Precise experimental investigation of eigenmodes in a planar ion crystal

H. Kaufmann,1 S. Ulm,1 G. Jacob,1 U. Poschinger,1 H. Landa,2 A. Retzker,3 M.B. Plenio,4 and F. Schmidt-Kaler1

1QUANTUM, Institut für Physik, Universität Mainz, Staudingerweg 7, 55128 Mainz, Germany
2Raymond and Beverly Sackler School of Physics and Astronomy, Tel-Aviv University, Tel Aviv, Israel
3Racah Institute of Physics, The Hebrew University of Jerusalem, Jerusalem 91904, Great Ram, Israel
4Institut für Theoretische Physik, and Center for Integrated Quantum Sciences and Technology, Universität Ulm, Albert-Einstein-Allee 11, 89069 Ulm, Germany

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The accurate characterization of eigenmodes and eigenfrequencies of two-dimensional ion crystals provides the foundation for the use of such structures for quantum simulation purposes. We present a combined experimental and theoretical study of two-dimensional ion crystals. We demonstrate that standard pseudo-potential theory accurately predicts the positions of the ions and the location of structural transitions between different crystal configurations. However, pseudo-potential theory is insufficient to determine eigenfrequencies of the two-dimensional ion crystals accurately but shows significant deviations from the experimental data obtained from resolved sideband spectroscopy. Agreement at the level of $2.5 \cdot 10^{-3}$ is found with the full time-dependent Coulomb theory using the Floquet-Lyapunov approach and the effect is understood from the dynamics of two-dimensional ion crystals in the Paul trap.

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Accurate control of ion crystals is of major importance for spectroscopy, quantum simulation or quantum computing with such experimental platform. Since the invention of dynamical trapping by Wolfgang Paul [1], this versatile instrument has been adapted and optimized for specific purposes. Charged particles, more specifically singly charged ions, are confined in a radio frequency (RF) potential, which is formed by tailored electrode structures. In the case of the linear Paul trap, one aims for a quadrupole field along one z-axis, such that a harmonic pseudopotential in x- and y-direction is formed. This radial potential only confines the ions, while an additional weaker axial potential in z-direction is generated with static (DC) voltages applied to endcap electrodes. Trapped ions are cooled by laser radiation [2] in the potential described by three trap frequencies $\omega_{x,y,z}$ eventually forming a crystallized structure.

The conditions of operation are characterized by two anisotropy parameters where the radial confinement $\omega_{x,y}$ typically exceeds the axial DC confinement $\omega_z$. For sufficiently small values of $\alpha(x,y) \equiv \omega_z^2/\omega_{x,y}^2$ the ion crystal is linear and aligned along the weakest, the z-trap axis; all ions are placed in the node of the RF potential. Spectacular highlights using linear crystals of cold ions are the demonstration of quantum logic operations [3, 4], the generation of entangled states [5, 6], sympathetic cooling of ions of different species [7, 8], or the quantum-logic clock [9]. To reach the level of quantum control, as required in the experiments listed above, the first precondition was a complete understanding of eigenmodes and eigenfrequencies for such stored linear ion crystals [10, 12].

For larger numbers of ions, or for larger values of $\alpha$, the linear crystal undergoes a transition to a zigzag structure and eventually a fully crystalline two- or three-dimensional structure [13, 14]. Especially interesting are planar ion crystals, where usually one of the confining radial potentials is much tighter than the axial one, as such structures with non-degenerate radial frequencies do not rotate [13, 16] and allow to address and observe individual ions. Recent proposals have outlined how to achieve laser-induced spin-spin interactions on such 2D ion crystals and how to use their spatial arrangement for the realisation of spin lattices exhibiting frustration [17, 18]. It was also proposed how to study the spin-phonon interactions coupling the geometric structure of the ion chain to the spin-spin interaction of the chain to realize a Peierls instability [19] or the Jahn-Teller quantum phase transition [20, 21]. Topological defects in the zigzag configuration were proposed for simulation of quantum effects with solitons [22]. It was also proposed, that the dynamics of the structural transition from linear to zigzag configuration may be induced by electronic excitations or a fast change of the trap parameters [22, 20], and it would allow for the verification of the predicted scaling laws for defect formation when traversing the transition, that is the Kibble-Zurek mechanism [22, 23]. Furthermore, the double well structure realized by the two different configurations of the zigzag ion crystals can be manipulated and may thus allow for the creation of a coherent superposition between these two configurations and hence serve as a test bed for decoherence models [23, 24].

Despite these opportunities, no experiments have been successful in using planar crystals in a Paul trap. This is due to the high complexity of controlling such crystalline two-dimensional structures. Required is the knowledge of eigenmodes and eigenfrequencies of such crystals, since it is important for the cooling and for the design of ion-ion interactions which rely on the setting of laser parameters. These are tailored to induce spin-dependent light forces.
via Stark effects \cite{30,32}.

Here we present a combined experimental and theoretical study of eigenmodes of ions in planar crystals. We describe an accurate experimental determination of positions of ions and the frequencies of the eigenmodes of the crystal structures together with a comparison of these data with theoretical expectations based on pseudo-potential approximation (PPT) and a full dynamical classical theory for solving the linearized Coulomb problem in ion crystals using a Floquet-Lyapunov approach (FLT) \cite{33,34}. From observations of multiple sequential phase transitions between different structures of the ion crystal we map out the phase diagram and find good agreement with theoretical predictions following PPT for both the transition points and for the positions of individual ions in the different phases. The measurement of the eigenfrequencies relies on sideband spectroscopy and is applied here to the vibrational frequencies of a three-ion zigzag crystal. Surprisingly, some modes exhibit a significant 37 kHz deviation when compared to calculations in PPT. We can understand the measured data quantitatively only if the full time dependent solution of the trapping potential is taken into account, with the eigenfrequencies calculated using FLT.

\begin{equation}
\frac{d^2y}{d\xi^2} + [a - 2q \cdot \cos(2\xi)]y = 0.
\end{equation}

One derives stable solutions with characteristic exponents $\beta_i \approx \sqrt{\Omega_i + q_i^2/2}$, and obtains a harmonic time-independent pseudo-potential with frequencies $\omega_i = \beta_i \Omega_i/2$.

In the pseudo-potential approximation, the ions arrange in positions corresponding to a stable minimum of the time-independent potential, the determination of which follows the method for linear crystals \cite{10}, extending it to three dimensions. In the time-dependent potential, the equivalent of a minimum configuration crystal is a periodic solution with the ions oscillating at the RF-frequency $\Omega$, about well-defined average locations (the oscillation denoted as micro-motion). The ion coordinates in the radial direction obey

\begin{equation}
y_n(t) = \bar{y}_n[1 - \frac{q_n}{2} \cos(\Omega t)] + O(\frac{q_n^2}{4}),
\end{equation}

while in the axial z-direction, the micro-motion is negligible \cite{33}.

In the experiment, the resonance fluorescence of Doppler cooled $^{40}$Ca$^+$ ion crystals near 397 nm is imaged on a CCD camera \cite{36}. We use a linear Paul trap consisting of four cylindrical rods, diameter 2.5 mm and at diagonal distance between centres of 4.7 mm as well as two DC endcaps at 10 mm distance. For these measurements we operate with a RF-amplitude of $U_{RF} \sim 300$ V$_{pp}$ at $\Omega/(2\pi) = 14.62$ MHz, and endcap voltages of 350 V, yielding secular frequencies $\omega_{x,z}/(2\pi)\approx(316, 111)$ kHz and a much larger $\omega_x$. All secular frequencies are determined from the resonant electric excitation of the com modes, observed as a blurring of the ion crystal image \cite{37}.

Equilibrium positions are determined from CCD images such as shown in Fig.\textbf{1} with a 7-ion crystal. Averaging over 100 exposures and applying a Gaussian fit to the data we determine the ion locations, see Fig.\textbf{1b}. As the planar crystal is observed at an angle of 45°, the y-axis is compressed by $\sqrt{2}$. The magnification of the imaging system is determined from the com mode frequency in z-direction and the z-distance in a linear two-ion crystal.

As the micro-motion amplitude is small compared to the diffraction-limited spots of the ion fluorescence, and as it consists of a symmetric oscillation about the equilibrium positions, the observed fluorescence precisely indicates the ion positions. The measured data show perfect agreement with the theoretical expectations following PPT at the level of a few parts in $10^5$, see Fig.\textbf{1}, for a crystal of 7 ions, and similar agreement is found for crystals with 6 to 17 ions.

Structural phase transitions are induced when the value of $\alpha$ is varied. While so far the first linear-to-zigzag

![FIG. 1. (color online) Determination of ion positions in planar crystals: a) The ion fluorescence near 397 nm is imaged on a CCD chip, b) The ion positions (black dots) are determined by averaging over 100 exposures and applying a Gaussian fit to the data we determine the ion locations, see Fig. 1b.]

The positions of the ions in a crystal are determined by the mutual Coulomb repulsion together with a static electric potential in the z-direction and a dynamic radial trapping potential. A single charged particle in the time-dependent potential of a quadrupole Paul trap, or the center-of-mass (com) modes of a general crystal of ions, obey decoupled, linear Mathieu equations of motion in each spatial direction.\cite{35}. The RF and DC trap voltages, together with the RF frequency $\Omega$ and the mass of the ions $m$, determine the dimension-free Mathieu parameters in each direction of space $i \in \{x,y,z\}$ as $a_i = 4eU_{DC}/\gamma_i m\Omega^2$ and $q_i = 2eU_{RF}/\gamma'_i m\Omega^2$, where $\gamma_i$ and $\gamma'_i$ are geometrical factors denoting the curvature of the respective potentials and $\xi = \Omega t/2$. The Mathieu equation is solved

\begin{equation}
\frac{d^2y}{d\xi^2} + [a - 2q \cdot \cos(2\xi)]y = 0.
\end{equation}
transition was investigated, we observe multiple consecutive critical \( \alpha \), see Fig. 2. The experimentally determined positions and the critical values of \( \alpha \) agree with the prediction of PPT. Comparing the PPT prediction and the solution of the full time-dependent equations, with \( q \lesssim 0.1 \), the relative differences for critical \( \alpha \) are only about 1\%, not resolved in the experimental data.

Normal mode frequencies result from the Coulomb forces between ions at their equilibrium positions. For the calculation in PPT these forces are expanded in small excursions about the equilibrium positions, linearized, and the Hessian matrix is solved. Laser spectroscopy provides a precise tool to determine eigenfrequencies with a relative accuracy of 0.2 percent or better, and we do not find agreement between the measurements and the PPT calculation. The corresponding values for mode frequencies for the three-ion crystal under study, in a zigzag configuration with \( \alpha \approx 0.53 \), are in Tab. 1. Only recently, the influence of micro-motion on the frequencies of secular frequencies, superimposed on micro-motion at the RF-frequency. This effect resembles the Lamb shift where the Zitterbewegung of the bound electron leads to a modification in the Hydrogen energy levels. A short description of the method of solution, together with code for the calculations presented in this Letter, is available online.

We test the FLT prediction experimentally using a three-ion crystal and performing resolved sideband spectroscopy on the narrow \( S_{1/2} \) to \( D_{5/2} \) transition near 729 nm. In the spectrum (Fig. 3) we identify the vibrational frequencies of a three-ion zigzag crystal. When testing PPT, we use the com modes in axial and radial y-direction to generate predictions for the zigzag frequencies, which deviate from the experimental values by 37 kHz, and 15 kHz, respectively. For FLT we fit the five measurements of the 3 com modes and the 2 lowest planar zigzag modes using a weighted-least-squares fit, with 3 parameters \( q_y \), \( a_x \) and \( a_y \) (imposing \( a_x = -a_x - a_y \) as required from the Laplace equation). The weighted-least-squares fit norm is a random variable distributed like \( \chi^2 \) with 2 degrees of freedom, see [10]. The theoretical values coincide exactly with both the eigenfrequencies, fitting the data with about 22\% probability with all the eigenfrequencies. A similar fitting procedure for...
the PPT results in a negligible probability of the order $10^{-11}$, reflecting that PPT frequencies do not agree with the experimental finding.

The strong influence of the trap drive on the eigenfrequencies of zigzag modes is further explained by a theoretical analysis, where the Mathieu parameter $q$ is varied and the variation of the mode frequencies $\omega_{zz(a)}$ and $\omega_{zz(b)}$ is plotted, see Fig. 3. In the experiment, the $q$-value is fixed and can not varied easily over a large range. The data points, near $q=0.2$, for the $zz(a)$ and $zz(b)$ mode hit the FLT prediction and exclude the PPT frequencies. The fractional frequency shift reaches for high $q$-values a level of up to 20%. Examining Fig. 3, it is understandable why the $zz(a)$ mode is affected the most by the micro-motion, and why its frequency increases monotonically with $q$. It can be seen from Eq. 2 that the micro-motion amplitude of each ion is proportional to the negative of its $y$-equilibrium position, so that this motion has a large projection on the eigenvector of the $zz(b)$ mode. At each point along the periodic trajectory, the restoring forces are larger for this mode than at the center, and thus its frequency increases with the amplitude of the micro-motion, hence with $q$.

**Conclusion** — We calculated and characterized experimentally the behavior of two-dimensional crystals under the influence of micro-motion. We show that while the pseudo-potential theory fails to explain the experimental results to an accuracy within $3\sigma$-deviation, the newly established method predicts the experimental finding correctly. We could show that FLT provides an experimentally verified tool for understanding the static and dynamic properties of mesoscopic Coulomb crystals, and might prove to be a key ingredient for establishing these systems as a new experimental platform. The presented results are an essential prerequisite for the success of quantum simulation in two-dimensions where the effective Hamiltonian depends on the crystal frequencies of all normal modes in the presence of micro-motion. Based on our results, a theory could be constructed that takes into account such effects in the effective Hamiltonian.

Moreover, the precise knowledge of the behavior of the crystal under the influence of micro-motion is crucial for quantum-to-classical simulation experiments that use such transitions in condensed matter or high energy effects, such as the Kibble Zurek mechanism. The deviation of critical $\alpha$ values from the pseudo-potential predictions is expected to be larger for larger values of $q$, and merits further experimental and theoretical investigation.

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