Evidence for Highly Charged Ion Coulomb Crystallization in Multicomponent Strongly Coupled Plasmas

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Multicomponent non-neutral ion plasmas in a Penning trap consisting of Be^+ and highly charged Xe ions, having different mass-to-charge ratios than Be^+, are cooled to form strongly coupled plasmas by applying a laser-based collisional cooling scheme. The temperature of the plasma was determined from a Doppler broadened transition in Be^+. For the Xe ions, which are centrifugally separated from the Be, the Coulomb coupling parameter was estimated to be $\approx$1000. Molecular dynamics simulations of the ion mixture show ordered structures, indicating crystallization of the Xe.

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We report the formation of strongly coupled, non-neutral highly charged ion (HCI) plasmas in Retrap, a cryogenic Penning trap. The estimated temperature and density permits the conclusion that Coulomb crystallization of the HCIs has been achieved. This conclusion was confirmed by molecular dynamics (MD) simulations of related plasmas. The study of these and similar [1–3] plasmas refines our understanding of basic physics in various areas. As examples, (i) laser accessible fine and hyperfine magnetic dipole transitions (e.g., in H-like high-Z ions) [4] can potentially be measured to high precision in cold HCI plasmas [5,6]. This allows the measurement of nuclear properties with accuracies of high resolution laser spectroscopy in high-Z nuclei. (ii) White dwarf star interiors consist of mixed strongly coupled plasmas in the liquid and solid phase [7]. An analog for such star matter can be produced and investigated, leading to a better understanding of energy release mechanisms in such stars. (iii) Using multiply, instead of singly, charged ion plasmas for ion trap based quantum computing schemes could have advantages due to stronger ion-ion coupling, larger inter-ion distances, narrower laser transition linewidths, and larger separation of sideband resonances due to higher cyclotron frequencies.

Earlier experiments on strongly coupled ion plasmas and Coulomb crystals were done with singly charged ions using laser cooling in ion traps [8–10]. One component plasmas are called strongly coupled if the Coulomb coupling parameter Γ fulfills the following condition:

$$\Gamma \equiv \frac{q^2}{4\pi\epsilon_0 a_0}\frac{1}{k_B T} \geq 1\,, \qquad (1)$$

where $a_0 = [3/(4\pi n)]^{1/3}$ is the Wigner-Seitz radius, q is the charge of one particle, T is the temperature, n is the density, and ϵ_0 and k_B are the permittivity of free space and the Boltzmann constant, respectively. The plasma is expected to crystallize if $\Gamma \geq 172$ [11].

Since laser cooling techniques cannot generally be applied directly to HCIs due to a lack of suitable transitions accessible with lasers, a cooling scheme combining laser cooling of $^9Be^+$ ions and sympathetic cooling [12] of HCIs by Coulomb collisions with the cold Be^+ ions has been chosen. In a Penning trap, theory predicts centrifugal separation at low temperature of ion species with different mass-to-charge ratios and no centrifugal separation for ion species with matching mass-to-charge ratios [13]. The present experiment studied a mixture of $^9Be^+$ and ^{136}Xe ions with charge states as high as $^{136}Xe^{44+}$.

The HCIs are produced by successive electron impact ionization in an electron beam ion trap [14]. The extracted ion bunch [15] (typical: 1.5×10^4 Xe^{44+} ions in a 6 μs pulse at 2 keV/u) is guided with static electric and magnetic ion optics to Retrap [16]. At Retrap the ions are decelerated upon entering a tube biased at approximately the extraction potential of about 6.5 kV, thereby reducing the ion kinetic energy to $\approx$70 eV/u. The tube potential is then rapidly ($\leq$50 ns) switched to ground so that the ions are not reaccelerated upon exiting the tube. Some of these ions can then be caught in the hyperbolic trap of Retrap by briefly switching the voltage on the capture electrode (Fig. 1) to ground, allowing the ions to enter the trap. For this experiment the magnetic induction in Retrap was set to $B = 4$ T and for most of the observations the potential V_0 between the ring and the end cap electrodes was set to 540 V. The hyperbolic shape of the electrodes produces a quadratic dc potential along the z axis, and, in addition, allows for a compact design. The motion of an ion with charge q and mass m in such a potential is a superposition of three independent harmonic oscillations: an axial oscillation along the magnetic field lines with the angular frequency ω_z (dependent on V_0) and two radial oscillations

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with the angular frequencies ω_+ and ω_-. The radial angular frequencies are related to the cyclotron angular frequency ω_C in the following way:

$$\omega_+ = \omega_C - \omega_- = \frac{q}{m} - \omega_- . \qquad (2)$$

The magnetron motion of a single ion with the angular frequency ω_- is caused by an $\vec{E}_r \times \vec{B}$ drift, where $\vec{E}_r$ is the radial component of the quadrupole electric field. This frequency is modified by the space charge potential of the ion cloud when more particles are trapped and can therefore give information about the density of the cloud. The ring electrode is split into four sectors and has six holes. Two opposing holes each were used to pass a cooling and a probe laser beam and the two remaining holes permit scattered photon detection using a photomultiplier tube (PMT) and, simultaneously, a cryogenically cooled charge-coupled-device (CCD) camera [17,18]. PMT signal and CCD images are continuously recorded during the experiment.

Be^+ and Be^{2+} ions are produced in a metal vapor vacuum arc ion source. After momentum analysis by a 90° bending magnet, Be^+ ions are decelerated and caught in the hyperbolic trap of Retrap [18]. Laser cooling of the Be^+ ions [19] in Retrap is done by shining a laser beam through the trap, perpendicular to the magnetic field. The laser beam, initially tuned several GHz to the red side of the Be^+ $2s\,^2S_{\frac{1}{2}}(m_j = -\frac{1}{2}, m_I = -\frac{3}{2})$-$2p\,^2P_{\frac{3}{2}}(m_j = -\frac{3}{2}, m_I = -\frac{3}{2})$ transition at $\lambda \approx 313$ nm, reduces the temperature of the ion cloud.

Once the Be^+ ions are cold (indicated by the fluorescence signal on the PMT dropping almost to background), a pulse of HCIs is caught into the Be plasma.

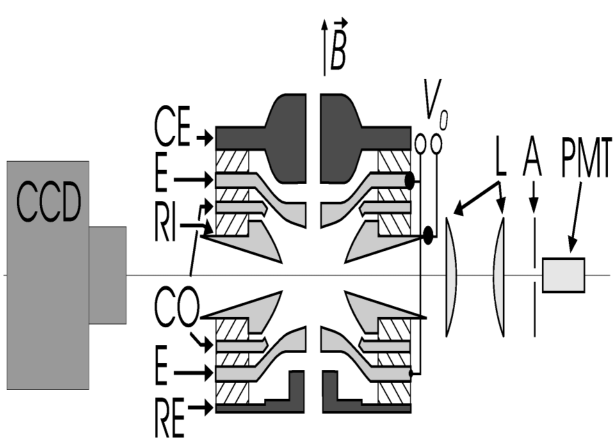

FIG. 1. Schematic of the hyperbolic trap used in the present experiments. CCD is a cryogenically cooled CCD camera, CE is the capture electrode, E are the end cap electrodes, RI is the ring electrode, CO are the compensation electrodes, RE is the release electrode, L are optical lenses, A is an aperture to block stray light, and PMT is a photomultiplier tube. V_0 is the voltage applied between E and RI to create the quadratic electric potential along the magnetic field.

To avoid releasing the Be plasma, a potential difference of $V_0 \approx 300$ V is maintained between the ring and the end cap electrodes. The injection of the Xe^{44+} ions heats the Be^+ ions, due to the introduction of the "hot" Xe ions and, to a lesser degree, the switching of the electrode potentials. As a result, an increase of the fluorescence signal from the Be^+ ions on the PMT is observed. The time dependence of that signal follows the heating and recooling of the mixed plasma. The cooling of the HCIs occurs via Coulomb collisions with the continuously laser cooled Be^+ ions. The fluorescence signal in most cases falls to a steady state near background level after about 20 s, indicating an equilibration time between the two species of significantly less than this time scale. As the mixed ion plasma reaches a thermally steady state, both ion species will assume temperatures, which are determined by the laser cooling rate, the sum of all heating rates (e.g., electrical noise, trap imperfections) and the collisional coupling of the two species. Reduction of the cooling laser detuning can lower the Be^+ temperature into the sub-Kelvin regime. At these and higher temperatures the CCD images show a dark gap, indicating a clear separation of the Be^+ ions from the ions with a different mass-to-charge ratio. If Xe^{15+} ions, with a $\frac{m}{q}$ nearly equal to Be^+, are injected, no separation is seen. This measurement also shows that the numbers of any background ions associated with the separation, other than Xe ions, are negligible.

To probe the trap content nondestructively, radio frequency voltages were applied to opposing 90° sectors of the ring electrode to excite the modified cyclotron motion ω_+. The excitation frequency was scanned from 14 to 19 MHz across the expected range of resonances of the HCIs. As an ion species is excited, the Be^+ plasma is heated due to Coulomb collisions and therefore the photon scattering rate increases. The number of detected photons scattered by the cloud as a function of the excitation frequency is shown in Fig. 2a. The equidistant peaks are attributed to different charge states of Xe (Xe^{32+}–Xe^{40+}). The peak at 15.1 MHz might be caused by He^+ ions, which can be created by charge exchange between Xe^{q+} and neutral He. The detection measurement of these ions was started about 2000 s after the trap had been loaded with the HCIs and lasted 1800 s. Since the lifetime of Xe^{44+} due to charge exchange with the residual gas was on the order of 100 s, after 2000 s the trap contains a broad distribution of lower charge state Xe ions.

If the resonance frequencies are plotted as a function of the charge-to-mass ratio of the ion which is expected to cause the resonance (see Fig. 2b), it can be seen that all of the points (but one) are on a straight line within their error bars. A linear regression returns according to Eq. (2) the magnetic induction $B = 4.15(2)$ T and the angular frequency $\omega_- = 2\pi \times 584(70)$ kHz related to the rotation of the cloud $\omega_{\rm rot}$ [20]. For an axial potential well depth of $V_0 = 540$ V the minimum rotation frequency of the cloud is $\omega_{-,\min} = 2\pi \times 460$ kHz. Assuming the fluid model

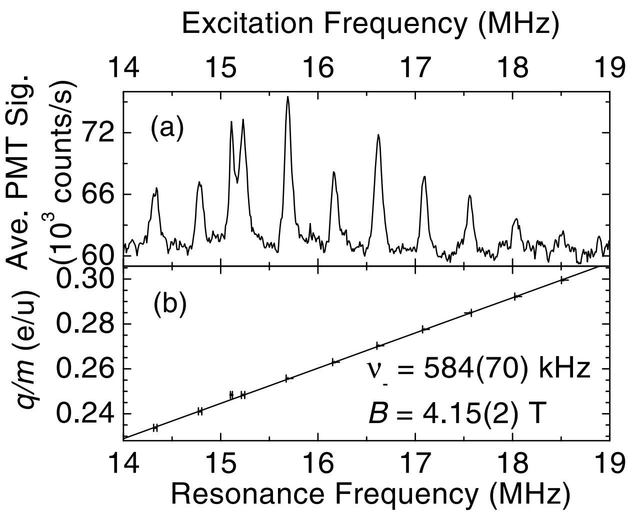

FIG. 2. The number of 313 nm photons scattered by the Be^+ of the mixed plasma as the modified cyclotron motion of different ion species is excited. (a) Ions with different mass-to-charge ratios are excited sequentially and heat the Be^+ which is then cooled by the laser, indicated by the higher scatter rate. (b) Fitting the positions of the resonances as a function of the charge-to-mass ratio of the associated ions with a straight line, yields the magnetic induction B and frequency ν_-, which is nearly the rotation frequency of the cloud. The resonance, believed to be caused by He^+, does not fit the model.

is valid it is possible to calculate a density with the relation [19]

$$n = \frac{3\epsilon_0 m \omega_{\rm rot}(\omega_C - \omega_{\rm rot})}{q^2}. \tag{3}$$

For Xe^{35+} and $\omega_{\rm rot} = \omega_{-,\min}$ the minimum density $n_{\min} = 3.7 \times 10^{13}\ \mathrm{m}^{-3}$ is determined.

The temperature of the Be plasma was determined in the following way: A second, much weaker probing laser beam was scanned across the $2s\,^2S_{\frac{1}{2}}(m_J = -\frac{1}{2}, m_I = -\frac{3}{2})$-$2p\,^2P_{\frac{3}{2}}(m_J = +\frac{1}{2}, m_I = -\frac{3}{2})$ transition depopulating the ground state of the cooling transition in a Be^+ one component plasma [8]. This caused a decrease of the scatter rate at resonance, allowing the temperature to be deduced from the Doppler width. The temperature was calculated from a Voigt profile analysis neglecting saturation broadening but correcting for a finite probe beam waist and is therefore an upper limit for the real temperature. The lowest measured temperature was $T_{\rm Be} = 100(100)$ mK.

By measuring the equilibration temperature of a Be^+ plasma as a function of the cooling laser detuning, a constant ratio of 1.18(5) was determined between the laser detuning and the Doppler width of the scattering transition. Using the overlap of the cooling laser with the ion cloud (3×10^{-3}), laser power (1.1 mW), laser polarization (linear), laser beam diameter (180 μm), and an expression for laser cooling power [8], it was possible to determine the constant overall heating rate to be 770(80) $\frac{\mathrm{K}}{\mathrm{particle \cdot s}}$. The scattered signal level is consistent with a rate less than $10^3 \frac{\mathrm{K}}{\mathrm{particle \cdot s}}$. If this heating is due to electrical noise we expect this rate to scale no faster than the motional frequencies ($\frac{q}{m}$ at worst). This order of magnitude heating rate, in conjunction with the Be^+ temperature measurements and molecular dynamics simulations made it is possible to estimate the Xe temperature. A Verlet integration of the equations of motion was performed for each particle with mass m_i and charge q_i, moving in the field of the $N-1$ other particles with charges q_j, confined by the electric trap potential $\Phi_T(\vec{r})$ and the magnetic induction $\vec{B}$:

$$m\ddot{\vec{r}}_i = \frac{q_i}{4\pi\epsilon_0} \sum_{\substack{j=1 \\ j \neq i}}^{N} \frac{q_j \vec{r}_{ij}}{r_{ij}^3} + q_i(\dot{\vec{r}}_i \times \vec{B}) - q_i \vec{\nabla}\Phi_T(\vec{r}_i), \tag{4}$$

with $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ being the distance between the ith and the jth particle. The simulation was performed for a small number (typically 64 Xe^{44+} and 64 Be^+) of Coulomb interacting ions in a Penning trap with equivalent parameters to the described experiment. The cyclotron motion was not neglected. A more detailed description of these MD simulations will be presented [21]. The Xe ions were cooled only by interacting with the Be^+ ions. Using the experimentally determined heating rate of 1000 $\frac{\mathrm{K}}{\mathrm{particle \cdot s}}$, the Xe ions were heated to 1.1 K (Fig. 3) if the Be^+ temperature was maintained at 1 K. Since in the experiment there were typically 10^4 times more Be^+ ions and about 5 times more Xe ions in the trap than in the simulations, these results represent upper limits. The existence of charge exchange and the distribution of ion charge states should also provide better heat transfer between the remaining highest charge state ions and the laser cooled Be^+ ions. A snapshot of

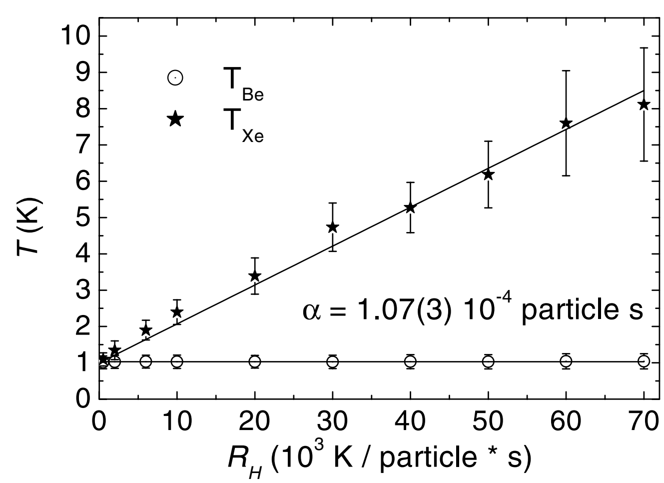

FIG. 3. Result of different MD simulations of a cold mixture of Be^+ and Xe^{44+} with different heating rates. The Be^+ are kept at 1 K and the Xe temperature rises to a fixed value $T_{\rm Xe}$. This equilibrium temperature is shown as a function of the heating rate R_H. The line is a linear fit to the data and α is the slope.

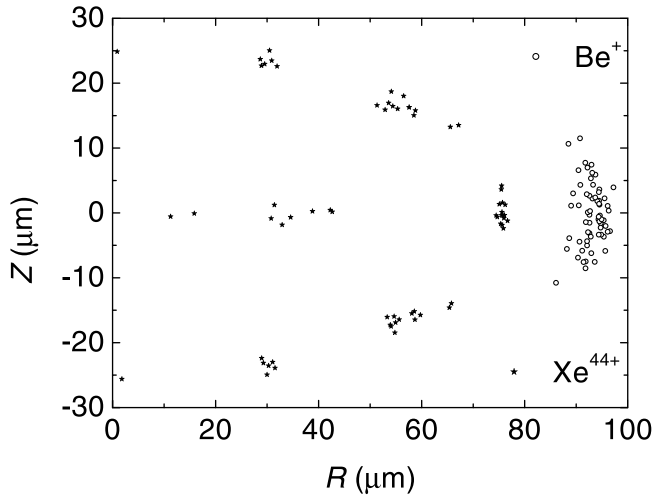

FIG. 4. Final configuration of ions after a molecular dynamics simulation of a mixture of 64 Be^+ ions (hollow circles) and 64 Xe^{44+} ions (solid stars). The ion positions are displayed as a function of radius R and the axial coordinate z. The Be ions were forced to an axial temperature of 1 K and the Xe ions equilibrate to the same axial temperature within the error bar. The magnetic field is parallel to the z axis. Ordered structures in the Xe^{44+} ions can be seen.

the simulated trap content showing ordered structures [1] is shown in Fig. 4.

The sympathetic cooling of highly charged Xe ions with laser cooled Be^+ ions has been demonstrated for the first time. The temperature and the density of a cold Xe plasma were determined to be $T_{\mathrm{Xe}} \leq 1.1(2)$ K (for $T_{\mathrm{Be}} \leq 1$ K) and $n_{\min} \geq 3.7 \times 10^{13}$ m^{-3}, respectively. These parameters indicate the formation of a strongly coupled plasma consisting of highly charged ions with a Coulomb coupling parameter of $\Gamma \geq 1000$, assuming an average charge state of $q = 35$. This coupling suggests the conclusion that Coulomb crystallization for highly charged ions has been achieved; however, a disordered but frozen glassy state cannot be ruled out. A direct observation of the existence of an ordered structure of HCIs was not possible at this time, but fine and hyperfine structure transitions of certain HCIs can be accessed with a laser and might make future imaging of crystalline structures feasible.

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