International Symposium on Stored Ions for Precision Measurements

The Progress on Highly charged Ni^{12+} Clock and $^{40}Ca^{+}$ Optical Clocks

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Innovation Academy for Precision Measurement Science and Technology, CAS

International Symposium on Stored Ions for Precision Measurements—Shanghai,2023.11.24

Highly charged Ni^{12+} Clock

Highly charged ions(HCIs): Promises and benefits

REVIEWS OF MODERN PHYSICS

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Highly charged ions: Optical clocks and applications in fundamental physics

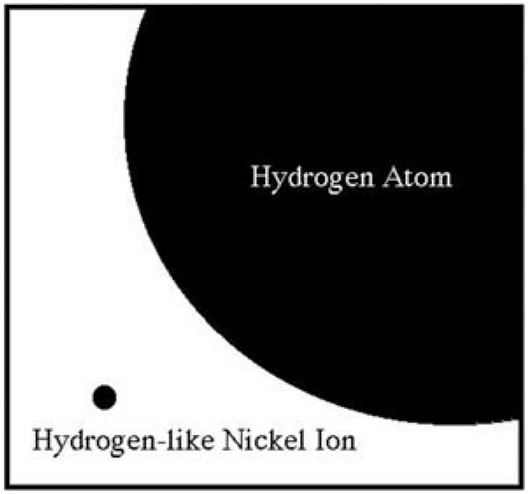
M. G. Kozlov, M. S. Safronova, J. R. Crespo López-Urrutia, and P. O. Schmidt

Rev. Mod. Phys. **90**, 045005 – Published 4 December 2018

For fundamental physics		For atomic clocks	
K	$Z^2(Z_i+1)^2$	2nd-order Stark shift	Z_i^{-4}
Fine structure	Z^4	Electric-quadrupole shift	Z_i^{-2}
Hyperfine structure	Z^3	Black-body radiation shift	Z_i^{-4}
QED shift	Z^4	Collisional shift	Negligible
		……	……

- **Novel optical clock candidates**
- **probing possible variation of α**
- **Test understanding of QED and nuclear effects**

Highly charged ions(HCIs): Promises and benefits



Feature	
Black-body radiation shift	Z_i^{-4}
Electric-quadrupole shift	Z_i^{-2}
……	……

Ions	Al^{+}	Yb^{+}	Ni^{12+}
Clock transition (nm)	267 (E2)	467 (E3)	498 (E2)
BBR shift (300 K)	3×10^{-18}	1×10^{-16}	2×10^{-20}

Compact size,

Insensitive to external electric and magnetic field perturbations

A. D. Lodlow, *et al*, Rev. Mod. Phys. 87, 637 (2015)
Y.-M. Yu, *et al*, PRA 97, 041403(R) (2018)
S. Liang et al., PRA 103, 022804 (2021)

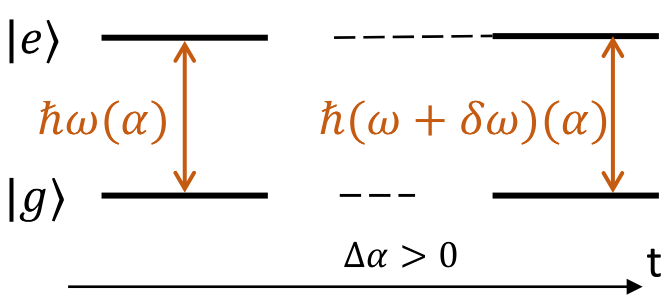
➢ **HCI—One of the best candidate system for optical clock with an uncertainty of E-19 or even less**

J. C. Berengut, *et al*, PRL 105, 120801 (2010)

HCI optical clock

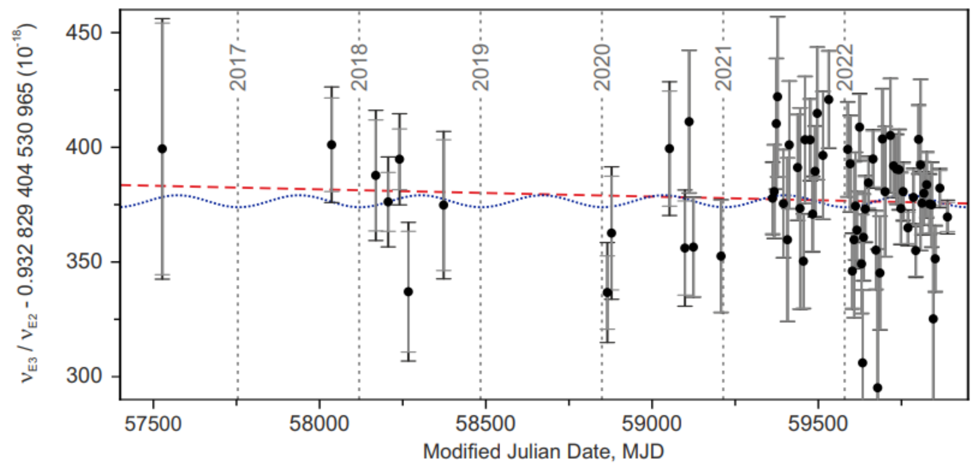
➢Physics beyond the Standard Model

- variation of fundamental constants?



$$\frac{\delta\omega}{\omega} = K\frac{\delta\alpha}{\alpha}$$

Atomic Clock Comparisons



Yb^{+}(E2)--Yb^{+}(E3)

Relative variation of optical clock frequency

$$\boxed{\frac{\delta\omega_1}{\omega} - \frac{\delta\omega_1}{\omega}} = \boxed{(K_1 - K_2)}\frac{\delta\alpha}{\alpha}$$

α constant sensitive factor

$$\frac{1}{\alpha}\frac{\mathrm{d}\alpha}{\mathrm{d}t} = 1.8(2.5)\times 10^{-19}/year$$

No variation was found with an accuracy of E-19

J. C. Berengut , *et al.*, PRL 105, 120801 (2010)

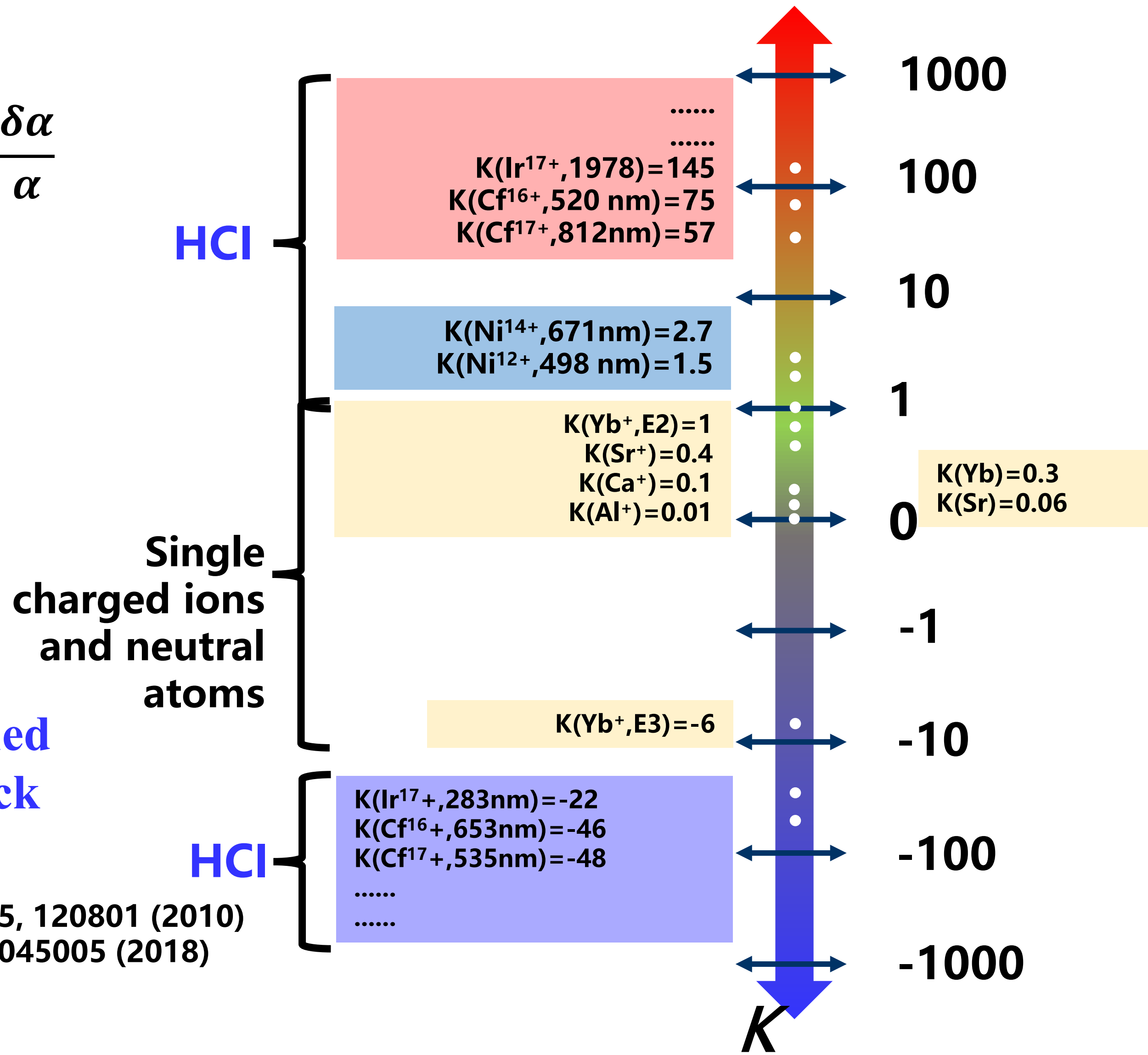
R. Lange *et al.*, arXiv:2301.03433

HCI optical clock

- variation of α

$$\boxed{\frac{\delta\omega_1}{\omega} - \frac{\delta\omega_1}{\omega}} = \boxed{(K_1 - K_2)}\ \frac{\delta\alpha}{\alpha}$$

$K\ factor \sim Z^2(Z_i + 1)^2$

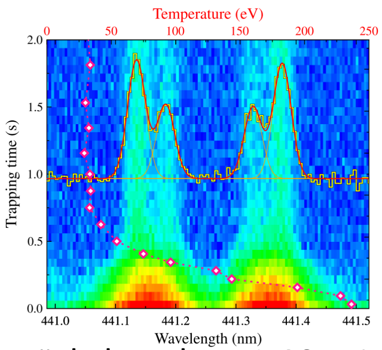


- **Larger Δ*K* can be obtained by using HCI optical clock**

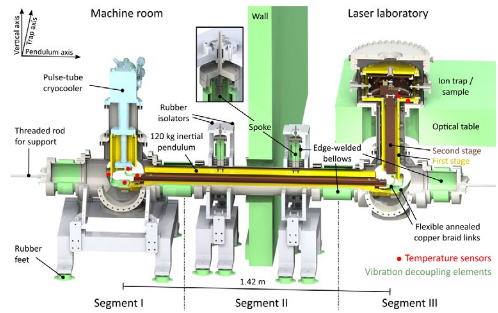
J. C. Berengut , *et al.,* Phys. Rev. Lett. 105, 120801 (2010)
M. G. Kozlov, *et al.,* Rev. Mod. Phys. 90, 045005 (2018)
S. Liang, *et al.,* Phys. Rev. A 103, 22804

HCI optical clock has special advantages to test the variation of α constant

HCI optical clock

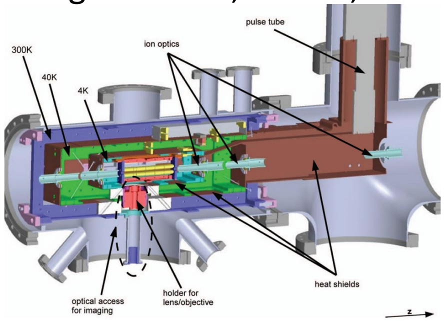


V. Mäckel et al., PRL **107**, 143002 (2011)
I. Draganić et al., PRL **91**, 183001 (2003)

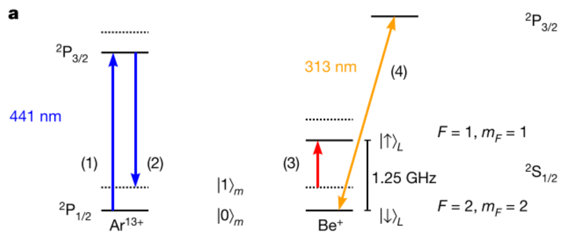


P. Micke et al., Rev. Sci. Instrum. **90**, 065104 (2019)

José R. Crespo López-Urrutia

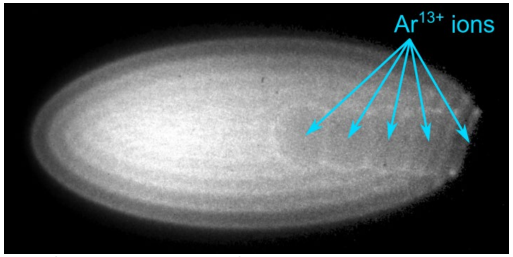


M. Schwarz et al., Rev. Sci. Instrum. **83**, 083115 (2012)

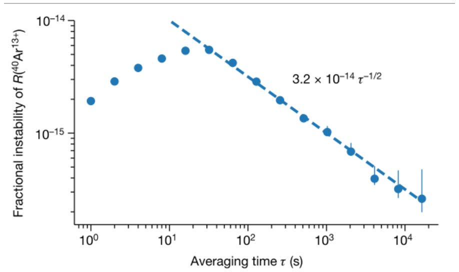


P. Micke et al., Nature **578**, 60–65 (2020)

Piet O. Schmidt



L. Schmöger et al., Science **347**, 1233–1236 (2015)



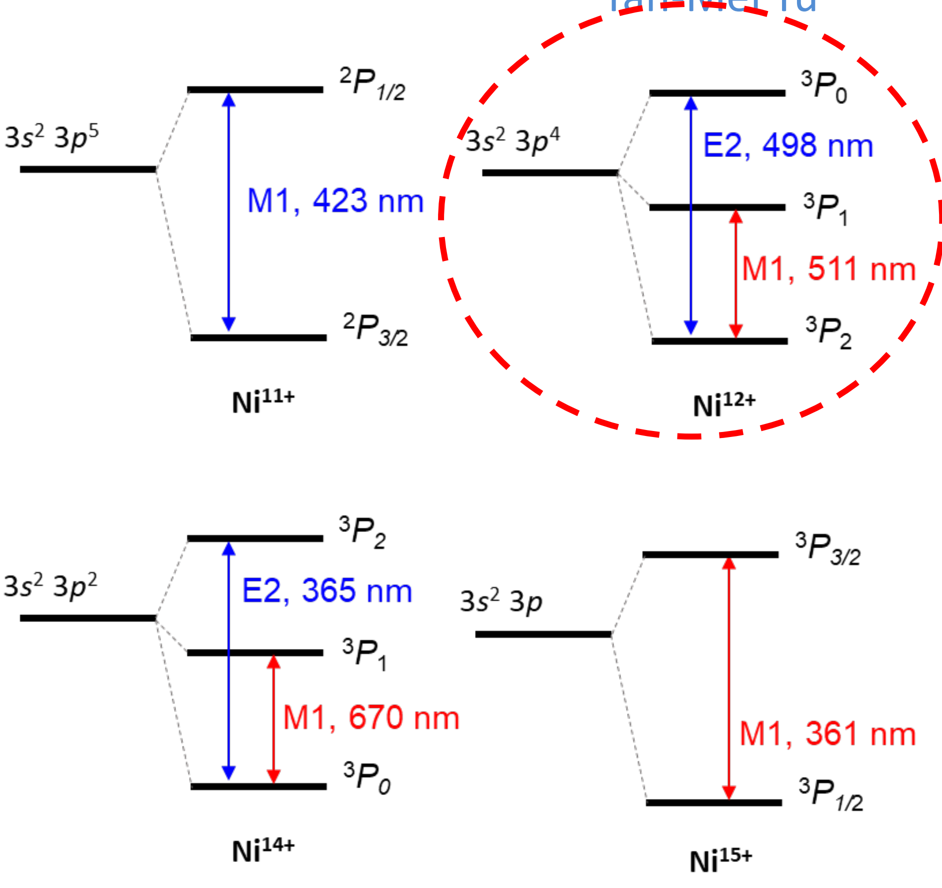
K. Steven A et al., Nature **611**, 43–47 (2022)

The first HCI clock (2022)
(Ar^{13+})

Highly charged Nickle ions

- Mutipule clock transition in visble light band, Various stable isotopes
- Simple energy level structure
- High Q factor of E2 transition

Yan-Mei Yu



Y.-M. Yu, *et al*, PRA 97, 041403(R) (2018)
S. Liang et al., PRA 103, 022804 (2021)

PTB –Ar^{13+},Ni^{12+}, Ca^{14+}

QSNET –Cf^{15+} Cf^{17+}

NIST –Pr^{9+}

MPIK –Ca^{14+}

Ni-HCI clock

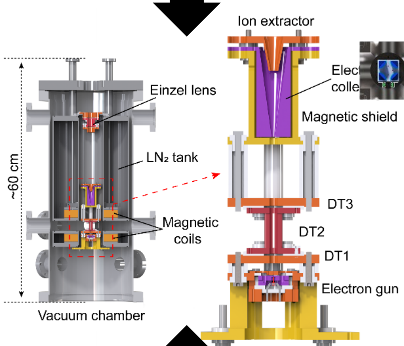
- **Steps of HCI optical clock**

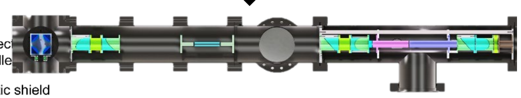
HCI Preparation → Selecting and Deceleration → Traping and Cooling → HCI optical clock

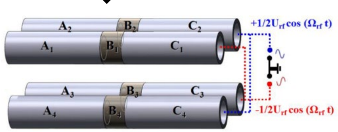
Electron Beam Ion Trap (EBIT)

HCI Beamline

Linear Ion Trap



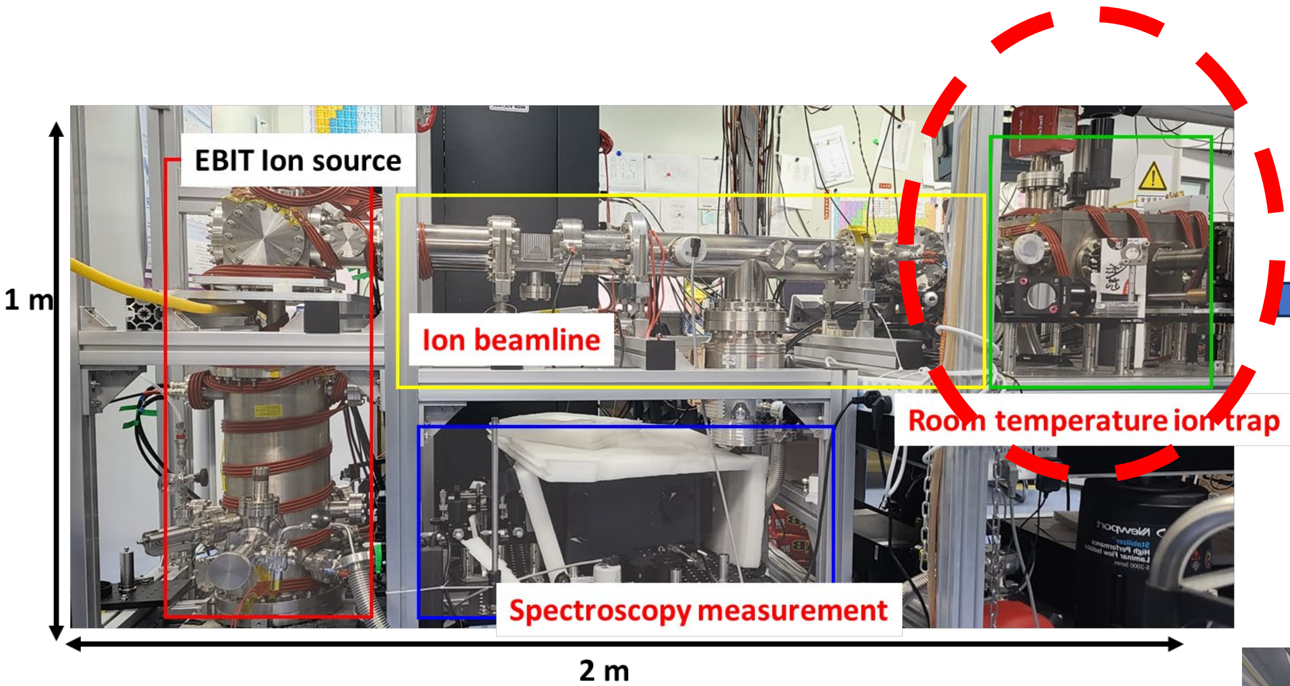




Preliminary measurement of Clock transition

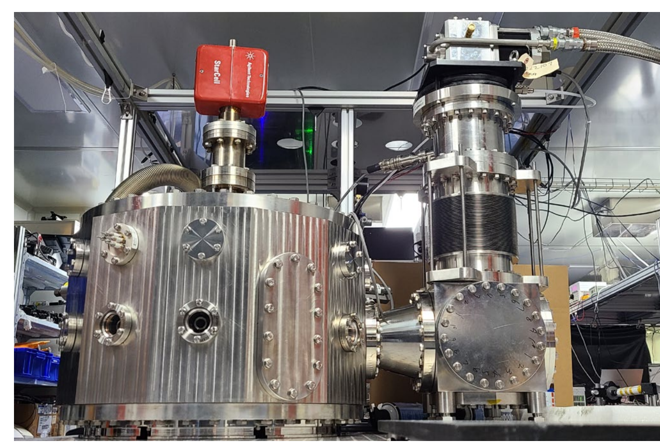
HCI experiment overview

- **Setup overview**



300K to 4K

Cryogenic ion
trap system



HCI preparation

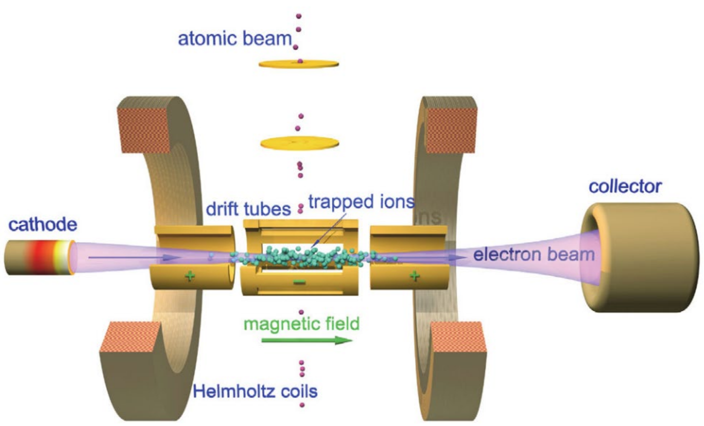
HCI Preparation → Preliminary measurement of Clock transition → Selecting and Deceleration → Traping and Cooling → HCI optical clock

□ HCI ion source

- **EBIT: Electron Beam Ion Trap**

  Radial confinement: Space charge effect

  Axial confinement: Electrostatic field



Ni^{12+} $3s^2\ {}^3p^4\ \ {}^3P_2$

>300 eV

Ni

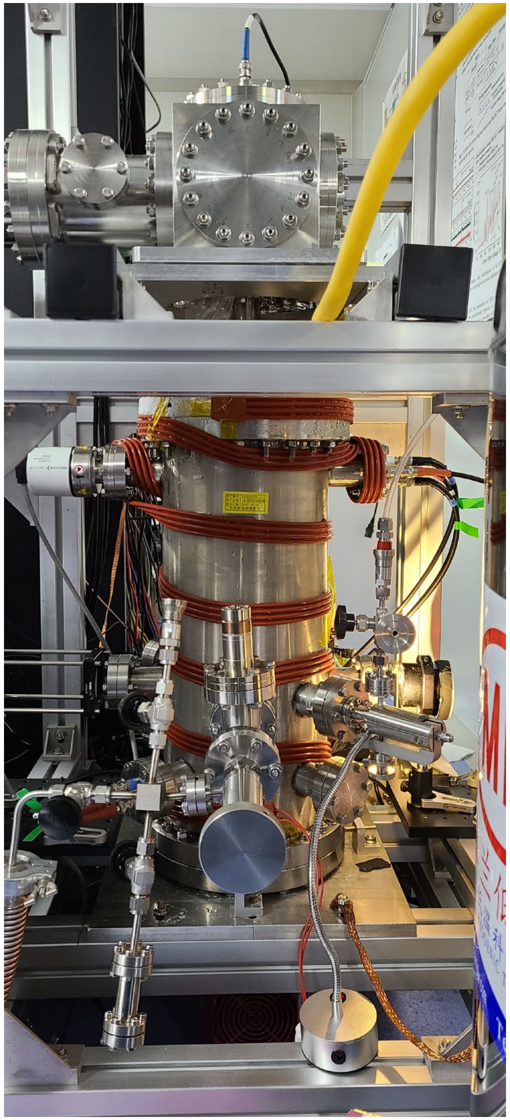
- **Compact**
- **Charged state adjustable**
- **Stable**
- **Has viewport for spectral detection**

M. G. Kozlov, *et al.*, Rev. Mod. Phys. 90, 045005 (2018)
M. A. Levine, *et al*, Physics Scripta T22, 157 (1988)

Optimal ion source for HCI clock

HCl preparation

SW-EBIT



(C5H5)2Ni

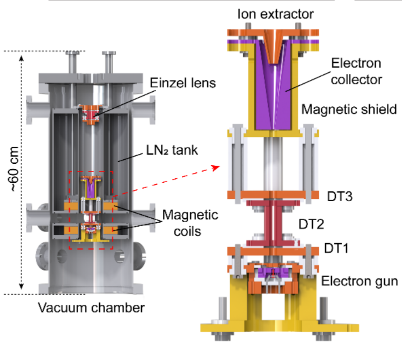
Ionization energy（eV)	**Ions**
0-200	W^{7+}
200-400	W^{14+}, Ni^{11+}, Ni^{12+}
400-600	Ni^{14+}, Ni^{15+}
600-800	Ar^{13+}
800-1000	Ar^{15+}, W^{27+}

Rev. Sci. Instrum. 90, 093301 (2019)

Preliminary measurement of Clock transition

● Clock transition of Ni-HCI

HCI Preparation → **Selecting and Deceleration** → Traping and Cooling → HCI optical clock



Ions	Types	Wavelength(nm, NIST)	Methods
Ni^{11+}	M1	423.12	Corona、Tokamak
Ni^{12+}	M1	511.58	Corona
	E2	498.37(248)	Theoretical calculations
Ni^{14+}	M1	670.17	Corona
	E2	365.173	Corona（Indirect measurement)
Ni^{15+}	M1	360.123(2)	Corona、Tokamak

Preliminary measurement of Clock transition

Low accuracy: tens of pm

- **Improving the accuracy of the wavelength measurement to sub-pm or even higher**

→ **crucial for constraining the range of laser wavelengths for finding the "needle in a haystack" narrow lines**

Preliminary measurement of Clock transition

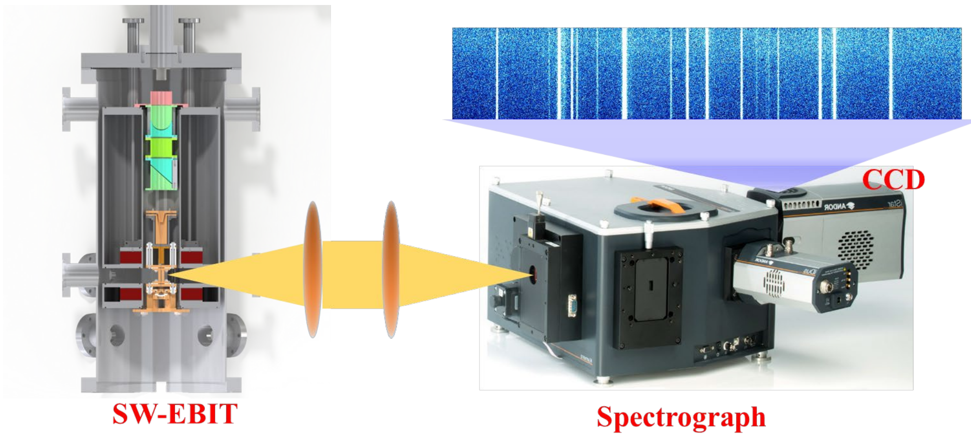


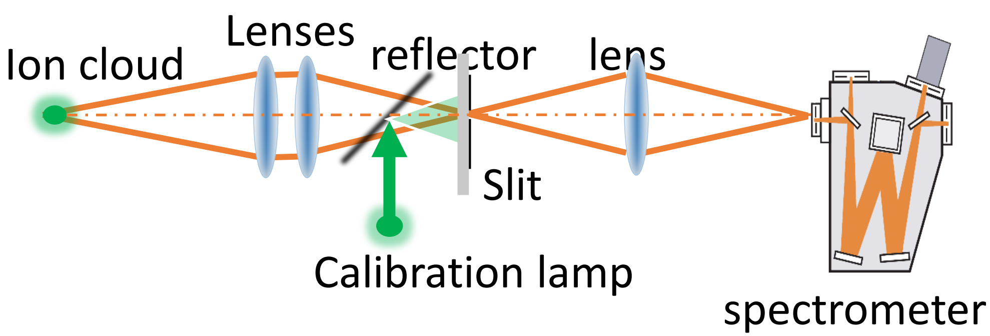
TABLE I. Observed and calculated wavelengths of magnetic-dipole ($M1$) and electric-quadrupole ($E2$) transitions, where the lines a through f are candidate clock transitions, in nm.

				NIST	This work		
Line	Ion	Transition	Type	Vacuum	Air (observed)	Vacuum	Theory
a	Ni^{11+}	$3s^23p^5\ ^2P_{1/2}-^2P_{3/2}$	$M1$	423.2	423.104(2)	423.223(2)	423.0(6)
b	Ni^{12+}	$3s^23p^4\ ^3P_1-^3P_2$	$M1$	511.724	511.570(2)	511.713(2)	511.8(6)
c	Ni^{14+}	$3s^23p^2\ ^3P_1-^3P_0$	$M1$	670.36	670.167(2)	670.352(2)	671.1(14)
d	Ni^{15+}	$3s^23p\ ^2P_{3/2}-^2P_{1/2}$	$M1$	360.22	360.105(2)	360.207(2)	359.9(9)
e	Ni^{12+}	$3s^23p^4\ ^3P_0-^3P_2$	$E2$	498.50(249)	–	–	496.9(24)
f	Ni^{14+}	$3s^23p^2\ ^3P_2-^3P_0$	$E2$	365.277	–	365.278(1)	365.0(3)
g	Ni^{14+}	$3s^23p^2\ ^3P_2-^3P_1$	$M1$	802.63	802.419(2)	802.639(2)	800.3(25)

S. Liang et al., PRA 103, 022804 (2021)

Preliminary measurement of Clock transition

- **Ni^{12+} M1 clock transition**



Error source	Uncertainty in wavelength (pm)
Line centroid	0.21
Isotope shift	**0.06**
Calibration systematic	**0.33**
Total	**0.40**

Wavelength: 511.58211 (40) nm

reaching sub-pm level with a fractional uncertainty of **8E-7**

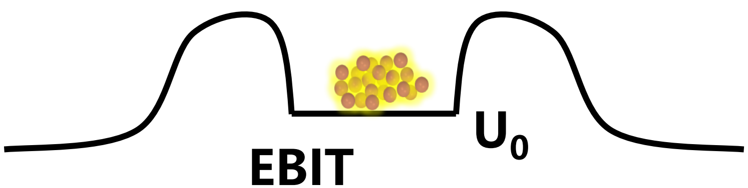
I. Draganić et al., PRL **91**, 183001 (2003)

arXiv:2303.04552

HCI-Extraction

- **Ni-HCI Extraction**

Raising the U_{extra} of EBIT



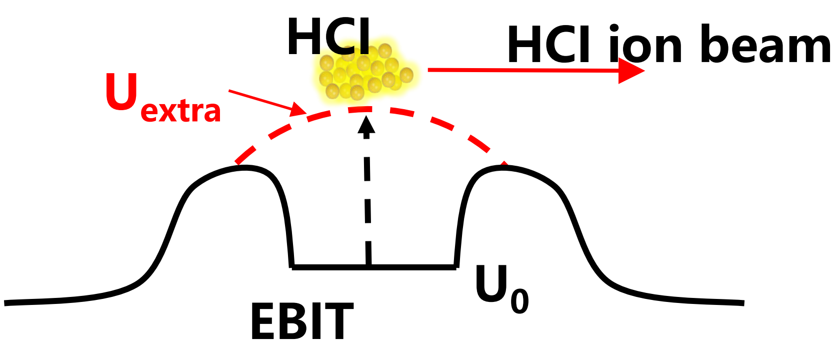
HCI extraction

HCI-Extraction

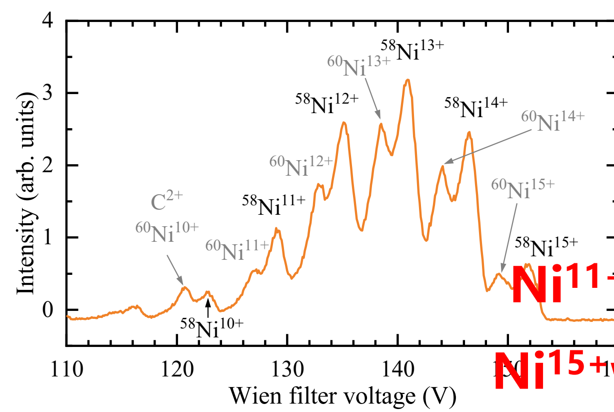
- **Ni-HCI Extraction**

HCI Preparation → **Selecting and Deceleration** → Traping and Cooling → HCI optical clock

Raising the U_{extra} of EBIT



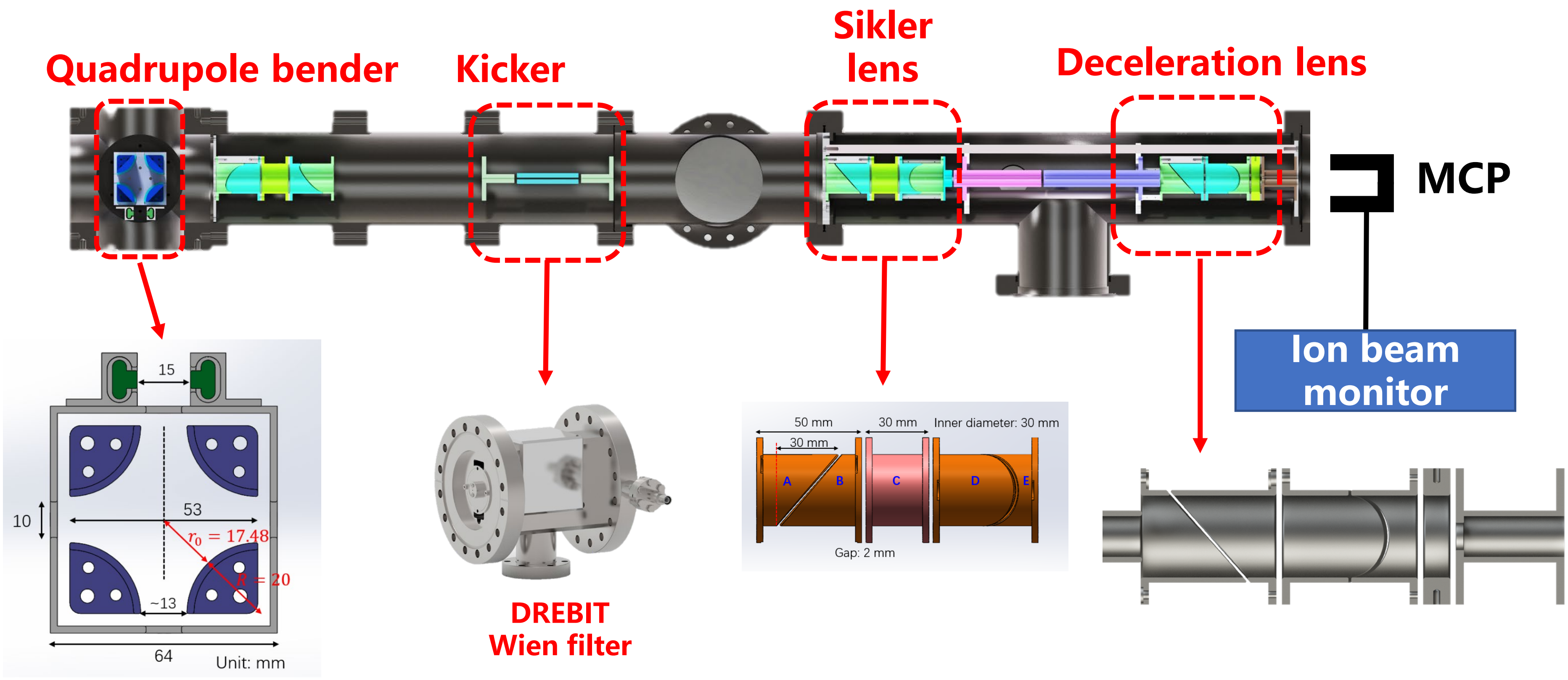
HCI Extration



Ni^{11+}、Ni^{12+}、Ni^{14+}、Ni^{15+} were successfully extracted

Valence distribution of Ni-HCI

Beam line



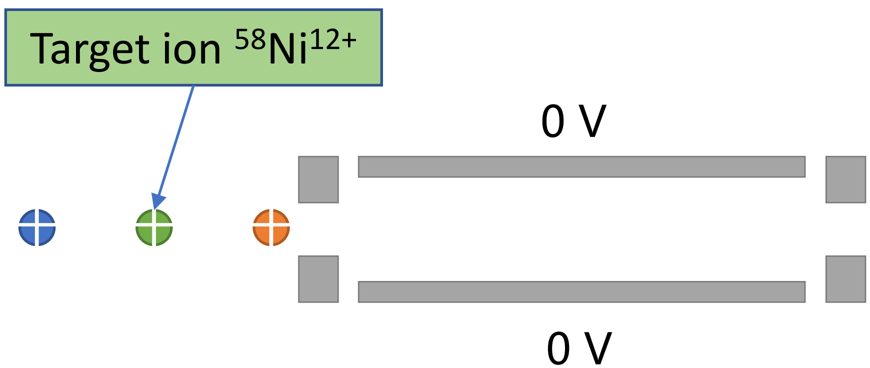
- Time of fight is used for ion selecting
- Wien filter is used as a Kicker

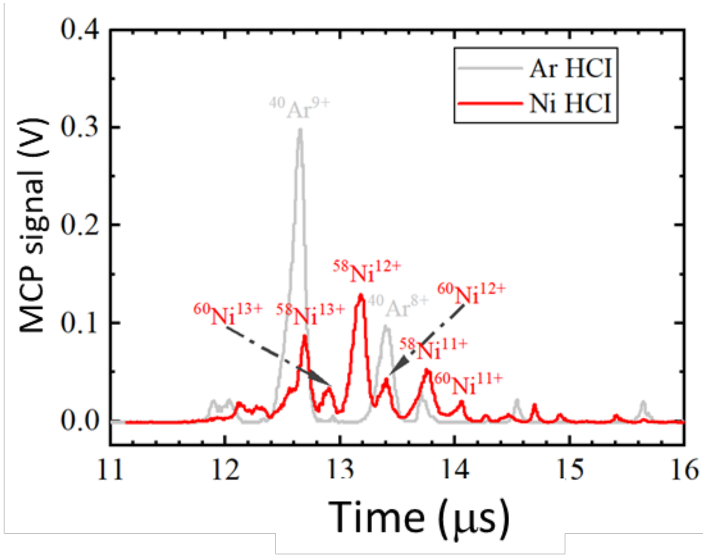
Ion selecting with TOF

The time of flight of ions:

$$\Delta t = \frac{-L\Delta R}{\sqrt{\frac{8R^3 e U_{extr}}{\mathrm{u}}}}$$

The distance L, Charge-to-mass ratio $R = Q/M$, Voltage at the extracted electrode U_{extr}, elementary charge e, atomic mass unit: u





Separation of pure $^{58}Ni^{12+}$ ions by changing the parallel plate potential at different moments

Ion selecting with TOF

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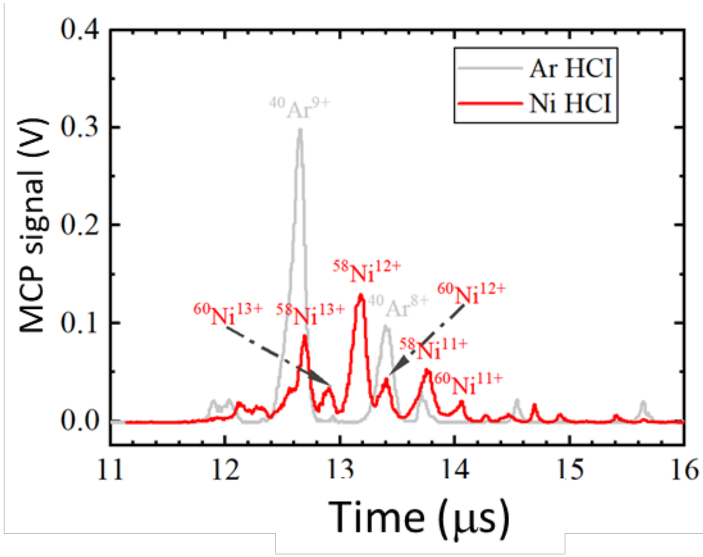

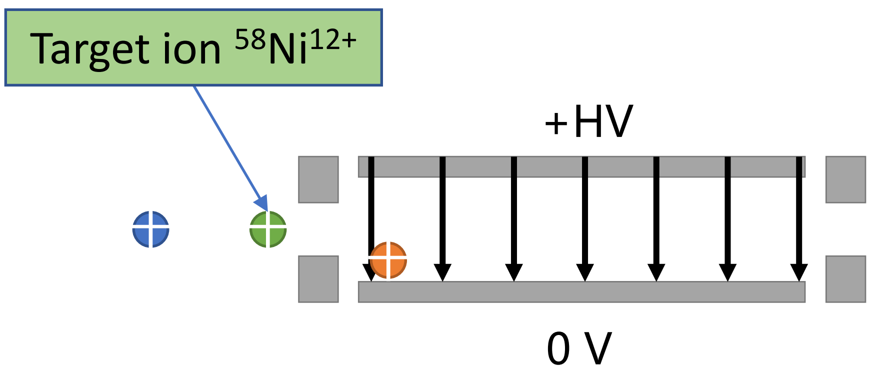

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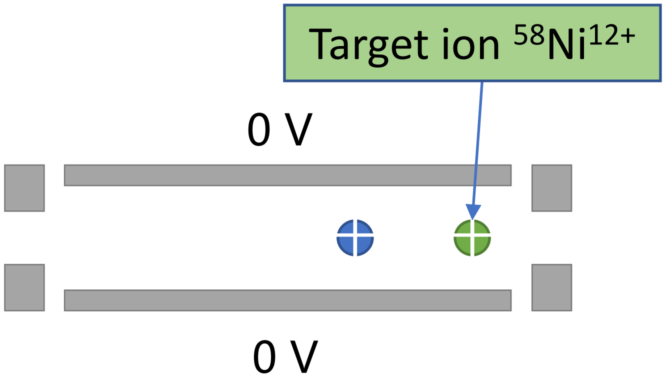

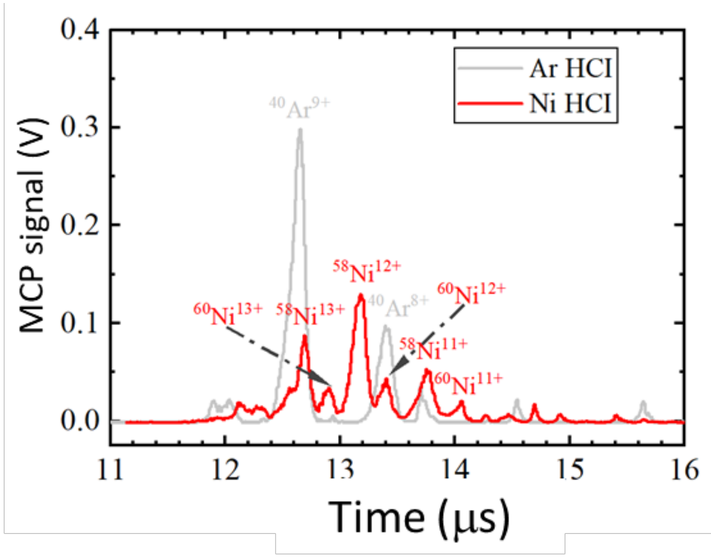

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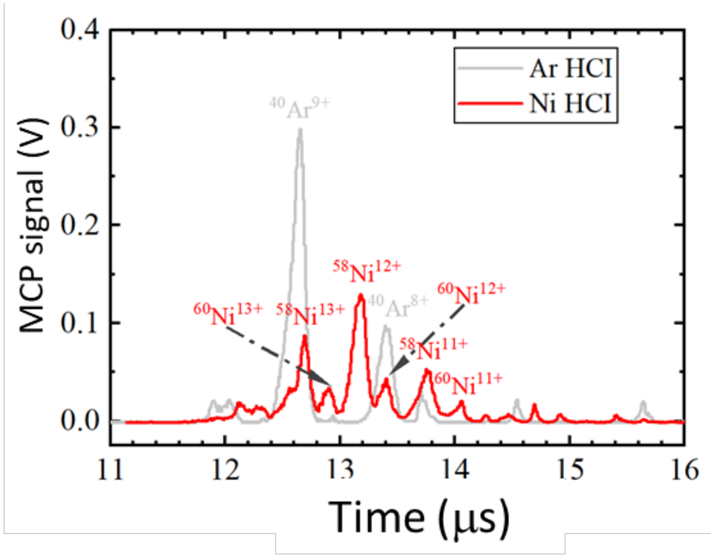

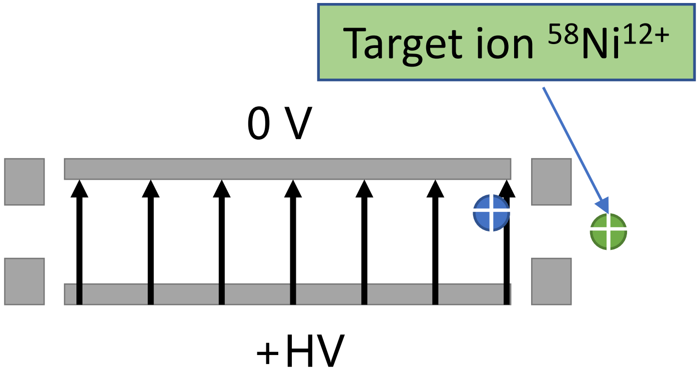

Separation of pure $^{58}Ni^{12+}$ ions by changing the parallel plate potential at different moments

Ion selecting with TOF

The time of flight of ions:

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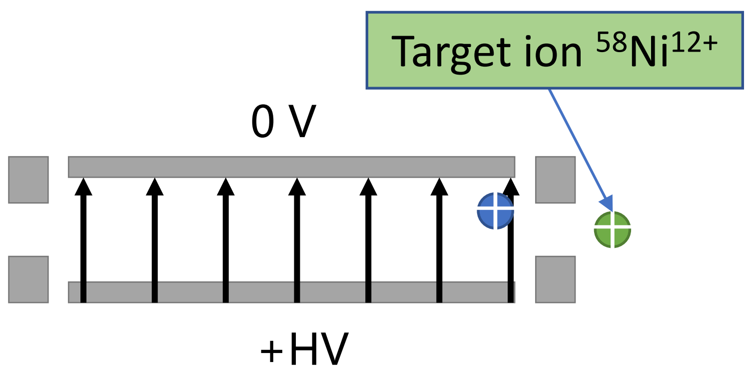
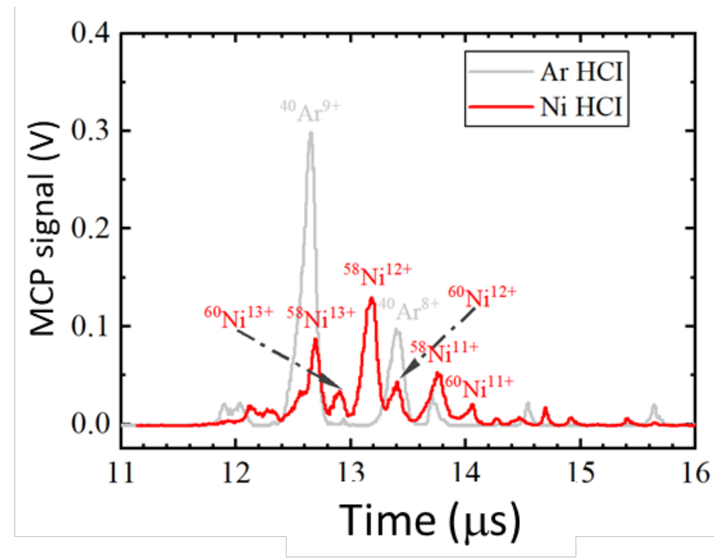
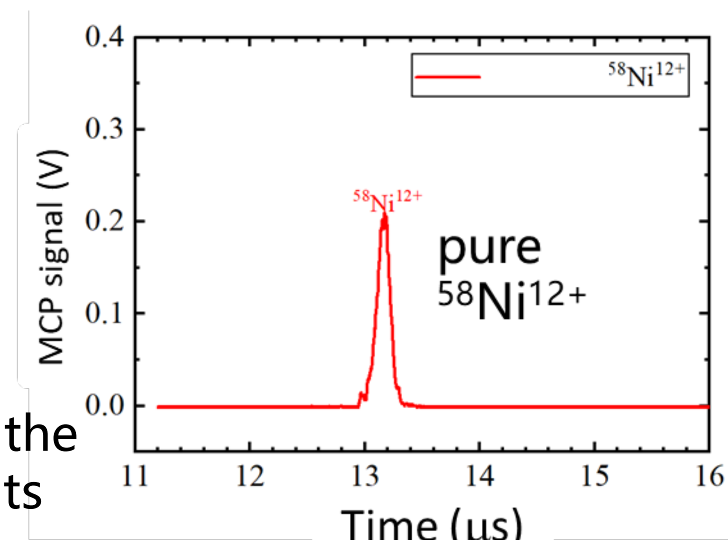




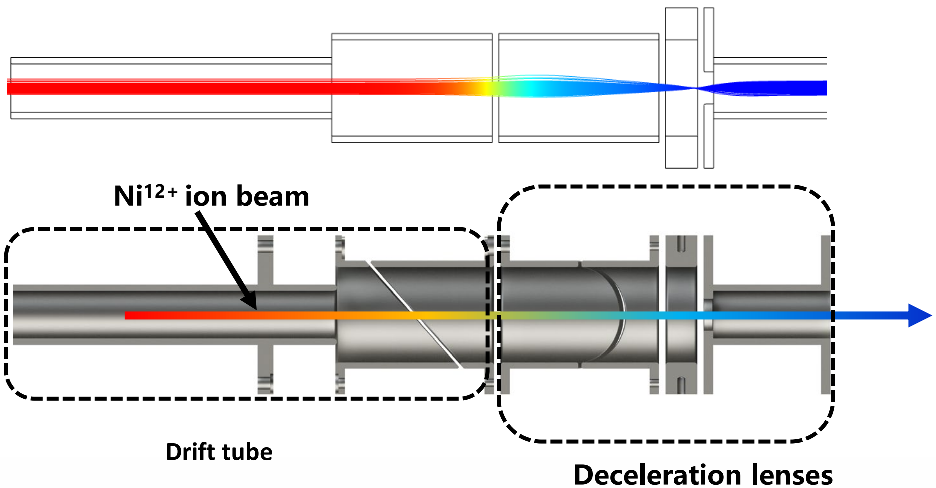


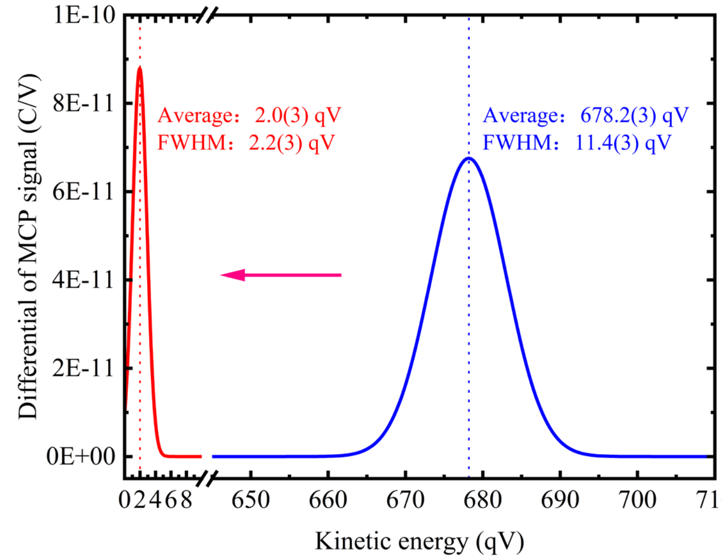
Separation of pure $^{58}Ni^{12+}$ ions by changing the parallel plate potential at different moments

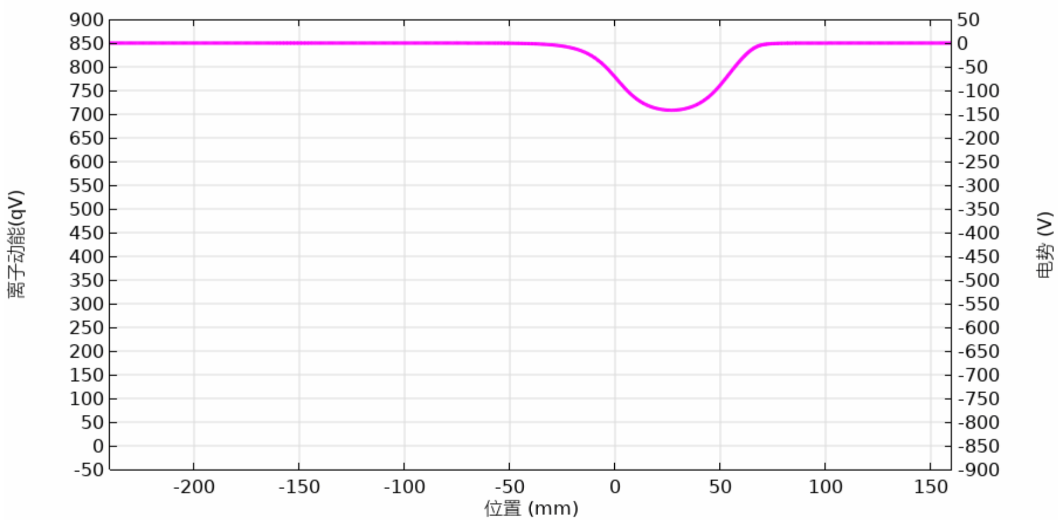
AIP Advances 12, 035220 (2022)

Ion deceleration

➢ Ion deceleration





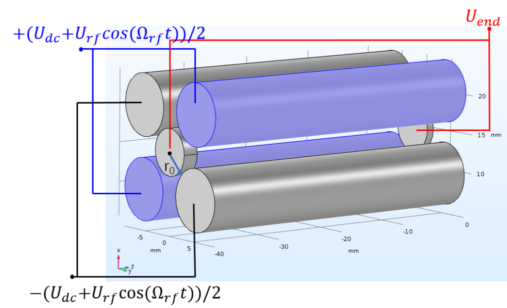


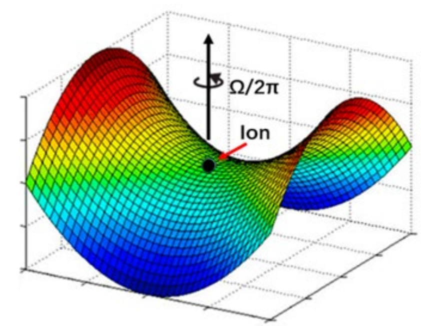
Kinetic Energy was decelerated from 678.2(3) qV to 2.0(3) qV

AIP Advances 12, 035220 (2022)

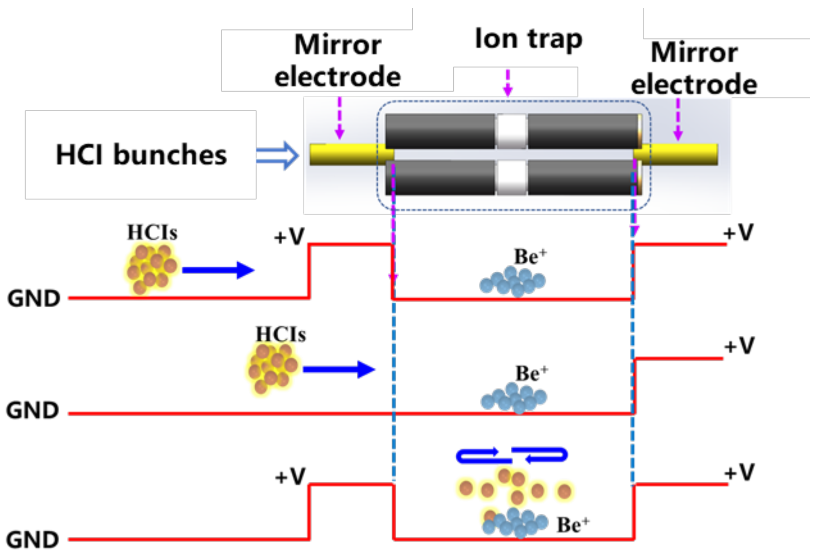
HCI sympathetic cooling

- **Ni-HCI injection**





Timing sequence for ion injection



Ni-HCI injected to ion trap

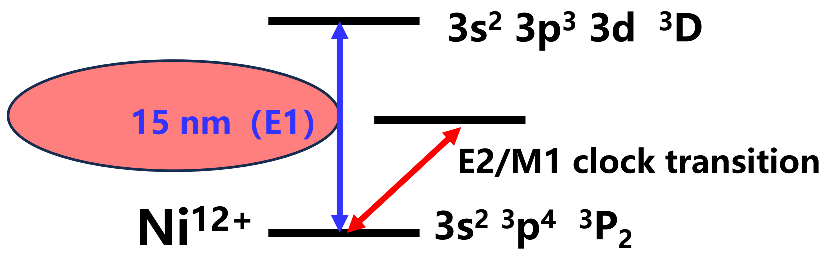
NEXT

Sympathetic cooling

HCI sympathetic cooling

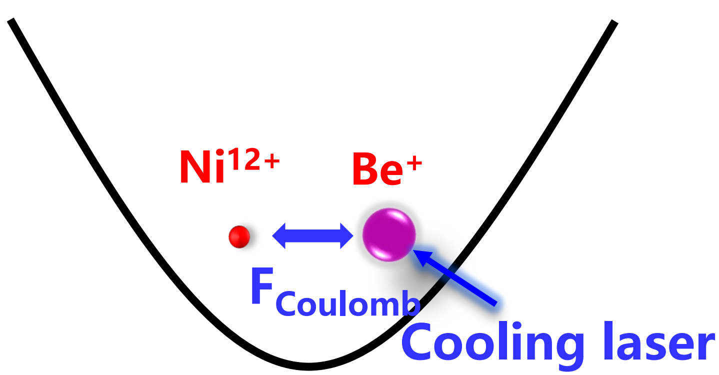
- **Ni-HCI cooling**

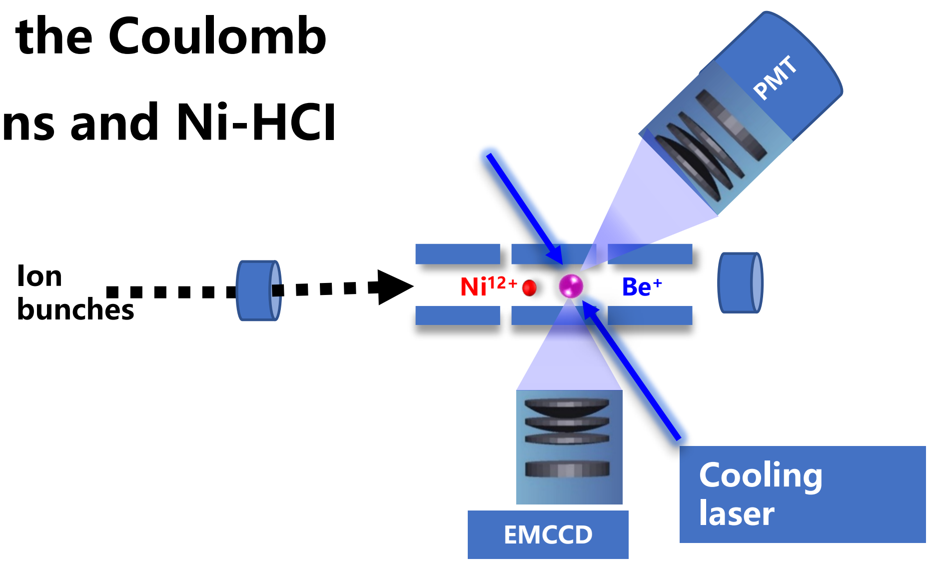
➢ **Laser cooling: E1 transition is located in EUV region**



➢ **Sympathetic cooling: Cooling with the Coulomb interaction between laser cooled ions and Ni-HCI**

Schematic diagram

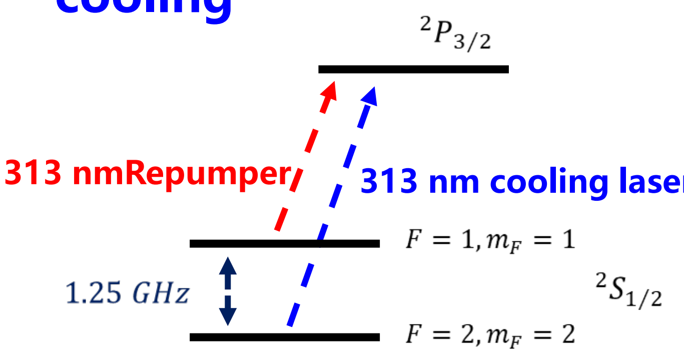




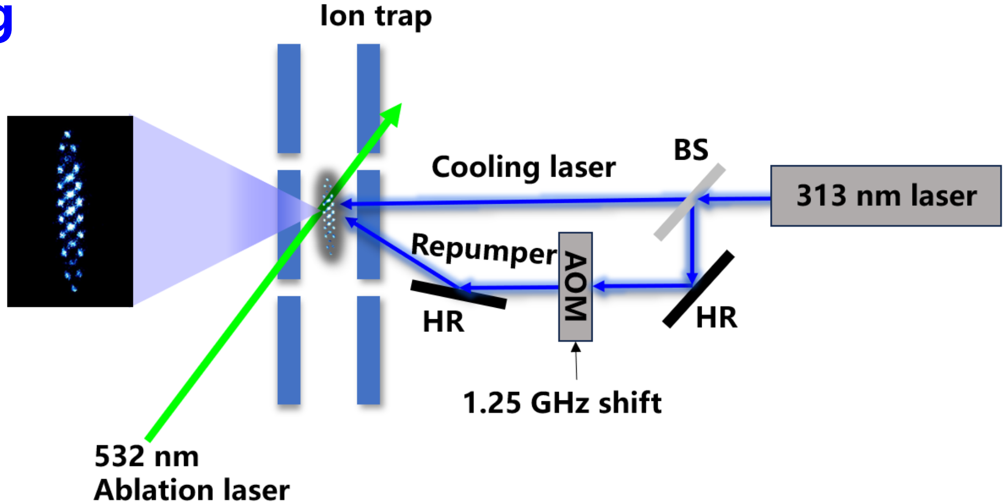
HCI sympathetic cooling

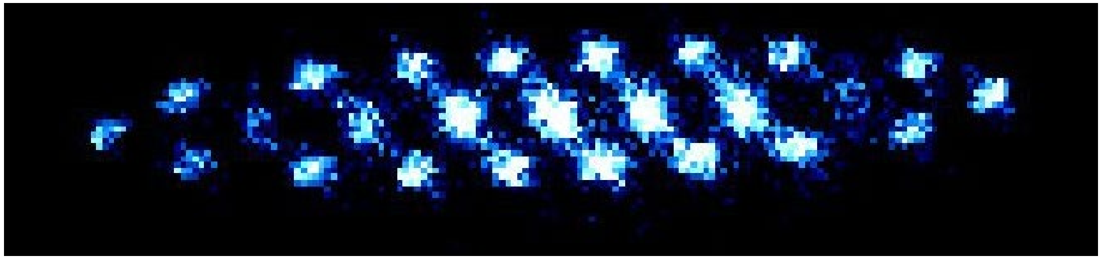
- **Ni-HCI sympathetic cooling**

Be⁺--Laser cooling



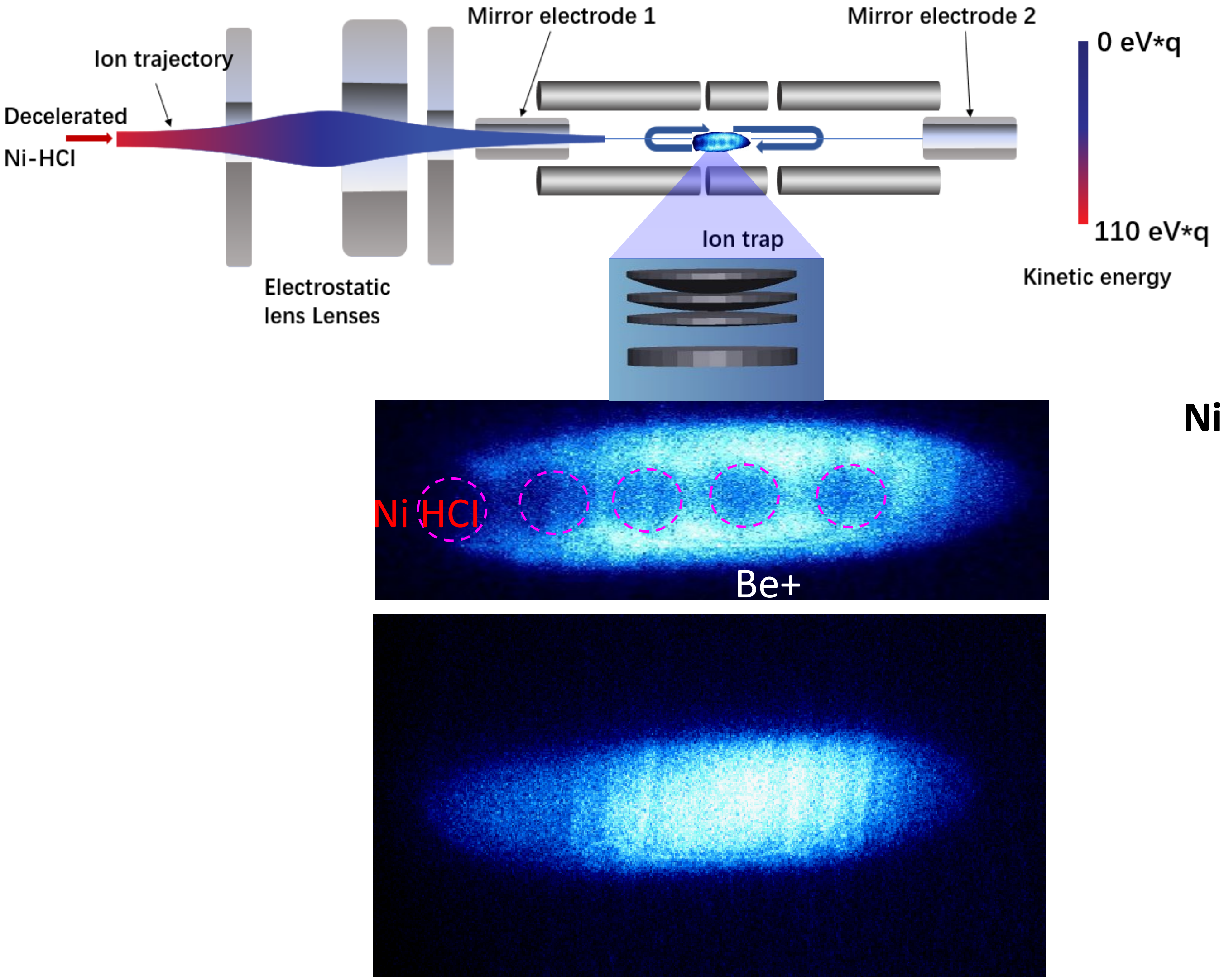
$^9Be^+$ energy level



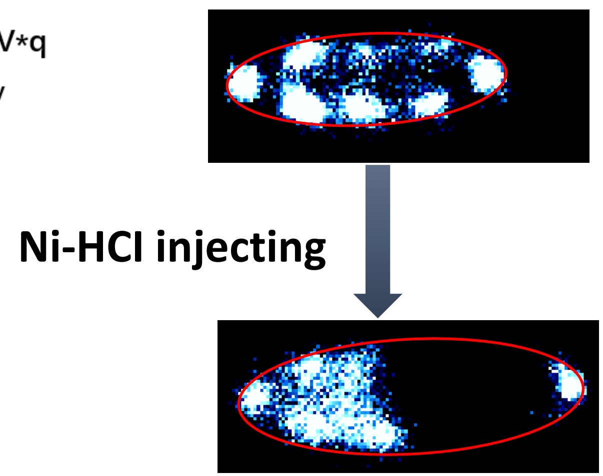


Ion crystal of Be^+

HCI sympathetic cooling



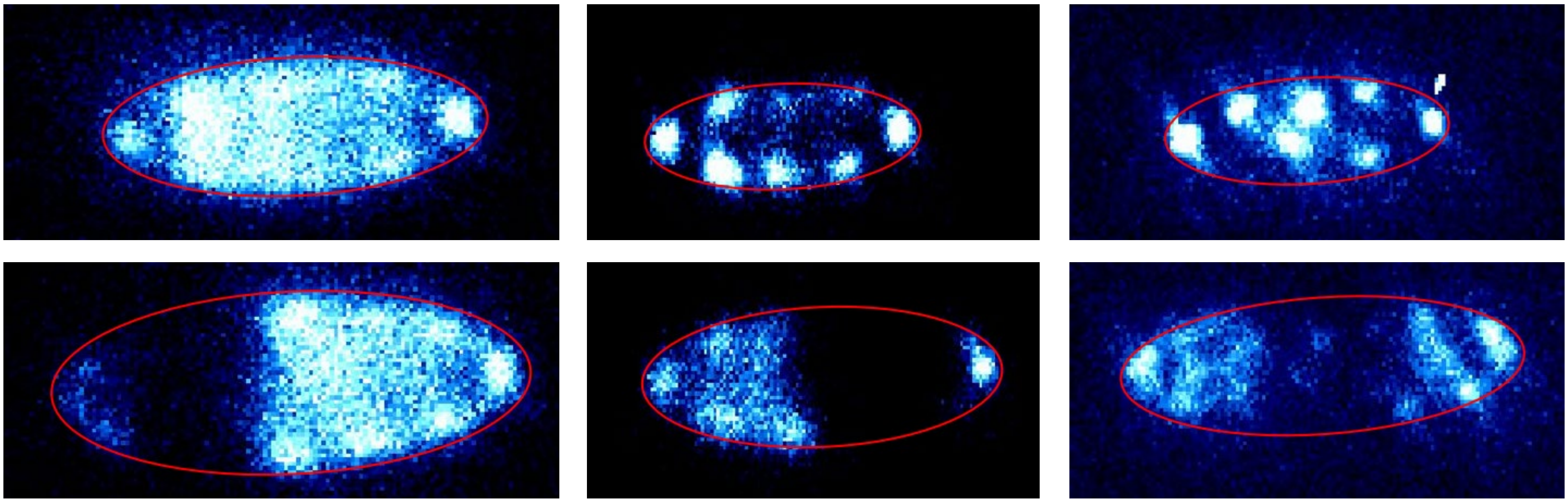
Be$^+$ crystal



Crystal of Ni-HCI and Be$^+$

HCI sympathetic cooling

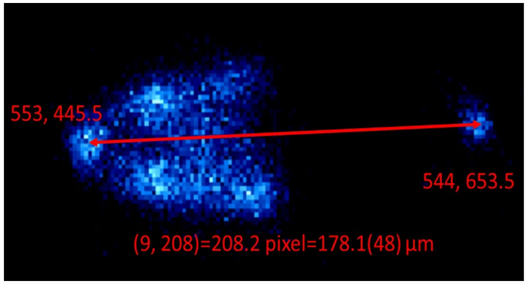
HCI charge state analysis：



$Q_{HCI}= 13\pm3$ Injecting Ni^{13+}

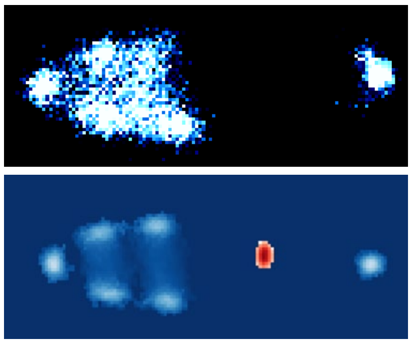
$Q_{HCI}= 12\pm3$ Injecting Ni^{12+}

$Q_{HCI}= 10\pm2$ Injecting Ni^{9+}



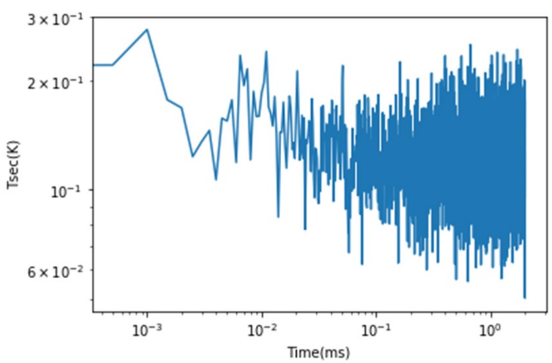
HCI sympathetic cooling

HCI temperature estimating:

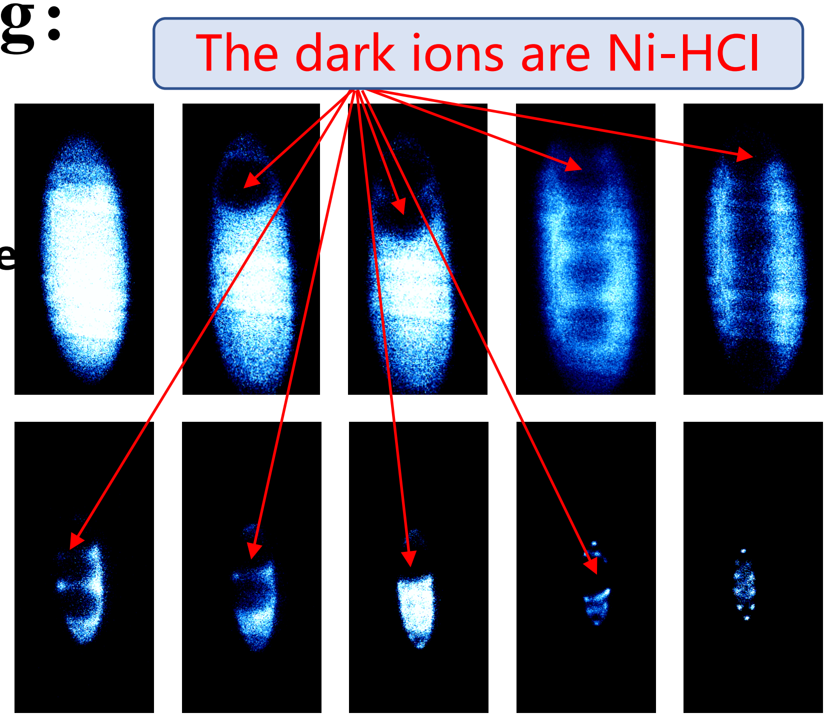


Experimental image

Simulation image



the order of 100 mK



Ni-HCI sympathetic cooling with Be+

The temperature of Ni-HCI was down to hundreds mK from MK, reduced by 7 orders of magnitude

Cryogenic ion trap system

Ion trapping with cryogenic system:

The lifetime of highly charged ion: $\tau = \dfrac{1}{\mathrm{n} \cdot k_L}$

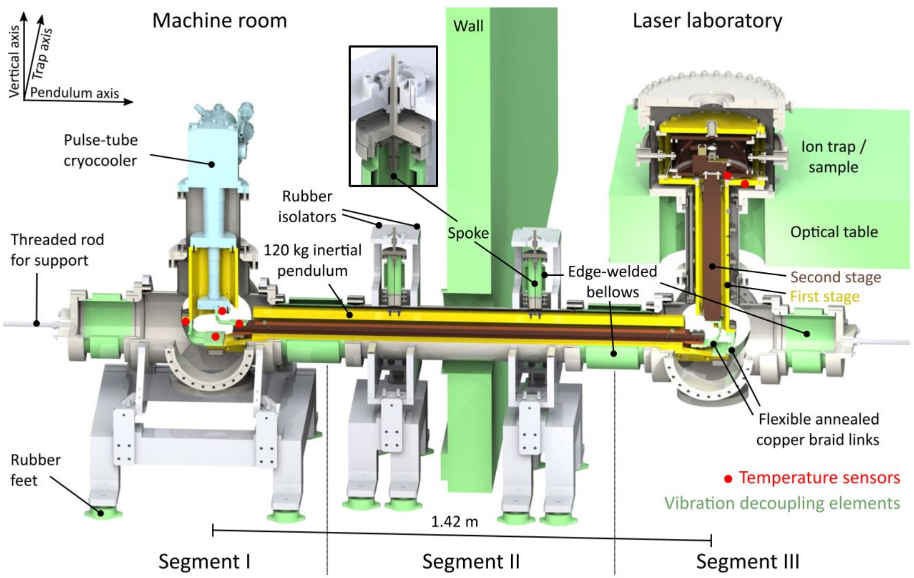
$$k_L = \frac{q}{2\varepsilon_0\sqrt{\alpha/\mu}}$$

$$\alpha = 4\pi\varepsilon_0(0.787 \times 10^{-24})cm^3$$

$$\mu = m_{H_2} \cdot m_{HCI}/(m_{H_2}+m_{HCI})$$

n是剩余气体粒子密度，$\mathrm{n} = p/(k_B \cdot T)$，

p为气压， T为温度



The cryogenic system in MPIK

Temperature of ion trap：4.6K

Vacuum pressure：<1.26 (−0.11/+0.12) ×10^{-12}Pa

Lifetime of $Ar^{13+}$is about 43.1(3.9) min

Ni^{12+}:

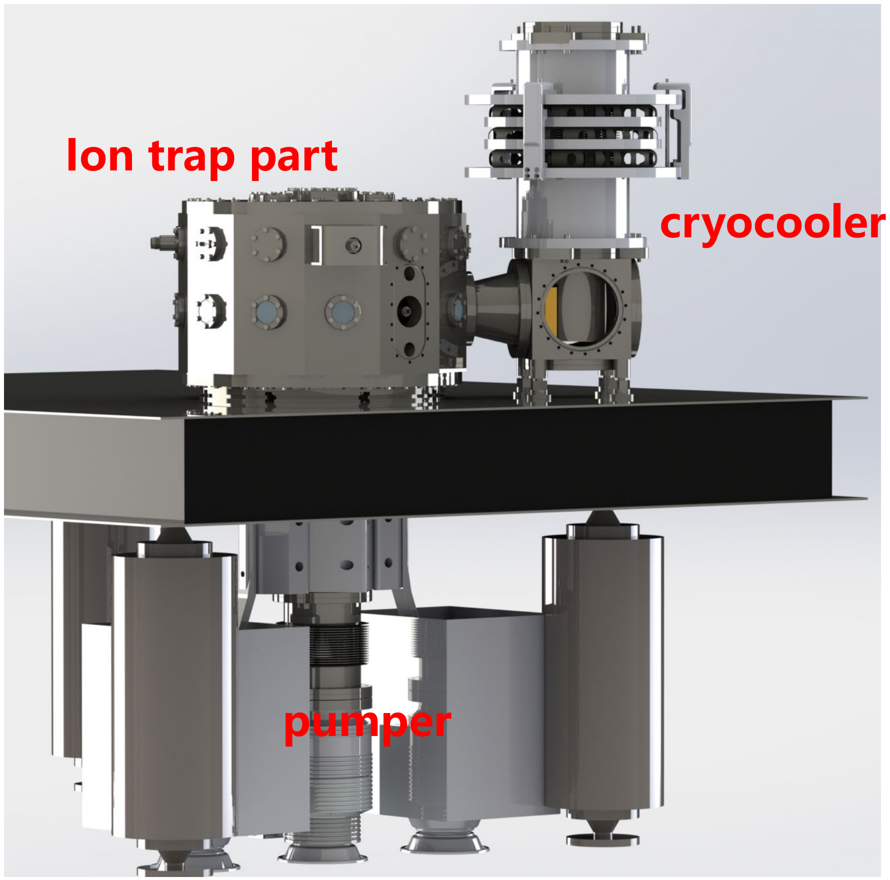
1×10^{-8}Pa，lifetime: about 22s

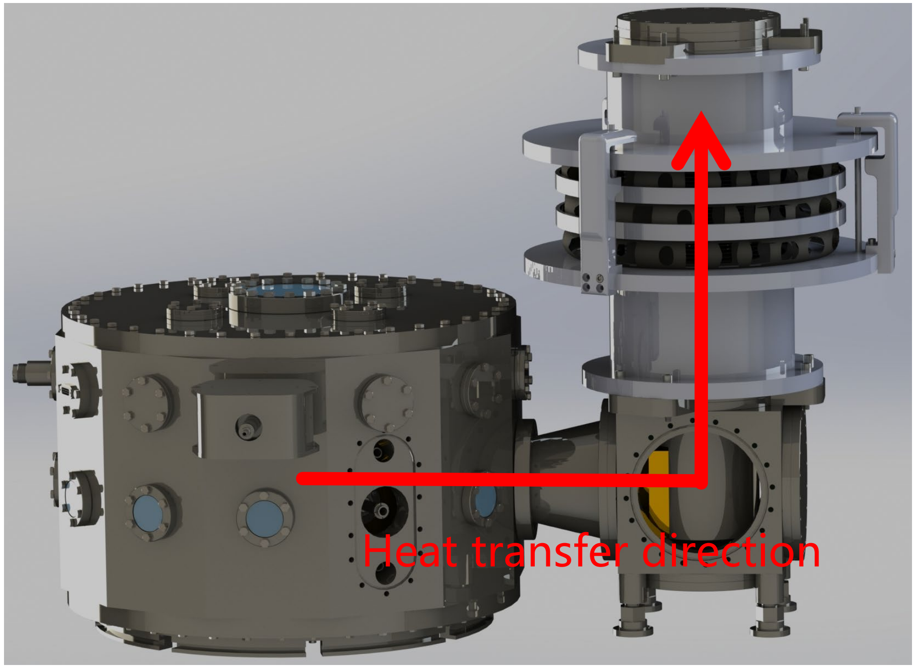
1×10^{-12}Pa， lifetime: more than 30min

P. Micke, *et al*, Rev. Sci. Instrum. **90**, 065104 (2019).

Cryogenic ion trap system

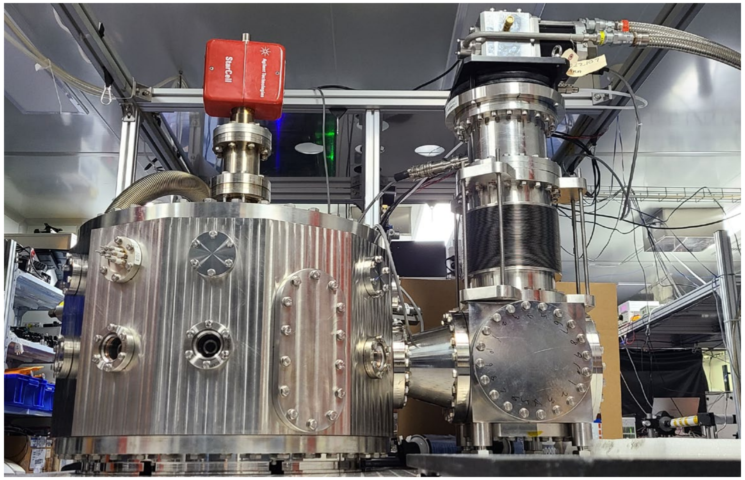
Our design of 4 K cryogenic system

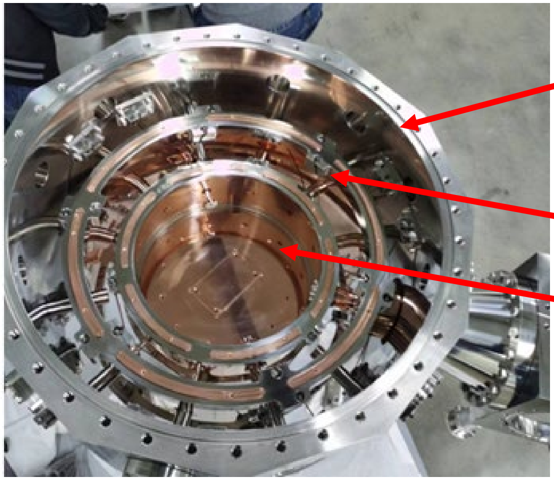




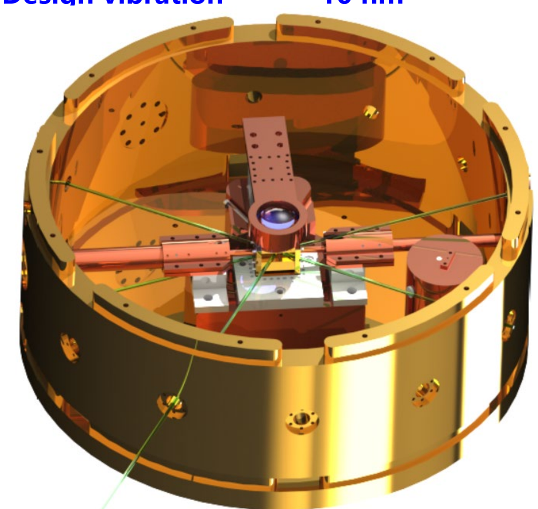
Cryogenic ion trap system

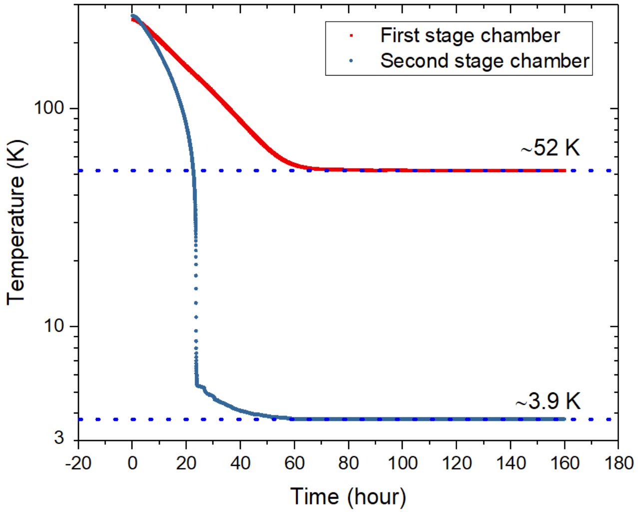
➢4 K cryogenic system





Heat capacity	**>35 W @40K, >1.5 W @4.2K**
Design temperature	**4.5 K**
Design vibration	**10 nm**





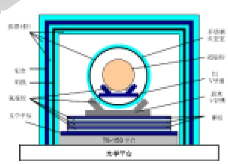
High-precision Ca^+ Optical Clock

Research Process of Ca^+ Optical Clocks at APM

2000

Single ion trapping

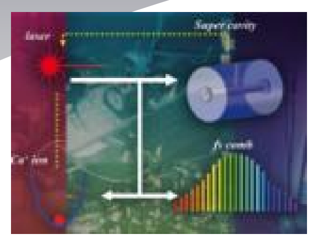
2004



Narrow linewidth laser

2011

The first domestic optical clock (E-16)



Phys. Rev. A, 84, 053841(2011)
Phys. Rev. A, 85, 030503(R) (2012)

2016

Uncertainty to E-17 level

- multiple pairs of Zeeman components
- multi-layer magnetic shields
- Hertz-level narrow linewidth laser
- High precision optical frequency comparison technology

Phys. Rev. Lett. 116, 013006 (2016)
Rev. Sci. Instrum. 87, 063121 (2016)

2022

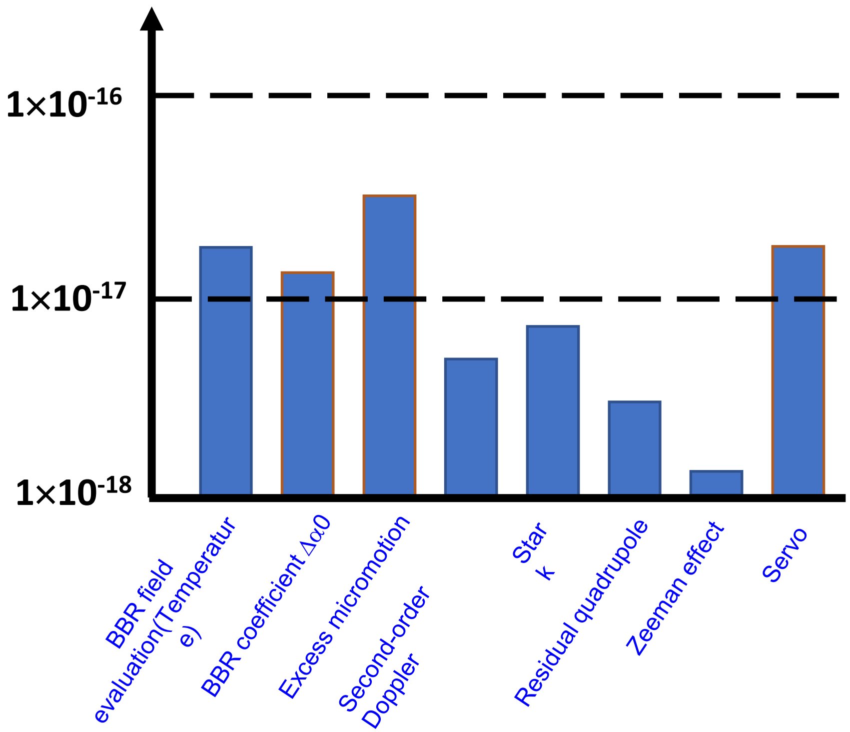
Uncertainty to E-18 level

- magic trap driving
- High precision measurement of the difference between static polarization
- liquid nitrogen cryogenic system
- Hyper Ramsey scheme

Phys. Rev. A 99, 011401(R) (2019)
Phys. Rev. A 102, 050802(R) (2020)
Nat. Sci. Rev. 7, 1799(2020)
Phys. Rev. Applied 17, 024041 (2022)
Metrologia 60, 035004 (2023)
Phys. Rev. Applied 19, 064004 (2023)

Improvement of Calcium Ion Clock

- Uncertainty Evaluation (2016) :



Uncertainty: 5.4×10^{-17}

Only gain/lose <1 s in 600 million years

Phys. Rev. Lett. 116, 013001 (2016)
Appl. Phys. B 123, 166 (2017)

Improvement of Calcium Ion Clock

- Suppress BBR shift – the measurement of the differential static polarizability

Micromotion-induced frequency shift：

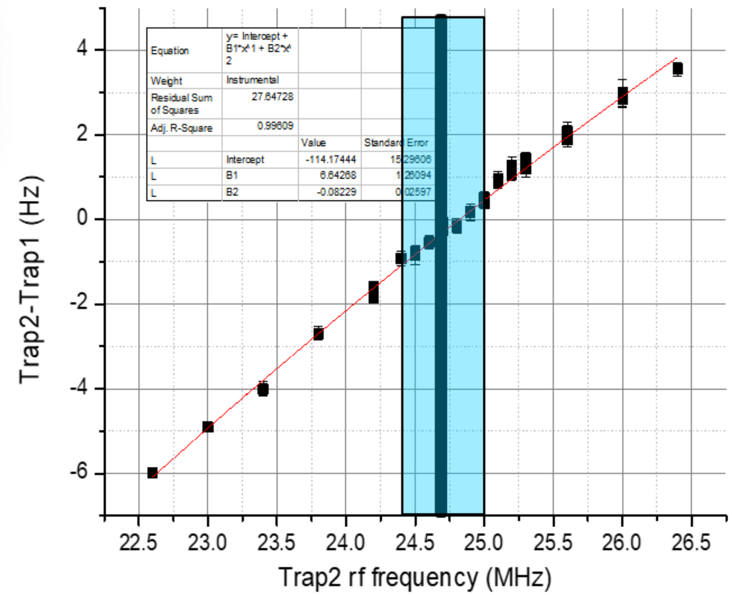
$$\boxed{\frac{\Delta\nu_\mu}{\nu_0} = -\left(\frac{\Omega}{\omega_0}\right)^2 \left[1 + \frac{\Delta\alpha_0}{\hbar\,\omega_0}\left(\frac{m\,\Omega\,c}{e}\right)^2\right] \sum_{x,y,z} R_i\,.}$$

Magic RF trapping drive frequency：

$$\boxed{\Omega_0^2 \simeq -\frac{h\nu_0}{\Delta\alpha_0}\left(\frac{e}{mc}\right)^2}$$

BBR shift ：

$$\boxed{\delta\nu_{BBR} = -2\times(831.9Vm^{-1})^2\left(\frac{\mathrm{T}}{300}\right)^3\Delta\alpha_0(1+\eta)\Delta\mathrm{T}}$$



- Theory: 24.7(6) MHz
- Experiment: 24.801(2) MHz

Madej *et al* , Phys. Rev. Lett. 109, 203002 (2012)
Dubé *et al,* Phys. Rev. Lett. 112, 173002 (2014)

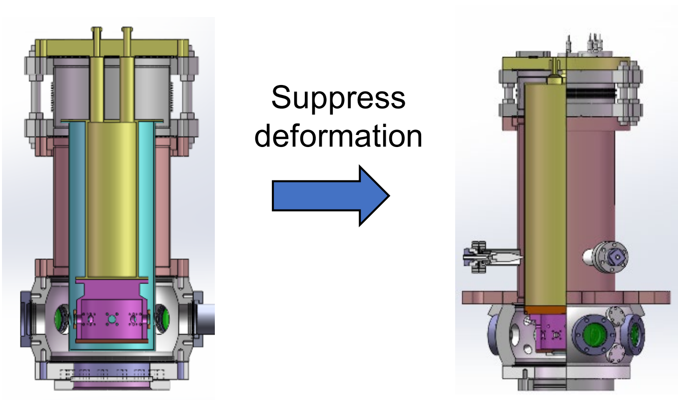
Safronova *et al,* Phys. Rev. A 83,012503(2011)

Phys. Rev. A 99, 011401(R) (2019)

Improvement of Calcium Ion Clock

➢ Suppress BBR shift – temperature

$$\delta\nu_{BBR} = -2\times(831.9Vm^{-1})^2\left(\frac{T}{300}\right)^3\Delta\alpha_0(1+\eta)\Delta T$$



- **Room temperature ~295K：**

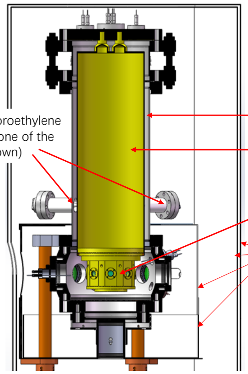
  BBR uncertainty with a temperature uncertainty of 3 K: 3.4×10^{-17}

- **Liquid nitrogen environment~77K：**

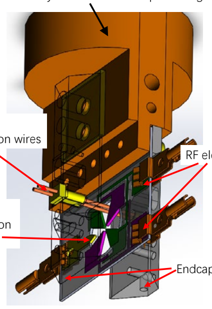
  BBR uncertainty with a temperature uncertainty of 3 K: 7×10^{-19}

Improvement of Calcium Ion Clock

➢ Suppress BBR shift – temperature



BBR environment	Solid angle percentage	Temperature (K)	Contributions to the total BBR shift uncertainty (10^{-18})
BBR shield (copper)	33.64%	82(5)	0.43
BBR shield (BK7 glass)	5.45%	82(5)	0.07
Ion trap (including the compensation wires)	14.19%	82(5)	0.18
Endcaps	46.63%	82(5)	0.59
Holes on the shield	0.09%	293.0(1.5)	1.46
Total	100%		2.7



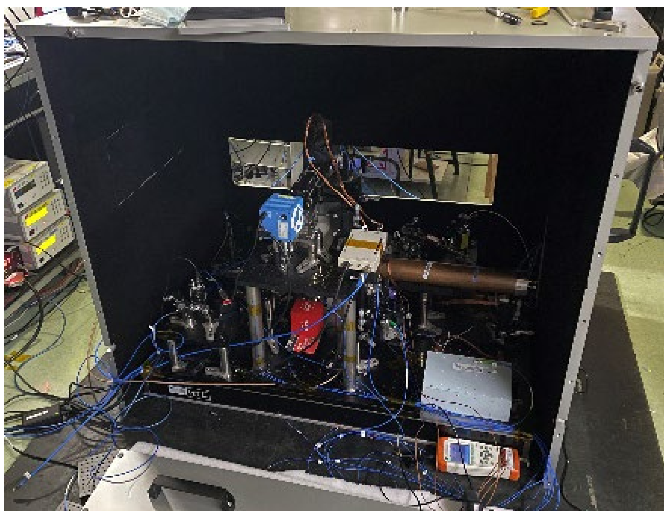
BBR shift uncertainty owing to the temperature uncertainty: 2.7×10^{-18}

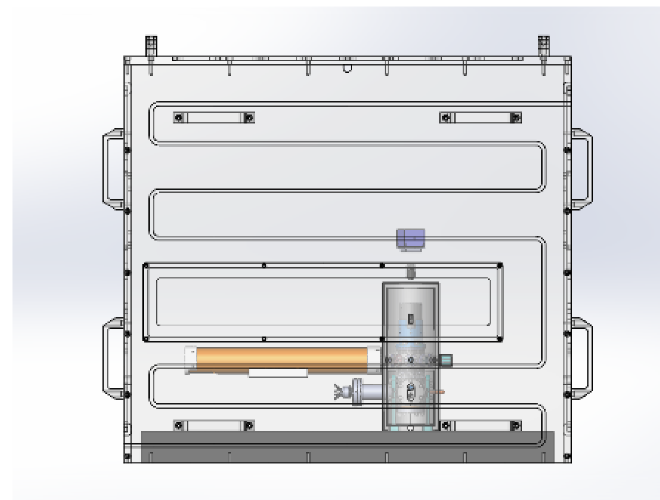
Phys. Rev. Applied, 17, 024041 (2022)

Improvement of Calcium Ion Clock

- Suppress BBR shift – temperature variation

$$\delta\nu_{BBR} = -2\times(831.9Vm^{-1})^2(\frac{T}{300})^3\Delta\alpha_0(1+\eta)\Delta T$$





Room temperature system with active temperature control

Temperature fluctuations ： ±0.3℃

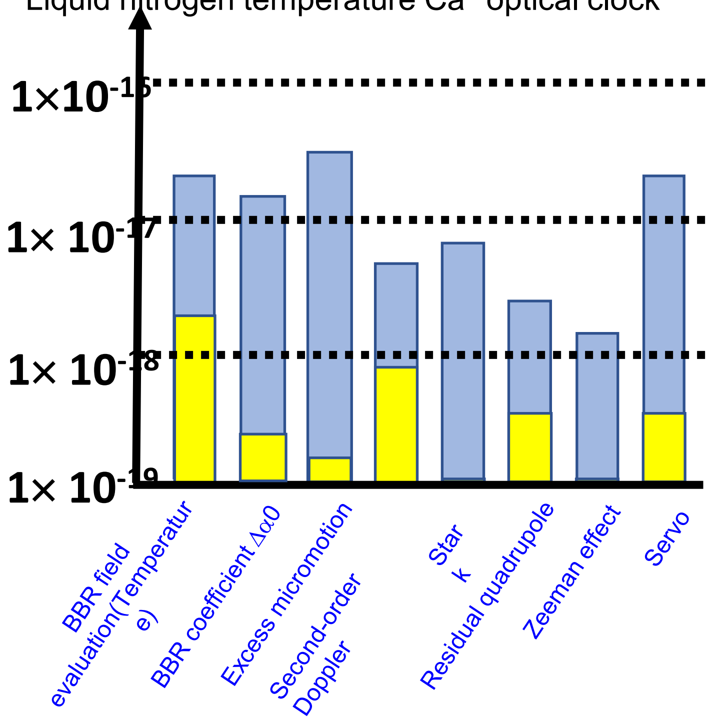
BBR Shift Uncertainty owing to

temperature variation: 4.6×10^{-18}

High-Precision Ca^+ Optical Clocks

Uncertainty:

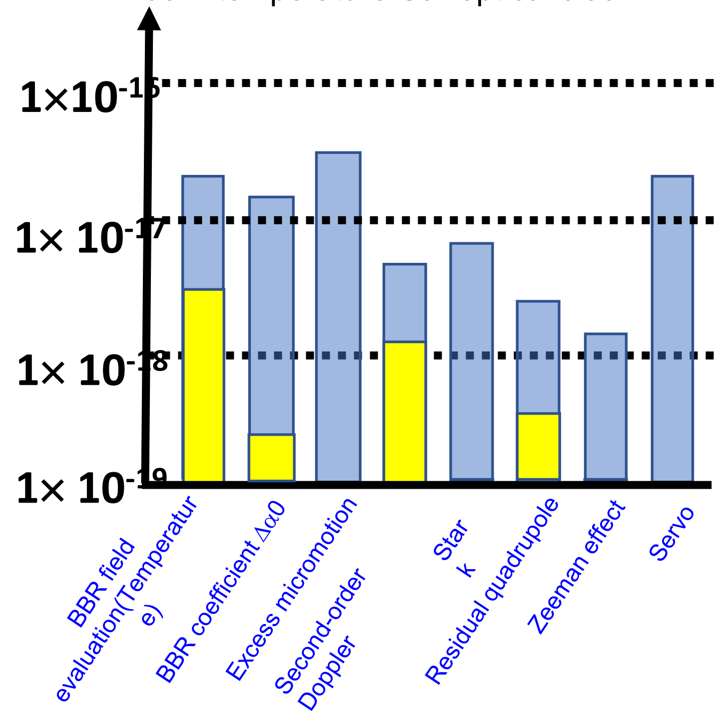
Liquid nitrogen temperature Ca^+ optical clock



Uncertainty 3×10^{-18}

Phys. Rev. Applied, 17, 024041 (2022)

Room-temperature Ca^+ optical clock



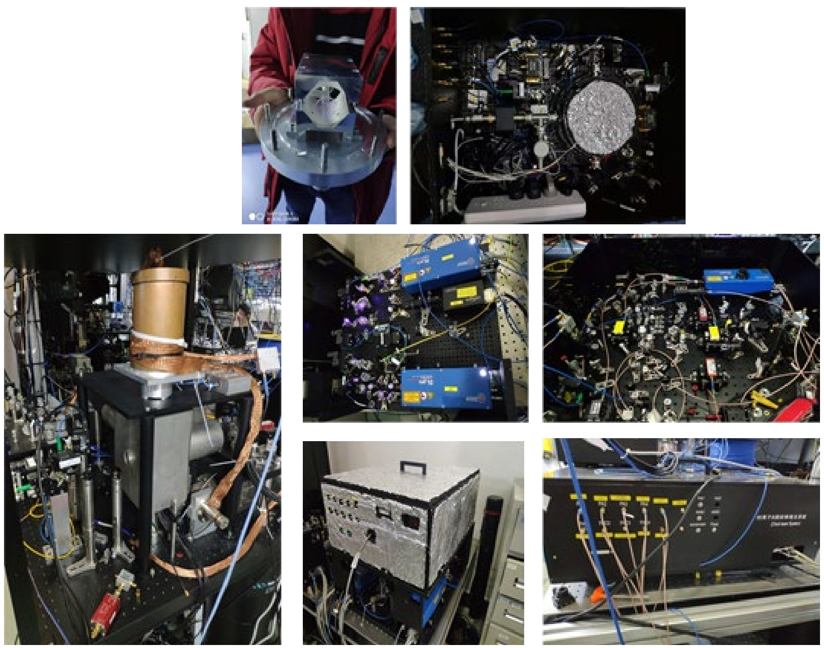
Uncertainty 4.8×10^{-18}

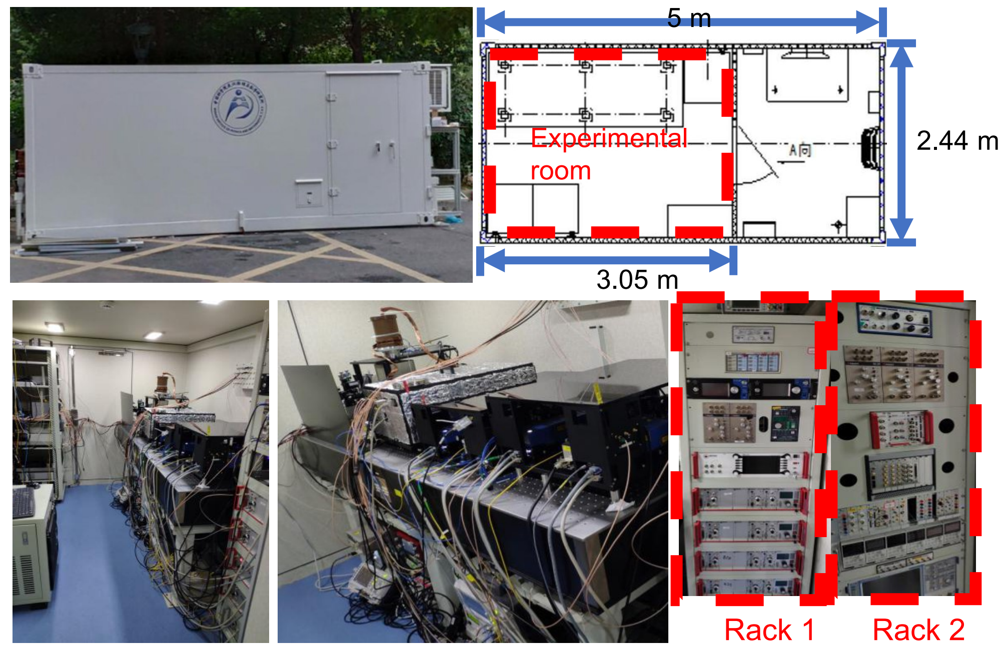
Phys. Rev. Applied 19, 064004 (2023)

Only gain/lose < 1 s in 10.5 billion years

Transportable Ca^+ Optical Clock

- system relatively simple and easy to move for application
- ensure high reliability and operating rate





Transportable Ca⁺ Optical Clock

Transportation:

Comparisons of transportable optical clocks

	Research Unit	Uncertainty	Transportable distance	operating rate
Sr	The University of Tokyo [1]	5.5×10^{-18}	< 10 km	40%
Ca^+	Our group [2]	1.3×10^{-17}	1200 km	75%
Sr	Physikalisch-Technische Bundesanstalt (PTB) [3]	7×10^{-17}	1000 km	20%

[1] Takamoto *et al*, Nat. Photon. 14, 411 (2020)
[2] Huang *et al,* Phys. Rev. A 102, 050802(R) (2020)
[3] Grotti *et al,* Nat. Phys. 14, 437 (2018)

The second optical clock in the world to achieve long-distance transportation of thousands of kilometers

Transportable Ca^+ Optical Clock

- Absolute frequency measurement accuracy at E-16 level with a transportable optical clock in 2020
- absolute frequency of the Ca^+ clock was adopted by CCTF, and endorsed it as a secondary representation of the definition of the second in 2021

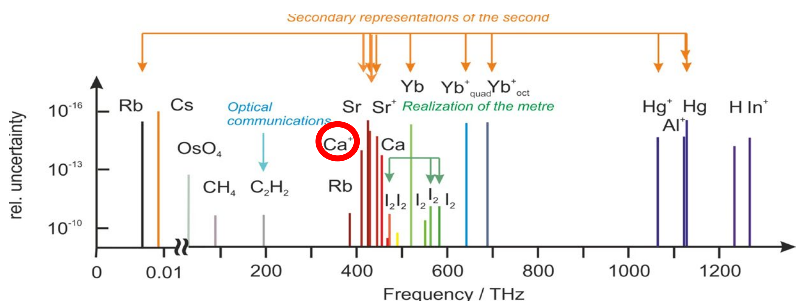
RECOMMENDATION CCTF PSFS 2 (2021)

Updates to the CIPM list of standard frequencies[1]

The Consultative Committee for Time and Frequency (CCTF), at its 22nd session in 2020 and 2021

- the unperturbed optical transition 4s $^2S_{1/2}$ – 3d $^2D_{5/2}$ of the $^{40}Ca^+$ ion with a frequency of f_{40Ca+} = 411 042 129 776 400.4 Hz and an estimated relative standard uncertainty of 1.8×10^{-15};

This radiation is now endorsed as a secondary representation of the second;



https://www.bipm.org/en/committees/cc/cctf/22-_2-2021

Bureau International des Poids et Mesures

Approved by the CCTF in March 2021

RECOMMENDED VALUES OF STANDARD FREQUENCIES FOR APPLICATIONS INCLUDING THE PRACTICAL REALIZATION OF THE METRE AND SECONDARY REPRESENTATIONS OF THE DEFINITION OF THE SECOND

CALCIUM 40 ION ($f \approx 411$ THz)

^{40}Ca +, 4s $^2S_{1/2}$ – 3d $^2D_{5/2}$ unperturbed optical transition

1. **Recommended value [1] of the frequency in the CIPM List of Frequencies**

$f(^{40}Ca^+)$ = 411 042 129 776 400.4 Hz

equivalent to

$\lambda(^{40}Ca^+)$ = 729 347 276.793 942 7 fm,

with a relative standard uncertainty of 1.8×10^{-15}.

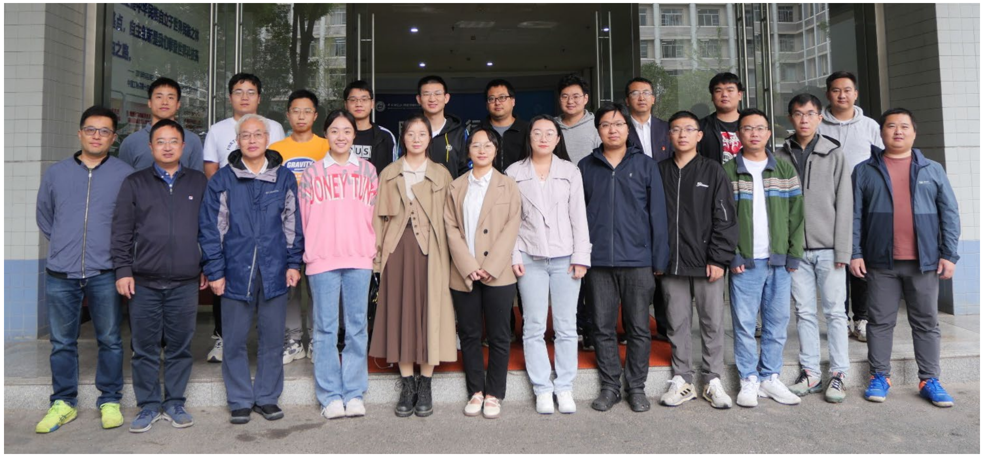
This radiation is endorsed by the CCTF as a secondary representation of the definition of the second [1].

a new atomic transition frequency to be a secondary representation of the SI second

Summary and Outlook

- Highly charged ion clock:
  - ✓ Preliminary measurement of Clock transition, sympathetic cooling of the Ni-HCI.
- Laboratory $^{40}Ca^{+}$ optical clock:
  - ✓ Uncertainty of E-18 level , Stability of E-18 level
- Transportable $^{40}Ca^{+}$ optical clock:
  - ✓ Transportation distance > 1200 km, Accuracy of clock transition frequency to E-16 level
- Targets：
  - ◆ Laboratory Clock: Uncertainty to the E-19 level
  - ◆ Transportable Clock: Uncertainty to the E-18 level, Miniaturization, High operating rate, and automated operation
  - ◆ Highly charged ion: Ni^{12+} highly charged ion clock with uncertainty to E-19 level.

Acknowledgement



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Theory: Tingyun Shi, Zongchao Yan, Liyan Tang, Jiguang Li, Yanmei Yu, Chenbing Li …

Chaohui Ye, Jun Luo, Yuzhu Wang, Yiqiu Wang, Jiaming Li, Li You, Zhengtian Lu …

J. Ye, P. Gill, J. Bergquist, D. Leibrandt, J. Pan, A. Derevianko, Piet O. Schmidt, E. Peik, P. Dube, B. K. Sahoo, José R. Crespo López-Urrutia, A. Derevianko …

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