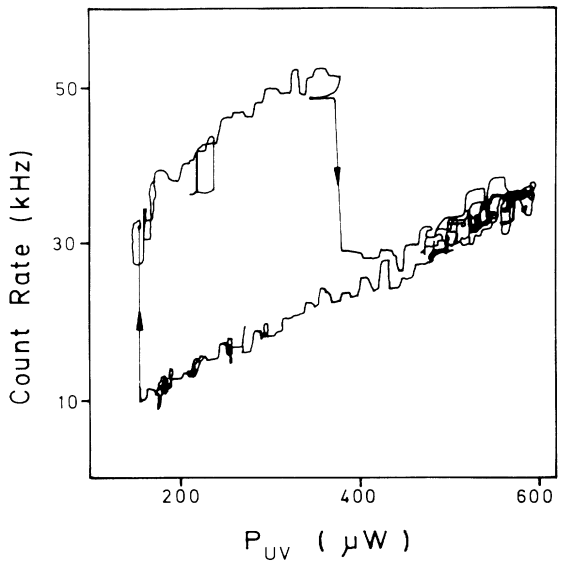

FIG. 5. Observation of the phase transition as a function of laser cooling. Plotted is the dependence of the scattered intensity upon the UV power, while the laser detuning and storage field amplitude were kept fixed at −400 MHz and 460 V, respectively. The time constant of the recording system was 0.1 s, and the time for one signal transient was about 10 s.

power for fixed detuning and storage-field amplitude is shown in Fig. 5. Here the detuning has been chosen so that because of the large Doppler width, the cloudlike state has a bigger fluorescence signal than the crystalline state. When the UV power is less than 154 μW, laser cooling is not sufficient any more and the ions are in the cloudlike state. When the UV power is increased, the ions stay first in the strong-scattering state, but, when the UV power exceeds 375 μW, the ions return to the ordered state with smaller fluorescence, and the hysteresis cycle can be repeated. Again we observe behavior typical of a phase transition (in the present case it resembles optical bistability).[15]

In this paper we have reported the first observation of a phase transition for laser-cooled ions in a Paul trap. The trapped ions represent a very neat model system. It is of considerable interest to investigate how the conditions depend on the stored-ion number. Certain ion configurations can be expected to be more stable than others and therefore will need stronger heating for "evaporation." Furthermore, the vibrational modes of the crystalline structures can be investigated.

The authors would like to thank W. Schleich, R. Blümel, and Y. R. Shen for many discussions.

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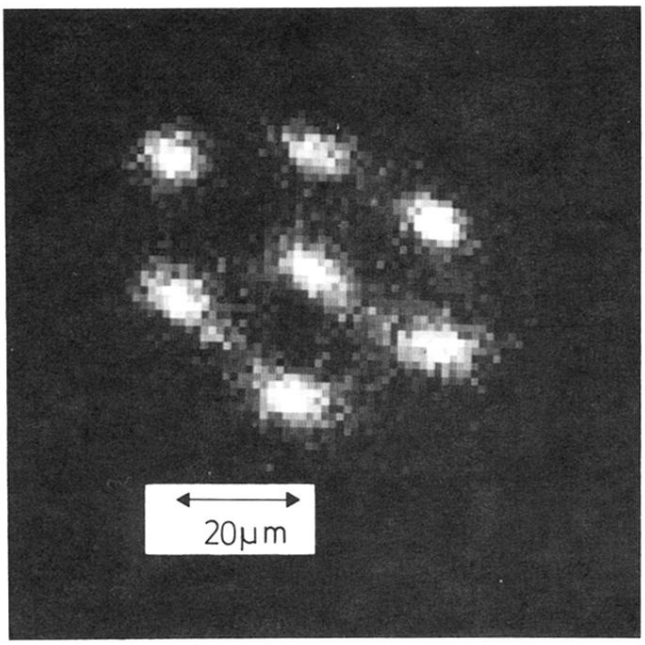

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