



Highly Charged Ion Optical Clocks to Test Fundamental Physics



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Physikalisch-Technische Bundesanstalt





- National Metrology Institute, founded 1887
- Tasks: determination of fundamental constants, dissemination of SI units, development of measurement techniques,...
- ca. **2100 employees**, of which are **>200 PhD candidates**
- 60% research: >600 publications per year

Quantum Logic Spectroscopy Group



www.quantummetrology.de/eqm

Overview

- Introduction to clocks
- Introduction to highly charged ions
- State-of-the art generation & spectroscopy of highly charged ions
- Quantum logic spectroscopy of Ar¹³⁺
- Ar¹³⁺ optical clock
- search for 5th forces: ^{40,42,44,46,48}Ca¹⁴⁺ isotope shift
- Summary & future



[Overview article: Schmidt & Crespo, Physik Journal **15** (2016) Nr. 10 Review: Kozlov, Safronova, Crespo, Schmidt, Rev. Mod. Phys **90**, 045005 (2018)]

Highly Charged Ions

Charge state dependence:

- Binding energy $\sim Z^2$
- Hyperfine splitting $\sim Z^3$
- QED effects $\sim Z^4$
- Stark shifts ~ Z⁻⁶



strongly relativistic systems with large QED effects

• optical transitions: fs, hfs, level crossings [Kozlov *et al.* Rev. Mod. Phys **90**, 045005 (2018]

- H \rightarrow U^{91+} (H-like)10 eV \rightarrow 140 keV
- $\mu eV \rightarrow eV$

μeV

 \rightarrow 300 eV



Applications of HCI

- abundant in the universe
 → probes for cosmic & stellar processes
- diagnostic tools for plasma physics









XUV light generation in Sn¹²⁺ @ 13.5 nm for semiconductor fabs

Testing fundamental physics with HCI

- few remaining electrons \rightarrow calculable systems
- features
 - strong QED effects: g-factor measurements \rightarrow QED in strong fields
 - strongly relativistic & high ionization potential:
 - \rightarrow highest sensitivity of all atomic species to $\dot{\alpha}$ & dark matter
 - strongly relativistic \rightarrow violation of local Lorentz invariance
 - electrons close to nucleus
 - \rightarrow isotope shifts (5th forces), nuclear physics, parity violation, ...





[Reviews: Indelicato, J. Phys. B 52, 232001 (2019), Safronova et al., RMP 90, 025008 (2018), Kozlov et al. RMP 90, 045005 (2018)]

Effects of dark matter on normal matter



Variation of Fundamental Constants & Dark Matter



$$\frac{\Delta\omega}{\omega} = \mathbf{K} \frac{\Delta\alpha}{\alpha}$$





Apparent changes in α may be induced by scalar fields ϕ , e.g. dark matter



Variation of Fundamental Constants



$$\frac{\Delta\omega}{\omega} = \mathbf{K} \frac{\Delta\alpha}{\alpha}$$



$$\Delta \omega$$
 $\Delta \alpha$

 System
 K
 λ (nm)

 Sr
 0.06
 699

 Yb⁺ E2
 0.91
 436

 Yb⁺ E3
 -6
 467

 Hg⁺
 -2.9
 281.5

 Al⁺
 0.01
 267

Combined data from clocks



Principle of Optical Clocks



Clock characterization

Two (main) notions: Stability & Accuracy



- (In-)Stability: How large is the scatter of data points?
 → Uncertainty type A (u_A, only statistical) "statistical uncertainty"
- (In-)Accuracy: How well is the unperturbed reference reproduced?
 - \rightarrow Uncertainty type B (u_B , outside information) "systematic uncertainty"

Problem: value of unperturbed reference a priori not known!

Why optical clocks?



Evolution of (estimated) clock accuracy



Highly charged ions as optical clocks?

- High accuracy
 - \rightarrow low sensitivity to resonance shifts
- HCI advantage: suppressed shifts



Hydrogen-like HCI:

Linear Stark shift	Z^{-1}			
Second order Stark shift	Z^{-4}			
Linear Zeeman shift	Z ⁰			
Second order Zeeman shift	Z^{-34}			
Electric quadrupole shift	Z^{-2}			
$[D_{1}, \dots, D_{n}] = [D_{1}, D_{2}, D_{2}, D_{2}, \dots, D_{n}]$				

[Berengut et al., PRA 86, 022517 (2012)]



electric & magnetic fields

Other clock species requirements can be fulfilled:

narrow, laser accessible transition, simple level structure, ...

[dozens of proposals, many summarized in: Kozlov et al., Rev. Mod. Phy. 90, 045005 (2018)]

Many HCI to choose from...



State-of-the-art HCI spectroscopy



- Problem #1: Electron beam ion trap (EBIT) is a noisy & hot environment
- Problem#2: No cycling transition for cooling & state detection



Doppler-limited resolution of $\sim 150 \text{ MHz}$

Solution #1: Transfer HCIs into a Paul trap

Features of trapped ions

- large trap frequencies
 recoil-free absorption
- long interrogation times
- trap ion in zero field
 small trap induced shifts
- isolated from environment
 + laser cooling
 + no interactions





 $d_{ion\text{-}electr}$ \sim 0.8 mm ω_z \sim 2 MHz, ω_r \sim 4 MHz

Yb⁺ single-ion clock: 3 × 10⁻¹⁸ [Sanner *et al.*, Nature **567**, 204 (2019)]

Solution #2: Quantum Logic Spectroscopy



Many more applications demonstrated, e.g.

- Most accurate clock: Al⁺ [Brewer *et al.*, PRL **123**, 033201 (2019)]
- Molecular ion state detection and preparation [Wolf et al., Nature 530, 457 (2016); Chou et al., Nature 545, 203 (2017)]
- ions in linear Paul trap \rightarrow high accuracy achievable
- logic ion provides sympathetic cooling & signal readout
- strong Coulomb interaction couples motional modes
- composite system: combine advantages of both species
- \rightarrow investigation of previously inaccessible species

[D.J. Wineland et. al., Proc. 6th Symposium on Frequency Standards and Metrology, 361 (2001); P.O. Schmidt et al., Science, 309, 749 (2005)]

Approach to precision HCI spectroscopy: CryPTEx-PTB



Specs vacuum system:

- Vacuum: < 10⁻¹⁴ mbar
 → HCI lifetime: ~ 100 min
- Temperature: < 5 K
- Vibrations: < 20 nm
- Magnetic field: < 200 pT

Specs EBIT:

- Magnetic field: 0.86 T (72 permanent magnets)
- Acceleration voltage: 10 kV
- Current: > 80 mA

Specs ion trap:

- 5 segments, Au-coated Al₂O₃, 0.7 mm ionelectrode distance
- Trapping frequencies:
 > 1 MHz
- Heating rates: $\sim 1 \text{ 1/s}$
- f/# ~ 1 imaging with bi-aspheric lens

Quantum Logic with Trapped Ions



Ω: Carrier Rabi frequency; $\eta = kz_0$: Lamb-Dicke factor

Doppler cooling & charge state identification



 single Be⁺ axial frequency: 0.995 MHz
 → Be⁺/Ar¹³⁺ axial frequencies: 1.47 MHz and 1.99 MHz



Sympathetic axial ground state cooling of Ar¹³⁺



- resolved Raman sideband cooling on Be⁺ + algorithmic cooling of radial modes
- Lamb-Dicke parameter: $\eta_z = 0.82 \sqrt{MHz/\nu_z}$

[Leopold et al., Rev. Sci. Instrum. 90, 073201 (2019); King et al., PRX 11, 041049 (2021)]



Quantum Logic State Transfer



[D.J. Wineland et. al., Proc. 6th Symposium on Frequency Standards and Metrology, 361 (2001); P.O. Schmidt et al., Science, 309, 749 (2005)]

Quantum Logic Spectroscopy of Ar¹³⁺



 spectroscopy laser transfer locked of Ar¹³⁺ to Si cavity-stabilized laser [Sterr & Benkler @ PTB: D. G. Matei *et al.*, Phys. Rev. Lett. **118**, 263202 (2017)]

How to turn this into a clock?

Evaluation of systematic frequency shifts

Time dilation shift: cooling challenges....

Large q/m mismatch between ions

→ large difference in radial amplitudes
→ inefficient cooling of radial modes



• radial mode x_2 : \rightarrow weak cooling





[King et al., PRX 11, 041049 (2021)]



Ground-state cooling using quantum logic



Algorithmic cooling: results



[King et al., PRX 11, 041049 (2021)]

Systematic shifts for Ar¹³⁺

Shift source	Mitigation	Shift (10 ⁻¹⁸)	$\begin{matrix} \text{Uncertainty} \\ (10^{-18}) \end{matrix}$		
Micromotion	Real-time measurement	-443	22	٦	
AC Zeeman shift	Calibration at much higher powers and extrapolation	0	2	no fundamental limitations	
First-order Doppler	Counter-propagating beams	0	< 1		40 Ar ¹³⁺ clock with
Electric quadrupole	Small coefficient, averaging over multiple Zeeman components	0	< 1		estimated systematic uncertainty
Linear Zeeman	Averaging over multiple Zeeman components	0	< 1		
Quadratic Zeeman	Small coefficient, small field	< 1	≪1		8 orders of magnitude
2 nd order Doppler	Algorithmic cooling	-1	< 1		Improvement

atomic data from: [Y-M. Yu and B.K. Sahoo, PRA **99**, 022513 (2019)]

[King, Spieß *et al.*, Nature **611**, 43-47 (2022)]

Clock operation



Frequency ratio measurement Ar¹³⁺/Yb⁺ E3



- Frequency ratio uncertainty limited by Ar¹³⁺ excited state lifetime to $\sim 3 \times 10^{-14} / \sqrt{\tau}$
- Measurements to $\sim 1 \times 10^{-16}$ statistical uncertainty for 40 Ar¹³⁺ and 36 Ar¹³⁺
- Yb⁺ E3 absolute frequency known with 1.3×10^{-16} fractional uncertainty

[King, Spieß et al., Nature 611, 43-47 (2022)]

Isotope shift ${}^{40}Ar^{13+} \leftrightarrow {}^{36}Ar^{13+}$



[King, Spieß *et al.*, Nature **611**, 43-47 (2022)]

Isotope shift ${}^{40}Ar^{13+} \leftrightarrow {}^{36}Ar^{13+}$



[Soria Orts *et al.,* PRL **97**, 103002 (2006)]

[see also: T. Sailer *et al.* Nature **606**, 479–483 (2022)]

[King, Spieß et al., Nature 611, 43-47 (2022)]

Search for 5th forces



$$\delta v_i^{A,A'} = F_i \delta \langle r^2 \rangle_{A,A'} + k_i \frac{A - A'}{A A'} + \alpha_{NP} X_i (A - A')$$



IS transition 2

[Stadnik, PRL **120**, 223202 (2018); Berengut *et al.*, PRL **120**, 091801 (2018); Frugiuele *et al.*, Phys. Rev. D **96**, 015011 (2017); Delaunay *et al.*, Phys. Rev. D **96**, 093001 (2017); Delaunay *et al.*, Phys. Rev. D **96**, 115002 (2017)]

-1970

Isotope shift spectroscopy of ^{40,42,44,46,48}Ca^{+/14+/15+}

need: pair of narrow transitions with different electronic character

promising approach: isotope shifts of clock transitions in Ca⁺ & Ca¹⁴⁺





related experimental work:

[Chang et al., arXiv:2311.17337 Figueroa et al., PRL **128**, 073001 (2022) Ohayon et al., NJP **24**, 123040 (2022) Hur et al., PRL **128**, 163201 (2022) Ono et al., PRX **12**, 021033 (2022) König et al., PRC **103**, 054305 (2021) Neely et al., PRA **103**, 032818 (2021); Solaro et al., PRL **125**, 123003 (2020); Counts et al., PRL **125**, 123002 (2020);

Bhatt *et al.*, PRA **101**, 052505 (2020); Müller *et al.*, Phys. Rev. Research **2**, 043351 (2020); Miyake *et al.*, Phys. Rev. Research **1**, 033113 (2019); Manovitz *et al.*, PRL **123**, 203001 (2019); Knollmann *et al.*, PRA **100**, 022514 (2019); Imgram *et al.*, PRA **99**, 012511 (2019); Gebert *et al.*, PRL **115**, 053003 (2015)]



[Rehbehn, et al., PRA 103, L040801 (2021)]

Frequency ratio measurements ^{40,42,44,46,48}Ca¹⁴⁺ / Yb⁺ E3



Comparison: E. Benkler, Yb⁺ E3 clock: M. Filzinger, N. Huntemann

- Yb⁺ E3 absolute frequency: 1.3×10^{-16}
- statistical uncertainty: $\sim 1 \times 10^{-16} \rightarrow 0.1$ Hz (improvement by $\times 10^{-9}$)
- systematic uncertainty: $\sim 3 \times 10^{-17}$

[A. Wilzewski et al., in preparation]

Higher order SM contributions



[also: Flambaum et al., PRA 97, 032510 (2018); Yerokhin et al., PRA 101, 012502 (2020); Counts et al., PRL 125, 123002 (2020)]

Outlook

Future: competitive clocks & fundamental physics

- Clock candidate: ⁵⁸Ni¹²⁺ [Yu & Sahoo, Phys. Rev. A 97, 041403 (2018); Chen *et al.*, Phys. Rev. Res. 6, 013030 (2024)] also: Gao (Wuhan) Challenge: transition energy uncertainty of several THz (few nm)
- dark matter & α -sensitive level-crossings:
 - Pr⁹⁺ [Bekker *et al.*, Nat. Commun. **10**, 5651 (2019)]
 - Ir¹⁷⁺ [Windberger *et al.*, PRL **114**, 150801 (2015)]
 - Cf¹⁵⁺ & Cf¹⁷⁺ [Porsev *et al.*, PRA **102**, 012802 (2020)]
 also: V. Schäfer (MPIK), G. Barontini (Birmingham)

Other ideas

- Ba⁴⁺, Pr¹⁰⁺: S. Brewer (Colorado State)
- XUV clocks: J. Crespo (MPIK)
- few-electron HCI: P. Micke (GSI/Jena)



better calculations: M. Safronova + new search strategies for narrow transitions



Summary

Summary

- precision spectroscopy of HCI addresses fundamental physics
- full quantum optical control over HCI achieved
- first coherent spectroscopy of HCI
- optical HCI clock with 2.2×10^{-17} syst. uncertainty
- improved absolute frequency & isotope shift by $10^8 \& 10^9$
- test of fundamental physics: verification of QED nuclear recoil & bounds on 5th forces
- new HCI clocks with high sensitivity to dark matter and α-variations
- "universal" spectroscopy scheme







Heraeus Seminar Announcement

Precision Atomic Physics Experiments to Probe for New Physics 817. WE-Heraeus-Seminar

23 Sep - 27 Sep 2024 at the Physikzentrum Bad Honnef

Confirmed speakers:

Maria Archidiacono, University of Milano Julian Berengut, University of New South Wales Liliane Biskupek, Leibniz University of Hannover **Diego Blas**, CERN **Dima Budker**, University of Mainz Eric Cornell, JILA, Boulder Sergei Eliseev, MPIK Heidelberg Alexey Elykov, KIT Melina Filzinger, PTB Braunschweig Elina Fuchs, PTB & Leibniz University of Hannover Jens Gundlach, University of Washington, Seattle Hans Hepach, University of Vienna Meike List, DLR-SI Hannover

Riccardo March, INFN Frascati **Christian Panda**, UC Berkeley Gilad Perez, Weizmann Institute Marianna Safronova, University of Delaware **D. Sheng**, University of Science and Technology of China Sven Sturm, MPIK, Heidelberg Yoshiro Takahashi, Kyoto University Michael Tobar, University of Western Australia Stefan Ulmer, University of Düsseldorf Vladan Vuletic, MIT (USA) **Peter Wolf**, OBSPM Paris (France)



WILHELM UND ELSE

HERAEUS-STIFTUNG



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Yb⁺ Clocks & frequency comb
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