Ion Coulomb crystals: Properties and applications

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1. Introduction

The intention of the present article is to give an introduction to the research of the Ion Trap Group at the Institute of Physics and Astronomy, University of Aarhus. The bases of the variety of themes that are investigated by this group are the so-called ion Coulomb crystals. After an introduction to these very unusual crystals, results and descriptions of the various research themes are given.

2. Ion Coulomb crystals

Coulomb crystals were initially studied theoretically by plasma physicists interested in understanding the properties of simple one-component plasmas (OCP), i.e., plasmas consisting of only one type of charged particles, under various conditions. It turns out that for an *infinitely* large OCP, the thermodynamical properties may be described by a single dimensionless parameter Γ, named the plasma coupling parameter, which is defined by

$$\Gamma \equiv \frac{E_{Coul}}{E_{kin}},$$

with E_{Coul} being the average Coulomb energy of nearest-neighbor particles and E_{kin} the average kinetic energy of a single particle. For Γ much smaller than one, the plasma is equivalent to an ideal gas, while for Γ of the order of unity the plasma behaves as a liquid. Finally, if Γ passes 173, the plasma undergoes a phase transition into a solid state, sometimes named a Coulomb crystal, with the charged particles forming a body-centered cubic (bcc) structure.[1] In nature, Coulomb crystals have not been directly observed, but theoretical models predict the presence of these crystals in such exotic sites as the interior of white dwarf stars[2] and in the crust of neutron stars,[3] where the presence of very dense fully stripped highly charged ions may lead to Γ exceeding 173 even at very high temperatures (up to millions of Kelvins). In the laboratory, the achievable charge densities are much lower than in the above exotic stellar objects, and hence temperatures of the order of tens of milliKelvins or lower are needed in order to form Coulomb crystals. With the advent of laser cooling[4,5] it became possible to realize such low temperatures by cooling singly charged ions, and during the past ten years larger ion Coulomb crystals have been investigated in details by a few groups around the world, including the Ion Trap Group in Aarhus.

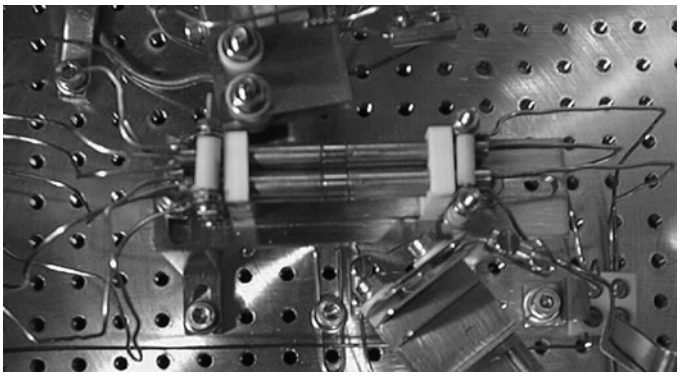

Fig. 1. The linear Paul trap in which the results presented are obtained. The trap consists of four gold-plated cylindrical rod electrodes each sectioned into three parts (two of these rods are visible in the center of the picture), to which dc and radio frequency voltages may be applied. The length of the short central part of the electrode is 5 mm. When operated, the trap is situated in a vacuum chamber with a background pressure of about 10^{-10} torr.

In order to make laboratory studies of ion Coulomb crystals, first of all a trapping device has to be applied. In our group a so-called linear Paul trap is used, where the ions are confined along one axis (from now on referred to as the main trap axis) by an electrostatic field, while in the perpendicular plane (from now on referred to as the radial plane) ions are confined by a radio frequency field,[6] see Fig. 1. The effective trapping potential becomes harmonic in all directions with rotational symmetry around the main trap axis. The very open electrode structure of the trap makes it easy to introduce laser beams for laser cooling of the ions both along the main trap axis and in the radial plane. By Doppler cooling of Mg^+ and Ca^+ ions, we may experimentally reach values of Γ around or above 173, which is the crystallization value for infinite plasmas. While cold, three dimensional *finite* OCP plasmas trapped in the laboratory have an averaged constant density like the infinite plasmas, the ion Coulomb crystal structures are generally very different from a bcc-lattice due to the presence of a surface boundary, see e. g. Refs. 6 and 7. Hence, if the number of ions in the crystals are below ~100.000, the ions are arranging themselves in a series of concentric shells, in which the ions form quasi-hexagonal structures. In Fig. 2, a series of pictures of ion Coulomb crystals containing ~350 ions are presented. The pictures represent the projection of the fluorescence from laser-cooled Mg^+ ion crystals to the surface of a CCD-camera. The ion density of these crystals is a few times 10^8 cm^{-3}, which corresponds to a nearest-neighbor spacing of a few tens of microns. The various outer shapes of the crystals have been obtained by varying the ratio of the main axis to the radial oscillation frequency. From an extensive series of measurements with ion crystals containing more than 100 ions, one may conclude that the shape of these finite crystals for a large span of aspect ratios is equivalent to that of a homogeneously charged liquid under the same trapping conditions.[8] Only when the number of ions exceeds ~100.000, bcc-structures start to be formed in the core of crystals having more than 20 outer shells.[9] In the other extreme of very few trapped ions, the ions will line up on strings for sufficient strong radial confinement as shown in the pictures of Fig. 3.

If more than one ion species is present at the same time in the trap, new more complex orderings appear. In our group we have

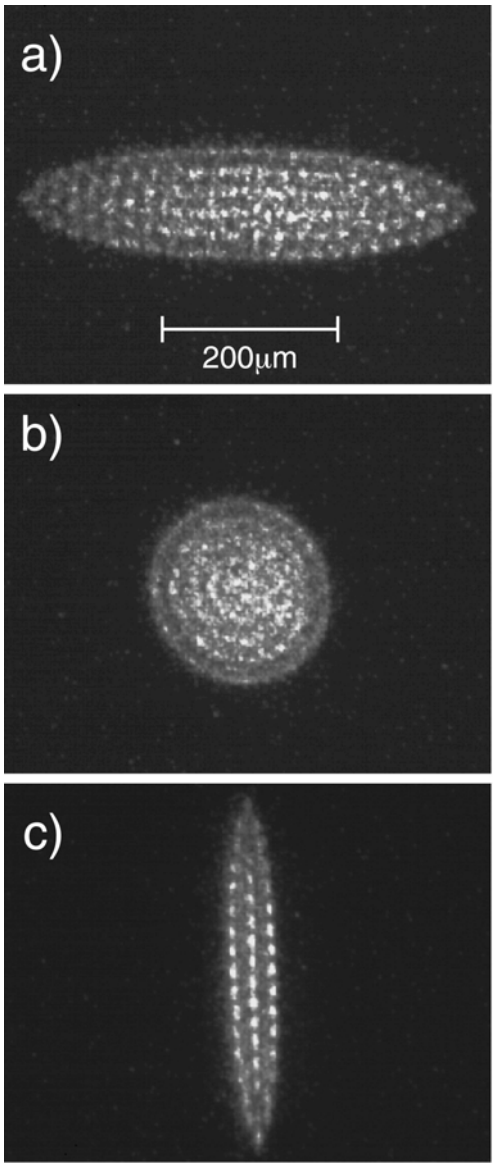

Fig. 2. The pictures represent the projection of the fluorescence from laser cooled Mg^+ ion Coulomb crystals (~350 ions) to the surface of a CCD-camera for three different trapping potentials. As in the pictures to follow, the main trap is in the left-to-right direction.

recently investigated the structures of ion Coulomb crystals containing both laser-cooled Mg^+ and Ca^+ ions.[8] Examples of pictures of such bi-crystals are shown in Fig. 4, where the blue color code and the red color code correspond to fluorescence from Ca^+ and Mg^+ ions, respectively. The reason for the obvious radial separation of the two ion species is that for singly-charged ions the oscillation frequency along the static field axis of the trap is independent of the ion mass, while in the radial plane the oscillation frequency increases with decreasing mass of the ions. Hence, in the radial plane it is energetically most favorable for the ions to segregate with Mg^+ ions closest to the main axis. The structures of the Mg^+ ions in these crystals are found to be surprisingly identical to those expected for an infinitely long crystal that is radial confined by a harmonic potential. This similarity is perhaps most obviously seen in Fig. 4b, where one observes a string of 47 Mg^+ ions which are equidistantly spaced instead of being more closely placed at the center of the trap, as seen in Fig. 3.

Apart from being interesting objects on their own, due to the extremely small Doppler shifts and interrogation times of up to several hours, ion crystals of a few ions, see Fig. 3, have for many years been valuable in high resolution spectroscopy[10] and lifetime measurements of long-lived electronic states.[11] The accuracy of specific atomic transition frequencies makes a string of a few ions a serious candidate for a future time standard.[12] Several quantum optics experiments have as well been performed, including demonstrations of simple quantum logic gates needed for future quantum computers.[13,14] This last theme will be discussed shortly in a later section.

3. Sympathetic cooling

One application of bi-crystals is to laser cool only one ion species and let the other species be sympathetically cooled through the Coulomb interaction with the directly cooled ions. Since the Coulomb interaction is long ranging, one may very effectively achieve sympathetic cooling at time scales of seconds, which is much shorter than the typical storage time (up to hours) of the ions in the trap. An example of the result of sympathetic cooling is shown in Fig. 5. This figure shows the observed fluorescence from laser cooled Ca^+ ions (~2000 ions), cooling O_2^+ molecular ions (~500 ions) formed by electron bombardment of a gas of O_2 molecules let into the trap vacuum chamber. Since the O_2^+ ions are lighter than the Ca^+ ions, when cooled they form cylindrical structures equivalent to the Mg^+ ions in Fig. 4, and, indeed, a hollow (non-fluorescing) cylindrical structure is present in Fig. 5. It is not easy to obtain a value of the exact temperature of the sympathetically cooled ions, but from pictures like that in Fig. 5, where crystal structures of the laser-cooled Ca^+ ions are clearly

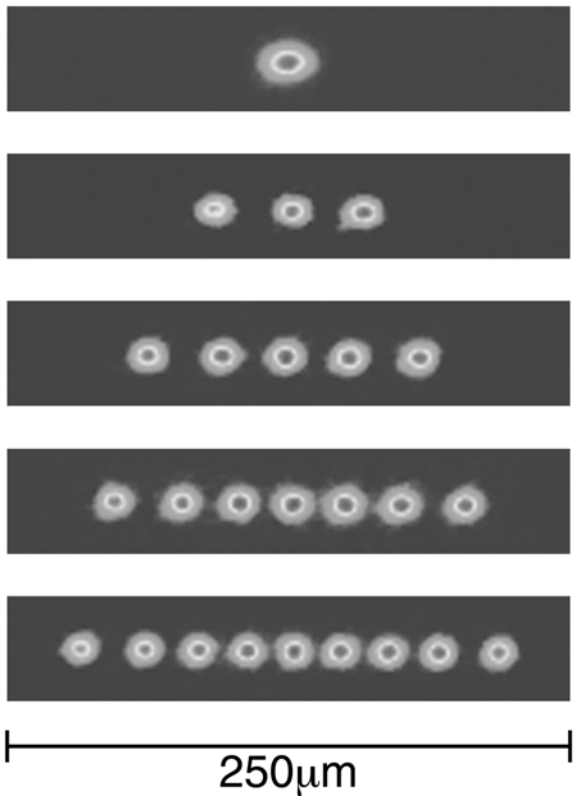

Fig. 3. Strings of cold Mg^+ ions containing 1, 3, 5, 7 and 9 ions, respectively.

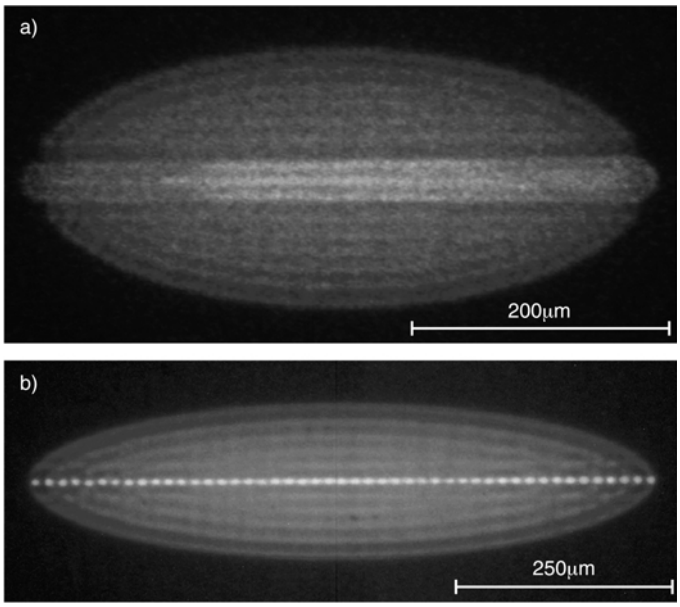

Fig. 4. Two-species Coulomb crystals (bi-crystals) composed of Ca^+ and Mg^+ ions. The blue color code and the red color code correspond to fluorescence from Ca^+ and Mg^+ ions, respectively.

observed, we may from simulations of such two-component ion plasmas conclude that the temperature cannot be above a few tens of milliKelvins.[15]

Since direct laser cooling may only be applied to a very limited number of singly-charged atomic ions, sympathetic cooling highly expands the possibilities of detailed investigation and manipulation of other ion species including molecular ions, as shown in Fig. 5, and multi-charged ions.[16] The spatial segregation of the ions with different masses makes it furthermore possible to, e.g., let a laser beam interact only with the ion species of interest.

4. Cold molecular physics

In the previous section it was shown that molecular ions produced by electron ionization could be trapped and cooled sympathetically to very low translational temperatures. With such a translational cold and spatially localized molecular ion target one can investigate various possibilities of cooling the molecular ion's internal degrees of freedom, i.e., rovibrational motions. From simple theoretical considerations[17] it is not believed that the Coulomb interaction will efficiently cool these degrees of freedom. Due to the multi-level structures of molecules, standard laser cooling cannot be applied. However, since the trapping time can be of the order of minutes or more, we plan to investigate various types of optical pumping schemes, which might bring the molecular ions into specific internal states. If this will be achieved, such cold state-specific molecular ions can be an interesting playground for performing various coherent-controlled molecular dynamics experiments.

Ion Coulomb crystals are furthermore interesting targets for reaction experiments. So far, we have studied a few reactions between laser-cooled ions in Coulomb crystals and neutral molecules. For larger crystals of Mg^+ ions we have, e.g., studied reactions with H_2 and D_2 gasses. Monitoring the change in the fluorescence rate from the remaining Mg^+ ions as a function of reaction time, reaction rates were determined. Furthermore, the reaction-product ions were deduced to be MgH^+ or MgD^+ in a non-destructive way by monitoring their radial position with respect to two Mg^+ isotopes in the Coulomb crystal.[15] We have very recently initiated experiments with the aim of studying reaction processes on the single molecule level. This may be achieved by, e.g., initially having a string of laser-cooled Ca^+ ions, which may individually be monitored by a CCD-camera as shown in Fig. 6a. By introducing a thermal O_2 gas at a very low pressure (typically $<10^{-9}$ torr) into the trap region, one may observe how the individual Ca^+ ions disappear by time and leave non-fluorescent spaces in the string, which from additional experiments are known to be CaO^+ ions, see Fig. 6b. Monitoring the fluorescence integrated over the whole string of ions as a function of time, one observes significant discrete changes in the fluorescence level revealing the fundamental discrete nature of the reactions. Wheth-

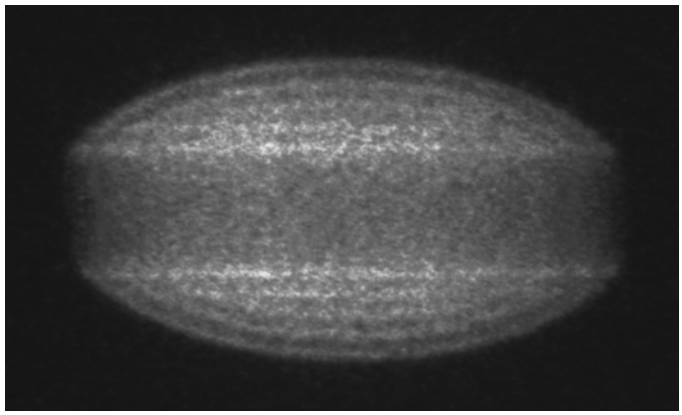

Fig. 5. A bi-crystal containing O_2^+ and Ca^+ ions. Since the O_2^+ ions are lighter that the Ca^+ ions, they form a cylindrical structure equivalent to the Mg^+ ions in Fig. 4a. This shows up in the picture as a hollow (non-fluorescing) cylindrical structure.

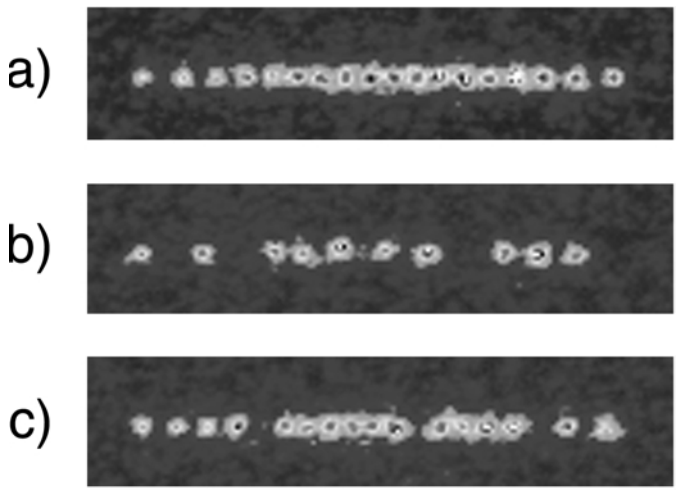

Fig. 6. a) A string of 18 laser-cooled Ca^+ ions. b) The same string as in figure a), but after reactions with thermal O_2 molecules, which lead to the production of non-fluorescing CaO^+ ions. c). The string of ions in Fig. b) after reactions with CO molecules. Reactions between CaO^+ ions and CO molecules lead to reappearance of fluorescing Ca^+ ions and CO_2. A few dark spaces are still present due to unknown impurity reactions.

er such "digital" information on the reaction processes will give fundamental new insight into chemical reactions is still unclear, but the technique could certainly be used in studies of extremely low-rate reactions, due to the long storage time of the cold ions. Since the product ions may typically be trapped and cooled, multi-step reactions may furthermore be studied. An example of the product of a two-step reaction is shown in Fig. 6c. Here the ion string of Fig. 6b has been subjected to a CO gas, which in reaction with CaO^+ leads to reappearance of fluorescing Ca^+ ions and CO_2. A few dark spaces are still present in Fig. 6c due to unknown impurity reactions.

5. Quantum optics

As mentioned earlier, few-ion Coulomb crystals have already proven to be interesting objects in the field of quantum optics. Below is given a short description of two projects within this field which are currently being developed in our group, and which will be essential parts of the research within a recently formed Quantum Optics Center funded by the Danish National Research Foundation.

A. Entangled ions and quantum gates

Entanglement is an intrinsic quantum phenomenon, which has no classical physics counterpart. In short, two physical observables are entangled if the wave function describing both quantities cannot be written as a direct product of parts describing each of the observables alone. An example of an entangled state of two identical atoms (numbered 1 and 2) could be: $\phi_1 \phi_2 + \varphi_1 \varphi_2$, where ϕ and φ are the wave functions describing different energy states of a single atom. Measuring the energy of just one of the atoms in an experiment would lead to a 50/50 chance of finding the atom in one of the two states, while when first a measurement has been performed on one atom, the energy state of the other atom is immediately fixed. Such very non-intuitive correlations have historically given rise to extensive debates, e.g., between Albert Einstein and Niels Bohr.

Besides being a very interesting intrinsic quantum phenomenon to study, multi-particle entangled states such as the one described above is a key element for creating quantum gates needed in quantum computing.

For several reasons Coulomb crystals in the form of ions on a string are excellent physical systems for studies of such multi-particle entangled states. First, the ions may be spatially localized to within 100 nm or less, and typically be separated by a distance of about 10 μm, see Fig. 3. This may provide individual access to the ions in a preparation process by means of focussed laser beams as well as in detection by high resolution imaging systems. Second, due to the extreme spatial localization, the trap vibrational modes can, together with near-resonant light, serve as a means for creating entangled states between the ions.[18,19] Third, the low-pressure trap environment leads to coherence times of up to several tens of seconds due to infrequent interactions with particles from the surroundings.

At present, several schemes have been proposed for creating multi-ion entanglement, see, e.g., Refs. 18 and 19. So far, however, only one group has been able to experimentally create and study quantum entanglement of ions.[13,14]

In our experiments, $^{40}Ca^+$ ions in a linear Paul trap will be the starting point. Apart from having appropriate internal electronic structures, the $^{40}Ca^+$ ions have previously been isotopic selectively loaded and Doppler laser cooled in a linear Paul trap in our group.[7] In order to carry out few-ion entangled state experiments, the necessary laser cooling to near the ground state of the trap potential will be achieved by Raman side band cooling using a Zeeman splitting of the ground level of the $^{40}Ca^+$ ion. Raman laser-pulses frequency tuned close to specific sideband transitions will be used to prepare the entangled states. The detection of the state of the ions will be performed in two steps. First, the part of the ion's wave function corresponding to one of the two Zeeman states of the ground level $^2S_{1/2}$ will be shelved in the $^2D_{5/2}$ level by a stimulated, rapid addiabatic transfer process. Second, lasers resonant with the $^2S_{1/2} - {}^2P_{3/2}$ and the $^2D_{3/2} - {}^2P_{3/2}$ transitions will be applied and presence or absence of fluorescence will reveal the state of the ion.

As mentioned above, creation of few-ion entanglement is very closely related to experimental implementation of quantum gates, and we plan to implement various types of universal gates between two ions as well as to study gates involving more ions useful for some specific quantum computing problems.

B. Quantum memory for light pulses

In order to make future quantum information networks operational, one will need to store quantum information temporarily. Since quantum information most likely will be transferred in the form of light, naturally, one seeks to find appropriate quantum memory devices for quantum states of light.

Recently, two independent experiments have demonstrated that a pulse of coherent light may be temporarily stored as a collective excitation of a cold atomic ensemble.[20,21] The idea behind these experiments suggests that not only coherent light pulses but also light with non-classical characters may be stored. Both experiments relied on a very large reduction of the group velocity of the light pulse as well as on the atomic medium being optically thick in the absence of a control laser beam. It has, however, been shown that these requirements may be relaxed if the atomic ensemble is placed within a high-finesse optical cavity,[22] where the light pulse, due to multiple reflections, will pass the atoms several times. In short, the proposal is based on dynamical impedance matching of the light pulse of interest to an optical cavity with absorption in the form of a stimulated rapid adiabatic transfer process in the atomic media. The absorption rate of the light carrying the quantum information is controlled via a strong classical laser pulse. By the adiabatic transfer process, the initial quantum information of the light pulse is transferred into collective coherent excitation of the atomic ensemble. By a time reversal of the classical light pulse, the initial quantum state of light will be regenerated as an output from the cavity.

In our view, the atomic sample in the optical cavity could advantageously be a Coulomb crystal of a few thousand ions. Ions in such crystals may have storage life times of tens of minutes, and have internal state coherence times of the order of a second or more. Using bi-crystals as the ones shown in Figs. 4 and 5, one species may be used as the storage medium, while the other may be directly laser cooled and sympathetically cool the "memory" ions.

In such experiments we plan to use $^{40}Ca^+$ ions as "memory" ions since they have suitable electronic transitions for the quantum storage of light in the near infrared where non-classical light sources are available. The $^{40}Ca^+$ ions will be cooled sympathetically by $^{44}Ca^+$ ions, which we plan to load in appropriate amounts us-

ing isotope-selective resonant two-photon ionization of an atomic beam.[7]

6. Summary

Apart from being fascinating objects to study on their own, it has been shown that ion Coulomb crystals may indeed be exploited in a large variety of experiments and more applications will probably appear in the future.

Acknowledgements

The author would like to thank all current and previous members of the Ion Trap Group, (homepage www.ifa.au.dk/iontrap/), who has taken or is taking part in the work presented. In particular, the extensive work by Kristian Mølhave, Liv Hornekær, and Niels Kjærgaard is greatly acknowledged. The presented work has been possible due to financial support from the Danish National Research Foundation, the Danish Research Council, and the Carlsberg Foundation.

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