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PHASE TRANSITIONS OF STORED LASER-COOLED IONS

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Phase transitions of stored laser-cooled ions

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Single ions in miniature traps can be imaged by using laser light to stimulate fluorescence radiation. At the same time, radiation pressure can be used to bring them nearly to rest. When a small number of ions are trapped, phase transitions can be observed between a chaotic cloud and an ordered crystalline structure, depending on the degree of laser cooling.

THE behaviour of charged particles in an external potential has been discussed frequently in different contexts. Examples include the Thomson 'raisin model'[1] and strong electrolyte solutions, treated by Debye and Hückel[2]. The first quantum treatment of the problem was presented by Wigner[3] who proposed a new phase of the degenerate electron gas, the Wigner crystal.

Experimental studies of charged particles in an external potential reached a new stage after the invention of ion-traps[4,5]. The first observation of crystallized charged particles stored in a Paul-trap was reported for aluminium microparticles (20 μm diameter, 10^5 times the elementary charge) by Wuerker *et al.*[6], when cooling was achieved by a light background gas. Repeated crystallization of the particles to a regular array and its subsequent melting were obtained by variation of the storage potential and the cooling force. Later, experiments were reported[7,8] with Be^+-ions in a Penning-trap[9,10]. The ions were cooled by radiation pressure[11-13] and it was claimed[7,8] that plasma parameters[14,15] (namely the ratio between the potential and kinetic energies of the ions) were high enough for crystallization to be feasible. The observation of the crystallization of Mg^+ ions in a Paul-trap to an ordered structure, its melting and recrystallization (that is, phase transitions) were then reported[16-19]. Later, the vibrational frequencies of small ion-clusters of Hg^+ ions were measured[20].

Pure electron plasmas have also been produced. They bear the exciting possibility that the quantum regime may be reached at low temperatures[21-24]. Recently the problem of ion-crystal formation[25-29] in strongly coupled Coulomb systems[30,31] found additional interest in connection with the new ion storage rings with electron- and laser-cooling[32-34].

The Paul-trap has so far been directed mainly towards using a single ion[35,36] for high-resolution spectroscopy[37-42] and as a frequency standard[43], which could easily surpass the accuracy of modern Cs atomic clocks[44].

Here we concentrate mainly on few-body phase transitions that occur for a small number of ions in a Paul-trap between a chaotic cloud and an ordered crystalline structure. We describe our recent experimental and theoretical work on this subject. In particular, we focus on the following questions: what keeps ion-clouds and crystals stable? What are the parameters controlling the transitions between these two phases, and what is the mechanism?

Description of the trap

Central to our experiment is a radio frequency (r.f.) Paul-trap[4]. This device grew out of the work on quadrupole mass filters[45]. Our Paul-trap (see Fig. 1 and ref. 46 for technical details) is larger than most other traps used in laser experiments. The frequency of the r.f. field, f, had a value of $f = 11$ MHz. For the measurement of the secular and vibrational frequencies of ion crystals, an additional a.c.-voltage can be applied between the ring and the two end-cap electrodes.

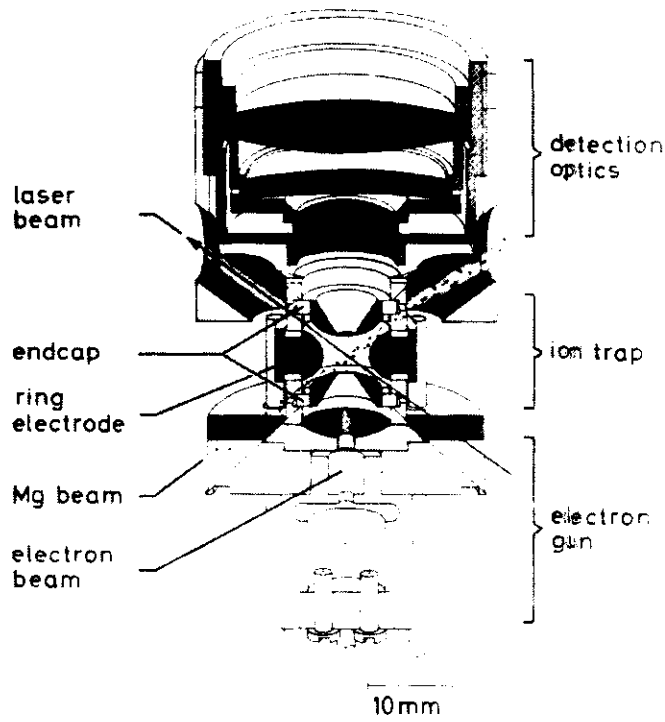

Fig. 1 Sketch of the ion-trap. The fluorescence light is observed through a hole in the upper end-cap. The ionizing electron beam enters through a hole in the lower end-cap. Both holes are covered by a fine molybdenum mesh in order not to distort the trap-potential (for details see ref. 46).

Our trap is mounted inside an ultra-high vacuum chamber and is loaded by a thermal beam of neutral Mg atoms which are ionized close to the trap's centre by an electron beam entering the trap through a small hole in the lower end-cap. The neutral Mg beam and a laser beam are passed through the gaps between cap and ring electrodes. The laser excitation serves two purposes: on one hand it detects the ions by laser-induced fluorescence on the transition $3S_{1/2} \rightarrow 3P_{3/2}$ of Mg^+, and on the other it cools the ions by radiation pressure[11-13]. For this purpose the laser frequency is red-shifted by an amount Δ with respect to the resonance transition (negative detuning). The large size of the trap affords a large solid angle for detecting the fluorescence radiation either with a photo-multiplier or a photon-counting imaging system (Hamamatsu, ARGUS or PIAS). To observe the ions, the cathode of the imaging system was placed in the image plane of the microscope objective attached to the trap (Fig. 1).

Outline of the theory

We now analyse the equations of motion of ions in a Paul-trap. The ions are subjected to essentially four different forces: the

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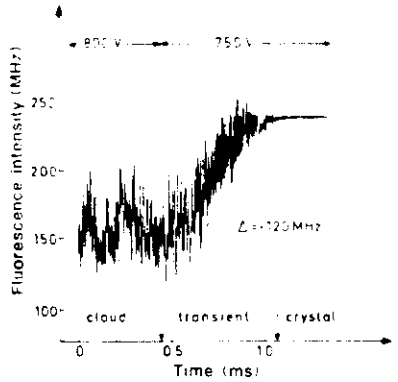

Fig. 4 Time-resolved dynamics of the phase transition of Fig. 3*b* (with identical trap parameters) extracted from three-dimensional molecular dynamics calculations. Horizontal arrows indicate the time periods over which the r.f. voltage, V_0 has the value of 800 V and 750 V respectively.

the ions perform an erratic motion of much larger spatial extension and mean kinetic energy (cloud phase). As a result of the cooling and randomizing effect of the forces $\mathbf{F}_i^{\text{laser}}$ and $\mathbf{F}_i^{\text{rand}}$, the equilibrium energy is independent of the particular choice of initial conditions. For those trap parameters, however, where both phases occur and are stable (bistability), the phase space of initial conditions is divided into two domains representing the basins of attraction for the crystalline phase and the cloud phase, respectively.

Experimental and theoretical results

In this section we discuss our few-ion phase transition experiments in detail and compare them with the results of three-dimensional molecular dynamics simulations of equation (8). Care has been taken in the experiments to ensure that only $^{24}Mg^+$ ions are in the trap. The effects of the other Mg isotopes, $^{25}Mg^+$ and $^{26}Mg^+$, have been studied in detail and will be published elsewhere.

The phase transitions manifest themselves experimentally in abrupt changes in fluorescence intensity when the laser frequency is scanned over critical negative detunings[18,19]. The transitions between the cloud and the crystalline phase show hysteresis (discussed below). Furthermore, the two phases can be observed directly with the help of a highly sensitive imaging system. It is also possible to identify the crystalline structure by an excitation of vibrational modes using an additional r.f. voltage applied to the trap.

The behaviour of the ions in the trap is governed essentially by three parameters: the trapping voltage V_0, the laser detuning Δ and the laser power P. Hysteresis in the fluorescence intensity occurs whenever two of these are kept constant and the third is swept. This is shown for the detuning Δ in Fig. 2 and for the r.f. voltage V_0 in Fig. 3. Such hysteresis behaviour can be expected with laser-cooled ions, because the cooling power of the laser is strongly dependent on the details of the velocity distribution of the ions. The jumps from the cloud to the crystal reproduce well in the experiment, as well as in our simulations. The transitions from the crystal to the cloud state, namely the melting of the crystal, occurs only in the experiment (see Fig. 3*a*), but not in our simulations (see Fig. 3*b*, for example), where the crystals are stable until the critical voltage corresponding to the instability condition of the Mathieu equation is reached. On the other hand, the experimental data show that the location of the transition from crystal to cloud is not well defined, and the melting of the crystal could be caused by fluctuations in the laser intensity. We note that, depending on the detuning, either the crystal or the cloud can have a higher fluorescence intensity.

What is the timescale on which the cloud-crystal phase transitions occur? To study theoretically the dynamics of a particular

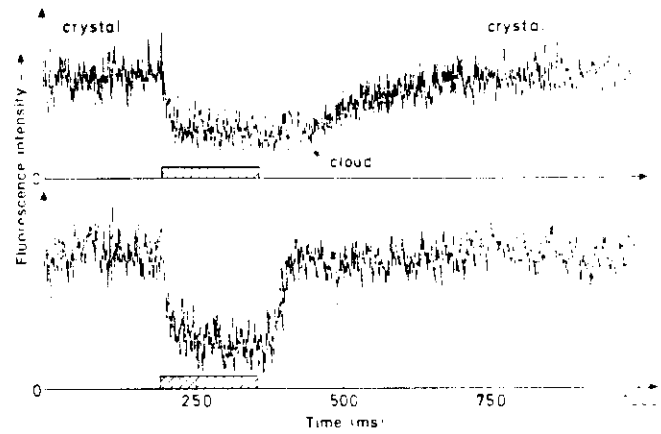

Fig. 5 Time-resolved dynamics of an experimentally obtained phase transition of three ions. $U_0 = 0$, $P = 300\,\mu W$; *a*, $\Delta = -40$ MHz, *b*, $\Delta = -80$ MHz. The r.f. voltage V_0 is suddenly switched from its value 140 V (crystal) to 420 V (cloud), and back to 140 V (crystal). The period over which the r.f. voltage of 420 V is applied is marked by the hatched rectangles. Note that here the number of ions is different from the number in Fig. 4.

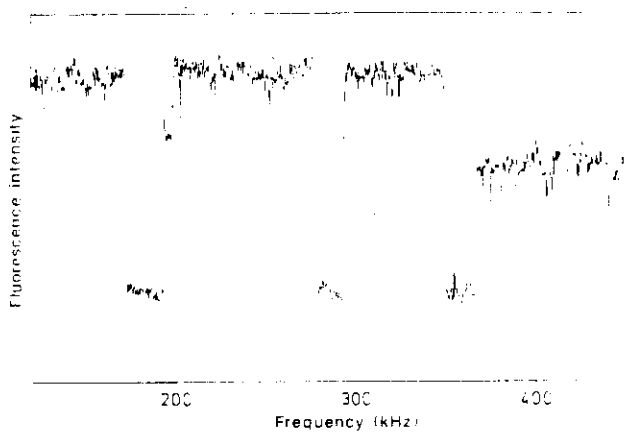

Fig. 6 Spectroscopy of vibrational modes of a two-ion crystal: fluorescence intensity as a function of the frequency of an additionally applied a.c. voltage for $U_0 = 0$, $V_0 = 165$ V, $\Delta = -60$ MHz and $P = 500\,\mu W$. The theoretical values of the secular frequencies are $\omega_x/2\pi = \omega_y/2\pi \approx 170$ kHz and $\omega_z/2\pi \approx 340$ kHz.

jump, such as the one shown in Fig. 3*b*, in Fig. 4 we analyse the fluorescence as a function of time, when we suddenly switch the r.f. field from $V_0 = 800$ V (cloud state) to $V_0 = 750$ V (crystal state). After a transient of about 0.5 ms, the value of the fluorescence typical for this cloud state settles to its crystalline value. Experimental time-resolved phase transitions shown in Fig. 5 demonstrate that the transient time strongly depends on the laser detuning. In contrast to Fig. 4, the results in Fig. 5 represent an average over 60 such switching experiments, whereupon the large intensity fluctuations in the cloud state are washed out. The residual fluctuations in Fig. 5 result mainly from counting statistics.

The experimental results described above represent only a small fraction of the entire body of data accumulated. Ion-crystals, clouds and phase transitions of two to approximately 100 ions have been observed.

The most basic ion-crystal—the two-ion crystal—enjoys two kinds of normal modes: (1) three centre-of-mass oscillations with secular frequencies[37] $\omega_x = \omega_y$ and ω_z, where the distance between the ions is fixed and, (2) vibrational modes, in which

Fig. 7 The large amplitude vibration of a single ion (maximal visible excursion from the trap-centre is 160 μm) for the bistable situation $U_0 = 0$, $V_0 = 410$ V, $\Delta = -100$ MHz and $P = 220$ μW. Increasing the r.f. voltage to $V_0 = 630$ V, the line collapses to a dot. Decreasing V_0 back to its original value, the dot remains stable.

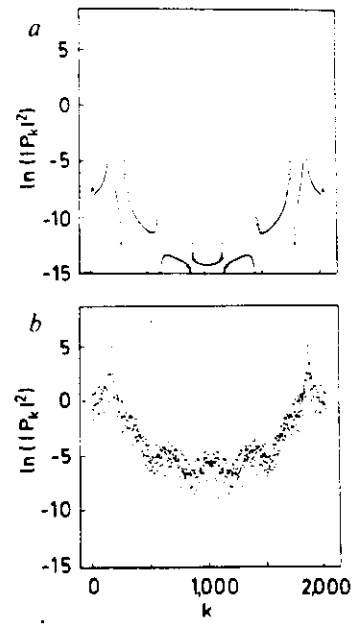

Fig. 8 Theoretical power spectrum of the x coordinate of one of the two ions in a Paul-trap. *a*, Ions started close to the equilibrium position of the corresponding crystalline state. The discrete spikes in the power-spectrum are typical for quasi-periodic motion. *b*, Two ions in the cloud phase: the spectrum is noisy, a characteristic feature of chaotic motion.

the two ions oscillate against each other with an approximate frequency[20,21] of $\sqrt{3}\omega_x$, or $\sqrt{3}\omega_z$, according to the alignment of the crystal, as determined by U_0. When we apply an additional a.c. voltage to the electrodes of the trap (15 mV), we excite the centre-of-mass motion as well as the oscillation between the two ions (Fig. 6). The resonant excitation of one of these modes results in an additional heating and in a reduction in the fluorescence intensity. The dip in the spectrum at ~170 kHz thus corresponds to $\omega_{x,z}$, and the one at ~340 kHz to ω_z. The one in the middle corresponds to the mode in which the ions oscillate against each other. At the end of the recording time, one ion left the trap leaving the fluorescence intensity at half its original value. Figure 6 demonstrates the feasibility of this experimental technique to determine the frequencies of the vibrational modes of ion crystals.

In Fig. 7, a single ion shows a stable large-amplitude vibration orthogonal to the cooling laser beam. According to equation (5), there is only a cooling effect in the **k** direction. Motion orthogonal to this direction is damped only in a minor way giving to laser-focus effects, contact potentials or trap-imperfections that couple the different degrees of freedom. When we increase the r.f. voltage, the amplitude of vibration reflected in the length of the white line (integrated fluorescence signal) in Fig. 7 is reduced until it suddenly collapses into a dot representing the single ion sitting at the centre of the trap. When we now decrease the voltage again to its original value where the large-amplitude vibration was observed, the ion will still be located at the centre of the trap. This means that there is bistability, even for the case of a single ion. Theoretical calculations confirm this observation and will be published elsewhere.

Discussion

The cooling effect of the laser is balanced by a heating mechanism. Heating of the ions by the r.f. field has been known[9] since the early days of r.f. traps. Its understanding, however, remained incomplete.

Does the most elementary of all conceivable one-dimensional models for ions in an ion-trap, namely the one-dimensional version of equation (8), taking into account only $\mathbf{F}^{\text{trap}}$ and the Coulomb interaction force $\mathbf{F}^{\text{Coul}}$, reveal the origin of the r.f. heating mechanism? To answer this question, we have monitored the average energy of two to five ions as a function of time in such a one-dimensional trap. For less than five ions we could not observe any gain in energy over several tens of milliseconds. For five ions, a slow increase in energy was recorded, but it was too small to account for the stability of ion-clouds in the presence of laser cooling. A fast Fourier transform of the positions $x_n(j)$ of the n-th member of the one-dimensional ion-chain taken at time $t = jT$

$$P_k^{(n)} = \frac{1}{\sqrt{N}} \sum_{j=0}^{N-1} x_n(j) \exp\left[-i\frac{2\pi}{N} jk\right];$$

$$k = 0, 1, \ldots, N-1 \qquad (9)$$

($N = 2{,}048$ in the present case) shows a small number of discrete frequencies dominating the spectrum, and thereby qualifies this model as being close to integrable and lacking a heating mechanism.

Two-dimensional ion-traps behave essentially like one-dimensional traps if the motion is restricted to the x–y plane. In the x–z (y–z) plane, however, there is strong heating. To study this observation in more detail, we have calculated the power-spectrum $|P_k|^2$ of the positions of two ions in a two-dimensional trap. We have chosen an initial separation of the ions that was about twice the equilibrium separation of the two-ion crystal, and a set of discrete frequencies appeared, as shown in Fig. 8*a*. In this case, the ions perform a quasi-periodic motion. They are unable to extract energy from the r.f. field and, as a result of the cooling laser, they eventually end up in the crystalline state. Such a power-spectrum characterizes phase space domains that act as basins of attraction for the crystal. But when we choose initial conditions that correspond to typical separations in a cloud state, the spectral power of any one of the two ions is a very noisy, quasi-continuous band which resembles the power spectrum of a turbulent fluid[50] (see Fig. 8*b*).

There is an interesting connection between the r.f. heating in a Paul-trap and the ionization mechanism of Rydberg atoms. Both are subjected to strong time periodic fields. For a given field strength, microwave frequency and small diameter of the Rydberg atom, the motion of the Rydberg electron is regular and no ionization occurs. An increase of the atom's diameter (by increasing the principle quantum number for example) makes chaotic phase space domains accessible. The motion of the Rydberg electron becomes erratic, resulting in a diffusive gain of energy and ionization of the atom. We encounter an analogous situation for ions in a Paul-trap. If we prepare ions in the vicinity of their equilibrium positions with small initial velocities, they are located in regular regions of phase space. No heating occurs and after a while the cooling laser establishes a stable crystalline configuration. If, however, we prepare an ionic array with small initial velocities, but with a diameter which amounts to about four times the typical dimensions of a crystal, this configuration will be located in a chaotic domain of phase space. Strong heating occurs by a cooperative effect of r.f. driving field and Coulomb non-linearities. Furthermore, our calculations show that the heating rate reduces with the diameter of the clouds. As a result, for very large clouds, the heating stops and the ions will still be confined in the trap. This is confirmed by experiments in which, even in the absence of a cooling laser, large clouds of ions can be stored in a Paul-trap over several hours without being heated out of the trap.

The above considerations and observations suggest a simple explanation for the surprisingly sharp phase transitions found in our experiments. Starting with a large cloud, and increasing the laser cooling power, the cloud reduces in diameter. The increased cooling power of the laser is counter-balanced by the larger heating-rate of the smaller cloud. But as soon as the cloud diameter reaches the critical value separating regular and chaotic domains in phase space, the smallest increase in laser power beyond this value will result in a collapse of the cloud into the crystal state. Diffusion models similar to the ones developed for Rydberg atoms[51-53] could be designed to calculate the energy diffusion coefficient, which would allow for an analytical calculation of the steady-state size of ion-clouds and the exact location of the phase-transition points.

Summary and conclusions

In conclusion, we have shown that ions confined in an ion-trap can be found in two modifications: in a cloud phase and in a crystalline phase. Phase transitions characterized by a discontinuous change in the fluorescence intensity can be induced by appropriately adjusting the r.f. voltage, the detuning and the power of the cooling laser. The transitions show hysteresis as a function of these three essential control parameters. With the help of three-dimensional molecular dynamics calculations, we have reproduced qualitatively all the essential features in recent experiments, such as the size of ion clouds and crystals, as well as transitions between these two phases. The dynamics of the phase transitions were explored experimentally and numerically by time-resolved fluorescence intensity recorded for the transients of specific phase transitions. In detailed studies of one-, two- and three-dimensional simulations of ions in a Paul-trap, we have identified the occurrence of deterministic chaos[50] in the cloud state as the predominant source of heating. The mechanism of r.f. heating, therefore, relies only on the deterministic features of the system, rather than on the effect of external randomness, such as noise in the amplitude and frequency of the r.f. trapping field, or collisions with the atoms of the background gas.

The static as well as the dynamic features of laser-cooled ions in a Paul-trap is a very promising subject for the study of few-body non-equilibrium phenomena. With the help of present-day powerful imaging systems, crystals, clouds and phase transitions between them can be studied as a function of particle number.

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