PHASE TRANSITIONS OF STORED LASER-COOLED IONS

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By means of laser induced fluorescence studies of two to about a hundred laser cooled ions stored in a Paul-trap it was demonstrated that the ions occur in two stable modifications: clouds and crystals. This was also verified by means of a photon imaging system. We observe transitions between these two phases at specific values of the laser power, the laser detuning and the rf voltage applied to the trap. Hysteresis is observed as a function of any of the three parameters, keeping the other two fixed. Even a single ion shows bistability and hysteresis. All the experimental results are reproduced qualitatively with the help of three-dimensional molecular dynamics calculations which take photon recoil, laser polarization and saturation of the atomic transition into account. Satisfactory quantitative agreement was obtained whenever it was attempted. For a few representative cases, the dynamics of the phase transition is presented both experimentally and theoretically. A model for the \( rf \) heating mechanism of the ions in the trap is presented.

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2. Introduction

Single ions at rest and unperturbed by its environment, once only a physicists dream, has now been realized by the combination of an electromagnetic Paul-trap and laser technology. Laser-cooling allows to bring the ion practically to rest and to reach the Dicke-regime with disappearing first and second order Doppler effects. Since also the interaction time with the laser beam is practically infinite, the set-up does not impose any limitation to the resolution [1, 2]. Therefore, laser-cooled stored ions bear the potential for a frequency standard able to surpass the accuracy of modern Cs atomic clocks. This, however, is not the only field where spectroscopy of stored ions is useful. In the arena of research for manifestations of nonclassical effects of the radiation field, single ion experiments have provided a source of anti-bunched light of sub-Poissonian photon statistics [3] and have provided insight into the nature of quantum jumps [4, 5]. In the present paper we will show that the Paul-trap is also a good instrument to gain insight into the physics of “few-body phase transitions”.

The behavior of charged particles in an external potential has been discussed many...

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times in different context in the past ranging from Thomson’s “rainin model” to the Wigner crystal (for a review see Ref. [5]). The experimental studies reached a new stage after the ion traps had been invented [7, 8]. The first observation of crystallized charged particles stored in a Paul-trap were reported for aluminium micro-particles (≈ 20 μm diameter, 10⁵ times the elementary charge) by Wuerker et al. [9]. The particles were cooled by a light background-gas. Repeated crystallization of the particles to a regular array and melting of it were obtained by variation of the storage potential and the pressure of the background gas. The first observation of the crystallization of ions in a Paul-trap to an ordered structure (see Fig. 1), its melting and recrystallization, i.e. a phase transition, was then reported by Diedrich et al. [10–13]. Later small ion-clusters of Hg⁴⁺-ions were investigated by Wenzel et al. [14]. The quantitative interpretation of the phase transitions observed by Diedrich et al. [10–13] was given by Blümel et al. [15]; see also Hoffrage et al. [16] for comparison. Later also the formation of shell-structures of a large number of ions stored in a static Penning trap was observed by Gilbert et al. [17].

In the present article we are going to concentrate mainly on the collective aspects of ions in a Paul-trap and review and highlight our recent experimental and theoretical work on this subject [10–13, 15]. In particular we will focus on the following questions: What keeps ion-clouds and crystals stable? What are the parameters controlling the fast transitions between these two phases and what is the mechanism? What is the heating mechanism of the ions in the trap by the rf field?

2. Description of the trap

The heart of our experiment is a radio frequency (rf) Paul-trap [7, 8] which is larger than most of the other traps used in laser-experiments (see Ref. [3] for technical details). The frequency of the rf field, \( f \), had a value of \( f = 11 \) MHz.

Our trap which is mounted inside an ultra-high vacuum chamber is loaded by means of a thermal beam of neutral Mg-atoms, which are ionized close to the trap’s centre by an electron beam and enter the trap through the gap between cap- and ring-electrodes. The laser excitation serves two purposes: on one hand it is used for the detection of the ions via laser-induced resonance on the transition \( 3S_{1/2} \rightarrow 3P_{1/2} \) of Mg⁰ and on the other hand it cools the ions by radiation pressure [18, 19]. For this purpose the laser frequency is red-shifted by an amount \( \Delta \) with respect to the resonance transition (negative detuning). The large size of the trap affords a large solid angle for detecting the fluorescence radiation. This was done either by a photo-multiplier, or by means of a photon-counting image system (Hamamatsu, ARGUS or PIAS). For the observation of the ions, the cathode of the image system was placed in the image plane of the microscope objective attached to the trap.

3. Outline of the theory

In this section, we analyze the equations of motion of ions in a Paul-trap. The ions are subjected to essentially four different forces: The force \( \vec{F}_{\text{ion}} \), arising from the dynamical rf trapping field, the Coulomb interaction between the ions \( \vec{F}_{\text{Coul}} \), the laser cooling force \( \vec{F}_{\text{cool}} \), and a random force \( \vec{F}_{\text{rand}} \), arising from the spontaneously emitted photons.

We now discuss these forces in detail. For the case of \( n \) particles, the resulting trapping force reads

\[
\vec{F}_{\text{trap}} = -2e \left( U_0 + V_0 \cos(2\pi f t) \right) \frac{\vec{r}_i - \vec{r}_j}{r_i^2 + 2a^2} \left( \vec{r}_i - 3\vec{r}_j \right),
\]

where \( r_i \) denotes the \( i \)-th ion's position, and \( \vec{r}_j \) is a unit vector in z-direction. The DC and AC part of the trapping voltage are \( U_0 \) and \( V_0 \), respectively, and \( f \) is the driving frequency. The trap radius and the separation of the end-caps are \( r_0 \) and \( 2a \), respectively. The Coulomb interaction between the particles results in the force

\[
\vec{F}_{\text{Coul}} = \frac{e^2}{4\pi \varepsilon_0} \sum_{n \neq i} \frac{\vec{r}_i - \vec{r}_n}{|\vec{r}_i - \vec{r}_n|^3}.
\]

In the process of laser cooling [18, 19], every scattered photon changes the momentum of an ion on the average by an amount \( h\vec{k} \). An ion at position \( \vec{r} \) and velocity \( \vec{v} \) scatters photons at a rate \( R \) [20], with

\[
R(\vec{r}; \vec{v}) = \frac{2\pi I_s}{1 + 2S(\vec{r}; \vec{v})}.
\]

Here

\[
S(\vec{r}; \vec{v}) = \frac{I_0}{I_s} \left( \frac{\gamma/2}{\pi} \right)^2 \left( d - \frac{k \cdot v}{2\pi} \right)^2 + (\gamma/2)^2
\]

is the saturation parameter and \( \Delta \) and \( \gamma (= 42 \) MHz) denote the laser detuning and the natural line width (FWHM) of the \( 3S_{1/2} \rightarrow 3P_{1/2} \) transition of \( ^{24}\text{Mg}^0 \), respectively. For this particular transition, the theoretical value of the saturation intensity, \( I_s = \frac{ek^2 \gamma}{\pi} \), is 1.53 W/cm². The beam intensity

\[
I(\vec{r}; \vec{v}) = \frac{2P}{\pi D_r D_w} \exp \left[ -2(\vec{r} \cdot \vec{D}_r)^2 \right] \exp \left[ -2(\vec{v} \cdot \vec{D}_w)^2 \right]
\]

is assumed to have a Gaussian profile with beam waists \( D_r \) and \( D_w \) in polarization direction, \( \vec{v} \), and \( \vec{D}_r \) and \( \vec{D}_w \) respectively. Here \( \vec{D}_r \) and \( \vec{D}_w \) are unit vectors and \( P \) is the laser power. In the present experiments as well as in the theoretical calculations \( D_r \approx 50 \) μm and \( D_w \approx 20 \) μm. The ensemble average of the resulting laser cooling force acting on the \( n \)-th ion is thus

\[
\langle \vec{F}_{\text{cool}} \rangle = \hbar \vec{\mathbf{k}} R(\vec{r}; \vec{v}) \langle \vec{v} \rangle.
\]
The number, $N(t)$, of spontaneously emitted photons from the $i$-th ion during the $i$-th cycle of period $T = 1/f$ is given by

$$N(t) = \frac{\bar{N}}{\langle N \rangle} \int_{t-1/T}^{t} \langle \hat{R}(\hat{t}) \rangle \, d\hat{t}. \tag{7}$$

In our simulations, we emit these $N_i$ spontaneous photons, one by one, at the end of each rf cycle, rather than emitting them, one by one, according to the appropriate photon-statistics during the cycle. The direction $\theta_i$ of the $i$-th emitted photon is chosen at random, weighted by a cos$^2$-distribution with respect to the laser polarization axis. This way, only the linearly polarized spontaneously emitted photons have been simulated in our calculations. The circularly polarized ones, occurring less frequently with a sin$^2$-distribution, have been neglected. The random force, $F_{\text{ran}}(\theta)$, is thus

$$F_{\text{ran}}(\theta) = \hbar \omega \sum_{\theta_i} \frac{N_i(t)}{\langle N \rangle} \sum_{\beta} \delta_{\theta_i} \beta. \tag{8}$$

Altogether, the motion of the $i$-th particle is governed by the sum of the above forces, that is,

$$\dot{\vec{r}}_{i,t+1} = \vec{F}_{\text{las}}(\theta) + F_{\text{ran}}(\theta). \tag{9}$$

Given appropriate initial conditions, the equations of motion (9) are integrated forward in time by a standard Runge-Kutta procedure to obtain the trajectories of the $n$-ion system. These trajectories provide all the information necessary to find the quantities of interest, such as the occupation probability density of the $i$-th ion in phase space (i.e., the probability density of finding the $i$-th ion at a given position and with a given velocity), the (total) kinetic energy and the fluorescence intensity. In order to eliminate transient effects arising from initial conditions, we integrate the equations until the total kinetic energy has reached a steady state.

In the experimental situation, the imaging system is focused on the $z = 0$-plane. Therefore, in order to allow for a comparison between the theory and the experimental results, we project the 6-dimensional phase space to the $z = 0$-plane. That is, only the $x_i$ and the $y_i$-components of the $i$-th particle are recorded as a dot in the $x$-$y$-plane at the end of each rf cycle (see insets of Fig. 3). In one of the figures, we have also plotted the motion of the ions in the $x$-$z$-plane. Usually, these positions are recorded over $\approx 2000$ cycles. We also calculate the total fluorescence, $\tilde{F}$, from the $n$ ions during $n$ rf cycles via

$$\tilde{F} = \frac{1}{n} \sum_{i=1}^{n} \sum_{t-1/T}^{t} N(t), \tag{10}$$

where $N(t)$ is given by Eq. (7).

Depending on external parameters, such as the laser power $P$, the detuning $\Delta$ and the rf voltage $V_0$, the solutions of Eq. (9) can be divided into two classes: in the first the ions perform an erratic motion of much larger spatial extension and mean kinetic energy (cloud

phase). In the mono-stable situation (crystalline phase), the equilibrium energy is independent of the particular choice of initial conditions which is due to the cooling and randomizing effect of the forces $F_{\text{las}}$ and $F_{\text{ran}}$. For these trap parameters, however, where both phases occur and are stable (bistability), the phase space of initial conditions is divided into two domains forming the basins of attraction for the crystalline phase, and the cloud phase, respectively.

We conclude this section by briefly comparing our theoretical approach to tackle the few body phase transition problem with a method developed in Ref. [15]. Here, Hoffmang et al. confine themselves to the case of a two-ion crystal. They investigate the radial and z-component of the separation vector, $\vec{r} = \vec{r}_1 - \vec{r}_2$, of the two ions. A spatially isotropic damping in the direction of and proportional to the velocity $\vec{v} = \vec{r}$ damps out the azimuthal, i.e. the $\phi$-motion. Thus Hoffmang et al. treat a purely two-dimensional problem. Unfortunately this method is restricted to the two-ion case. In contrast, our method is able to treat any number of ions. Moreover, the elimination of the $\phi$-component of $\vec{r}$ relies on the isotropic damping, whereas the real laser-cooling force (Eqs. (3)-(6)) is anisotropic and depends nonlinearly on the velocities $v_1$ and $v_2$ of both particles. In addition, the presence of noise caused by spontaneously emitted photons produces $\phi$-components of angular momentum (compare the "crystal" inset of Fig. 3). As a result, the positions of the ions are smeared out to the shape of crescent moons. Thus noise makes the reduction of the three-dimensional problem to the two-dimensional problem questionable. In contrast, the method outlined in this section, is a microscopic description to the many-particle situation incorporating spatially anisotropic, velocity-dependent laser-cooling as well as a realistic modelling of the spontaneous emission noise (see Eqs. (7) and (8)). For a more detailed discussion on laser-cooling see Ref. [21].

3. Experimental and theoretical results

A manifestation of the existence of phase-transitions in a Paul-trap is provided by striking jumps in the fluorescence intensity (photon counts per second) of the ions as a function of the detuning $\Delta$ (see Fig. 2, where the discontinuities in the spectrum are indicated by vertical arrows). The jumps occur between two types of spectra: a broad spectrum, similar to the spectrum completed with the dashed line in Fig. 2, and a narrow spectrum, similar to the fluorescence spectrum of a single cooled ion. When we interpret the broad spectrum as a fingerprint of an ion-cloud and the narrow spectrum as being characteristic for an ordered many ion situation with a "single ion signature", these jumps clearly indicate a transition from a state of erratic motion to a situation in which the ions arrange themselves in regular structures similar to the Wigner-crystal. The regime of detunings in which such crystals exist is depicted in Fig. 2 by the horizontal arrow. The conjecture of the existence of these two phases has been verified by direct observation of crystals (see Fig. 1) and clouds with the help of a highly sensitive imaging system, by analyzing the ion-trajectories produced by our calculations (see insets of Fig. 3), or by exploring the different response of crystals and clouds to an additional rf voltage [15].

The experimental as well as the theoretical excitation spectra, and the jumps in them
expected with laser-cooled ions because the cooling power of the laser is strongly dependent upon the details of the velocity distribution of the ions (see e.g. equation (4)).

The behavior of the ions in the trap is governed by essentially three parameters: the trapping voltage $V_0$, the laser detuning $\Delta$ and the laser power $P$. Hysteresis-loops similar to the ones for $\Delta$ appear also whenever two of these are kept constant and the third is swept. This is shown for the case of the laser power, $P$, in Fig. 4. The experimentally observed jumps from the cloud to the crystal are well reproduced in our simulations. The transitions from the crystal to the cloud state, i.e., the melting of the crystal occur only in the experiment. For the cases studied (up to five ions), our simulations show that in the absence of noise, the crystals cannot be melted by an adiabatic change of any of the three control parameters. On the other hand, the experimental data show that the location of the transition from crystal to cloud scatters over a wide range and might be caused by fluctuations in the laser intensity, spontaneous emission noise or contact potentials.

![Diagram](image1)

**Fig. 2.** Experimental excitation spectrum of five ions as a function of the laser detuning $\Delta$, with $V_0 = 0$. The vertical arrows indicate the detunings where phase-transitions from crystal to crystal ($\Delta \approx -300$ MHz) and from cloud to cloud ($\Delta \approx -100$ MHz) occur. The horizontal arrow shows the range of detunings in which a stable five-ion crystal is observed. The spectrum was scanned from left to right.

![Diagram](image2)

**Fig. 3.** Theoretical excitation spectrum of five ions as a function of the laser detuning $\Delta$, with $V_0 = 700$ V and $P = 500$ \(\mu\)W. The solid arrows on the fluorescence curves indicate the scanning direction. Two phase-transitions (and bistability) are apparent. The insets demonstrate directly the existence of a transition from a cloud phase to a crystalline state. They are the results of three-dimensional molecular dynamics calculations for detunings slightly before and shortly after the jumps at $\Delta = -300$ MHz. The axes of the insets are in units of \(\mu\)m.

![Diagram](image3)

**Fig. 4.** a) Experimental hysteresis loop in the fluorescence intensity of five ions as a function of the laser power ($V_0 = 0$) (for details see Ref. [12]). b) Theoretical hysteresis in the fluorescence intensity of five ions as a function of the laser power ($V_0 = 0$).
It is instructive to compare this observation to the results of Ref. [16]. Here, the authors suggest that the melting of the crystal is an order-chaos transition, appearing "when a bifurcation introduces a third frequency at a critical value of a control parameter". However, as the authors admit, "chaos cannot occur via this route" unless noise is present "to displace the ions from equilibrium". On the other hand, Fig. 3 of Ref. [16] shows that Hofstalge et al. cannot produce a transition from the cloud to the crystalline phase in their simulations. In contrast, our calculations do contain this transition with the transition point being located approximately at the experimental value [15].

What is the time scale on which the cloud-crystal phase transitions occur? To study the dynamics of a particular jump, the rf voltage $V_0$ corresponding to a five-iron crystal (140 V) was suddenly switched to 410 V corresponding to the cloud phase and the fluorescence was monitored as a function of time. Fig. 5 shows that the cooling from the cloud to the crystal is a very sensitive function of the detuning. The cooling effect of the laser is balanced by a heating mechanism. This heating mechanism of the ions by the rf field is known [22] since the early days of rf traps. The understanding of the heating mechanism, however, remained incomplete.

In order to demonstrate the occurrence of rf heating, we have loaded approximately
100 ions into the trap. At time \( t = 0 \) the ions form a stable cloud, in equilibrium with the cooling laser. At \( t \approx 120 \) ms the cooling laser is switched off. After 300 ms the laser is turned on again. The fluorescence data of 60 such experiments have been averaged and the result is shown in Fig. 6. The initial low fluorescence at 500 ms, shortly after the laser was turned on again, indicates that the ions have been heated. The cloud is cooled down again to its initial temperature during the time interval 420 ms < \( t < 800 \) ms. Since the heating of the cloud occurs during the dark phase of this experiment, it cannot be due to the random forces caused by the spontaneously emitted photons. Since we estimate collisions with the background gas to occur at a negligible rate, the heating seems to be entirely due to the deterministic forces exerted by the Paul-trap.

4. **RF-heating mechanism in the Paul-trap**

The RF-heating mechanism is concealed in the properties of the equations of motion (9) and major results concerning this mechanism have already been presented in Refs. [15] and [21]. Here we attempt a more detailed and a more quantitative treatment and show new results, which support the general ideas outlined in Ref. [15]. In order to unravel the origin of RF-heating, one might think of investigating simpler versions of (9), e.g., a one-dimensional string of ions subjected to nothing but the RF trap force and the mutual Coulomb repulsion. Whether the simplified one-dimensional model of the ion trap reveals the origin of the RF-heating mechanism, is answered by the following numerical experiment: We integrated the x-component of (9) and monitored the average kinetic energy.
of two to five ions as a function of time. 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, also too small to account for the stability of ion-clouds in the presence of laser cooling. A Fast Fourier Transform of the positions $x_j(t)$ of the $n$-th member of the one-dimensional ion-chain taken at discrete times $t_j = jT$

$$P_k^N = \frac{1}{\sqrt{N}} \sum_{j=0}^{N-1} x_j(t) \exp \left[ -\frac{2\pi i}{N} jk \right]; \quad k = 0, 1, ..., N-1$$

$(N = 2048$ in the present case) shows a small number of discrete frequencies dominating the spectrum and thus qualifying this model as being close to integrable and lacking a heating mechanism. Although the one-dimensional chain of particles cannot be used to explain the heating mechanism in a Paul-trap, it is interesting in its own right and closely related to the integrable chain of particles interacting by two-body harmonic and 1/|x|^2-potentials (Calogero-Moser system).

Two-dimensional ion-traps behave essentially like one-dimensional traps if the motion is restricted to the $x$-$y$-plane. In the $x$-$z$-$(y=0)$ plane, however, strong heating occurs. In order to study this observation in more detail, we have calculated the power spectrum $P_k^N$ of the positions of two ions. As already shown in Ref. [21], there is always a small phase space region around the crystalline solution, in which no heating occurs. The power spectrum is discrete in this case, typical for quasi-periodic motion. In the quasi periodic phase, the ions are unable to extract energy from the rf field and if a cooling laser is present, they eventually end up in the crystalline state. Such a power-spectrum characterizes phase space domains which act as basins of attraction for the crystal. However, when we choose initial conditions, which correspond to typical separations in a cloud state, the spectral power of any one of the two ions shows continuous bands in frequency. This provides evidence for the occurrence of deterministic chaos [23] in the cloud phase.

For a quantitative investigation of chaotic rf heating, we calculated the work done by the rf field per unit time (angular brackets denoting ensemble averages)

$$\kappa = \langle f \sum_{i=1}^{n} \oint_{\text{paths}} \vec{F}_{\text{rad}} \cdot d\vec{u} \rangle$$

$$= R \sum_{r=1}^{\pi} \oint_{\text{paths}} \vec{F}_{\text{rad}} \cdot d\vec{u}$$

(12)

Fig. 7. Average heating rate $\kappa$ of five ions in a Paul-trap versus the rms radius of the ion configuration. The insets show the power spectrum and the corresponding stroboscopic Poincaré sections in the $x$-$z$ plane of relative separation for two ions in three characteristic domains: the quasi periodic regime, the chaotic regime and the Mathieu regime. All length scales are in units of $\mu m$.


The heating rate $\kappa$ of five ions in a Paul-trap is displayed in Fig. 15. For zero laser power and large $r$, we do not observe any net heating of the ions. This is confirmed by our 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 ions are far apart from each other, the Coulomb force is small and on short time scales the ions behave essentially like independent single stored ions. For this reason, we called this part of the heating diagram the "Mathieu regime". Turning on a small laser power, the rms radius $\sigma$ reduces drastically, but comes to a halt at about $14 \mu m$ where chaotic rf heating sets in and balances the cooling power of the laser. Increasing the laser power results in an even smaller cloud. The smaller cloud produces more chaotic rf heating as seen clearly by the negative slope of the heating curve (Fig. 7) in the range $8 \mu m < r < 14 \mu m$.

In the range $4 \mu m < r < 8 \mu m$ there is still chaotic heating but the slope of the heating curve is positive. As a consequence of the resulting triangular shape of the heating curve, at about $P = 150 \mu W$, corresponding to $r = 8 \mu m$, the chaotic heating power can no longer balance the cooling power of the laser and the cloud collapses into the "crystalline" state located at $r = 3.8 \mu m$. A "phase transition" has occurred. Since the rf heating power depends on $a$ and $g$ and the laser cooling power on $P$ and $\varphi$, the occurrence of the cloud to crystal transition is a rather complicated function of $U_0$, $\varphi$, $P$ and $\varphi$. Due to this collapse of the cloud state, the behavior of the heating rate in the range $3.8 \mu m < r < 8 \mu m$ cannot be studied by equilibrating laser cooling and rf heating. In this case we start out from the crystal state and slightly displace the ions to explore the "vicinity" of the crystal. We observe no heating for $3.8 \mu m < r < 4 \mu m$, but quasi periodic motion, and thus dubbed this regime the "quasi periodic" regime. We call the upper edge of the quasi periodic regime ($r \approx 4 \mu m$) the "chaos threshold". An initial condition beyond the chaos threshold,
i.e., satisfying \( r > 4 \mu m \), leads to heating and expansion of the ion configuration and numerical data relevant for the shape of the heating curve can be taken during this "explosion phase". The laser power \( P \) is set to zero for this type of experiment. We conjecture that, apart from the trivial case of a single stored ion, the heating curve is "universal", i.e., its qualitative shape, including the existence of the chaotic regime, does not depend on the number of simultaneously trapped ions and, as already mentioned in the introduction, even applies to systems as remotely connected as, e.g., Rydberg atoms in strong electro-magnetic fields.

For the quasi periodic, the chaotic and the Mathieu-regime, respectively, we display the corresponding type of power spectrum as the insets above the abscissa of Fig. 7. The data were actually taken for the case of two ions, but would not look much different in the five ion case. We obtain a discrete spectrum in the quasi periodic regime and a complicated noisy spectrum in the chaotic regime. The spectrum in the Mathieu regime is again quite simple and dominated by the secular motion frequency. We also show stroboscopic pictures of the locations of the ions in the \( x-y \) plane of the trap characterizing the three regions (insets below the abscissa of Fig. 7).

As a function of increasing \( rf \) voltage, the chaos threshold moves towards the radius of the crystalline configuration. However, before the chaos threshold reaches the crystal radius, the equations of motion (9) become unstable in 2 direction, indicating that in this particular situation the particles would fall out of the trap. In order to achieve proper melting without loosing particles, the crystal radius has to be enlarged artificially by noise so that the size of the distorted crystal overlaps with the region of chaotic heating. Proper melting without the assistance of noise should be possible if we start in a quasi periodic state typical in the bottom left inset of Fig. 7. Such configurations have a larger radius to begin with and the chaos threshold could be reached before the single particle Mathieu-instability sets in.

An interesting numerical experiment concerned replacing the 1/4 Coulomb repulsion between ions by a screened Coulomb (Yukawa) potential \( e^{-Q/r} \), where \( Q \) was set to 2 \( \mu m \). This replacement did not change the cloud \( \rightarrow \) crystal transition points and we conclude that the heating properties of an ion cloud do not depend sensitively on the long-range properties of the ion-ion potential. Heating, i.e., energy gain, seems to originate from very close ion-ion collisions.

This observation offers another possibility for future research: the investigation of the heating properties of a hard spheres gas and its comparison to the heating properties of the more conventional Coulomb gas confined in a trap.

We conclude this section by briefly comparing the \( rf \) heating mechanism in a Paul-trap with the ionization mechanism of Rydberg atoms in strong microwave fields (compare also Ref. [15]). In both cases heating, i.e., absorption of energy from the driving field, heavily relies on the occurrence of chaotic motion. But in contrast to the situation in ion traps, a proper order \( \rightarrow \) chaos transition marks the onset of strong ionization in the case of Rydberg atoms. Diffusion models similar to the ones developed for Rydberg atoms [15, 21] should be calculated in order to obtain the energy diffusion coefficient, which allows for an analytical calculation of the steady-state size of ion-clouds and the exact location of the cloud \( \rightarrow \) crystal (chaos \( \rightarrow \) order) phase-transition points.

5. 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 \( rf \) 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 was explored experimentally as well as numerically via time resolved fluorescence spectra recorded for the transients of specific phase transitions. In detailed studies of three-dimensional simulations of ions in a Paul-trap, we have identified the occurrence of deterministic chaos [23] in the cloud state as the predominant source of heating. The mechanism of \( rf \)-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 \( rf \) trapping field or collisions with the atoms of the background gas.

While chaotic heating is the dominant source of energy diffusion in the many-particle situation, heating by spontaneously emitted photons might explain the recently discovered bistability of a single ion in a Paul-trap: Beside a situation in which the ion is nearly at rest, a stable vibration of large amplitude in the direction orthogonal to the cooling laser has been found [15].

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 in detail.

REFERENCES

MONOLAYER ADSORPTION OF NONSPHERICAL MOLECULES ON SOLID SURFACES. THE MEAN FIELD TREATMENT OF ORIENTATIONAL TRANSITION IN COMMENSURATE FILM OF NITROGEN ADSORBED ON GRAPHITE*

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Orientational transition in commensurate two-dimensional solid phase of nitrogen adsorbed on graphite is considered. The effects due to anisotropic nitrogen-nitrogen interactions and periodicity of nitrogen-graphite interaction potential on the orientational ordering are discussed. Moreover, the influence of the third-order corrections to the nitrogen-nitrogen interactions, as well as the effects of next-nearest neighbor interactions in adsorbed film on the location of orientational transition and stability of ordered structure are considered. It is shown that both in-plane and out-of-plane effects must be taken into account to obtain agreement between theoretical predictions and experimental data.

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1. Introduction

Experimental and computer simulation studies of adsorption of multiatomic molecules on solid surface [1–11] showed that orientational effects in adsorbed layers may be of great importance. The first evidence of orientational transitions in monolayers came from the heat capacity measurements for nitrogen adsorbed on graphite [1]. It was observed that at low temperatures the heat capacity versus temperature curves exhibited maxima that were attributed to orientational transition. Further studies [2, 4, 7] confirmed that prediction and led to the discovery of several orientationally ordered structures in monolayer films of nitrogen on graphite. Stability regions of various ordered states were found to depend on the density of adsorbed film and temperature. At low and moderate densities, corresponding to the existence commensurate two-dimensional solid phase, the transition from orientationally ordered to disordered state was found to occur at the temperature about 27 K [2, 4]. Structure of the corresponding ordered state is $\sqrt{3} \times \sqrt{3}$

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