Rf-induced persistent long-range ordered structures in two-species ion Coulomb crystals in a linear Paul trap.

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We report on the observations of three-dimensional long-range ordered structures in the central $^{40}$Ca$^+$ ion component of $^{40}$Ca$^+$$^{44}$Ca$^+$ two-species ion Coulomb crystals in a linear Paul trap. In contrast to long-range ordering previously observed in single species crystals, the structures observed are strikingly persistent and always of one specific type in one particular orientation. Molecular dynamics simulations previously observed in single species crystals have been investigated in Penning and radio frequency (rf) traps (also named Paul traps), with structural findings in good agreement with theoretical predictions. Two-species crystals have been studied in much less detail, and experimentally mainly in linear Paul traps. For singly charged ions in such traps, the lighter species will in general segregate into a cylindrically shaped structure surrounded by the heavier ion species. In the case of $^{40}$Ca$^+$ and $^{24}$Mg$^+$ ions, the structures of the lighter species were found for the most part to be identical to the cylindrical structures of infinitely long one-species systems confined in two-dimensions by a rotational-symmetric harmonic potential. For two ion species with identical charge-to-mass ratios, mixing of the species is predicted. At present, Coulomb crystals find applications in such diverse fields as quantum computing and cold molecule ion research.

In this Letter, we present observations of persistent long-range ordered structures in the $^{40}$Ca$^+$ component of bicrystals consisting of $^{40}$Ca$^+$ and $^{44}$Ca$^+$ ions. The observed structures deviate from those expected to exist in a fully rotational symmetric harmonic potential, but are in close agreement with results from molecular dynamics (MD) simulations including the quadrupole nature of the trapping fields.

The $^{40}$Ca$^+$ ions are confined in a linear Paul trap which is situated in a vacuum chamber at a pressure of $10^{-10}$ Torr, and are laser cooled as depicted in Fig. 1(a). The Paul trap used in these experiments has been described in detail elsewhere. In short, the linear Paul trap consists of four electrode rods placed in a quadrupole configuration. The electrode diameter is 8.0 mm and the minimum distance of the central trap axis is $r_0 = 5$ mm. The time varying voltages $\frac{1}{2}U_{\text{rf}} \cos(\Omega_{\text{rf}} t)$ are applied to the two sets of diagonally opposite electrode rods, respectively, to obtain confinement in the radial plane (z-plane in Fig. 1(a)). Axial confinement along the z-axis is accomplished by sectioning each of the electrode rods into three pieces and then applying a static voltage $U_{\text{end}}$ to the end-electrodes. The length of the center-electrode is 5.4 mm, while the outer pieces are 20 mm. In the present experiments $\Omega_{\text{rf}} = 2 \pi \times 3.88$ MHz, $U_{\text{rf}} \sim 540$ V and $U_{\text{end}} \sim 10–50$ V were used. The resulting confinement for an ion species of type $i$ with mass $M_i$ and charge $Q_i$ is often approximated by a harmonic pseudo potential $\Phi_{ps}(r, z) = \frac{1}{2} M_i (\omega_r^2 r^2 + \omega_z^2 z^2)$, where $\omega_r$ and $\omega_z$ are the radial and axial trap frequencies, respectively. The axial trap frequency is given by $\omega_z^2 = 2 \kappa Q_i U_{\text{end}}/M_i$, where $\kappa = 3.97 \times 10^4$ m$^{-2}$ is a constant related to trap geometry, and the radial trap frequency is given by $\omega_r^2 = \frac{1}{2} \left( \frac{1}{M_i} + \frac{1}{M_{\text{eff}}} \right) Q_i^2 U_{\text{rf}}^2/2M_i r_0^2 \Omega_{\text{rf}}^2$ is the contribution from the time varying quadrupole fields.

The dependence on the charge and mass of the ion species makes the lighter isotope ($^{40}$Ca$^+$) more tightly bound towards the trap axis than the heavier ($^{44}$Ca$^+$) and leads consequently to a total radial separation of the two ion species when sufficiently cooled. The zero-temperature ion density in the pseudopotential is given by $n_{\text{adh}} = e_0 U_{\text{rf}}^2 / M_i r_0^2 \Omega_{\text{rf}}^2$, where $e_0$ is the vacuum permittivity. Due to the spatial separation of the ions in two-species ion Coulomb crystals, this expression is also applicable to the individual components of such crystals.

The $^{40}$Ca$^+$ and $^{44}$Ca$^+$ ions used in the experiments are produced isotope selectively by resonant two-photon photo-ionization of atoms in an effusive beam of naturally
abundant calcium \(^{40}\text{Ca}^+\) and \(^{44}\text{Ca}^+\) ions. In Fig. 1(b), the transitions in \(^{40}\text{Ca}^+\) used for Doppler laser cooling of the trapped ions are shown. The main cooling transition is the dipole allowed \(^4S_{1/2} \rightarrow ^4P_{1/2}\) transition at 397 nm. To avoid optical pumping into the metastable \(^3D_{3/2}\) state, repumping is done either by using a single repump laser at 866 nm via the \(^4P_{1/2}\) state (\(^{44}\text{Ca}^+\)) or by using two repump lasers at 850 nm and 854 nm via the \(^4P_{3/2}\) state (\(^{40}\text{Ca}^+\)). Due to the isotope shifts of the cooling transitions \(^{40}\text{Ca}^+\) and \(^{44}\text{Ca}^+\), each isotope ion requires its own laser cooling frequencies. For both isotopes the final temperature is of the order of \(\sim 10\) mK, which is low enough for achieving Coulomb crystallization (\(\Gamma \sim 250\)). Imaging of the fluorescence from the trapped ions is achieved using an image intensified CCD camera placed above the trap.

FIG. 1: (a) Schematics of the laser and linear Paul trap setup. (D)M denotes a (dichroic) mirror, PBS denotes polarization beamsplitters, and DL denotes diode laser. (b) Optical transitions used for Doppler laser cooling of the \(^{40}\text{Ca}^+\) ions.

FIG. 2: (color online) Images of two-species ion Coulomb crystals containing \(\sim 1500\) \(^{40}\text{Ca}^+\) ions (red) and \(\sim 2000\) \(^{44}\text{Ca}^+\) ions (blue) at different settings of the end-cap potential, \(U_{\text{end}}\). The camera exposure time is \(\sim 100\) ms and the trap potentials are \(U_{\text{rf}} = 540\) V and (a) \(U_{\text{end}} = 46.1\) V, (b) \(U_{\text{end}} = 30.2\) V, (c) \(U_{\text{end}} = 20.0\) V, and (d) \(U_{\text{end}} = 13.8\) V, respectively.
Images of two-species Coulomb crystals consisting of $^{40}\text{Ca}^+$ and $^{44}\text{Ca}^+$ ions are presented in Fig. 2 for various settings of the static voltage $U_{\text{end}}$ on the end-electrodes. Since the ions are only fluorescing when they are directly laser cooled, the two isotopes can be imaged separately by alternatingly blocking (about 1 Hz rep. rate) the 850 nm ($^{40}\text{Ca}^+$ laser cooling off) and the 866 nm ($^{44}\text{Ca}^+$ laser cooling off) using mechanical shutters. The presented combined images are subsequently created with a red color coding for $^{40}\text{Ca}^+$ and blue for $^{44}\text{Ca}^+$. Due to sympathetic cooling, the crystal retains its shape and structure during the alternating laser cooling sequence. As expected, the lightest isotope $^{40}\text{Ca}^+$ is located as a cylindrical core closest to the trap axis, surrounded by the heavier $^{44}\text{Ca}^+$. It is immediately clear from the images in Fig. 2 that the projection of the actual three-dimensional structure of the $^{40}\text{Ca}^+$ ions is a two-dimensional rectangular lattice aligned with the trap axis. Since the depth of focus of the imaging system ($\sim50$ $\mu$m) is several times the inter-ion distance, we conclude the rectangular structure in the images must originate from a three-dimensional long-range ordering. In contrast to our previous observations of long-range order in spherical one-component ion crystals, where the orientation of the observed metastable ($\sim100$ ms) structures seemed to be arbitrary, the rectangular structures presented here are very persistent ($\sim10$ s) and always oriented the same way. Accordingly, the presence of the surrounding $^{44}\text{Ca}^+$ ions apparently has significant influence on the formation and appearance of the observed long-range structure in the $^{40}\text{Ca}^+$ part of the crystal. The image sequence in Fig. 2 illustrates additionally that despite changes in the outer shape of the $^{40}\text{Ca}^+$ core, the observed rectangular lattice of the ions is preserved, indicating that the observed long-range ordered structure is rather stable to changes in the boundary conditions of the crystal.

In order to understand the observations, a series of molecular dynamics (MD) simulations of two-species crystals with the same number of the two calcium isotopes as in the crystals shown in Fig. 2 have been performed. In Figs. 3(a) and 3(b), results from one simulation using a pseudo-potential corresponding to the trapping parameters of Fig. 2(b) is presented. As in the experiments, a clear radial separation of the two isotope ions is observed. Furthermore the radial projection (Fig. 3(a)) clearly reveals that the $^{40}\text{Ca}^+$ part of the crystal is organized in concentric cylindrical structures, resembling the structure of an infinitely long 1D cylindrically symmetric confined ion crystal. In Fig. 3(b), a projection corresponding to the focal region of the imaging system is shown. Neither this nor other sections, e.g., in the $yz$-plane, lead to projection images with rectangular structures. However, when the full rf potential is used in the MD simulations, some regular ordering in the central component of the crystal does appear, as is evident from the results presented in Figs. 3(c) and 3(d) for a specific phase of the rf field. From Fig. 3(d), it is seen that indeed a rectangular projection image is expected when the rf-quadrupole field, which breaks the rotational symmetry, is included in the simulations. Even when averaging over all phases of the rf field, the rectangular structure persists. However, some blurring of the position of the ions along the $z$-axis, as is seen in the images of Fig. 2, is found. Analysis of a much simpler two-ion system in a linear rf trap has previously shown similar preferred orientation effects with respect to the rf quadrupole field axes.

A closer analysis of the simulation results presented in Figs. 3(c) and 3(d) shows that the $^{40}\text{Ca}^+$ ions in the core organize themselves in a long-range ordered structure in the form of a face-centered tetragonal (fct) lattice as illustrated in Fig. 4(a), with the sidelengths related by $a = b = \sqrt{3}c$. The rectangular projection presented in Fig. 3(d) is obtained when the fct structure is viewed along the $b$ vector as illustrated in Fig. 4(b). This rectangular projection has a height to width ratio of $h_{\text{tet}}/w_{\text{tet}} = \sqrt{3} \approx 1.73$, which is not exactly the same as the $h/w = 1.62 \pm 0.07$ observed in the experiments. In fact, the observed rectangular structure is more in agreement with a face-centered cubic (fcc) structure viewed along the [211] direction (see Figs. 4(c) and 4(d)), which
would lead to $h_{\text{fcc}}/w_{\text{fcc}} = \sqrt{8/3} \simeq 1.63$. The reason for the deviation between the simulated results and the actually observed structures is probably that the difference in the potential energies of the two structures in the rf potential is very small, as is well-known to be the case for various long-range ordered structures in infinite systems without the presence of rf fields. In contrast to the two-species results above, no fixed orientation of the atomic ensemble to the light field can be controlled by shifting the position of the whole crystal.

Very stable aligned crystal structures as those discussed above, may find many future applications. For instance, for cavity QED studies, the situation where the axial periodicity of the Coulomb crystal is an integer multiple of the node spacing of the standing wave cavity field is very interesting, since the effective coupling of the atomic ensemble to the light field can be controlled by shifting the position of the whole crystal.

In conclusion, very persistent three-dimensional long-range ordered structures with one specific orientation have been observed in two-species ion Coulomb crystals in a linear Paul trap. MD simulations strongly indicate that these characteristics are a consequence of the co-axial cylindrical symmetry of the central ion component of the Coulomb crystal and the radio frequency quadrupole trapping field.

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