Progress Towards a Microwave Frequency Standard Based on Sympathetically-cooled $^{113}Cd^+$ Ions

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***Abstract*—A microwave frequency standard based on $^{113}Cd^+$ ions at Tsinghua University has been carried out for twelve years. To overcome the limitations of the Dick effect and the second-order Doppler frequency shift (SODFS), we propose to use heavy $^{174}Yb^+$ as the coolant ions to sympathetically cool the large $^{113}Cd^+$ cloud, which is expected to further improve the performance of the cadmium-ion microwave frequency standard. This paper reports our recent experimental and simulation progress towards the cadmium-ion microwave frequency standard.**

Keywords—microwave frequency standards; cadmium ions; sympathetic cooling; molecular dynamics

I. INTRODUCTION

Microwave frequency standards based on laser-cooled ions demonstrate great potential in areas such as time standards, telecommunications, and deep-space exploration [1-9]. Since 2010, our team has been committed to developing a microwave frequency standard based on $^{113}Cd^+$ ions. So far, the 0-0 ground-state hyperfine transition frequency of $^{113}Cd^+$ was determined to be 15199862855.02799(27) Hz with a fractional uncertainty of 1.8×10^{-14}, and the fractional frequency stability was measured to be $4.2 \times 10^{-13}/\sqrt{\tau}$ [9]. Nevertheless, it still doesn't reach the anticipated performance, which mainly suffers 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 trapped ions during interrogation. In this case, the technology of sympathetic cooling may be a solution.

In the sympathetic-cooling scheme, the sympathetically-cooled (SC) ions' temperature is reduced through long-range Coulomb interactions with laser-cooled (LC) ions. The application of sympathetic-cooling technology to microwave ion frequency standards was proposed by Wineland and collaborators from the National Institute of Standards and Technology (NIST), which was preliminarily applied in the development of a Be^+ frequency standard [10]. In the last decade, we have studied the microwave ion frequency standards based on sympathetically-cooled $^{113}Cd^+$ ions with different coolant ions ($^{24}Mg^+$, $^{40}Ca^+$) [11-12]. The minimum temperature of $^{113}Cd^+$ ions was reduced to tens of millikelvins. However, the excess micromotion of trapped ions leads to a large SODFS because the $^{113}Cd^+$ ions locate far from the trap axial as a shell outside of the light laser-cooled ions. If we use a kind of coolant ions with heavier mass than $^{113}Cd^+$ ions, more $^{113}Cd^+$ ions could locate close to the trap axial, which will bring smaller SODFS than that of the previous schemes.

In this paper, we propose to use heavy $^{174}Yb^+$ as the coolant ions to cool the large $^{113}Cd^+$ cloud. Compared to $^{40}Ca^+$ ions, the mass difference between $^{174}Yb^+$ and $^{113}Cd^+$ is smaller, which is helpful to improve the sympathetic cooling efficiency. Moreover, the light $^{113}Cd^+$ ions are distributed inside the heavy $^{174}Yb^+$ crystal due to the $1/M$ dependence of the pseudopotential of a Paul trap, where M denotes the ion mass, which could reduce the SODFS of trapped ions. The structure of this paper is as follows: we first introduce our experimental setup for the confinement of ions. Then, we report our recent experimental and simulation progress towards the cadmium-ion microwave frequency standard.

II. EXPERIMENTAL SETUP

The relevant energy level structures of $^{113}Cd^+$ and $^{174}Yb^+$ and our system setup are shown in Fig. 1(a) and Fig. 1(b), respectively. The ions are trapped in a linear quadrupole Paul trap. Details of the trap are given in [13]; here, a brief description suffices. It consists of four rod electrodes with a diameter of $r_e = 7.1$ mm, and each rod is segmented into three parts. The minimum distance between the nodal line of the trap and the electrode surfaces is $r_0 = 6.2$ mm. The length of the trapping part is $2z_0 = 40$ mm. For the confinement of ions, a RF voltage $U_{\rm rf}\cos(\Omega t)$ is applied to one pair of diagonal electrode rods, and the other pair are grounded. A DC voltage $U_{\rm end}$ is applied to endcap electrodes to confine ions axially.

The $^{174}Yb^+$ ions are cooled by a 369.5-nm laser beam, and the 935-nm laser is applied to repump the ions from the dark state of $^2D_{3/2}$. The $^{113}Cd^+$ ions are sympathetically cooled by $^{174}Yb^+$ ions via mutual Coulomb interaction and detected by a 214.5-nm red detuned laser via the cycling transition with circular polarization (100 μW). In addition, microwave radiation of 15.2 GHz is applied to repump the ions from the dark state of $|^2S_{1/2}, F=0\rangle$. Because the hyperfine splitting of $|^2P_{3/2}\rangle$ is only 800 MHz, the pump laser beam is generated by blue-shifting the cooling laser beam using acousto-optic-modulators (AOM). The frequency difference between the states of $|^2S_{1/2}, F=0, m_F=0\rangle$ and $|^2S_{1/2}, F=1, m_F=0\rangle$ is

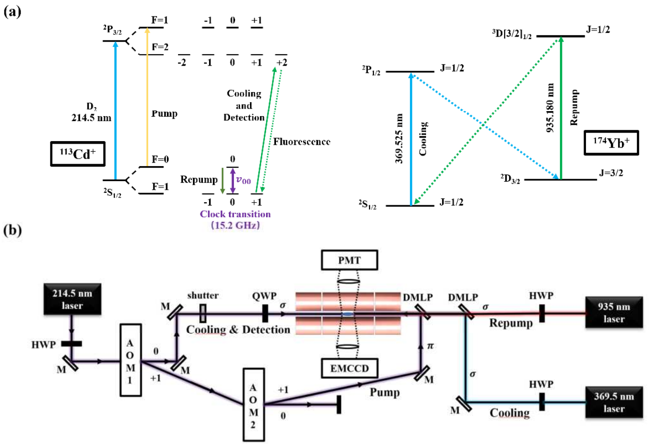

Fig. 1. (Color online) (a) Relevant energy levels of $^{113}Cd^+$ and $^{174}Yb^+$ (not to scale) and (b) schematic of the experiment system. M, mirror; DMLP, dichromatic mirror; HWP, halfwave plate; QWP, quarter-wave plate; AOM, acousto-optic modulator; PMT, photomultiplier tube detector; EMCCD, electron-multiplying charge coupled device.

the chosen transition frequency, which is approximately 15.2 GHz. The fluorescence signal from the ions is collected by a photomultiplier tube (PMT), and an electron-multiplying charge coupled device (EMCCD) images the structure of the ion cloud.

The radial motion of a single ion trapped in a quadrupole Paul trap can be described by the Mathieu equation

$$\frac{d^2r}{d\xi^2} + (a - 2q\cos 2\xi)r = 0, \quad (1)$$

where $r = x, y$. a, q, and ξ denote three dimensionless parameters given by

$$a_x = a_y = \frac{4Q\kappa U_{\text{end}}}{M\Omega^2 z_0^2},$$
$$q_x = -q_y = \frac{2QU_{\text{rf}}}{M\Omega^2 r_0^2}, \quad (2)$$
$$\xi = \frac{\Omega t}{2},$$

where Q denotes the charge of the ion, κ the axial geometrical factor related to the configuration of the quadrupole trap. To confine $^{174}Yb^+$ and $^{113}Cd^+$ ions in the trap simultaneously, the parameters a and q should be taken into account carefully.

As shown in Fig. 2, a and q differ for different ion species depending on their charge-mass ratio. For $^{174}Yb^+$ and $^{113}Cd^+$ ions, $a_{\text{Yb}}/a_{\text{Cd}} = q_{\text{Yb}}/q_{\text{Cd}} = 0.65$. Therefore, U_{rf} and U_{end} need be adjusted carefully considering the overlapping stability region (black part in Fig. 2). In general, in order to trap ions stably, the depth of the potential well in the center of the trap needs to be greater than 3 eV, and the adiabatic condition ($q < 0.3$) should be met. Considering the above conditions, the set of electrical parameters is as follows. The RF voltage U_{rf} is varied between 200-1000 V with a frequency of $\Omega = 2\pi \times 2$ MHz. The DC voltage U_{end} is varied between 10-200 V.

III. Preliminary Results

So far, we have separately obtained the images of $^{174}Yb^+$ and $^{113}Cd^+$ ion crystals, which are shown in Fig. 3. The exposure time of the cooled EMCCD is 0.12 s. The directions of the 369.5-nm laser and the 214.5-nm laser are opposite. In order to

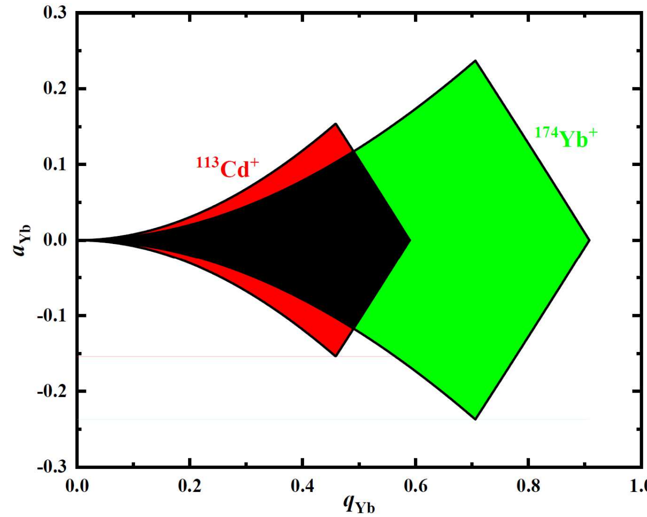

Fig. 2. (Color online) The lowest stability region for the radial plane, where a and q are present in the charge-mass ratio of $^{174}Yb^+$. Green part is the lowest stability region for $^{174}Yb^+$; red part is the lowest stability region for $^{113}Cd^+$; black part is the lowest stability region both for $^{174}Yb^+$ and $^{113}Cd^+$.

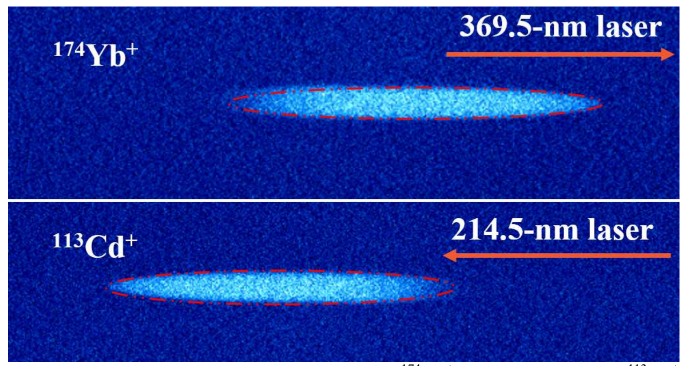

Fig. 3. (Color online) EMCCD images of the $^{174}Yb^+$ ion system and the $^{113}Cd^+$ ion system, respectively. The exposure time is 0.12 s. The directions of the 369.5-nm laser and the 214.5-nm laser are opposite.

achieve a stable $^{174}Yb^+$-$^{113}Cd^+$ two-component ion crystal in the next phase, we have applied molecular dynamics (MD) simulations to study the factors that affect the sympathetic cooling efficiency, including the electrical parameters and the number ratio between LC ions and SC ions.

With chosen electrical parameters of $U_{\text{rf}} = 500$ V and $U_{\text{end}} = 10$ V, we obtain simulation images of the sympathetic crystallization of 1024 $^{113}Cd^+$ ions by laser-cooled 4096 heavy $^{174}Yb^+$ and light $^{40}Ca^+$ ions, respectively, which are shown in Fig. 4. The exposure time is set to be 6 ms. As expected, the heavy $^{174}Yb^+$ ions are distributed outside the $^{113}Cd^+$ crystal, whereas the light $^{40}Ca^+$ ions are the opposite. In Fig. 4(a), the average secular energy and total energy per $^{113}Cd^+$ ion are 10.2(0.2) mK $\cdot\, 3k_{\text{B}}/2$ and 1245(1) mK $\cdot\, 3k_{\text{B}}/2$, respectively. In Fig. 4(b), the average secular energy and total energy per $^{113}Cd^+$ ion are 231(9) mK $\cdot\, 3k_{\text{B}}/2$ and 15734(89) mK $\cdot\, 3k_{\text{B}}/2$, respectively. As can be seen, compared to $^{40}Ca^+$ as coolant ions, the SODFS and its uncertainty are reduced by about an order of magnitude with $^{174}Yb^+$ as coolant ions, which may hopefully further improve the performance of the cadmium-ion microwave frequency standard.

A. Optimal electrical parameters

The electrical potentials experienced by ions in the trap

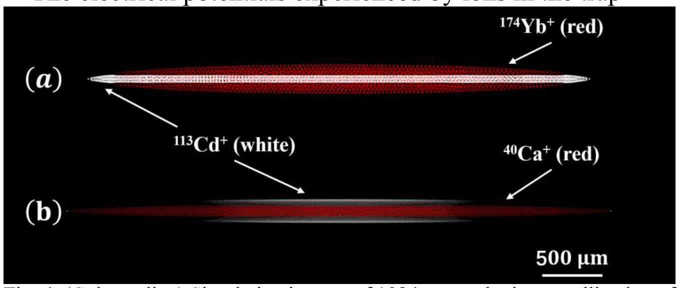

Fig. 4. (Color online) Simulation images of 1024 sympathetic crystallization of $^{113}Cd^+$ ions (white) by 4096 coolant ions (red). (a) With $^{174}Yb^+$ as coolant ions, the average secular energy and total energy per $^{113}Cd^+$ ion are 10.2(0.2) $\mathrm{mK} \cdot 3k_B/2$ and 1245(1) $\mathrm{mK} \cdot 3k_B/2$, respectively. (b) With $^{40}Ca^+$ as coolant ions, the average secular energy and total energy per $^{113}Cd^+$ ion are 231(9) $\mathrm{mK} \cdot 3k_B/2$ and 15734(89) $\mathrm{mK} \cdot 3k_B/2$, respectively. The electrical parameters are chosen as $U_{\mathrm{rf}} = 500$ V and $U_{\mathrm{end}} = 10$ V. The exposure time is set to be 6 ms.

decide the spatial configuration of the ion crystal, which is one of the most important factors that affect the sympathetic cooling efficiency and the temperature of SC ions. With this in mind, we have applied MD simulations to depict the relationship between the electrical parameters and the temperature of SC ions. In Fig. 5, we show simulated images of the $^{174}Yb^+$-$^{113}Cd^+$ ion system under the influence of different RF voltages U_{rf} and DC voltages U_{end}. The numbers of $^{174}Yb^+$ and $^{113}Cd^+$ ions are 768 and 256, respectively. It is clear that increasing U_{rf} and U_{end} lead to substantial compression and extension on the radial distribution of the ion system.

Figure 6 shows the equilibrium temperatures of sympathetically-cooled $^{113}Cd^+$ ions at different DC voltages and RF voltages. The graph of Fig. 6(b) shows that the optimum RF voltage corresponding to the lowest temperature of SC ions is approximately 500 V. Still, before the optimum RF voltage, the temperature of SC ions decreases with the increasement of U_{rf}, but increases after the optimum RF voltage. The results in Fig. 6(a) indicate that the temperature of SC ions and the DC voltage are positively correlated.

B. Optimal number ratio

The number ratio between LC ions and SC ions is also an important factor that affects the sympathetic cooling efficiency. To obtain the optimum number ratio of $N_{\mathrm{LC}}/N_{\mathrm{SC}}$, we have performed MD simulations by systematically changing the number of LC ions ($^{174}Yb^+$) under the influence of a fixed number of SC ions ($^{113}Cd^+$). In Fig. 7, the number of $^{113}Cd^+$ ions is fixed at 256. The difference of the $^{174}Yb^+$-$^{113}Cd^+$ ion system by the increase of N_{LC} is clearly shown. As N_{LC} increases, the $^{174}Yb^+$ crystal transforms from a hollow cylinder to a hollow ellipsoid whose inside is occupied by the $^{113}Cd^+$ ions. More importantly, the equilibrium temperature of $^{113}Cd^+$ ions decreases as N_{LC} increases, especially for $N_{\mathrm{LC}}/N_{\mathrm{SC}} < 2$. Therefore, the value of $N_{\mathrm{LC}}/N_{\mathrm{SC}}$ should be greater than 2 in order to obtain a stable low-temperature $^{113}Cd^+$ ion crystal ($<$ 10 mK).

IV. Conclusions

In summary, we propose to use laser-cooled $^{174}Yb^+$ ions to sympathetically cool the large $^{113}Cd^+$ cloud. Using MD simulations, we depict the relationship between sympathetically-cooled $^{113}Cd^+$ ions' secular temperature and electrical parameters applied to the linear Paul trap, including

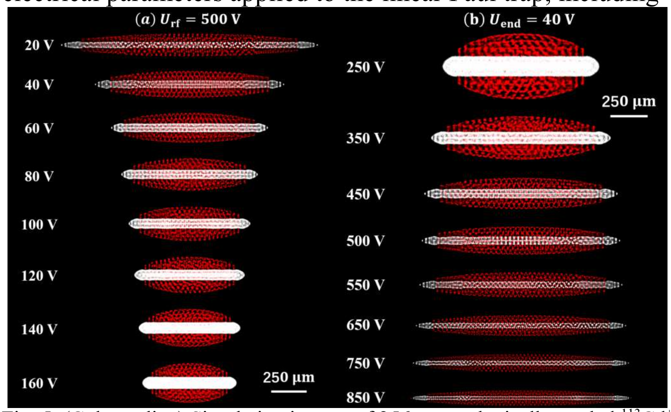

Fig. 5. (Color online) Simulation images of 256 sympathetically-cooled $^{113}Cd^+$ ions (white) by 768 laser-cooled $^{174}Yb^+$ ions (red). (a) Spatial configuration of the ion system changes with U_{end} at $U_{\mathrm{rf}} = 500$ V. (b) Spatial configuration of the ion system changes with U_{rf} at $U_{\mathrm{end}} = 40$ V.

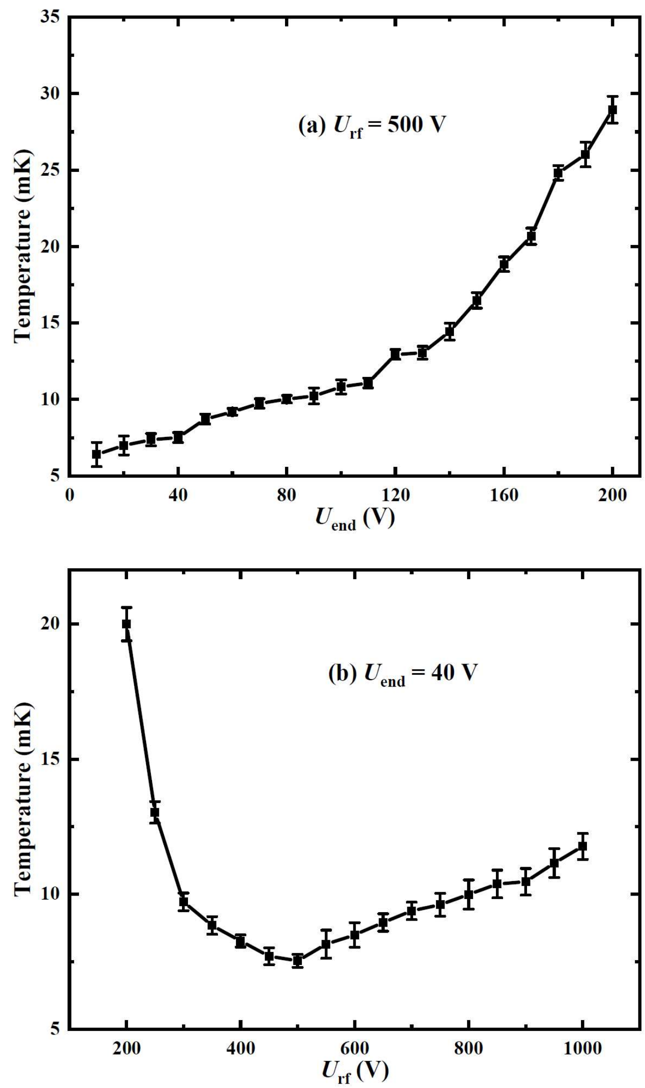

Fig. 6. (Color online) The secular temperature of SC ions in the thermal equilibrium state. (a) Secular temperature of $^{113}Cd^+$ at different DC voltages

with $U_{rf} = 500$ V. (b) Secular temperature of $^{113}Cd^{+}$ at different RF voltages with $U_{end} = 40$ V. The parameters are the same as those in Fig. 5.

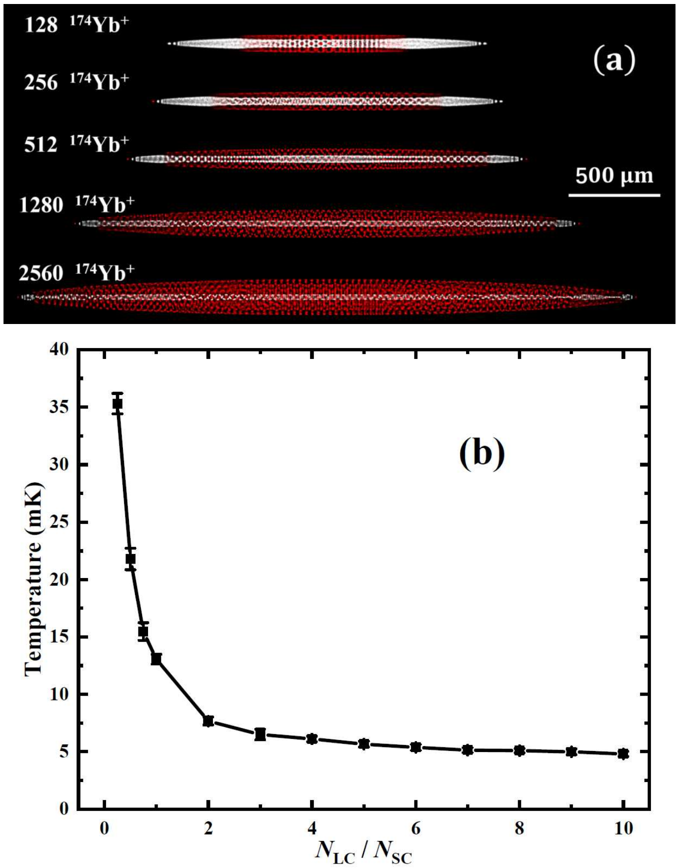

Fig. 7. (Color online) (a) Simulated images of the two-component ion crystal at different numbers of $^{174}Yb^{+}$ ions (red) under the influence of a fixed number of $^{113}Cd^{+}$ ions (256, white). (b) Secular temperature of $^{113}Cd^{+}$ ions at different number ratios of N_{LC}/N_{SC}.

radiofrequency (RF) voltage and endcap voltage, and the number ratio between sympathetically-cooled $^{113}Cd^{+}$ ions and laser-cooled $^{174}Yb^{+}$ ions. With these work, an improvement of the accuracy for the cadmium-ion microwave frequency standard limited by the second-order Doppler frequency shift and the Dick effect may be possible.

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REFERENCES

[1] Wineland, D. J., et al. "Progress at NIST toward absolute frequency standards using stored ions." *IEEE transactions on ultrasonics, ferroelectrics, and frequency control* 37.6 (1990): 515-523.

[2] Berkeland, D. J., et al. "Laser-cooled mercury ion frequency standard." *Physical Review Letters* 80.10 (1998): 2089.

[3] Tjoelker, R. L., et al. "Mercury trapped ion frequency standard for space applications." *Frequency Standards And Metrology*. 2002. 609-614.

[4] Burt, E. A., et al. "Demonstration of a trapped-ion atomic clock in space." *Nature* 595.7865 (2021): 43-47.

[5] Phoonthong, Piyaphat, et al. "Determination of the absolute microwave frequency of laser-cooled $^{171}Yb^{+}$." *Applied Physics B* 117.2 (2014): 673-680.

[6] Mulholland, S., et al. "Laser-cooled ytterbium-ion microwave frequency standard." *Applied Physics B* 125.11 (2019): 1-12.

[7] Xin, N. C., et al. "Laser-cooled $^{171}Yb^{+}$ microwave frequency standard with a short-term frequency instability of $8.5 \times 10^{-13}/\sqrt{\tau}$." *Optics Express* 30.9 (2022): 14574-14585.

[8] Miao, K., et al. "High accuracy measurement of the ground-state hyperfine splitting in a $^{113}Cd^{+}$ microwave clock." *Optics letters* 40.18 (2015): 4249-4252.

[9] S. N. Miao, et al. "Precision determination of the ground-state hyperfine splitting of trapped $^{113}Cd^{+}$ ions." Optics Letters 46.23 (2021): 5882-5885.

[10] Bollinger, J. J., et al. "Progress at NIST toward absolute frequency standards using storedions." *IEEE Trans. Instrum. Meas* 40 (1991): 126-128.

[11] Zuo, Y. N., et al. "Direct temperature determination of a sympathetically cooled large $^{113}Cd^{+}$ ion crystal for a microwave clock." *Applied Physics Letters* 115.6 (2019): 061103.

[12] J. Z. Han, et al. "Toward a high-performance transportable microwave frequency standard based on sympathetically cooled $^{113}Cd^{+}$ ions." Applied Physics Letters 118.10 (2021): 101103.

[13] S. N. Miao, et al. "Research On Sympathetic Cooling $^{113}Cd^{+}$-$^{174}Yb^{+}$ System By Molecular Dynamics Simulation." *2021 Joint Conference of the European Frequency and Time Forum and IEEE International Frequency Control Symposium (EFTF/IFCS)*. IEEE, 2021.