## Structural Properties of Two-Component Coulomb Crystals in Linear Paul Traps

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We report on structural properties of two-component Coulomb crystals in a linear Paul trap. The crystals consist of two laser cooled ion species,  ${}^{24}Mg^+$  and  ${}^{40}Ca^+$ . The lighter  ${}^{24}Mg^+$  ions form an inner cylindrical crystal structure surprisingly similar to that of an infinitely long single component crystal, while the outermost shell of the surrounding  ${}^{40}Ca^+$  ions have a spheroidal shape, which is highly insensitive to the presence of the  ${}^{24}Mg^+$  ions. Observed changes in the radial separation of the two ion species with the radius of the inner cylindrical crystal is explained by a simple model.

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Two-component Coulomb crystals are an example of the solid state of a strongly coupled plasma containing two types of particles, with the same sign of charge. Such bicrystals, consisting of mixtures of predominantly oxygen and carbon nuclei embedded in a neutralizing degenerate electron gas, are believed to be present in the interior of cooling white dwarf stars [1]. In laboratories, smaller ion bicrystals can be formed in ion traps, where magnetic and/or electric fields play the role of a neutralizing background charge. At present small ion bicrystals in a string configuration are regarded as a very promising medium for quantum computation [2,3], while larger bicrystals seem to be ideal sources of cold atomic or molecular ions [4] for spectroscopy and various chemical physics studies. The observed bicrystal structures presented in this Letter extend the range of possible applications of bicrystals. In combination with optical cavities the bicrystals presented here constitute unique systems for studying cavity QED effects and few ion lasers. Furthermore, large bicrystals might be a future storage medium for quantum information of light [5].

So far experimental studies of multicomponent strongly coupled plasmas have been limited to ion species where not all components could be imaged [4,6,7] or to plasmas in a liquidlike state [8,9]. In contrast, single component ion Coulomb crystals of various alkalilike ions have been studied extensively in both Paul [10-14] and Penning [15-17] ion traps. When the plasma coupling parameter,  $\Gamma$  (the ratio of the potential Coulomb energy between neighboring ions and the average kinetic energy per particle) exceeds 170, molecular dynamics (MD) simulations predict a transition from a liquid to a solid state, which for infinite plasmas takes the form of a bcc structure. Such structures have been observed in the center of very large ion crystals in Penning traps [17]. For smaller ion crystals, where surface effects dominate, the structures are governed by boundary conditions set by the trapping potentials and range from 1D ion strings, over 2D planar crystals, to 3D spheroidal crystals where the ions arrange themselves in concentric shells [10-16].

In this Letter, we present results on bicrystals with emphasis on crystals containing  ${}^{40}Ca^+$  and  ${}^{24}Mg^+$  ions. The experimental setup has been described in more detail elsewhere [18]. The ions are confined in a linear Paul trap [19] consisting of four parallel cylindrical electrodes in a quadrupole configuration (see Fig. 1). Confinement is achieved by applying a dc voltage,  $U_{dc}$ , to the eight end segments and an rf voltage,  $U_{rf}$ , to diagonally opposite rods, giving rise to an effective 3D harmonically confining potential with cylindrical symmetry. The axial potential is independent of ion mass and is well described by

$$\Phi_z(z) = \frac{1}{2} M_i \omega_z^2 z^2, \qquad \omega_z^2 = 2\kappa Q_i U_{\rm dc}/M_i,$$

where  $M_i$  and  $Q_i$  are the mass and charge of ions of type *i* and  $\kappa = 8.44 \times 10^4 \text{ m}^{-2}$  is a constant related to the trap geometry. The effective radial potential is given by

$$\Phi_{r,i}(r) = \frac{1}{2} M_i \left( \omega_r^2 - \frac{1}{2} \omega_z^2 \right) r^2, \qquad \omega_r^2 = \frac{Q_i^2 U_{rf}^2}{2M_i^2 r_0^4 \Omega^2},$$



FIG. 1 (color). Schematic overview of the experimental setup. The inset shows a small bicrystal containing approximately 75  $^{24}Mg^+$  ions (red) and 120  $^{40}Ca^+$  ions (blue).

where  $U_{\rm rf}$  and  $\Omega = 2\pi imes 5.1$  MHz are the amplitude and the frequency of the applied rf field and  $r_0 = 1.75$  mm is the distance from the trap center to the surface of the electrodes. Because of the mass dependence of the radial potential,  $\Phi_{r,i}(r)$ , lighter ions are confined radially more strongly than heavier ones. Typical trap frequencies are a few hundred kHz. The  ${}^{40}Ca^+$  and  ${}^{24}Mg^+$  ions are loaded into the trap from thermal beams of neutral atoms. <sup>40</sup>Ca<sup>+</sup> is produced by electron impact ionization and any produced impurity ions are ejected from the trap by changing the trap parameters to make their motion unstable. Subsequently, <sup>24</sup>Mg<sup>+</sup> is loaded using isotope selective resonanceenhanced two-photon ionization [18]. Laser cooling is done along the trap axis on the  $3s^2S_{1/2} \leftrightarrow 3p^2P_{3/2}$  tran-sition at 280 nm in <sup>24</sup>Mg<sup>+</sup> and on the  $4s^2S_{1/2} \leftrightarrow 4p^2P_{1/2}$ and  $3d^2D_{3/2} \leftrightarrow 4p^2P_{1/2}$  transitions in <sup>40</sup>Ca<sup>+</sup> at 397 and 866 nm, respectively [18]. The light, spontaneously emitted by the ions during the laser cooling cycles, is imaged onto an image intensified CCD video camera by a  $15 \times$  magnification lens system. As the fluorescent light is emitted at different wavelengths, the focus position of the lens system and the magnification for the two species differ. The time required to change focus position is typically less than 30 s. For absolute calibration of the magnifications a 125  $\mu$ m diameter optical fiber can be inserted into the trap center and imaged. Images of the two ion species are obtained separately by means of color filters, corrected for differences in magnification and combined using the following color coding: red for light emitted by <sup>24</sup>Mg<sup>+</sup> ions and blue for light emitted by  $^{40}Ca^+$  ions.

Because of the mass dependence of the radial trapping potential some spatial separation of different simultaneously trapped ion species is expected. For  ${}^{24}Mg^+/{}^{40}Ca^+$ bicrystals, such as those shown in Figs. 1 and 2a-2c, we observe complete spatial separation between the two species, with the <sup>24</sup>Mg<sup>+</sup> ions situated closest to the trap axis. While the presence of the  ${}^{24}Mg^+$  ions is observed to have only a weak influence on the shape of the outer envelope of  ${}^{40}Ca^+$ , the  ${}^{40}Ca^+$  ions force the  ${}^{24}Mg^+$  ions to order in nearly cylindrical structures. The ordering of the  ${}^{24}Mg^+$  ions resembles the expected shapes for a single component crystal in an infinitely long, cylindrically symmetric harmonic potential [20]. Axially this cylindrical structure continues right to the edge of the surrounding <sup>40</sup>Ca<sup>+</sup> crystal, from where curved end sections protrude. Though surprising, the resulting structure may be explained qualitatively by noting that the axial potential is the same for both species, whereas the radial potential is stronger for <sup>24</sup>Mg<sup>+</sup>.

The pictures of the large bicrystals in Figs. 2a–2c, containing approximately  $300^{24}Mg^+$  ions and  $3000^{40}Ca^+$  ions at an rf amplitude of  $U_{rf} = 60 \text{ V}$ , are snapshots from a series obtained while varying  $U_{dc}$ . Changing the trap potential in this way does not lead to variation in ion density, since the density of



FIG. 2 (color). A  ${}^{40}\text{Ca}^+/{}^{24}\text{Mg}^+$  bicrystal at three different end cap voltages. The crystal is symmetric under rotations around the trap axis, *z*, and contains approximately  $300 {}^{24}\text{Mg}^+$  ions (red) and  $3000 {}^{40}\text{Ca}^+$  ions (blue). The ratios of the axial and effective radial trapping frequencies for  ${}^{40}\text{Ca}^+$  and  ${}^{24}\text{Mg}^+$  ions in the three cases shown are (a)  $\omega_{z,\text{Mg}^+}/\omega_{r,\text{Mg}^+} = 0.4$  and  $\omega_{z,\text{Ca}^+}/\omega_{r,\text{Ca}^+} = 0.6$ ; (b)  $\omega_{z,\text{Mg}^+}/\omega_{r,\text{Mg}^+} = 0.7$  and  $\omega_{z,\text{Ca}^+}/\omega_{r,\text{Ca}^+} = 1.0$ ; (c)  $\omega_{z,\text{Mg}^+}/\omega_{r,\text{Mg}^+} = 1.1$  and  $\omega_{z,\text{Ca}^+}/\omega_{r,\text{Ca}^+} = 1.8$ .

ions of type *i*, given by  $n_i = (\epsilon_0 U_{\rm rf}^2)/(M_i r_0^4 \Omega^2)$ , is independent of  $U_{dc}$ . The ion densities for the crystal shown in Fig. 2 are  $n_{\rm Mg^+} = 3.3 \times 10^8 {\rm ~cm^{-3}}$  and  $n_{\rm Ca^+} = 2.0 \times 10^8 {\rm ~cm^{-3}}$ . The varying light intensity in the  ${}^{24}Mg^+$  core is due to the presence of a small amount of <sup>24</sup>MgH<sup>+</sup> ions. Because of the small mass difference, these do not influence the <sup>24</sup>Mg<sup>+</sup> structure. While the shape of the  ${}^{40}Ca^+$  outer envelope is similar to the single species crystal shape, the internal structure of the  ${}^{40}Ca^+$ part of the crystal changes from the outer shell shape which is spheroidal, to an inner cylindrical structure at the  ${}^{40}\text{Ca}{}^+\mathchar`-{}^{24}\text{Mg}{}^+$  interface. The radial separation at this interface,  $r_{Ca^+} - r_{Mg^+}$ , where  $r_{Ca^+}$  is the radius of the inner  ${}^{40}Ca^+$  shell and  $r_{Mg^+}$  is the radius of the outer  $^{24}Mg^+$  shell, increases with the radius of the  $^{24}Mg^+$ core. This feature can be accounted for by noting that the electrostatic force at  $r_{Ca^+}$  should be equal to the force at  $r = r_{Ca^+}$  found in a pure  ${}^{40}Ca^+$  crystal. Neglecting any axial components of the electric field between the  $^{24}Mg^+$  core and the surrounding  $^{40}Ca^+$ , this corresponds to assuming that the linear charge density of <sup>24</sup>Mg<sup>+</sup> ions in the core should be equal to the linear charge density of a <sup>40</sup>Ca<sup>+</sup> cylinder with radius  $r_{Ca^+}$ . In this model [21]  $r_{Mg^+}/r_{Ca^+} = (M_{Mg^+}/M_{Ca^+})^{1/2} = 0.77$ , in good agreement with the experimental result,  $r_{Mg^+}/r_{Ca^+} = 0.78 \pm$ 0.03, derived from the full scan series where  $U_{dc}$  is scanned from 3 to 13.8 V. Since the spatial separation increases with the radius of the central core and the square root of the ratio of the masses, this will presumably put a stronger limitation on the efficiency of sympathetic cooling than the one given by the mass requirements for stable trapping [7]. It should be noted that this radial separation is significantly different both in physical origin and in dependence on mass, from the centrifugal separation of different mass ions in Penning traps, see, e.g., Eq. (43) in Ref. [22].

In Fig. 3, we present the crystal aspect ratio, i.e., the ratio between radius and half length, R/L, of a spheroid fit to the outer <sup>40</sup>Ca<sup>+</sup> envelope, for various ratios of the axial and radial trap frequencies. These data may be compared with a theoretical model for zero temperature charged liquids [23], and to aspect ratios obtained in experiments with pure <sup>40</sup>Ca<sup>+</sup> crystals. No deviations from the liquid model or from experimental data for the pure crystals are observed. The figure also shows results for bicrystals containing <sup>40</sup>Ca<sup>+</sup> and <sup>24</sup>MgH<sup>+</sup> ions in the ratios 2:1 and 1:2. Only the latter case shows deviation from the pure crystal case.

The structure of the <sup>24</sup>Mg<sup>+</sup> core has been compared to the structures expected in the infinitely long, cylindrically

symmetric single component crystal case [20]. Here, MD simulations predict structure transitions for increasing linear ion density, from a string of equidistantly spaced ions on the axis to structures of multiple coaxial cylindrical shells of increasing radius. Figure 4 shows a comparison between the experimentally observed structure transitions in the central core of the bicrystal shown in Fig. 2 and the predictions for infinitely long single component crystals derived from MD simulations [20]. We find good agreement within our experimental uncertainties. Even for very few <sup>24</sup>Mg<sup>+</sup> ions, the infinitely long crystal behavior described above is observed. When few <sup>24</sup>Mg<sup>+</sup> ions are present in a  ${}^{40}\text{Ca}^+/{}^{24}\text{Mg}^+$  bicrystal, the  ${}^{24}\text{Mg}^+$ ions arrange themselves in a central string of equidistantly spaced ions. An example is shown in Fig. 5 where a bicrystal with a central line of 47 uniformly spaced <sup>24</sup>Mg<sup>+</sup> ions is observed. The interionic distance is given by 15.5  $\mu$ m  $\pm$  5%. In a linear Paul trap, such equidistantly spaced strings do not appear in single species plasmas, since the harmonic axial potential leads to larger ion densities at the center of the trap [19].

The two-component Coulomb crystals studied here not only expose interesting features of electrically confined strongly coupled plasmas, they are also potentially useful in other areas of physics. Sympathetically cooled [7,8] ions on a central string in the larger bicrystals, see Fig. 5, are an ideal source of cold atomic or molecular ions for spectroscopy or various chemical physics studies. Combined with an optical cavity they could be used for studies



FIG. 3 (color). The relation between the crystal aspect ratio R/L (see text) and the ratio,  $\omega_z/\omega_r$ , of the axial and effective radial trapping frequencies for  ${}^{40}\text{Ca}^+$ . The relation is shown for a pure  ${}^{40}\text{Ca}^+$  crystal, red  $\blacksquare$ ; a bicrystal containing  $10\% {}^{24}\text{Mg}^+$  and  $90\% {}^{40}\text{Ca}^+$ , black  $\bullet$ ; a bicrystal containing  $65\% {}^{40}\text{Ca}^+$  and  $35\% {}^{24}\text{MgH}^+$ , green  $\blacktriangle$ ; and a bicrystal containing  $33\% {}^{40}\text{Ca}^+$  and  $67\% {}^{24}\text{MgH}^+$ , green  $\bigtriangleup$ . Charged liquid model predictions for a pure  ${}^{40}\text{Ca}^+$  zero temperature plasma, red —, and a pure  ${}^{24}\text{MgH}^+$  zero temperature plasma, green ----, are also shown.



FIG. 4 (color). The number of shells in the <sup>24</sup>Mg<sup>+</sup> core as a function of the dimensionless linear particle density  $\lambda = a_{\rm WS}\sigma/e$ , where  $\sigma$  is the linear charge density in the <sup>24</sup>Mg<sup>+</sup> core and  $a_{\rm WS} = (4\pi n_{\rm Mg^+}/3)^{-1/3}$  is the Wigner-Seitz radius. The notation of *n* sh. + st. indicates a structure with *n* shells and a central string. Red dots are experimental data, while the dashed black line and single square indicate the structures predicted by MD simulations of infinitely long cylindrically symmetric single component crystals.



FIG. 5 (color). Bicrystal containing 47  $^{24}$ Mg<sup>+</sup> ions (red), and approximately 1300  $^{40}$ Ca<sup>+</sup> ions (blue). The  $^{24}$ Mg<sup>+</sup> ions are arranged in an equidistant string structure on the trap axis. The distance between the  $^{24}$ Mg<sup>+</sup> ions is 15.5  $\mu$ m.

of few atom lasers or, by matching the wavelength of a standing wave to the interionic spacing, they would constitute a unique system for studying cavity QED effects [24]. Furthermore, the cylindrical core of the even larger bicrystals, see Fig. 2, might be a future storage medium for quantum information of light [5].

In conclusion, we have studied several structural properties of completely spatially separated two-component Coulomb crystals at various aspect ratios and compositions. The light element formed cylindrical shell structures along the axis, similar to the structures expected for infinitely long, radial harmonically confined single component Coulomb crystals. The envelope of the heavier element was largely unperturbed by the presence of the lighter species, but close to the interface with the lighter element a transition to a cylindrical structure was observed. The radial separation between the two species was explained via a simple model and possible implications for the efficiency of sympathetic cooling were mentioned. Several new applications of bicrystals in other fields of physics were discussed.

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