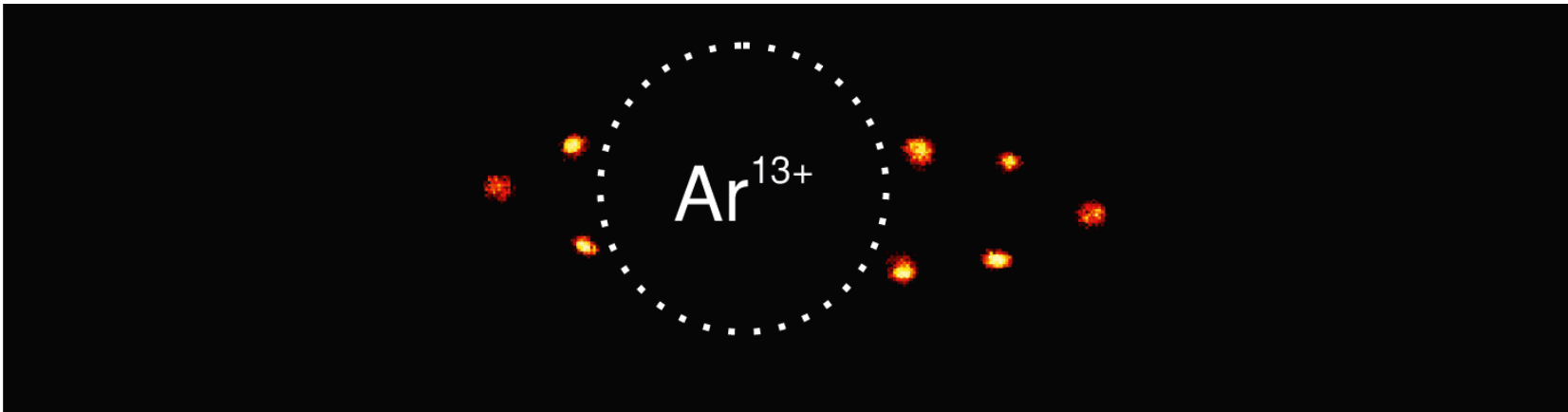


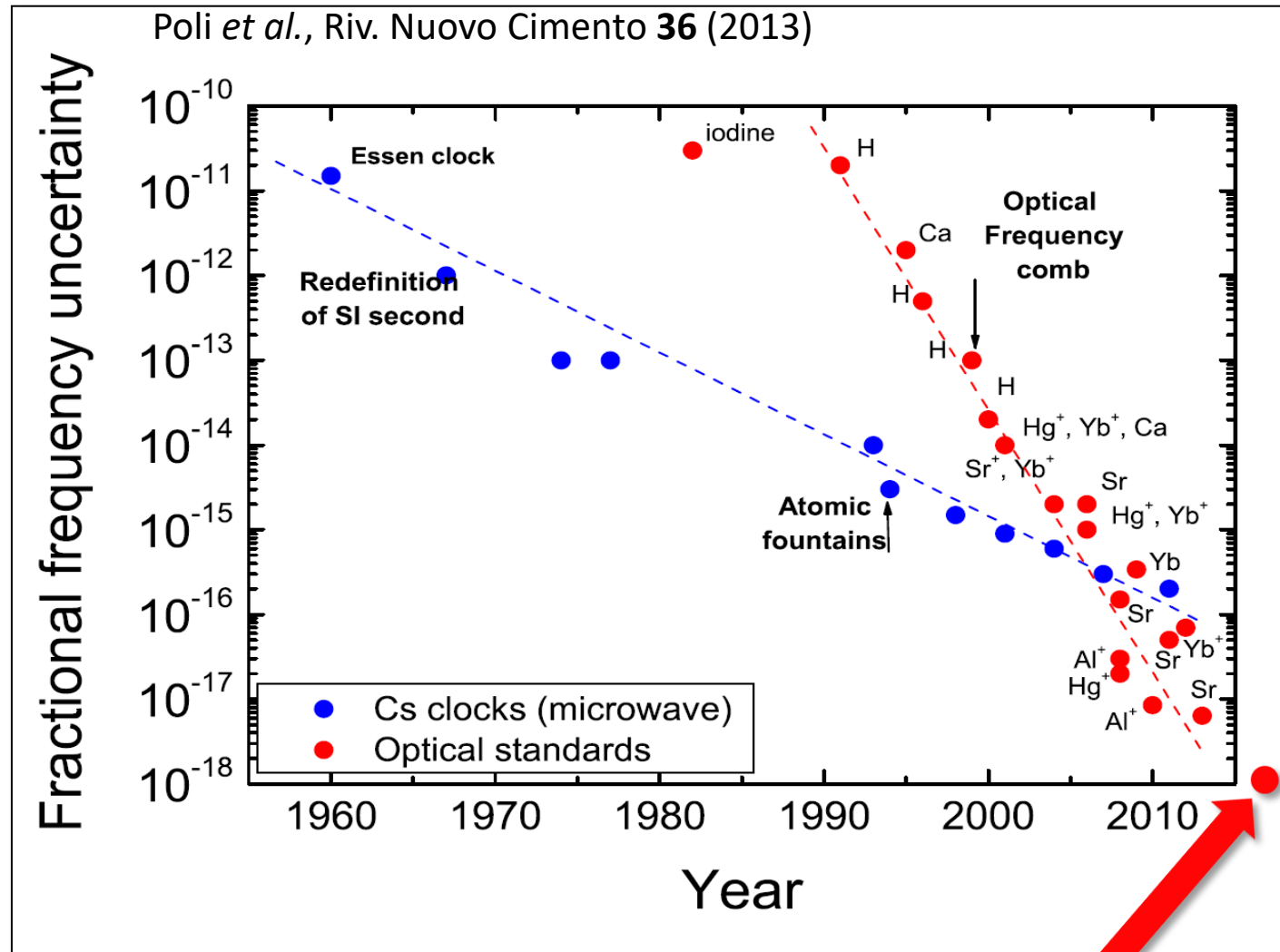
An optical clock based on a highly charged ion



Lukas J. Spieß

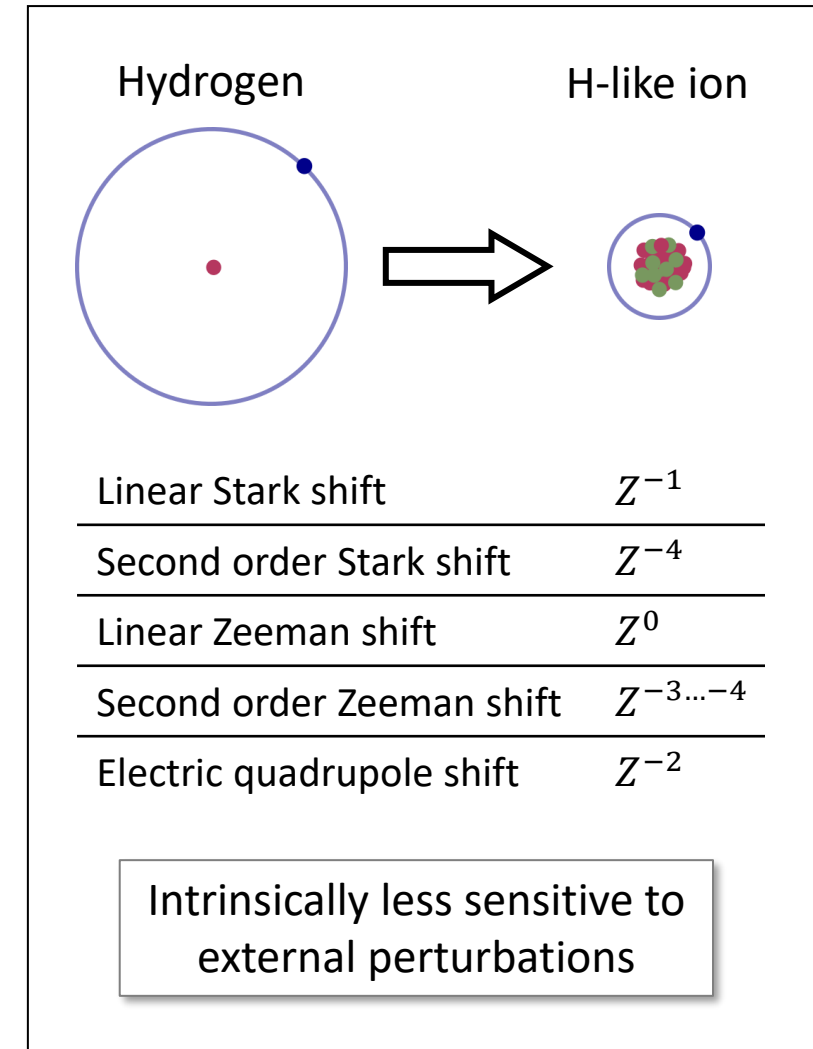
Group of Piet O. Schmidt
QUEST Institute for Experimental Quantum Metrology
PTB Braunschweig, Germany

International Symposium on Stored Ions for Precision Measurements, Shanghai, 24. Nov. 2023



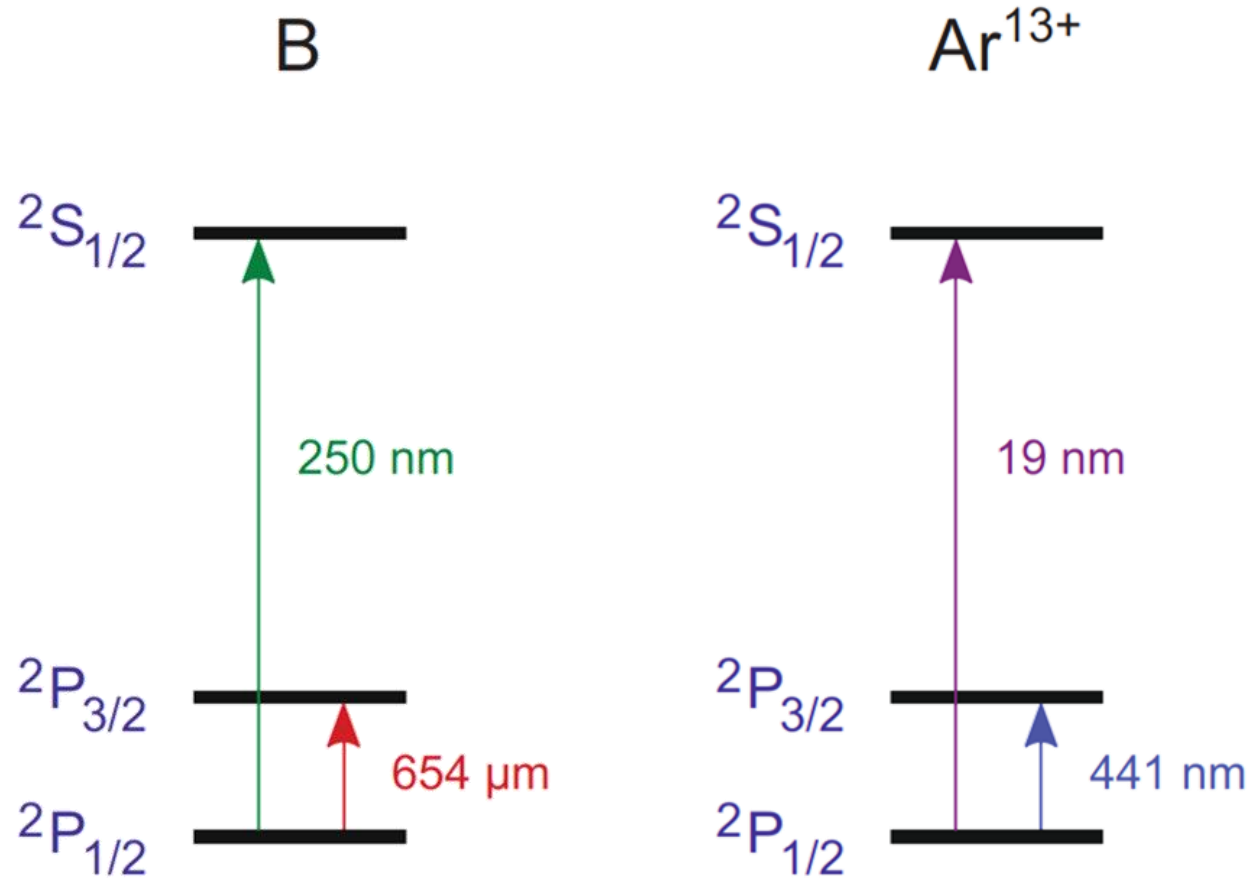
Al⁺ 9.4×10^{-19}
 Brewer *et al.*, Phys. Rev. Lett.
123, 033201 (2019)

Highly charged ions



Kozlov *et al.*, Rev. Mod. Phys **90**, 045005 (2018)

Fine and hyperfine structure transition



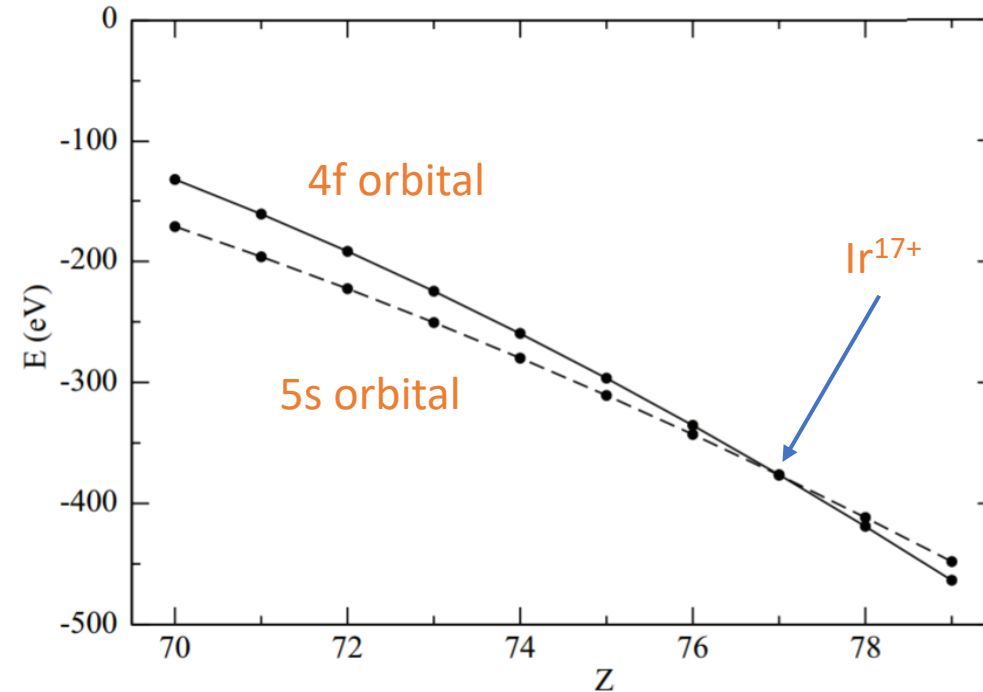
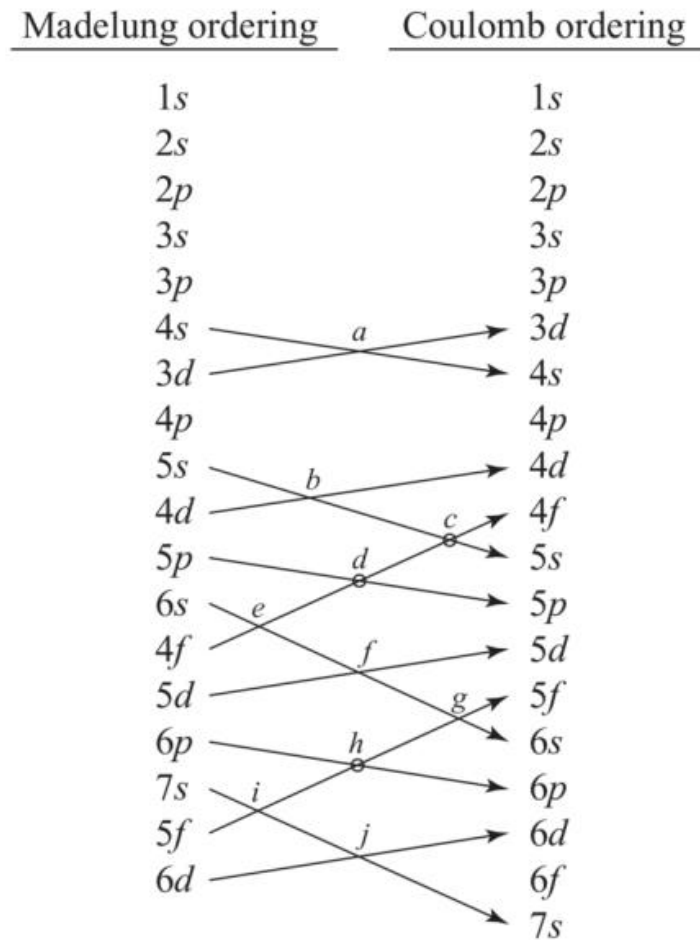
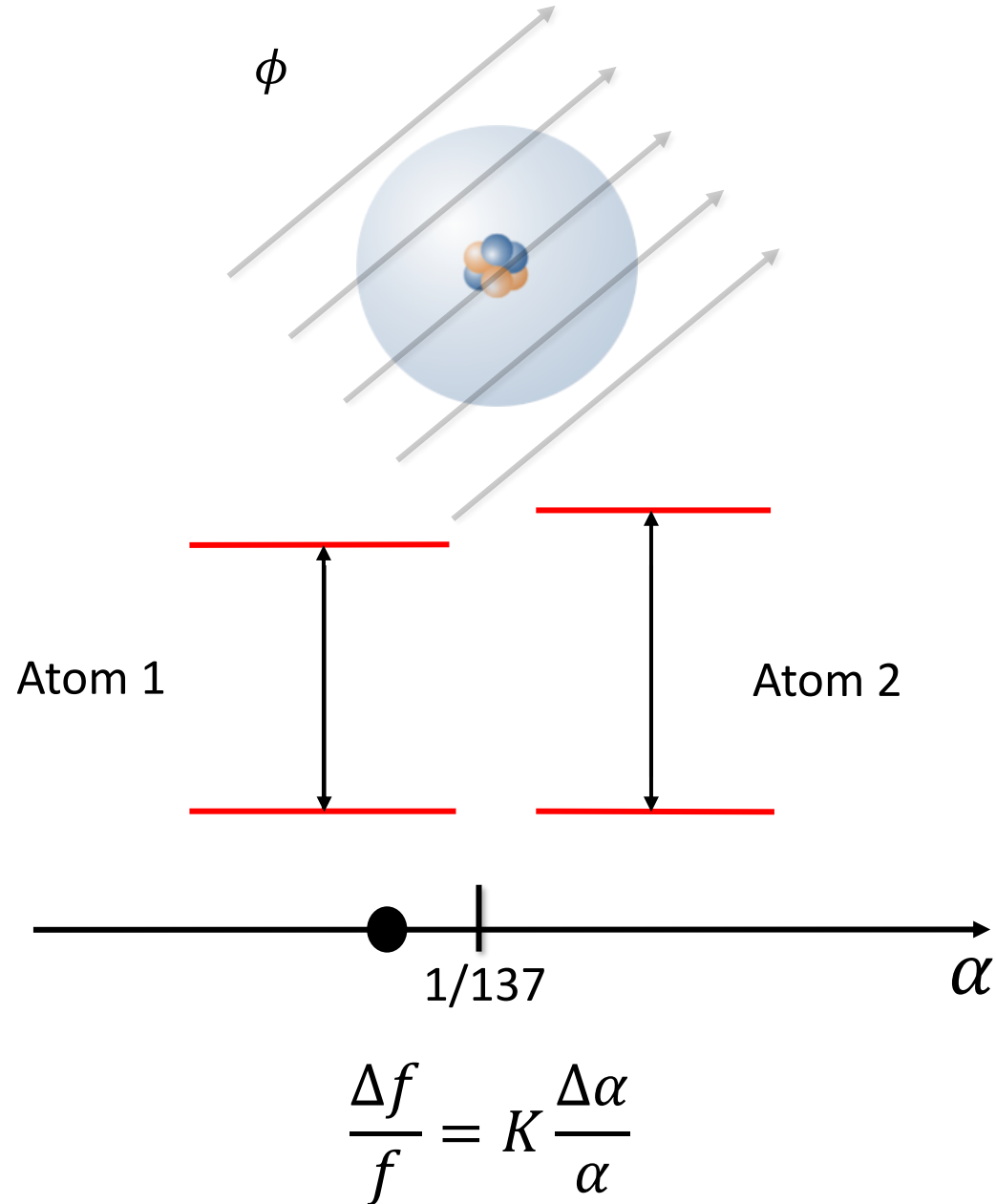
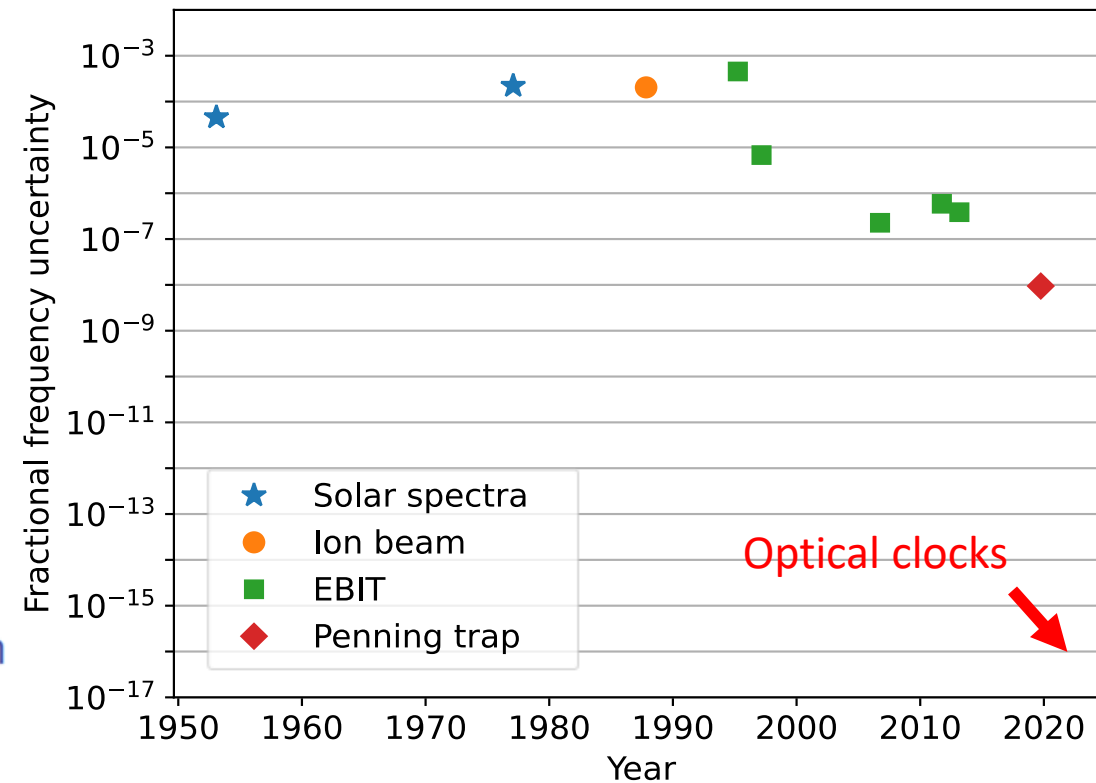
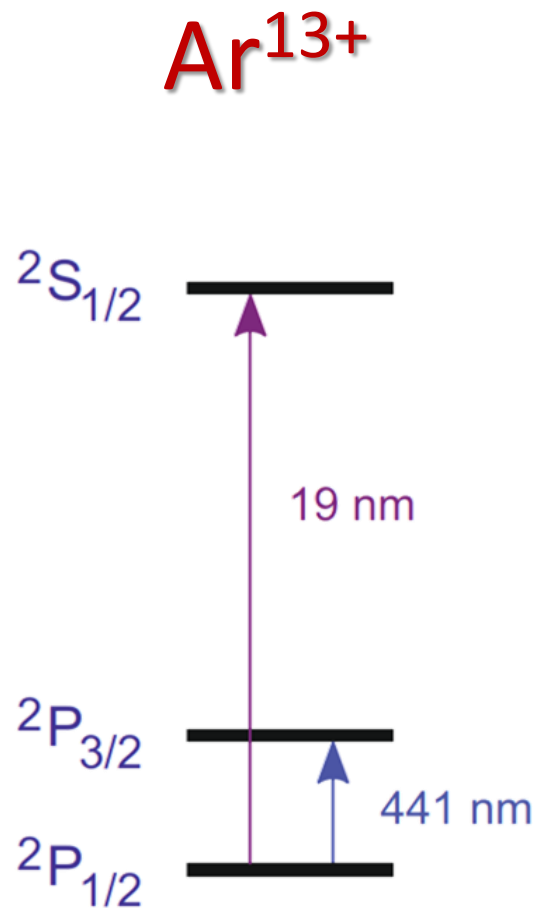
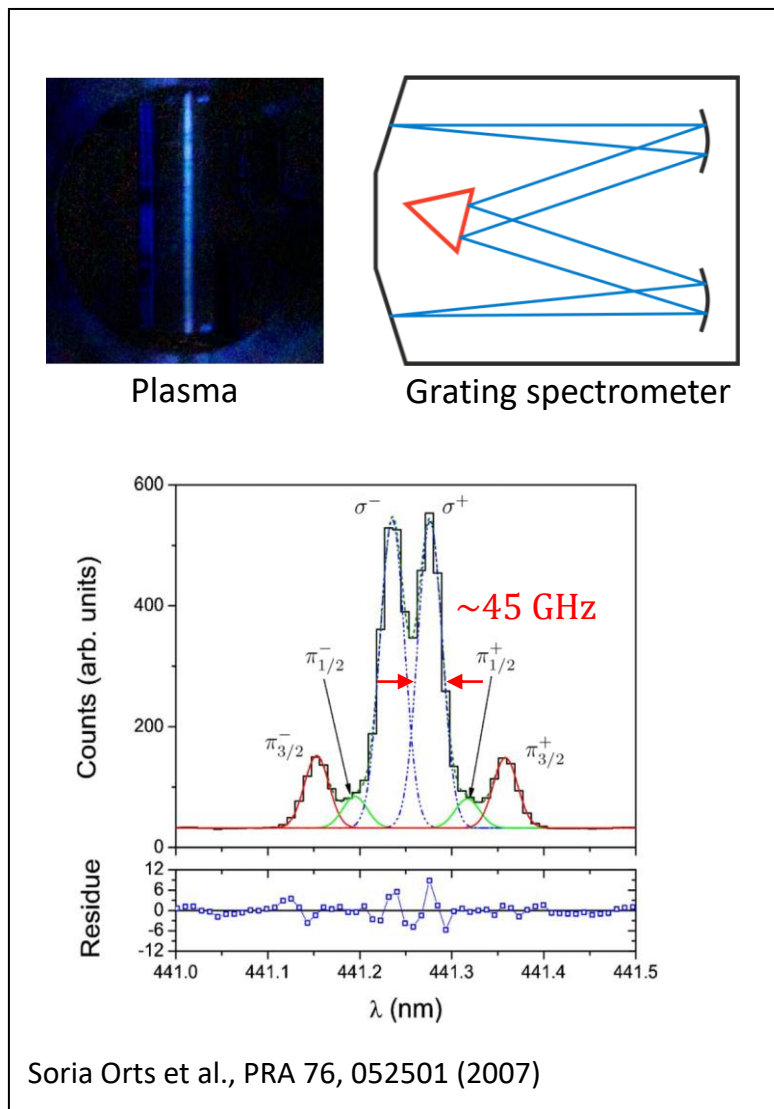


FIG. 2. Dirac-Fock energies of $4f_{7/2}$ (solid) and $5s$ (dashed) orbitals in the Sm isoelectronic sequence, $N = 62$, with closed external shells $5s^2 4f^{14}$.



System	K	λ (nm)
Sr	0.06	699
Yb ⁺ E2	0.91	436
Yb ⁺ E3	-6	467
Hg ⁺	-2.9	281.5
Ir ¹⁷⁺ T1	-22	ca. 280
Ir ¹⁷⁺ T2	145	ca. 1980
Cf ^{16+*} T1	59	ca. 775
Cf ^{17+*}	-48	ca. 535

$$\dot{\alpha}/\alpha = 1.8(2.5) \times 10^{-19}/\text{year}$$

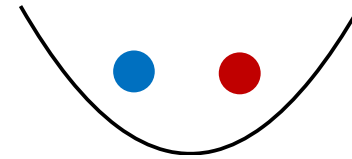


Uncertainty ~ 150 MHz

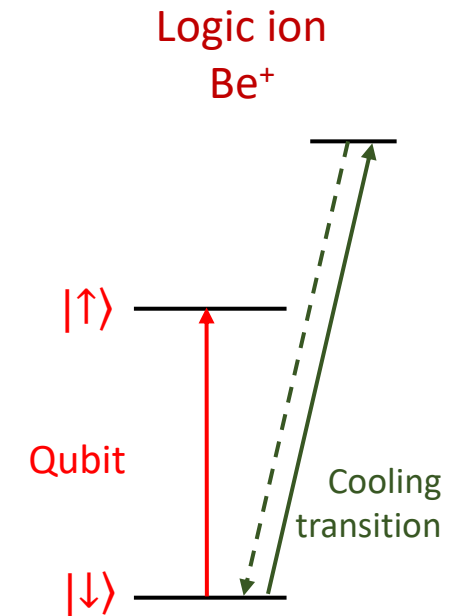
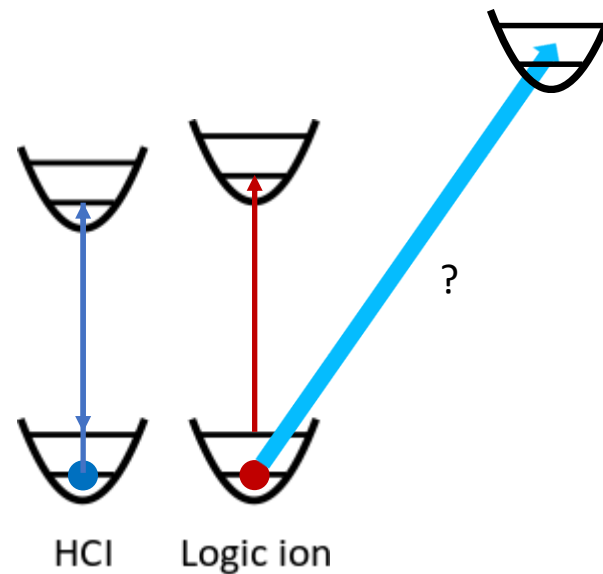
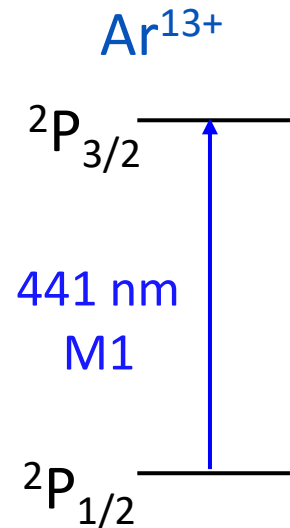
Dipole-allowed optical transitions for
laser cooling / electron shelving

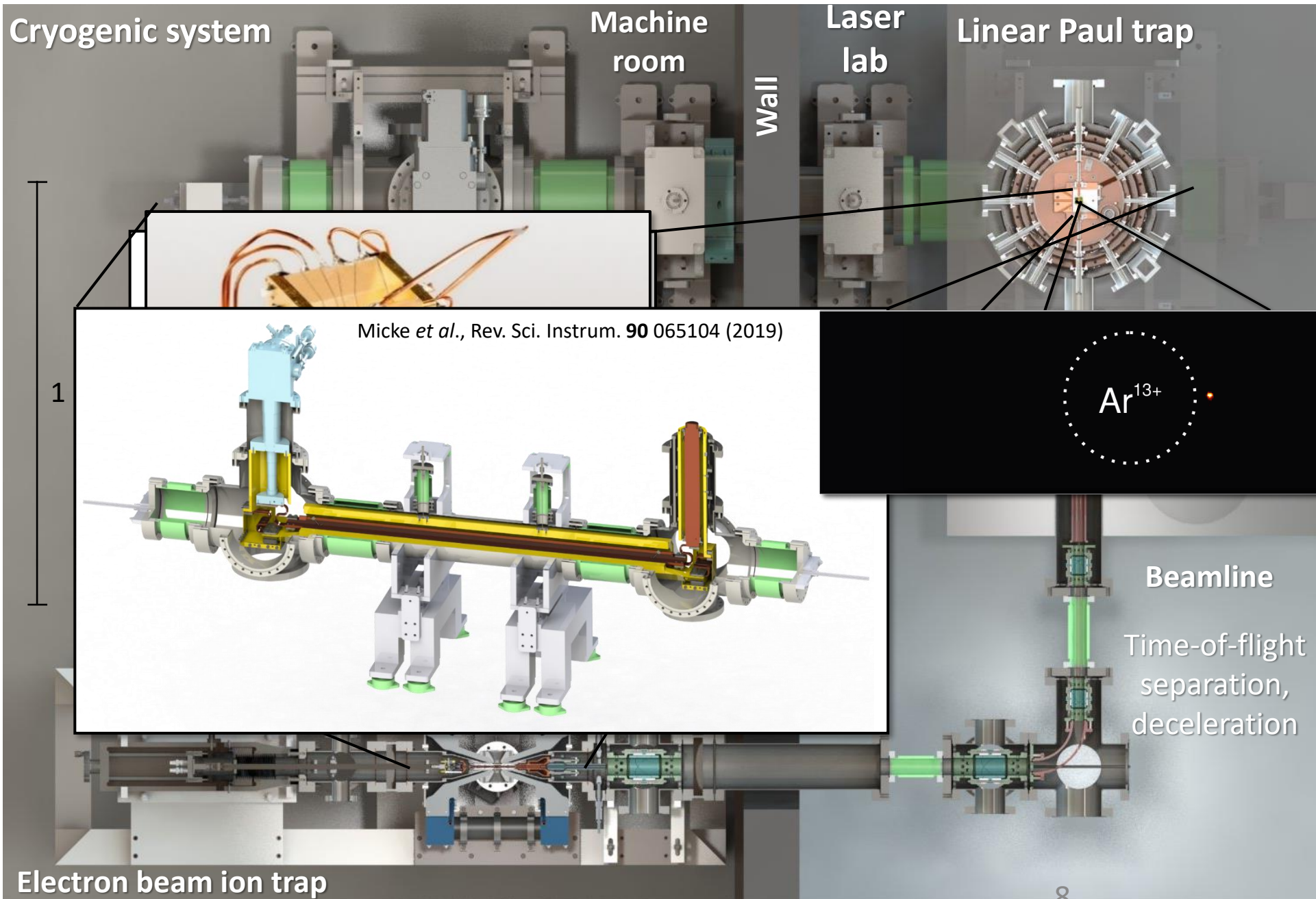
→ Sympathetic cooling and
quantum logic spectroscopy

Coupled motion:

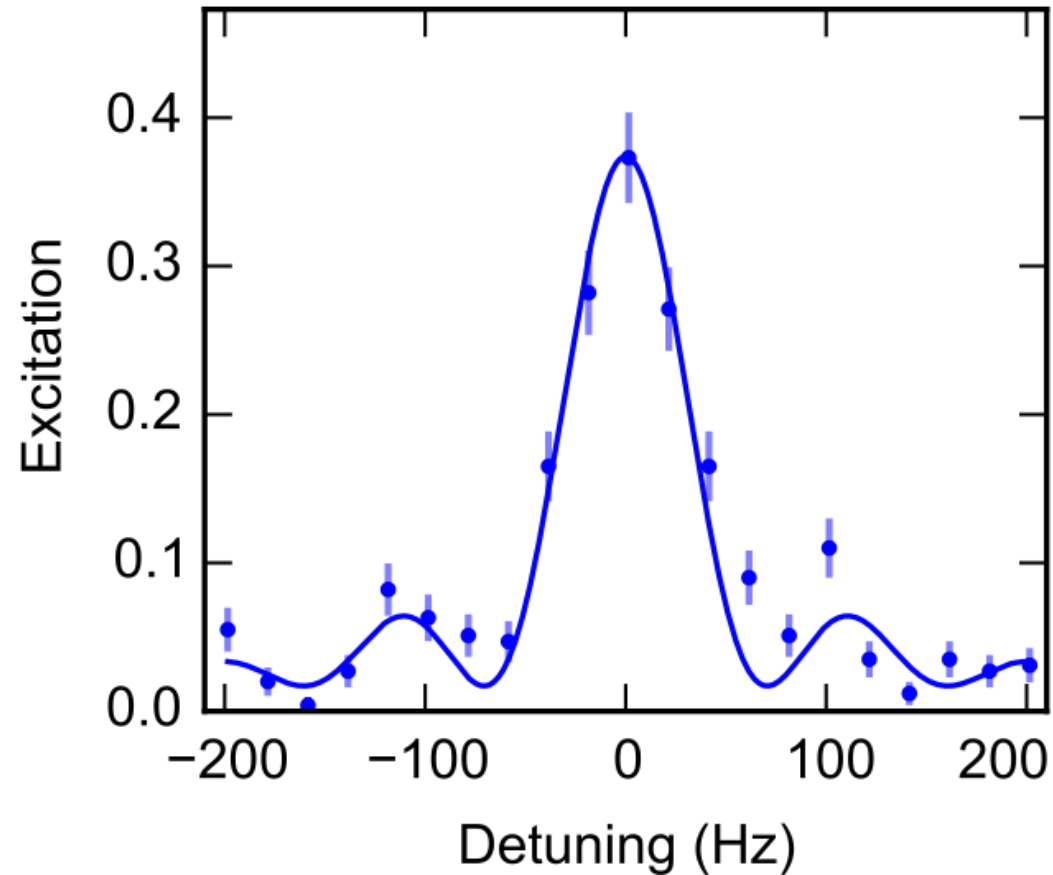


Spectroscopy ion

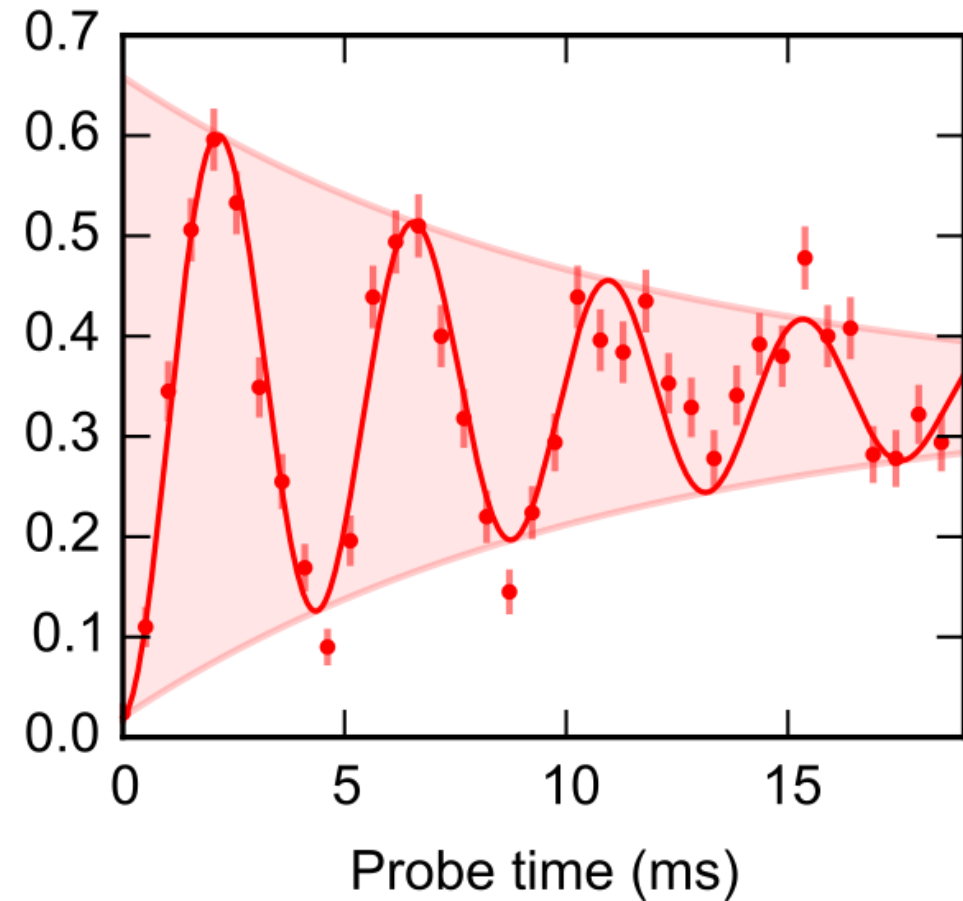




Fourier-limited linewidth: 65 Hz
(12 ms probe time) resolution: ~ 5 Hz



decoherence dominated by
excited state lifetime of 9.97(26)ms

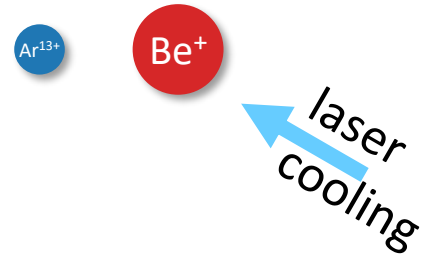


Large q/m mismatch between ions

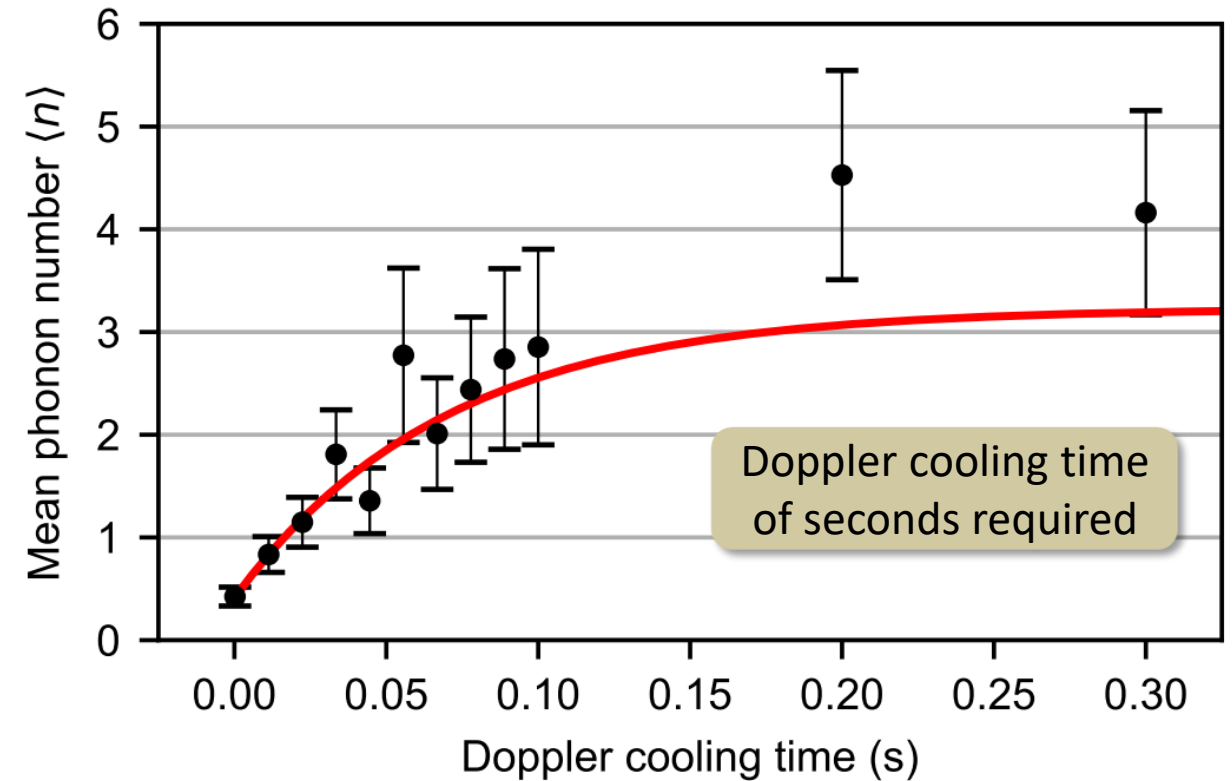
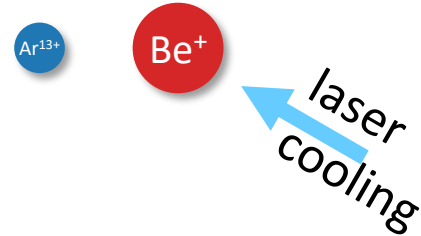
→ large difference in amplitudes

→ inefficient cooling of radial modes

- radial mode #1:
→ strong cooling

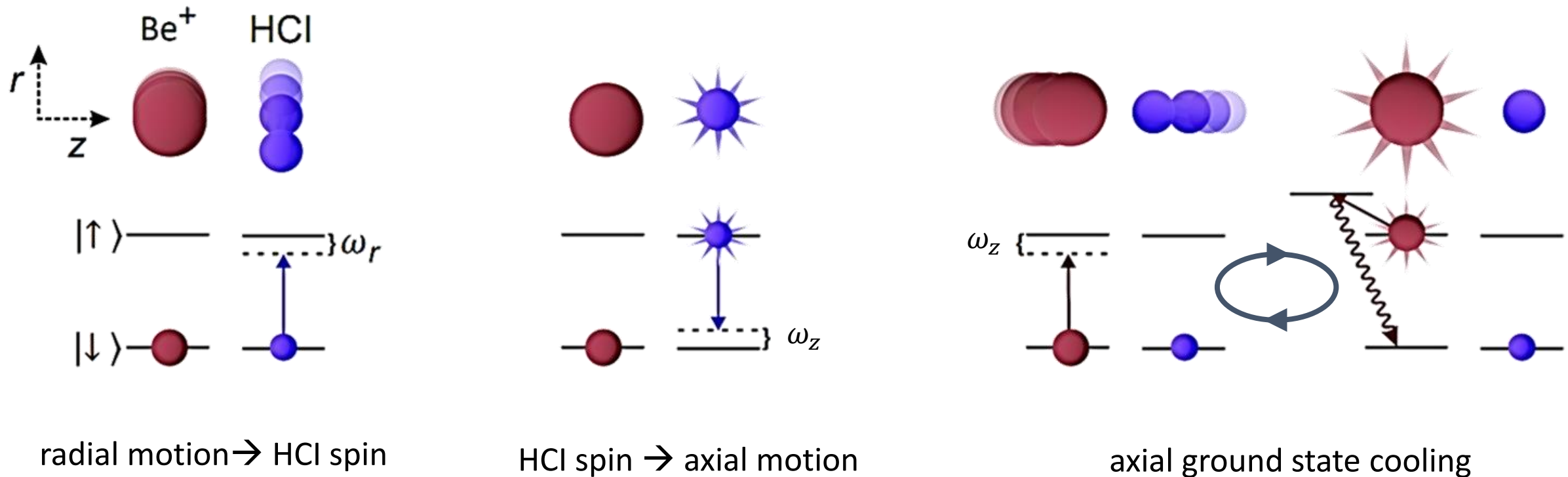


- radial mode #2:
→ weak cooling

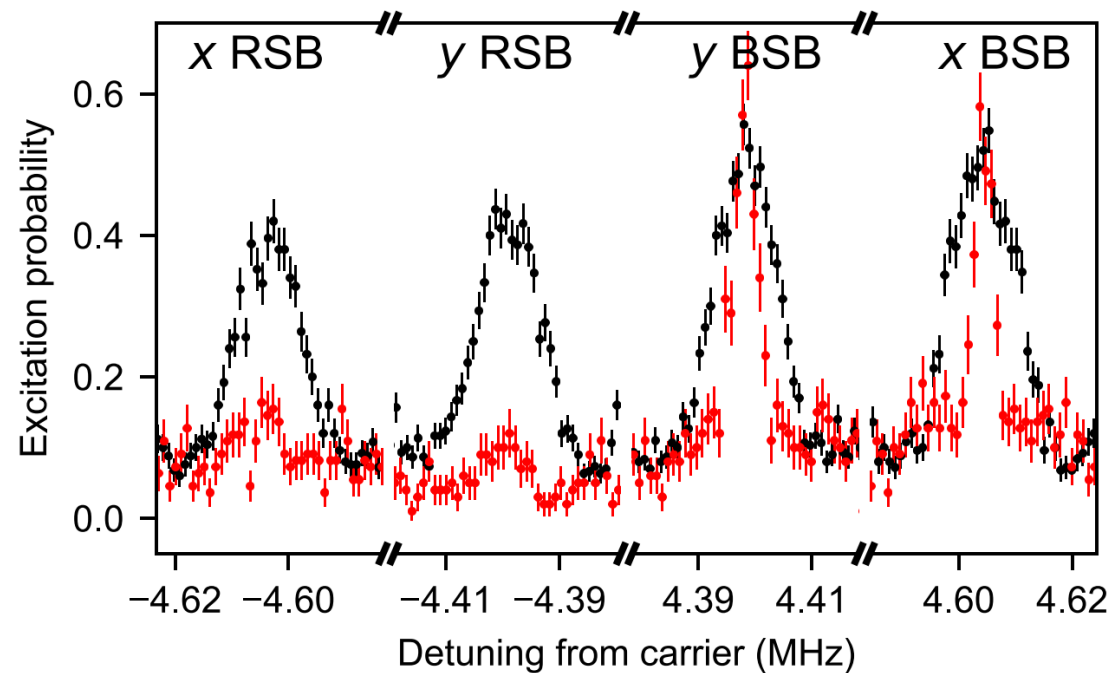


Coherently transfer radial mode phonons to efficiently-cooled axial modes

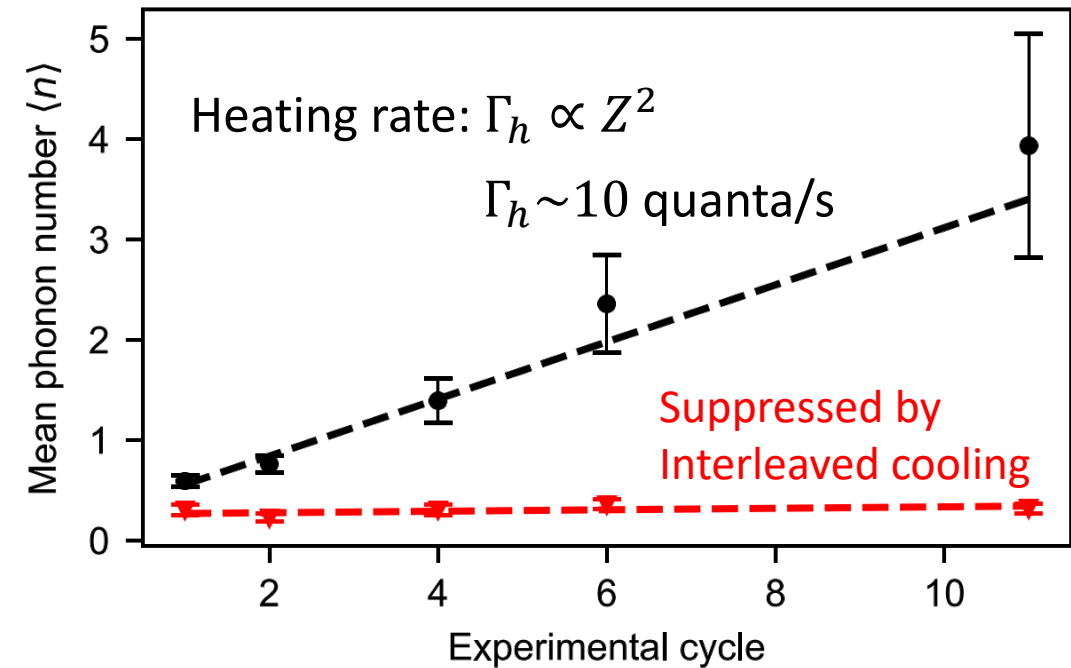
Algorithmic cooling



Universal applicable
Does not depend on q/m



Coldest HCI of all time!
 $T < 200 \mu\text{K}$ in all modes



Enables $< 10^{-18}$ levels of inaccuracy for
 HCI-based optical clocks

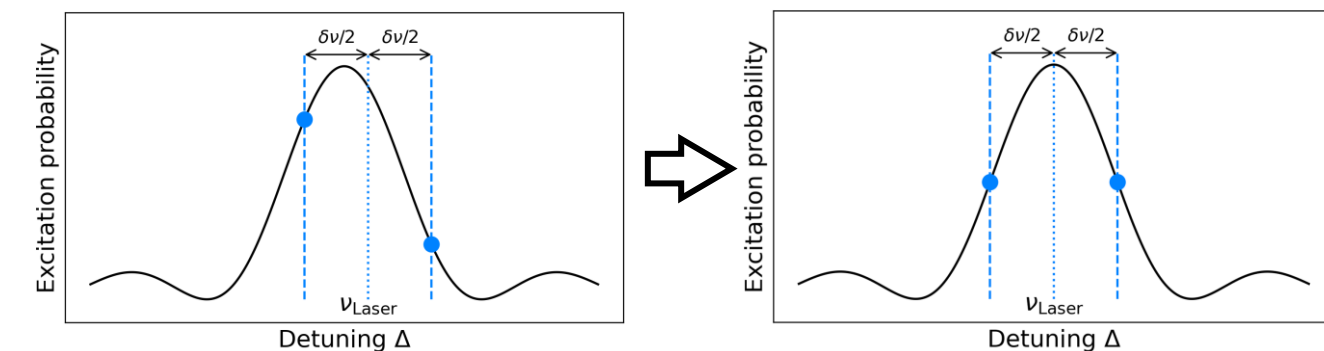
Shift source	Mitigation	Shift (10^{-18})	Uncertainty (10^{-18})
Micromotion	Real-time measurement	−443	22
Probe-laser-induced shift	Calibration at much higher powers and extrapolation	0	2
First-order Doppler	Counter-propagating beams	< 1	< 1
Linear Zeeman	Averaging over multiple Zeeman components	0	< 1
Quadratic Zeeman	Small coefficient, small field	< 1	$\ll 1$
Electric quadrupole	Small coefficient, Averaging over multiple Zeeman components	0	< 1
2 nd order Doppler	Algorithmic cooling	−1	< 1

} no fundamental limitations

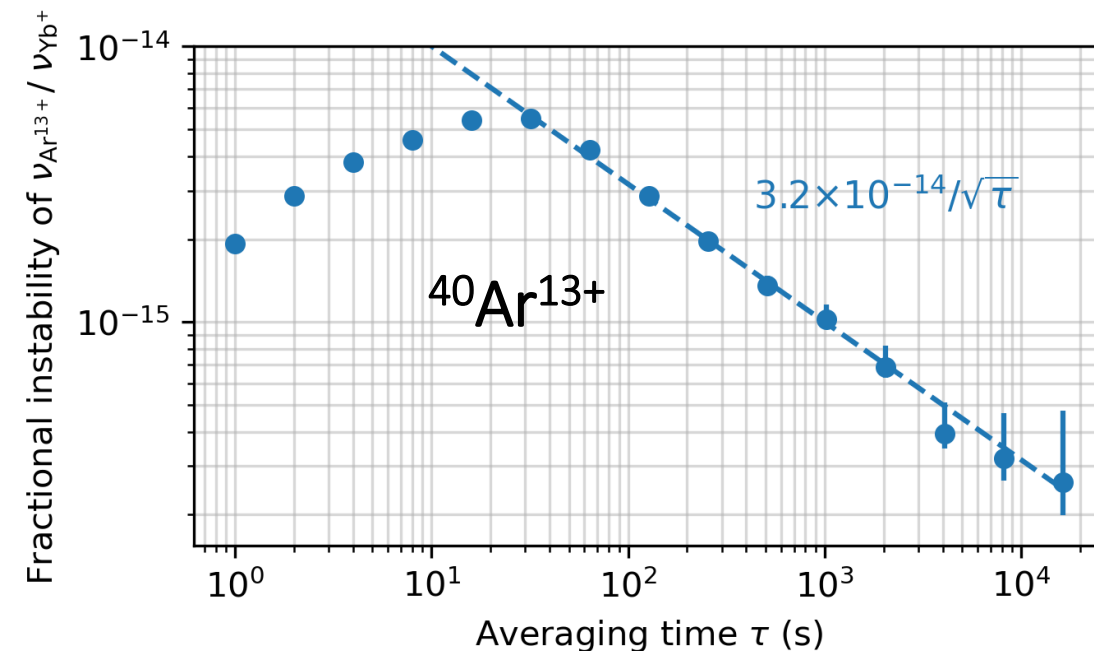
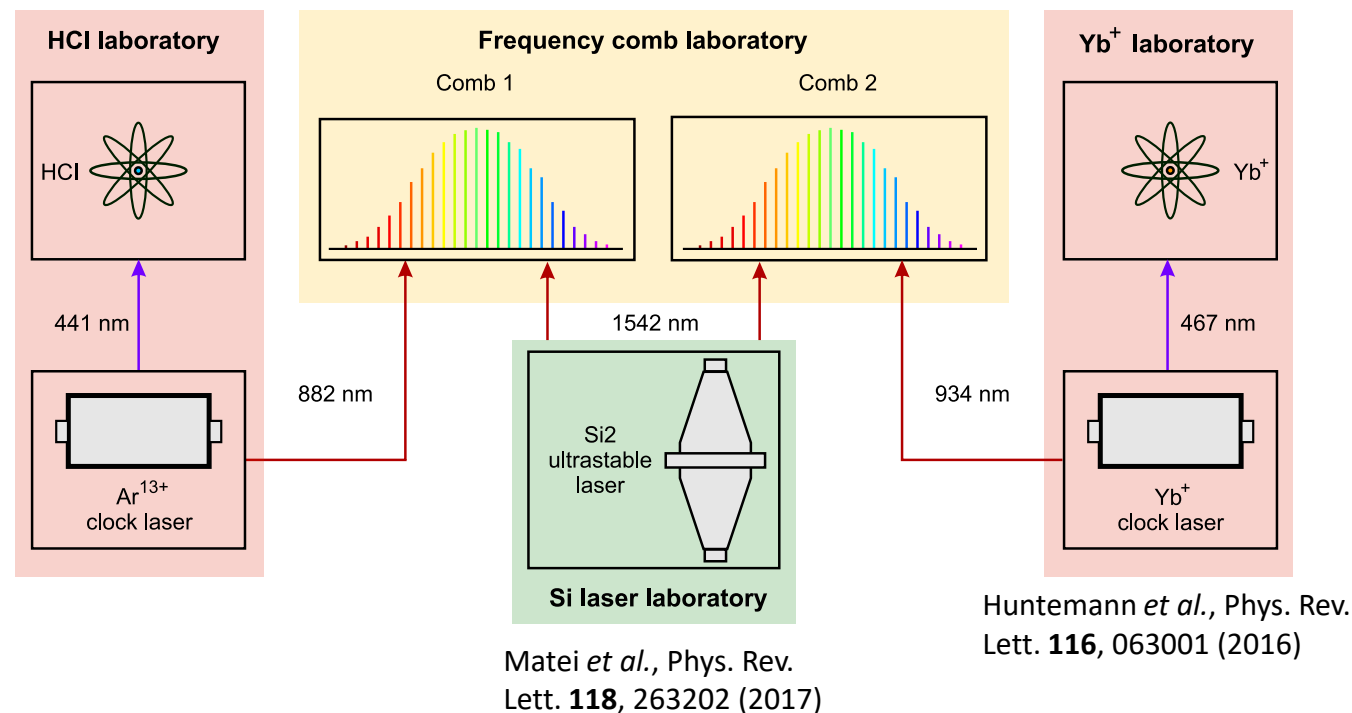
Systematic uncertainty
 $\sim 2 \times 10^{-17}$

Clock comparison

Two-point sampling



Peik *et al.*, J. Phys. B: At. Mol. Opt. Phys. **39** 145 (2006)



Statistical uncertainty
 1×10^{-16}

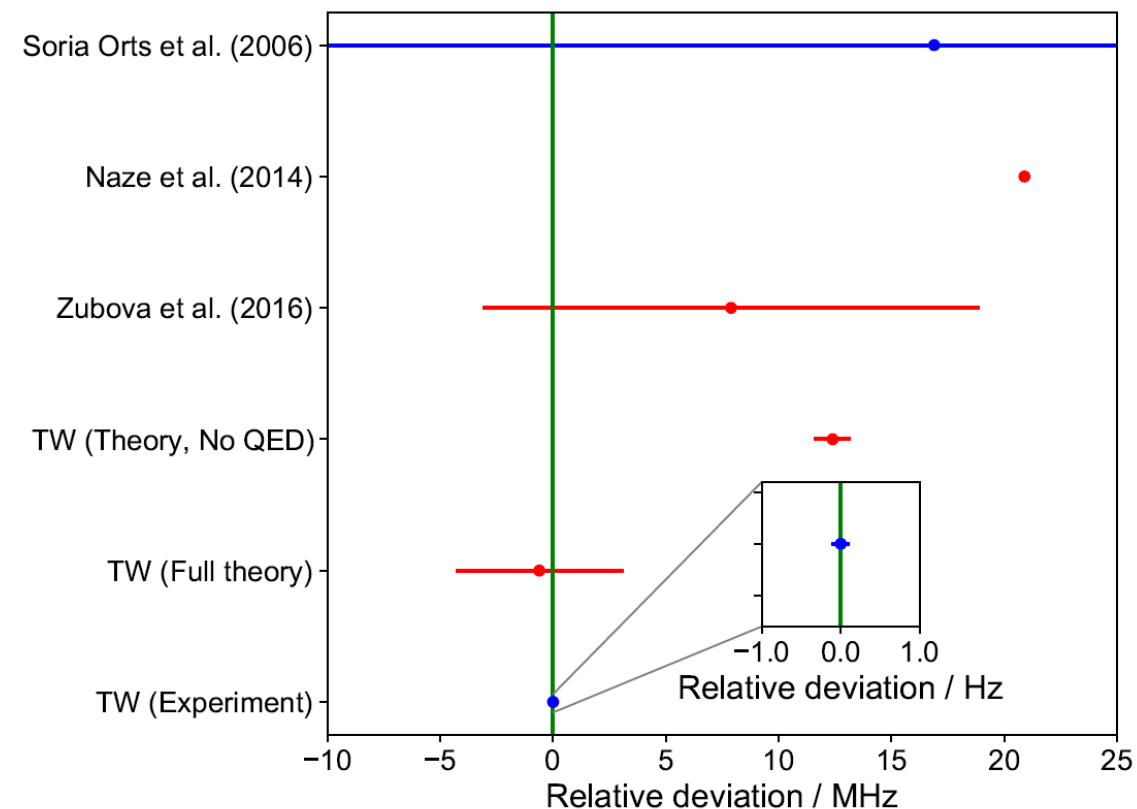
Limitation: Excited-state lifetime ≈ 10 ms

	Absolute frequency (GHz)	Relative uncertainty	Method
$^{40}\text{Ar}^{13+}$ [1]	679 216.464 (6)	8.8×10^{-9}	Penning trap
IS [2] $^{40}\text{Ar}^{13+} - ^{36}\text{Ar}^{13+}$	1.89 (9)	4.8×10^{-2}	EBIT
$^{40}\text{Ar}^{13+}$	679 216.462 397 957 42 (11)	1.5×10^{-16}	QLS
IS $^{40}\text{Ar}^{13+} - ^{36}\text{Ar}^{13+}$	1.878 110 532 52 (11)	5.7×10^{-11}	QLS

Best previously
published values

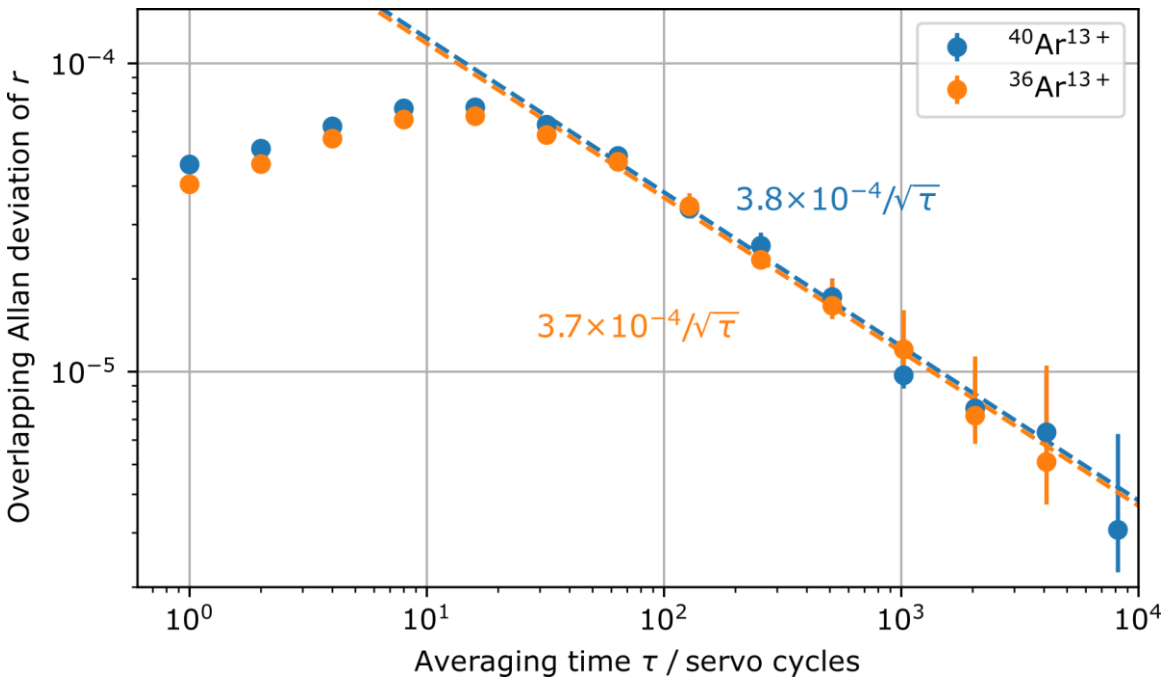
[1] Egl *et al.*, Phys. Rev. Lett. **123**, 123001 (2019)
[2] Soria Orts *et al.*, Phys. Rev. Lett. **97**, 103002 (2006)

Improvement by almost eight and nine
orders of magnitude respectively



King, Spieß *et al.*, Nature **611**, 43–47 (2022)

g-factor



$^{40}\text{Ar}^{13+}: g_{3/2} / g_{1/2} = 2.0075142(25)$

$^{36}\text{Ar}^{13+}: g_{3/2} / g_{1/2} = 2.0075154(32)$

$^{40}\text{Ar}^{13+} \text{ } ^2\text{P}_{3/2} \text{ g-factor}$	
Experiment	1.332 283 7 (17)
Theory	1.332 282 5 (14)

Agababaeu et al., X-Ray Spectrometry **49**: 143– 148 (2019)

Quadrupole moment Θ

	$\Theta \text{ (e a}_0^2\text{)}$
Ca ⁺	1.83
Sr ⁺	2.6
Yb ⁺ E2	1.95
Yb ⁺ E3	-0.0297
Hg ⁺	-0.510
Ar ¹³⁺ (Theory)	0.0235 [1]
$^{40}\text{Ar}^{13+}$ (Exp.)	-0.02 (9)
$^{36}\text{Ar}^{13+}$ (Exp.)	-0.03 (14)

$$\Delta\nu_{\text{QS}} \propto \Theta \frac{\partial E_z}{\partial z}$$

Ludlow et al., Reviews of Modern Physics **87**(2), 637–701 (2015)
Lange et al., Phys. Rev. Lett. **125**, 143201 (2020)

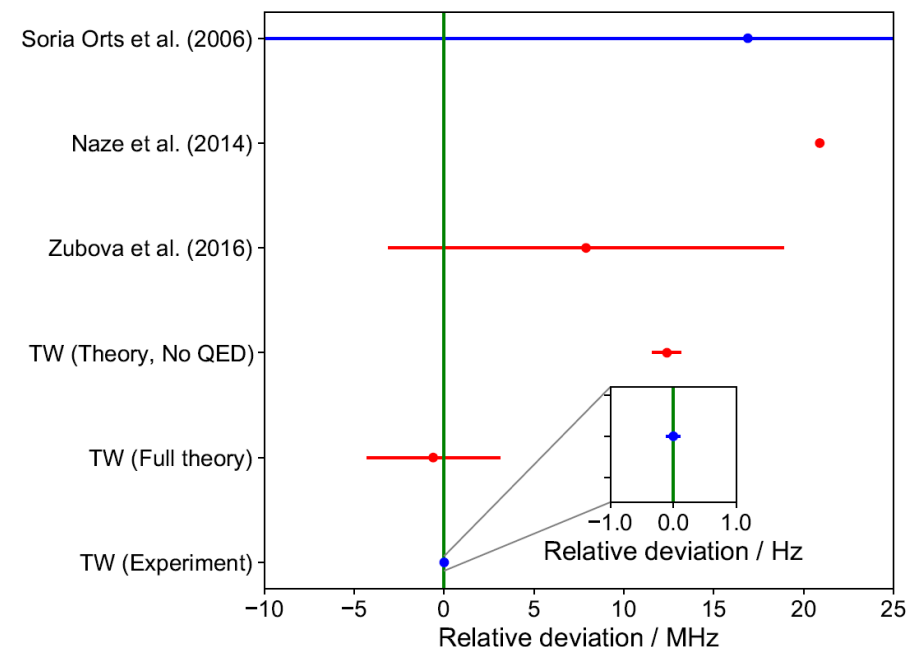
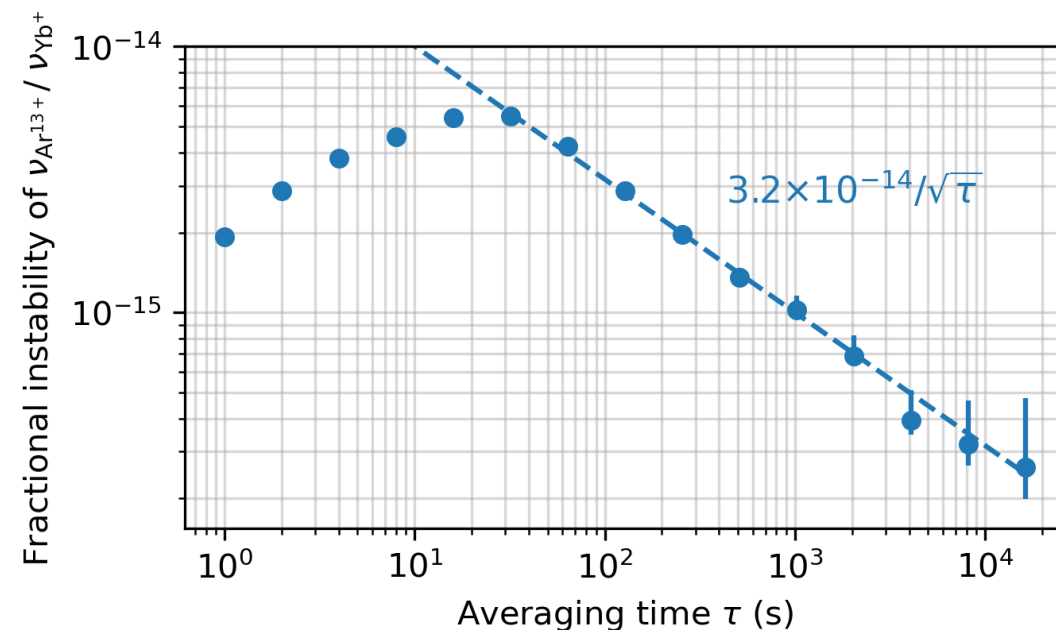
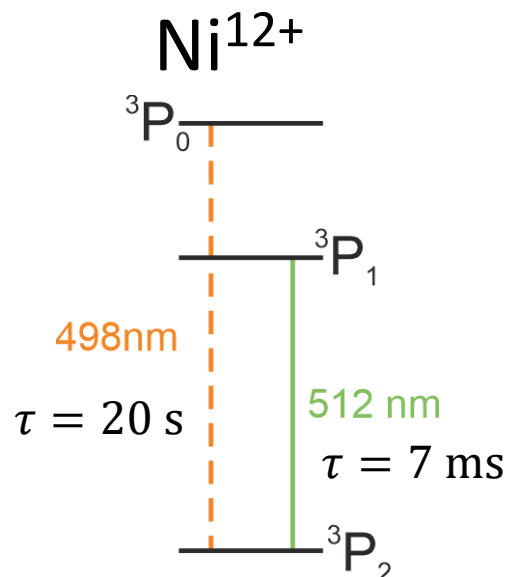
[1] Yu and Sahoo, PRA **99**, 022513 (2019)
Note: units for quadrupole moment in Tab. VI. should read $\times 10^{-7} \text{ Hz/(V/m}^2\text{)}$ (Y-m. Yu, priv. comm.)
King, Spieß et al., Nature **611**, 43–47 (2022)

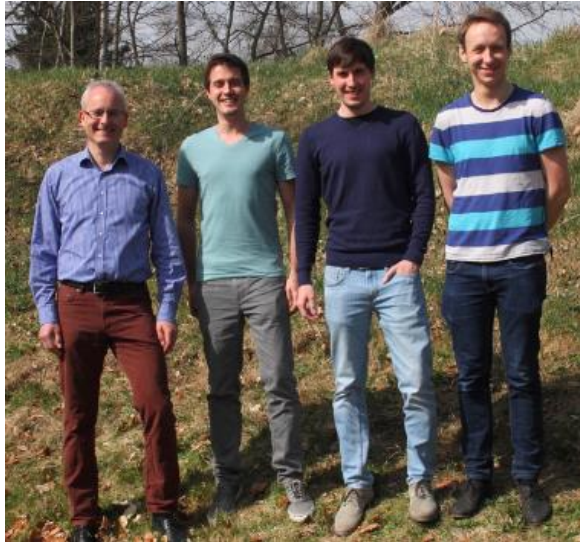
Summary

- Optical clock based on HCl with statistical uncertainty of 1×10^{-16}
- Systematic uncertainty of $\sim 2 \times 10^{-17}$
- Demonstrated optical clock comparison and improved the absolute frequency by eight orders of magnitude
- Isotope shifts for fundamental physics tests
- Atomic parameters test ab-initio calculations

Outlook

- Further reduce systematic uncertainties
- Investigating finding narrow transitions in HCl
- HCl optical clock based on Ni^{12+}





HCI Experiment

- PTB: Piet O. Schmidt, Steven A. King, Tobias Leopold, Peter Micke, LJS, Alexander Wilzewski, Malte Wehrheim, Shuying Chen
- MPIK: José R. Crespo López-Urrutia, Lisa Schmöger, Maria Schwarz

Yb⁺ Experiment

- Richard Lange (PTB)
- Melina Filzinger (PTB)
- Nils Huntemann (PTB)

Frequency comb

- Erik Benkler (PTB)

Theory

- Robert Müller (PTB)
- Vladimir Yerokhin (MPIK)
- Andrey Surzhykov (PTB & TU Braunschweig)

