Direct temperature determination of sympathetically cooled large \(^{113}\text{Cd}^+\) ion crystal for microwave clock

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Abstract

This paper reports the direct temperature determination of sympathetically cooled \(^{113}\text{Cd}^+\) ions with laser-cooled \(^{24}\text{Mg}^+\) in a linear Paul trap. The sympathetically cooled ion species distribute in the outer shell of the large ensembles containing up to \(10^5\) ions. With optimized parameters, the minimum temperature of sympathetically cooled \(\text{Cd}^+\) ions is measured to be tens mK. These results indicate promising performance for microwave atomic clocks with 2 magnitudes lower (from \(10^{-14}\) to \(10^{-16}\)) second-order Doppler frequency shift and suppressed Dick effect.

Introduction

Charged particles stored in ion traps feel stronger bound compared to neutral atoms in the magneto-optical traps. With the laser cooling (LC) techniques, trapped ions could be translationally cooled to milli-Kelvin only by Doppler cooling, or cooled to vibrational ground state at micro-Kelvin by sideband cooling [1][2]. Hence, trapping ion technique is widely used in precision measurements, such as frequency metrology[1], mass spectrometry [3], precision spectroscopy [4], quantum information processing [5], physical constants measurement [6], and chemical physics [7]. Frequency standards based on ion traps occupy important roles in frequency metrology. Especially, microwave frequency standards based on trapped ions have good prospects in the applications of compact atomic clocks, deep-space navigation, ultra-stable timekeeping and space-borne clocks [8].

Microwave clock based on laser-cooled \(^{113}\text{Cd}^+\) has obtained short-term frequency instability of \(6.1\times10^{-13}/\sqrt{T}\) and frequency uncertainty of \(6.6\times10^{-14}\) [9]. Nevertheless, it still doesn’t reach the anticipated performance [10]. This kind of microwave frequency standards based on laser-cooled ions mainly suffer from the Dick effect due to the dead time of laser cooling, and the second-order Doppler frequency shift (SODFS) due to the temperature rising of ions during clock interrogation [11]. The technique of sympathetic cooling (SC) is promising to overcome both of these two limits. Sympathetic cooling is a method to cool the translational motion of the target atomic or molecular ions by the coolant ions via mutual Coulomb interaction, and it was first proposed on isotopic ions in a Penning trap [12]. It has been demonstrated that laser-cooled alkaline-earth or alkaline-like-earth metal ions, like Be\(^+\), Mg\(^+\), Ca\(^+\), Ba\(^+\), and Yb\(^+\), as coolants could cool down other ions in a Penning or Paul traps. Especially for the ions without suitable direct cooling lasers, sympathetic cooling is an irreplaceable method. Doppler limit (< \(1\) mK) was reached by small ion crystals consists of about 10 ions, which was verified by spatial thermometry [13].

We proposed to use \(^{24}\text{Mg}^+\) ions as the coolant to sympathetically cool the large Cd\(^+\) cloud [14]. Compared to other ions, only one laser is needed to cool \(^{24}\text{Mg}^+\) ions.
The big mass difference between Cd\(^+\) and Mg\(^+\) ions make a large separation between these two kind crystallized ions, which is helpful to reduce the light shift caused by the cooling laser of Mg\(^+\). To estimate the SODFS, the exact temperature of ions has to be obtained. One can measure the Doppler broadening of a special transition to determine the ions’ temperature [15][16][17]. However, for the ions without suitable direct cooling lasers, it is difficult to directly measure their temperature. There is an indirect method to derive ions’ temperature by comparing the ions’ CCD photographs to the simulated statistical images generated by Molecular Dynamics (MD) [18]. Nevertheless, the accuracy of this method is limited by the hypotheses of simulation models and computational accuracy. In the published literature, there are few reports about the direct measurement of temperature for the SC ions. For example, in Penning ion traps, D. J. Wineland and his coworkers measured the temperature of \(^{199}\text{Hg}^+\) ions SC by \(^9\text{Be}^+\) ions to 0.4-1.8 K [19], and H. Imajo et. al. demonstrated SC Cd\(^+\) ions by isotopic Cd\(^+\) to 1 K [20]. In Paul trap, the direct temperature determination was achieved in Ca\(^+-\)Mg\(^-\) bi-crystals and obtained 40 mK of inner Mg\(^+\) core [21]. The reported experiment also showed that it was very challenging to sympathetically cool target ions by the coolant ions with a large mass difference due to the limited cooling efficiency [21].

In this letter, we report the direct measurement of the temperature of \(^{113}\text{Cd}^+\) ions sympathetically-cooled by \(^{24}\text{Mg}^+\) ions for developing a high-accuracy microwave atomic clock. To our knowledge, it is the first direct measurement of the temperature of sympathetically-cooled ion crystal with a large number of ions up to 3.3×10\(^5\) and a large mass ratio (4.71) to sub-Kelvin in a Paul trap. Compare to the ion clouds with LC ions, the SODFS is depressed 2 orders of magnitude [9]. And the Dick effect is also reduced owing to the short dead time since no additional cooling step is needed. It is a milestone to build high-performance microwave atomic clocks based on sympathetically cooled ions. This result can also be treated as an upper limit for the translational temperature of the outer shell SC ions to refine the MD simulation model.

II. EXPERIMENT

The experiment setup is similar to previous reports [4, 14, 15] with upgrades, and additional parameters are shown in Appendix I. The linear quadrupole Paul trap made of oxygen-free copper consists of four parallel cylindrical electrodes with a radius of \(r_e = 7.1\) mm. Each electrode is divided into three segments, and the lengths of the segments are 20, 40 and 20 mm, respectively. The minimum distance from the trap center axis to the surface of the electrodes is \(r_o = 6.2\) mm. The ratio of \(r_e\) and \(r_o\) is close to the optimized value to an ideal quadrupole field [23]. A radio frequency (RF) voltage \(V_{\text{RF}}\) is applied to one pair of diagonal electrodes, and the other pair is grounded. The driving frequency is \(\Omega/2\pi = 2\) MHz. A static voltage \(U_{\text{end}}\) is added to endcap electrodes, which can be adjusted between 0 and 120 V to manipulate the shapes of the Coulomb crystals. With \(V_{\text{RF}} = 280\) V the trapping parameters [24] of the well-known Mathieu equation are \(q_{\text{Mu}} = 0.03\) and \(q_{\text{Cd}} = 0.15\), and the trap depths are \(d_{\text{Mu}} = 2.11\) eV and \(d_{\text{Cd}} = 0.45\) eV, respectively.

To manipulate Cd\(^+\) and Mg\(^+\) ions at the same time, two frequency-quadrupled lasers with wavelengths of 214 and 280 nm counter-propagate along the trap axis. The diameters of the two laser beams are both 1 mm, and the power is 6 mW/mm\(^2\) for 280 nm and 1 mW/mm\(^2\) for 214 nm, respectively. The frequencies are stabilized with high precision wavemeters to the transition lines of \(|3s^2S_{1/2} \leftrightarrow 3p^2P_{1/2}>\) of \(^{24}\text{Mg}^+\) ions and \(|5s^2S_{1/2} \leftrightarrow 5p^2P_{3/2}>\) of \(^{113}\text{Cd}^+\) ions. The natural line-width of the D\(_1\) line for \(^{24}\text{Mg}^+\) is
2π × 42.7 MHz which gives a minimum Doppler temperature of 1.0 mK [25], while the natural line-width of the D₂ line for ¹¹³Cd⁺ is 2π × 60.1 MHz corresponding to 1.4 mK [26]. The images of ions are captured by an electron multiplying charge coupled device (EMCCD) camera. The camera system mounted on a precision motion stage consists of a home-made lens, tunable pinhole, and ultraviolet (UV) filters. The magnification factors of the lens can be adjusted from 4 to 8. The exposure time is set to be 0.3 s for one shot. In addition, a photomultiplier tube (PMT) with optional filters at the opposite side of the trap axis is applied to monitor the fluorescence intensity.

To load ions, neutral atoms are evaporated from pure metal (Mg metal is natural abundance, and Cd metal is ¹¹³Cd isotopic enriched) in homemade ovens. Magnesium atoms are ionized by electron bombardment, and Cadmium is ionized by resonance-enhanced two-photon ionization is by a fourth harmonic generation (FHG) laser at 228 nm. Thus photo-ionization for ¹¹³Cd⁺ is adopted to avoid disturbing and heating the loaded Mg⁺ ions [27]. The Mg⁺ ions are first loaded, and Cd⁺ is ionized and loaded then. The photo-ionization has advantages of steerable high loading efficiency, isotopic selection and avoidance of the stray electric field due to residual charge from E-gun. Concerns should be taken on appropriate ionization energy to avoid ²⁴Mg²⁺, whose ionization energy is 22.7 eV [15],[28]. While loading Mg ions, the V_RF is set to a higher value than the value during normal detection to get a deeper potential depth for loading more ions. The compensation voltages applied to the electrodes have to be tuned carefully after loading to obtain symmetric ion crystals. During the loading time, quite a part of ²⁴Mg⁺ ions was converted to ²⁴MgH⁺ dark molecular ions through chemical reactions with H₂ molecules involved in background gas [29]. Nevertheless, the rates for ¹¹³CdH⁺ production are quite low.

![Schematic of a typical temperature determination sequence. (1) Doppler cooling of Mg⁺. (2) tuning compensated voltages applied to the electrodes to decrease RF heating. (3) loading and cooling for Cd⁺. (4) measurement.](image)

To measure the temperature of sympathetically cooled Cd⁺ ions, the Doppler broadening of the transition of ⁵s²S₁/₂, F=1→⁵p²P₃/₂, F=2 is measured. During this measurement, the power density of 214 nm laser is set to 20 μW/mm² to avoid cooling or heating the ions. The frequency of 214 nm laser is scanned around the resonant frequency with a range of 2 GHz. The measured line profiles are fitted by a Voigt function. The Lorentz linewidth of the Voigt function is set as the natural linewidth of the D₂ transition of ¹¹³Cd⁺ ions, which is 60.13 MHz (2.647 ± 0.010 ns) [26]. The fitting deduced Gaussian linewidth represents the velocity distribution of SC Cd⁺ ions. Thus, the temperature can be calculated by
\[ T = \frac{Mc^2}{8k \ln 2} \left( \frac{\Delta \nu_G}{\nu_0} \right)^2, \]  

where \( M \) is the mass of \(^{113}\text{Cd}^+\) ions, \( c \) is the speed of light, \( k \) is the Boltzmann constant, \( \Delta \nu_G \) is the Gaussian linewidth, and \( \nu_0 \) is the resonant frequency of \( D_2 \) transition of \(^{113}\text{Cd}^+\) ions. Typical Voigt fitting of the LIF spectra is shown in Fig. 2., and the intensity of the LIF signals is proportional to the resonant ion numbers.

![Fluorescence spectrum of sympathetically cooled \(^{113}\text{Cd}^+\) ions. The frequency of probing laser scans past the \( |5s^2S_{1/2}, F=1\rangle \leftrightarrow |5p^2P_{3/2}, F=2\rangle \) transition of \(^{113}\text{Cd}^+\) ions with a range of 1 GHz. The laser power is set low enough to avoid cooling or heating effects. Experimental data are marked by squares and circles for \( U_{\text{RF}} \) at 185 V and 200 V, respectively. The curves are fitted by Voigt function with a Lorentz linewidth of 60.13 MHz, which is the natural linewidth of the \( D_2 \) transition of \(^{113}\text{Cd}^+\) ions. The obtained temperature of \(^{113}\text{Cd}^+\) is 31 mK and 136 mK, respectively. The inset shows the fitting of 31 mK result.](image)

### III. RESULTS AND DISCUSSIONS

There are two types of ion motion in linear Paul quadrupole trap, the secular-motion, and the micro-motion. The former one is an intrinsic motion under the time-averaged harmonic potential and could be suppressed by cooling techniques. The latter one is driven by the RF field and increases with the distance raise from the center of the trap. We minimize the micromotion by making the symmetry axis of the ion crystal insensitive to the trapping voltage [30]. Ions with different charge-to-mass ratios exhibit different displacements due to the mass dependence of the pseudopotential. In large Coulomb crystals, such displacements can result in a complete separation radially [18]. And the different radiation pressure of the cooling lasers will segregate ions axially.

To estimate the total number of ion crystals, the whole configuration of the multi-component ion cloud is shown as Fig. 3. The diameter of the 214.5 nm laser beam is of 1 mm. The diameters of \( \text{Cd}^+ \) ion clouds are larger than the laser beams'. Taking account of the relatively weak axial confinement field, the crystal has a large aspect ratio. Thus, only a part of the whole \( \text{Cd}^+ \) ions could be imaged due to the limited EMCCD area. In Fig. 3, the photograph of the whole ion cloud is obtained by picture stitching of 12 pieces of EMCCD photos. And due to the chromaticity of the lens at 214.5 nm and 280 nm, the focus positions are different for \( \text{Mg}^+ \) and \( \text{Cd}^+ \) ions. This effect has been taken into account during picture stitching.
Fig. 3 Photographs of Cd\(^{+}\) and Mg\(^{+}\) ions captured by the EMCCD with 0.3 s exposure time and UV filters respectively. Each picture is completed by stitching of 12 pieces of EMCCD photos taken separately. (a) is a combination of (b) and (c), \(U_{RF} = 240 \text{ V}\) and \(U_{end} = 10\text{ V}\). (b) and (c) represent Mg\(^{+}\) ions and Cd\(^{+}\) ions separately. These two kinds of ions are pushed to different sides because of the unidirection of laser incidence. The little difference of magnification factor caused by the chromaticity is taken account. The right end part of (d) is obtained by overlying 10 photos with different positions of Cd\(^{+}\) cooling laser beam. The profile of the entire ion cloud is marked by the dotted line. The ion number is estimated to be \(2.8 \times 10^3\) for Cd\(^{+}\) ions, and \(5.8 \times 10^4\) for Mg\(^{+}\) ions.

The numbers of the two visible atomic ions were determined by the size of ion crystal from CCD images and the estimated ion number density. Using the zero temperature charged liquid model, the number density \(n_0\) under the pseudopotential approximation is derived by

\[
n_{0_i}(r, z) = n_i = \frac{\varepsilon_0 V_{RF}^2}{M \Omega^2 r_0^2},
\]

where \(\varepsilon_0\) is the permittivity of the vacuum, \(V_{RF}\) is the voltage applied on the electrodes, \(M\) is the mass of ions, \(k\) is the Boltzmann constant, \(\Omega = 2\pi \times 2.02 \text{ MHz}\) is the trap driving frequency, and \(r_0\) is the minimum distance from the trap center to the electrodes’ surface. It is derived that \(n_{0_{Mg}} = 7.3 \times 10^{11} \text{ m}^{-3}\) and \(n_{0_{Cd}} = 1.6 \times 10^{13} \text{ m}^{-3}\) with \(V_{RF} = 280 \text{ V}\). For estimating the volume of ion crystals, the Mg\(^{+}\) crystal in the core was approximated to a cylinder, and the Cd\(^{+}\) crystal to an ellipsoid shell. The species of the dark ions were not clarified and calculated. According to Fig. 2, the three radii of the ellipsoid shell are 12.9/2 mm, 0.89 mm and 0.89 mm, respectively. And the length of the inner cylinder is 12.9 mm and radius is 0.14 mm. The gap between the inner surface of the ellipsoid shell and the outer surface of the inner cylinder is proportional to the root of the mass ratio and the inner radius \([15][31]\). It is obtained to be 0.16 mm approximately. Thus, the number of ions is estimated to be about \(N_{ion} = 3.3 \times 10^5\), \(N_{ion,Mg} = 5.8 \times 10^4\), and \(N_{ion,Cd} = 2.8 \times 10^7\). It was difficult to determine the accurate number of each ion species under the present conditions due to the large volume of ions.

To insight the separation of two species of ions, Figure 4 shows complete spatial separation between \(^{24}\text{Mg}\(^{+}\) and \(^{113}\text{Cd}\(^{+}\) ions without lens filters. Fig. 4 (a) and (b) show images when the 214.5 nm laser is tuned to bright the bottom and upper part of Cd\(^{+}\) ions respectively. In Fig. 4 (c), the profile of Mg\(^{+}\) ions in the core is clear with 3 s integration time, but the profile of Cd\(^{+}\) ions is much fuzzy due to the chromatic aberration. And Fig. 4 (d) is the image of small Mg\(^{+}\) ion crystal under well crystallization condition.
Fig. 4 EMCCD images of the multi-ion crystal with 3 s exposure time. (a), (b) and (c) are taken without UV filters to image two kinds of ions simultaneously. The red arrow indicates the propagating direction of the 214 nm laser, and the blue one indicates the 280 nm laser. (c) shows the right end part of the ions crystal shown in (a) and (b). And (d) is the unambiguous EMCCD image of Mg$^+$ ion crystal with 280 nm filter.

Although the first-order Doppler effect is suppressed by reaching the Lamb–Dicke criterion, the second-order Doppler shift (SODFS) is still not negligible for microwave clocks based on ion trap. Lower temperature helps reduce SODFS. The dependence of the temperature of outer shell SC ions on variable trapped parameters is measured and shown as Fig. 5. Each point is the average of three times of measurement to ensure valid and reaching equilibrium situation. After preparing a large multi-component crystal, the electrodes’ voltages tuned in a small range. And changing voltages in two opposite ways was used to avoid heating effects. The results indicated that ion temperature varies with different operating parameters. The maxim measured temperature is less than 200 mK, and the minimum temperature is around tens mK. Meanwhile, the number of total ions and SNR decreases with time under the same intensity of the probe laser. The temperature difference and the decrease are influenced by heating of inhomogeneous field, chemical reaction with residual gas and oscillations caused by voltages changing. And it also implies that sympathetic cooling of outer shell can’t completely counteract the RF heating and the collision of the background gas. The signal becomes stronger under large $V_{RF}$ voltages due to the number density of ions increasing with $V_{RF}$ voltages.
Fig. 5 The RF amplitudes $V_{RF}$ influence on sympathetic cooling. The temperature of Cd$^+$ ions while scanning the RF Voltages at different endcap voltages. (a), (b) and (c) show the variation of temperature with RF voltage, while (d), (e) and (f) show the signal intensity under these relevant detections.

The ion temperature dependence to endcap voltages is investigated as shown in Fig. 6. The ions’ temperature reached a minimum value at 60 V. The same phenomenon was observed in the experiment of small ion crystal (contains 34 Ca$^+$ ions) [32]. The spatial radius of every single Ca$^+$ ion also reaches the minimum at a relatively low voltage. Moreover, the number of stable trapped ions will decrease with rising endcap voltages, leading to the intensity of signal decrease as shown in Fig. 6.

Fig. 6 The Endcap voltage amplitudes $U_{end}$ influence on sympathetic cooling with the RF voltage $V_{RF}$ is fixed at 200 V.

The accurate evaluation of the SODFS would require MD simulations from which the velocity distribution of the ions could be extracted. But it can be roughly estimated using the simple models used for microwave clocks. The total SODFS due to both the
secular motion and the micromotion of the trapped ions is estimated as [33]

\[
\Delta f = f_{0, \text{SODFS}} = \left( \frac{-E_{\text{Kinetic}}}{M c^2} \right) = -\frac{3 k T}{2 m c^2} \left[ 1 + \frac{2}{3} \left( N_d^K \right) \right],
\]

where \( f_0 \) is the central frequency of \( ^{113}\text{Cd}^+ \) ion’s transition, \( N_d^K \) is a coefficient of the SODFS due to the micromotion averaged across the whole ion cloud [34]. Hence, the SODFS is less than \( 10^{-16} \) for the SC \( ^{113}\text{Cd}^+ \) ions. Illumination by 280 nm laser fluorescence could cause AC stark shift to the clock transition. Fortunately, the light shift can be controlled to a low value by adjusting the beam to be thin enough [35].

For the cadmium ion microwave clock, the dead time is unavoidable during the interrogation, which results in the Dick effect degrading the short-term frequency stability. Owing to the sympathetic cooling, the dead time is reduced since no additional cooling step is needed. Thus, the Dick-effect-limited Allan deviation is suppressed from \( 4E^{-13}/\sqrt{\tau} \) to \( 2E^{-13}/\sqrt{\tau} \) if the same local oscillator and microwave synthesizer are applied as in [10].

**IV. Conclusion**

In summary, we report a sympathetically cooled large ion crystal with a number up to \( 3.3 \times 10^7 \) and a large mass ratio up to 4.71 in Paul trap, whose temperature is measured to sub-Kelvin. The mass ratio of ions of the results is based on the obtainable laser wavelengths, without optimization. We obtain lower temperature even with larger mass ratio at outer shell. Even the efficiency of sympathetic cooling lacks a clear definition, it can intuitively be qualified by the mass ratio, acquired temperature, and the total number of SC ions. To our knowledge, it is the lowest direct measurement of outer shell SC ions of such a large number and mass ratio. These results bring verification to the translational temperature obtained by MD simulation. The second-order Doppler frequency shift of microwave atomic clocks is suppressed 2 orders of magnitude (from \( 10^{-14} \) to \( 10^{-16} \)). And the Dick effect is also suppressed. These results are important to build sympathetically-cooled Cd+ frequency standard. This result is also useful for the research of sympathetic cooling of large ion cloud.

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Appendix I. Experimental apparatus and relevant energy levels

An ultrahigh vacuum (UHV) chamber enclosing the ion trap is evacuated by an ion pump and a getter pump a vacuum of $<10^{-7}$ Pa. A stainless steel oven enclosed Mg metal is added, and it is labeled as (8) in Fig. 7. And a static magnetic field is applied parallel to the electrodes by Helmholtz coils.

![Fig. 7 The vacuum chamber with the trap. Labels in the diagram: (1) 280 nm laser window; (2) Electrode feedthrough; (3) and (6) E-guns; (4) To vacuum Pumps; (5) 214 nm laser window; (7) Cd oven; (8) Mg oven.](image)

The simplified energy levels are shown as Fig. 8. The cooling laser for Mg$^+$ ions is linear polarized, while the cooling laser for $^{113}$Cd$^+$ is circularly polarized.

![Fig. 8 Energy level diagram of the lowest relevant energy levels for $^{24}$Mg$^+$ and $^{113}$Cd$^+$ ions (not to scale).](image)