A $^{176}\mathrm{Lu}^{+}$ frequency reference

Murray Barrett
9SFM 2023

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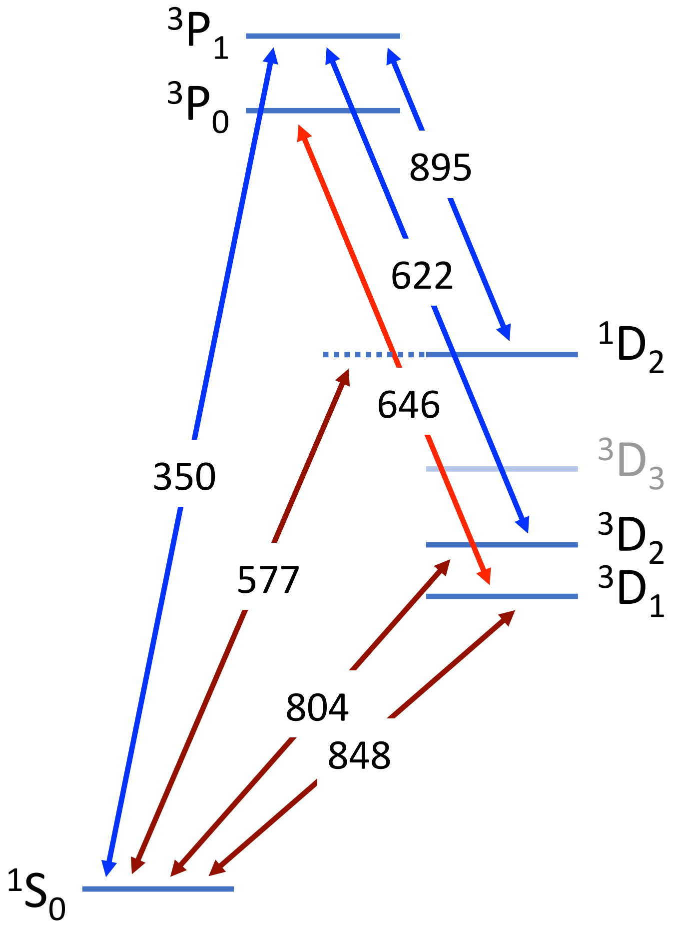
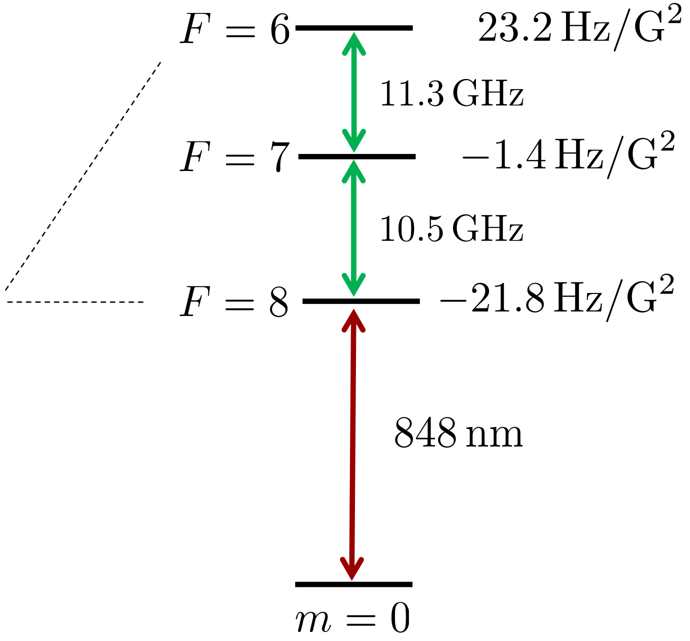
Lutetium ^{176}Lu^{+} : $\quad {}^1S_0 \leftrightarrow {}^3D_1$



- BBR Shift at 300K

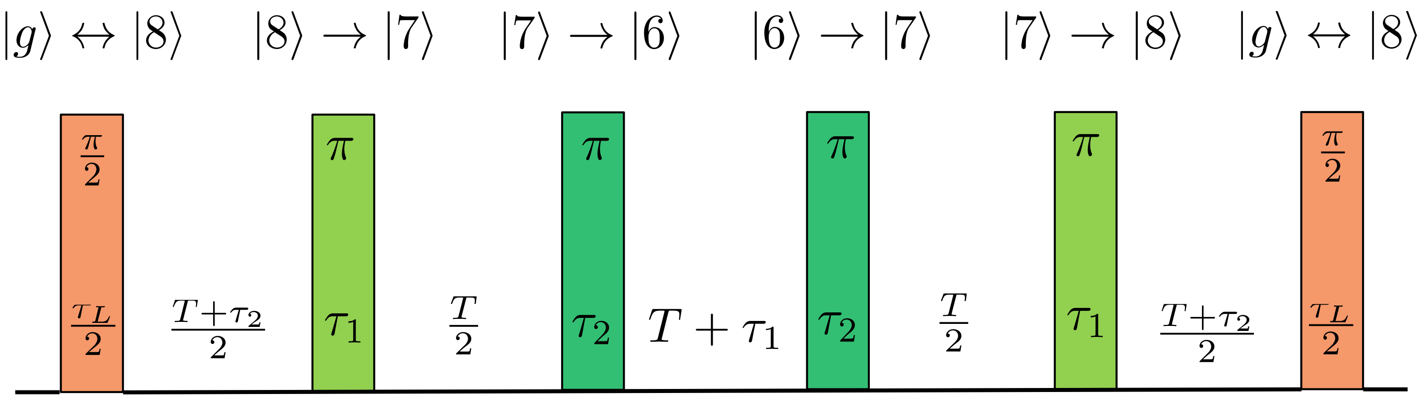
$$\frac{\Delta f}{f_c} \approx -1.36 \times 10^{-18}$$

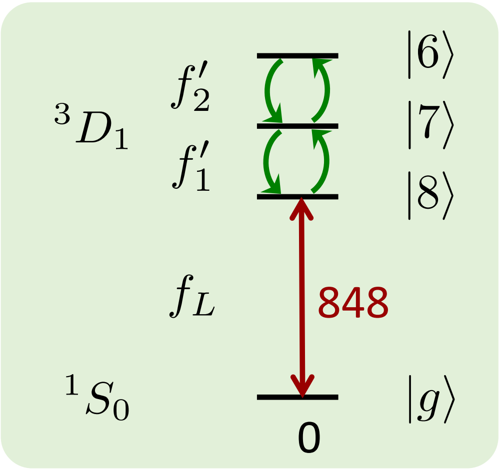


- All wavelengths diode laser-based
- Long lived D states
  - ^{1}D$_2$ (≈0.18s : E2)
  - ^{3}D$_2$ (≈18s : E2)
  - ^{3}D$_1$ (≈ 170 h : M1)
- Large hyperfine and fine-structure splittings
  - Magnetic field insensitivity
- Narrow cooling line
  - $\Gamma = 2\pi \times 2.45\,\mathrm{MHz}, \quad (T_D = 60\,\mu\mathrm{K})$

$$\frac{3k_B T_D}{2mc^2} \approx 5 \times 10^{-20}$$

Hyperfine-averaging



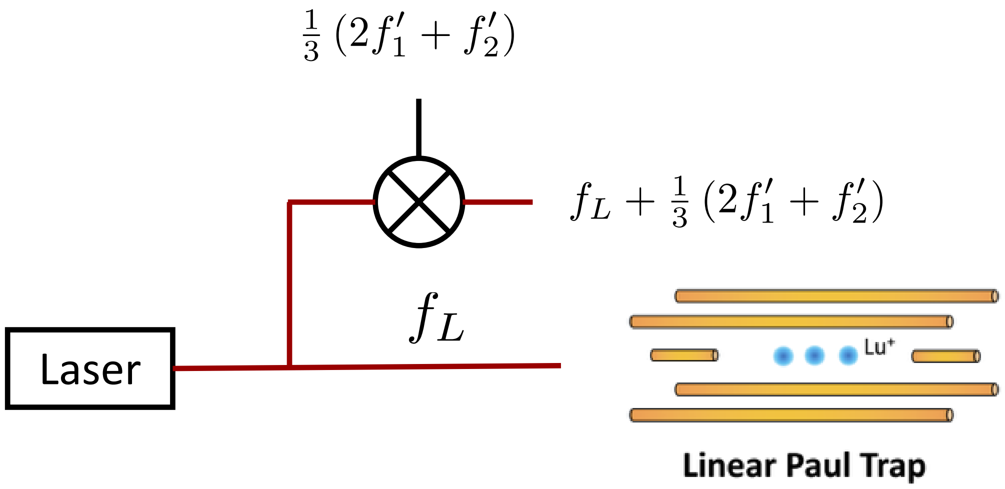


Hyperfine-averaged Ramsey spectroscopy (HARS)

Interrogation time: $T_R = 3(T + \tau_1 + \tau_2)$

Clock frequency:

$$\begin{aligned} f_c &= \tfrac{1}{3}(f_6 + f_7 + f_8) \\ &= f_8 + \tfrac{1}{3}(2f_1 + f_2) \\ &= f_L + \tfrac{1}{3}(2f_1' + f_2') \end{aligned}$$



R. Kaewuam, et al, PRL, **124**, 083202, 2020

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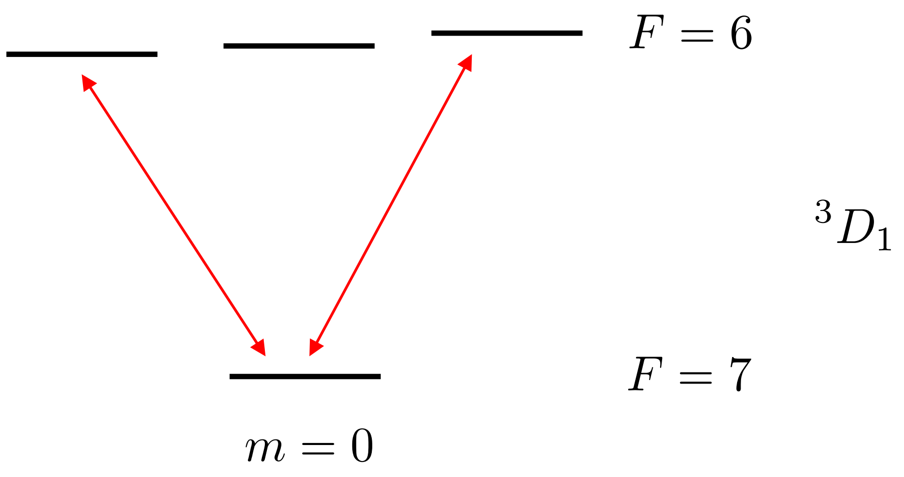
Systematics – Quadratic Zeeman

Electromagnetic

$\langle B^2 \rangle$ Dominated by the applied dc field.

$$\alpha_z = -4.89264(88)\,\mathrm{Hz/mT^2}$$

$$(\approx -1.4 \times 10^{-16} \text{ at } 0.1\,\mathrm{mT})$$



- Measured to high accuracy
    - Science Advances, Vol 9, No. 18, (2023)
- Monitored via microwave spectroscopy

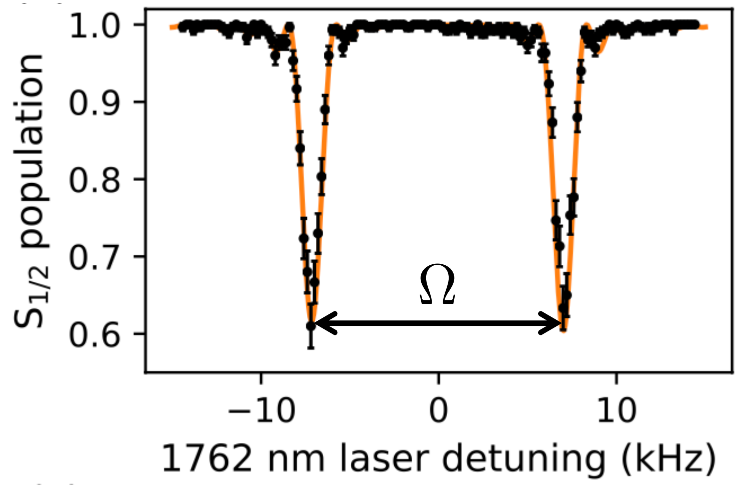
Systematics – Quadratic Zeeman

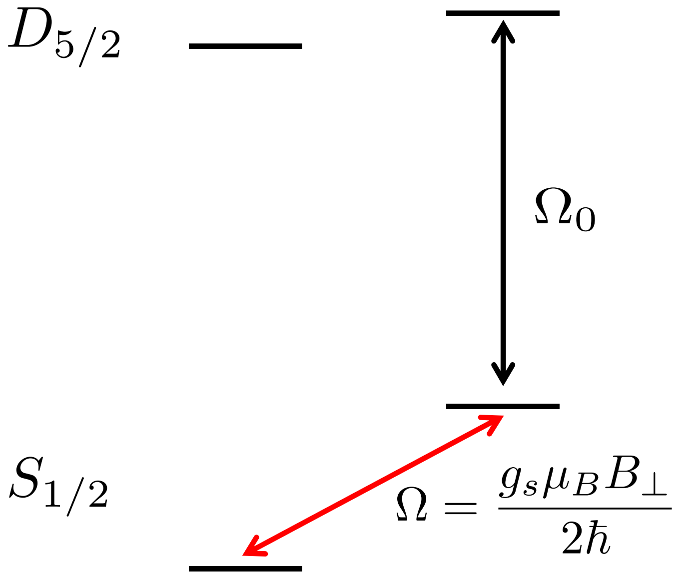
Electromagnetic

$\langle B^2\rangle$ Dominated by the applied dc field.

$\langle \widetilde{B}_\perp^2\rangle$ Coupling to trap-induced rf currents.

$$5.7\times 10^{-19}/\mu\mathrm{T}^2$$





K. J. Arnold. et al., PRL **124**, 193001 (2020).
H. C. J. Gan et al, PRA **98**, 032514 (2018)

Systematics – Quadrupole

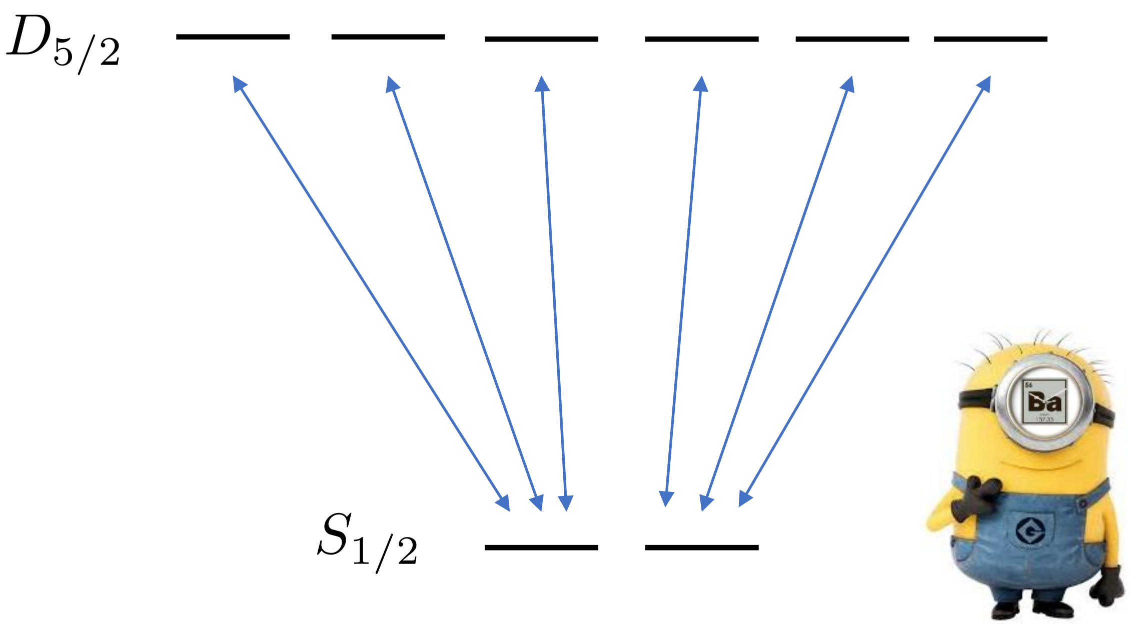
Electromagnetic

$\langle B^2 \rangle$ Dominated by the applied dc field.

$\langle \widetilde{B}_\perp^2 \rangle$ Coupling to trap-induced rf currents.

∇E Hyperfine mediated quadrupole moment

$$\langle \delta\Theta(J,F,m) \rangle = -2.54 \times 10^{-4} \mathrm{ea}_0^2$$

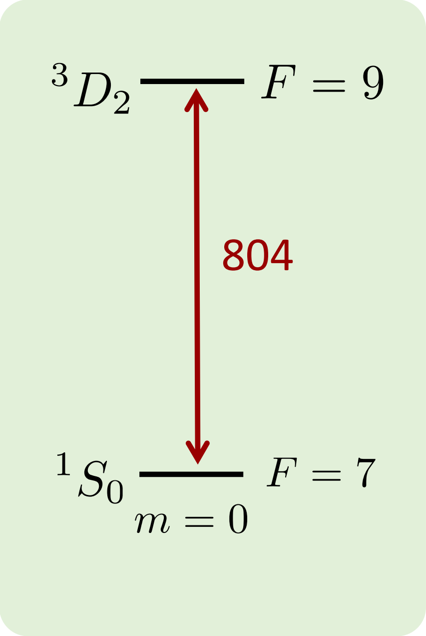


- Shift assessed via Ba⁺ clock
  - PRL, **124**, 19, 193001, 2020
- Residual moment from g-factors and theory
  - PRA, **102**, 5, 052834, 2020

Systematics

Electromagnetic

$\langle B^2 \rangle$	Dominated by the applied dc field.
$\langle \widetilde{B}_\perp^2 \rangle$	Coupling to trap-induced rf currents.
∇E	Hyperfine mediated quadrupole moment
$\langle E^2 \rangle$	Micromotion, ac-Stark shift, BBR shift



- Micromotion via sideband spectroscopy
- Probe stark shift suppressed via hyper-Ramsey

Systematics

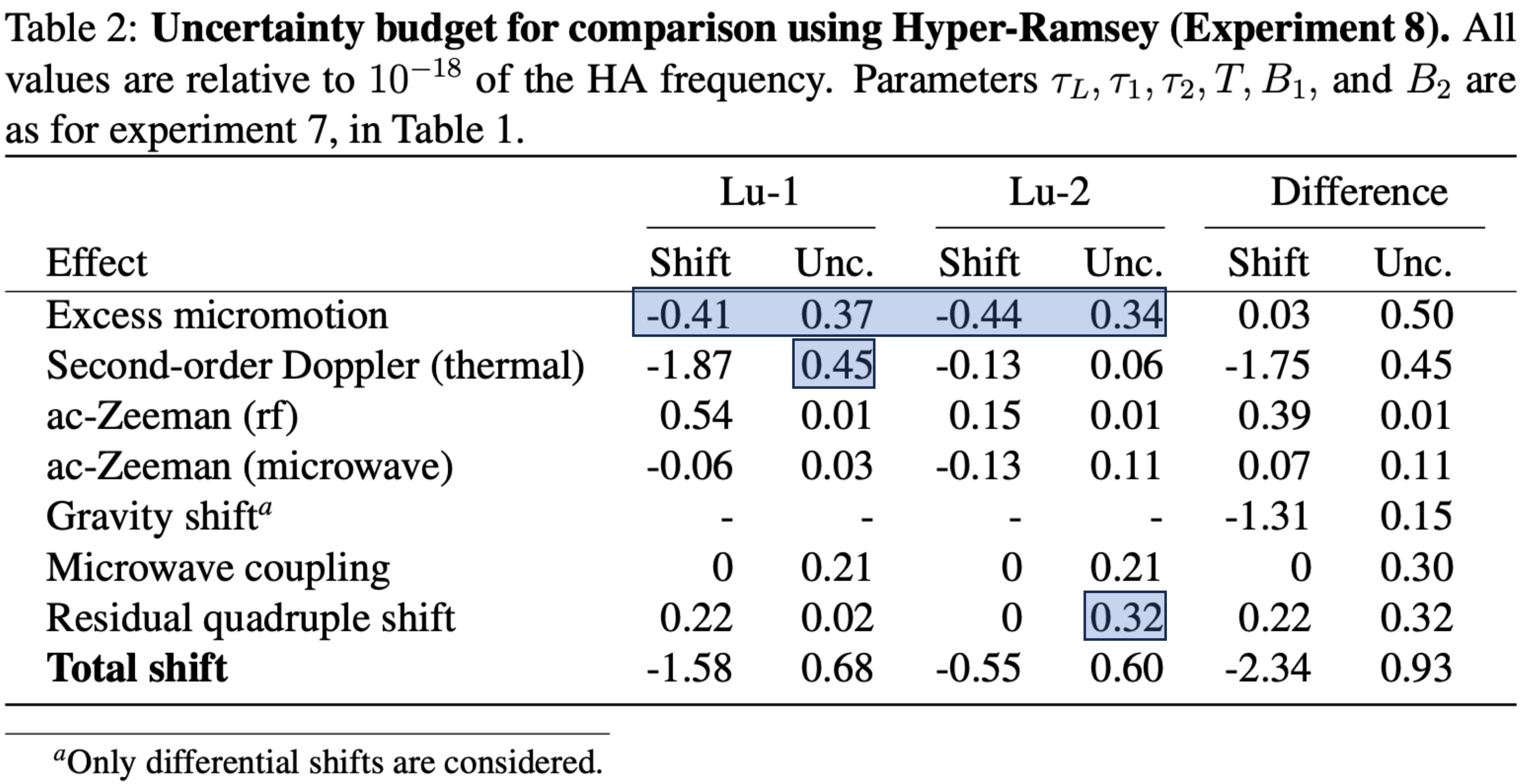


Table 2: **Uncertainty budget for comparison using Hyper-Ramsey (Experiment 8).** All values are relative to 10^{-18} of the HA frequency. Parameters $\tau_L, \tau_1, \tau_2, T, B_1$, and B_2 are as for experiment 7, in Table 1.

	Lu-1		Lu-2		Difference	
Effect	Shift	Unc.	Shift	Unc.	Shift	Unc.
Excess micromotion	-0.41	0.37	-0.44	0.34	0.03	0.50
Second-order Doppler (thermal)	-1.87	0.45	-0.13	0.06	-1.75	0.45
ac-Zeeman (rf)	0.54	0.01	0.15	0.01	0.39	0.01
ac-Zeeman (microwave)	-0.06	0.03	-0.13	0.11	0.07	0.11
Gravity shift[a]	-	-	-	-	-1.31	0.15
Microwave coupling	0	0.21	0	0.21	0	0.30
Residual quadruple shift	0.22	0.02	0	0.32	0.22	0.32
Total shift	-1.58	0.68	-0.55	0.60	-2.34	0.93

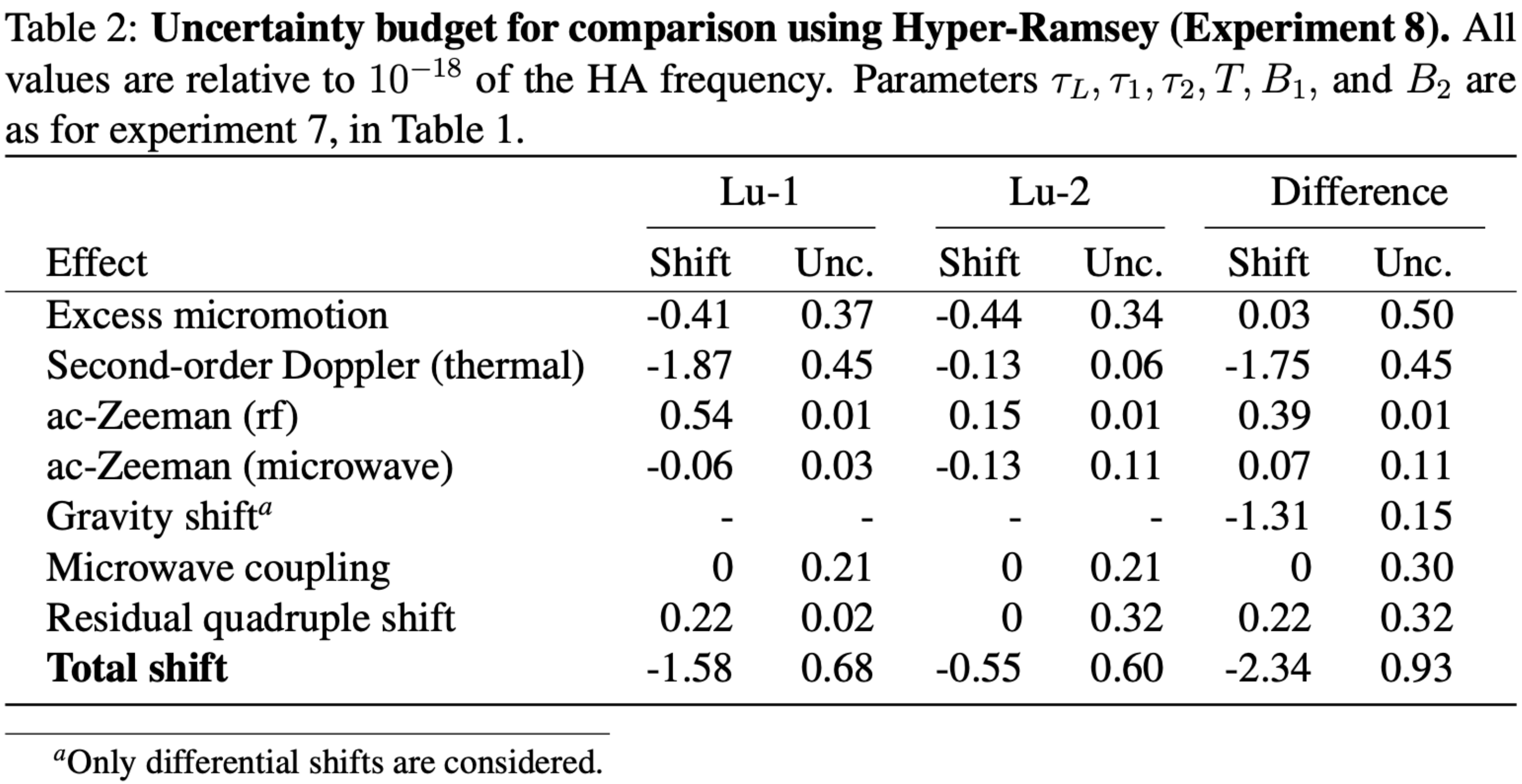
---

[a] Only differential shifts are considered.

$$(-2.0 \pm (3.7)_{\mathrm{stat}} \pm (0.9)_{\mathrm{sys}}) \times 10^{-18}$$

BBR shift: $T = 30 \pm 5\,^\circ\mathrm{C} \implies \dfrac{\Delta f}{f} = -1.44(16) \times 10^{-18}$

So how accurate is a Lu$^+$ clock?



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Segal’s Adage

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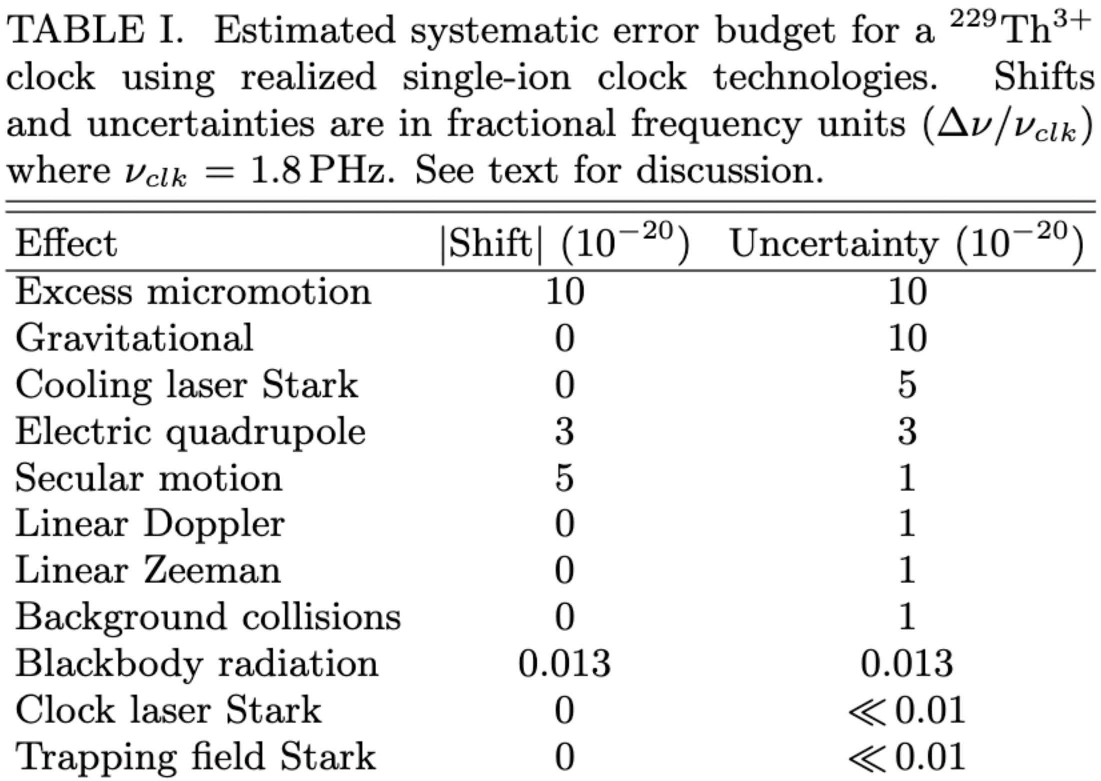
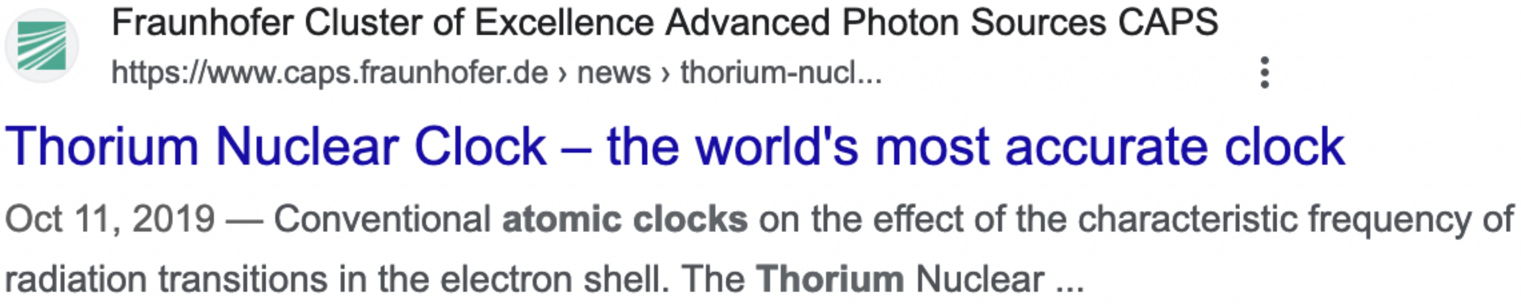


TABLE I. Estimated systematic error budget for a $^{229}\mathrm{Th}^{3+}$ clock using realized single-ion clock technologies. Shifts and uncertainties are in fractional frequency units ($\Delta\nu/\nu_{clk}$) where $\nu_{clk} = 1.8$ PHz. See text for discussion.

Effect	$\lvert\mathrm{Shift}\rvert$ (10^{-20})	Uncertainty (10^{-20})
Excess micromotion	10	10
Gravitational	0	10
Cooling laser Stark	0	5
Electric quadrupole	3	3
Secular motion	5	1
Linear Doppler	0	1
Linear Zeeman	0	1
Background collisions	0	1
Blackbody radiation	0.013	0.013
Clock laser Stark	0	$\ll 0.01$
Trapping field Stark	0	$\ll 0.01$



Fraunhofer Cluster of Excellence Advanced Photon Sources CAPS

https://www.caps.fraunhofer.de › news › thorium-nucl...

Thorium Nuclear Clock – the world's most accurate clock

Oct 11, 2019 — Conventional **atomic clocks** on the effect of the characteristic frequency of radiation transitions in the electron shell. The **Thorium** Nuclear ...

Stability

Loss of contrast

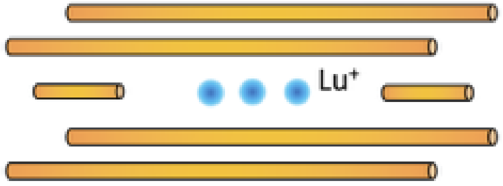
Because you make a comparison

$$\sigma = \frac{2\sqrt{2}}{2\pi f_0 \sqrt{NT\tau}}$$

$$N = 1, \quad T = 750\,\mathrm{ms}, \quad \tau = 48\,\mathrm{hr} \implies \sigma \approx 3.5 \times 10^{-18}$$

So, more ions and/or longer interrogation

Multiple ions?



Linear Paul Trap

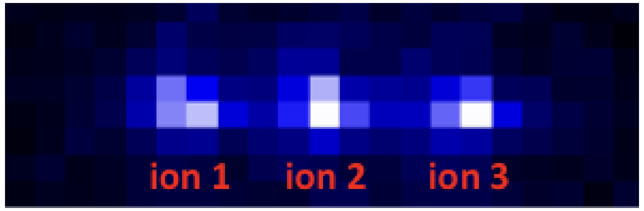
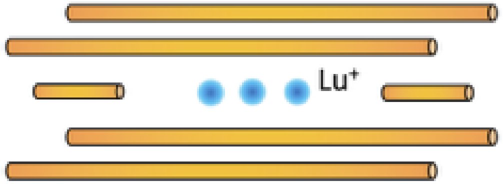


Image by EMCCD

- Inhomogeneous broadening
  - Micromotion
  - Magnetic fields
  - Quadrupole moment
  - AC magnetic fields

Multiple ions



Linear Paul Trap

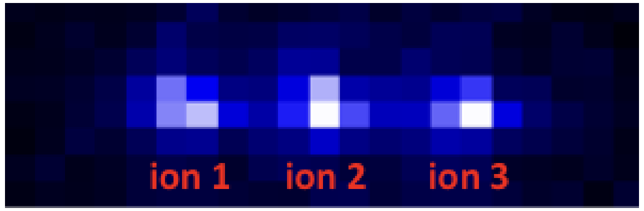
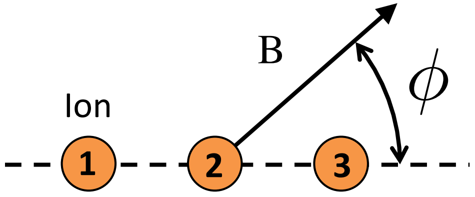
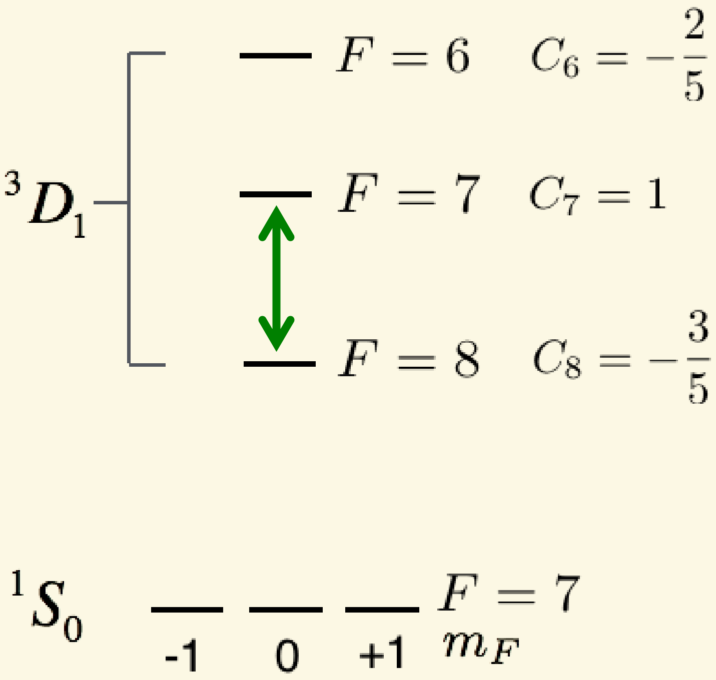
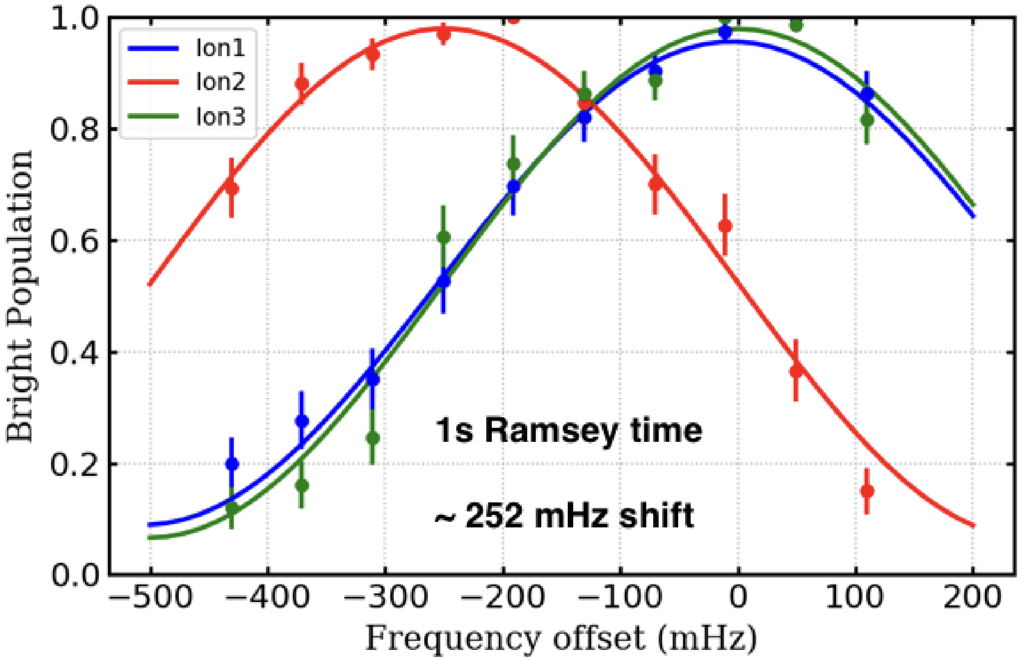


Image by EMCCD

Quadrupole: $\delta\nu_i = Q_i C_F (3\cos^2\phi - 1)\Theta(J)$



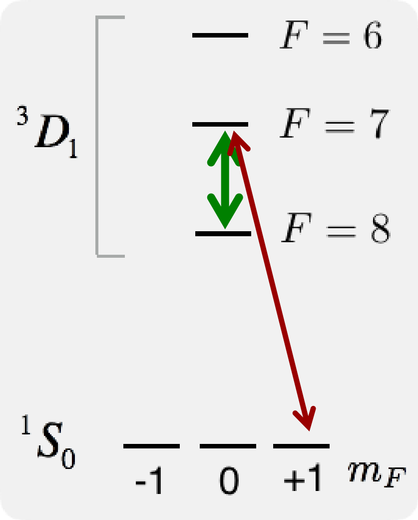




1s Ramsey time

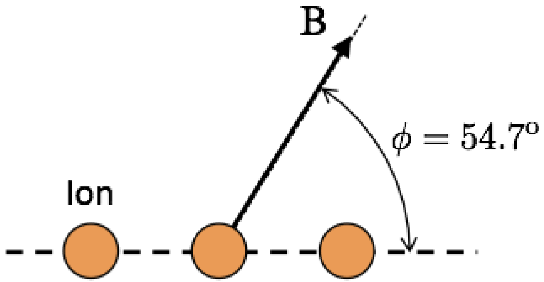
~ 252 mHz shift

Multiple ions

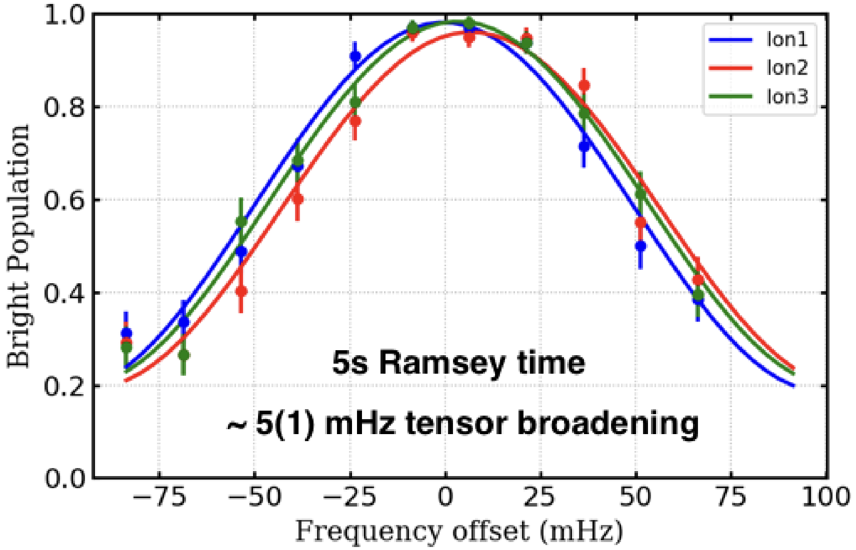


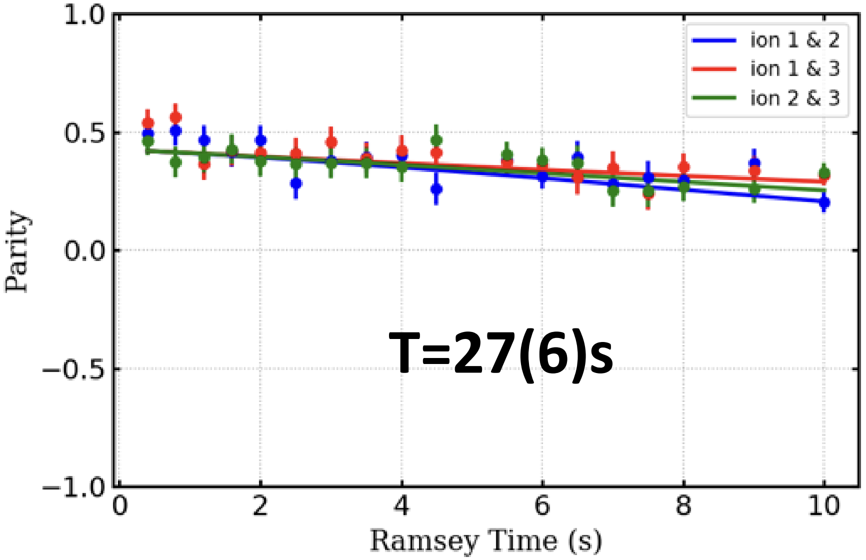
Tensor shifts

$$\delta\nu \propto \left(3\cos^2\phi - 1\right)$$



Magnetic Field is **nearly aligned**





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Stability

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Because you make a comparison

$$\sigma = \frac{2\sqrt{2}}{2\pi f_0 \sqrt{NT\tau}}$$

$$N = 1, \quad T = 750\,\mathrm{ms}, \quad \tau = 48\,\mathrm{hr} \implies \sigma \approx 3.5 \times 10^{-18}$$

$$N = 10, \quad T = 20\,\mathrm{s}, \quad \tau = 48\,\mathrm{hr} \implies \sigma \approx 2.2 \times 10^{-19}$$

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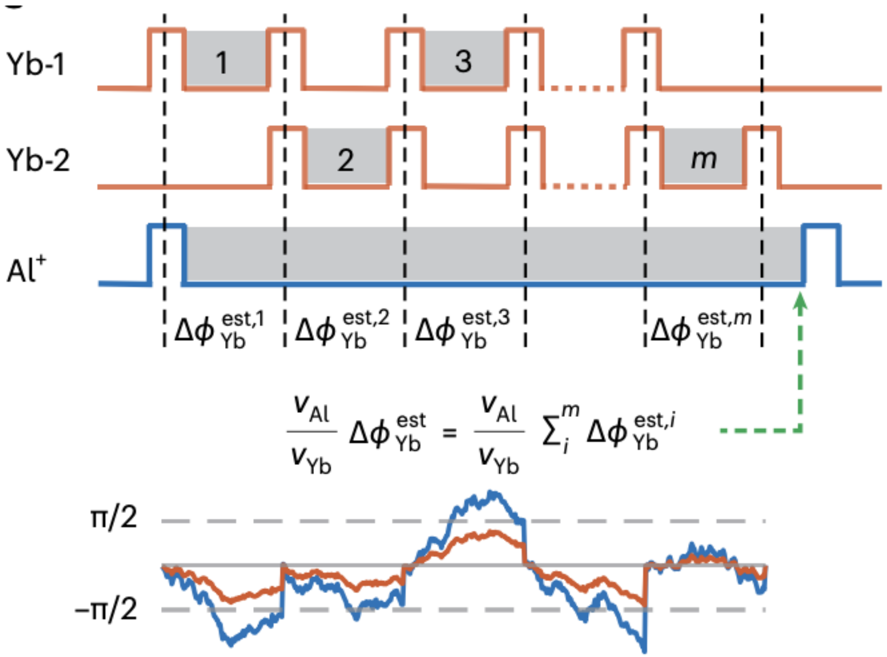
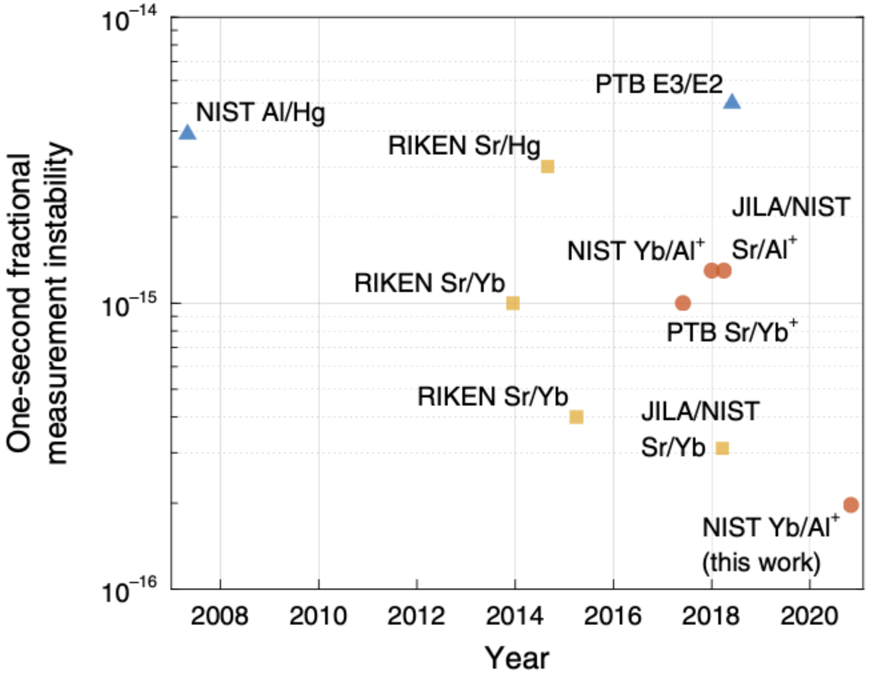
Stability

Letter

https://doi.org/10.1038/s41567-022-01794-7

Improved interspecies optical clock comparisons through differential spectroscopy





Verifying clock performance.

Measured clock frequency

$$f = f^{(0)}(1 + \mathbf{p} \cdot \mathbf{e})$$

In situ ratio measurement

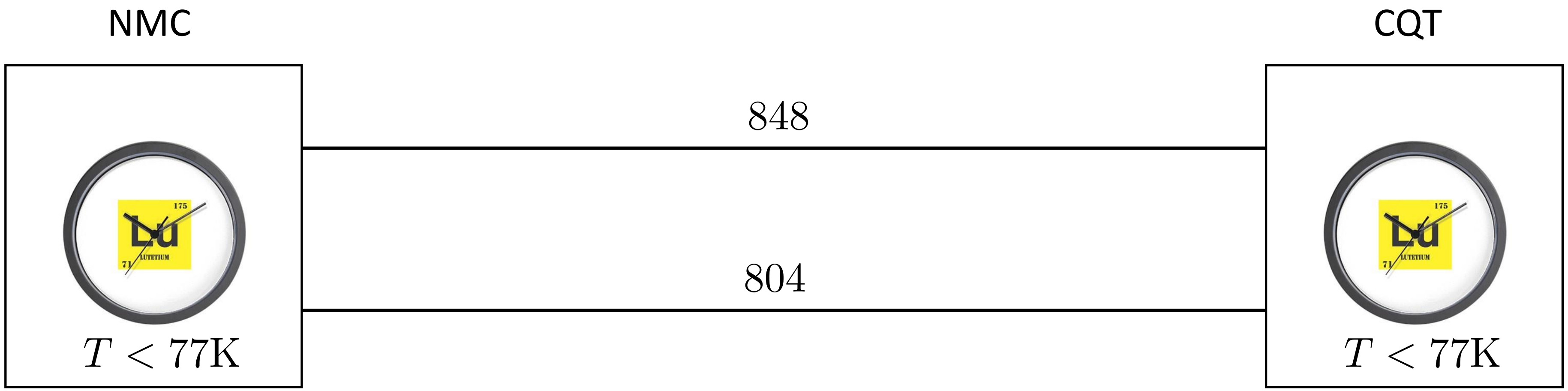
$$\frac{f_2}{f_1} = R^{(0)} \left(\frac{1 + \mathbf{p}_2 \cdot \mathbf{e}}{1 + \mathbf{p}_1 \cdot \mathbf{e}} \right)$$
$$= R^{(0)} \left(1 + (\mathbf{p}_2 - \mathbf{p}_1) \cdot \mathbf{e}\right)$$

Shift = sensitivity factor x magnitude of perturbation
(atomic property) (environment)

- determine atomic properties with sufficient precision
- rigorous measurement-based methodologies to assess environment

$\langle B^2 \rangle$	Dominated by the applied dc field.
$\langle \widetilde{B}_\perp^2 \rangle$	Limited by $^3D_2$ $-1.1 \times 10^{-17}/\mu\mathrm{T}^2$
∇E	Hyperfine mediated quadrupole moment ($^3D_2$?)
$\langle E^2 \rangle$	Micromotion, ac-Stark shift, BBR shift
	$^3D_1$ $^3D_2$ 2.7×10^{-17} at $300\,\mathrm{K}$

A local frequency reference



NMC

CQT

848

804

$T < 77\mathrm{K}$

$T < 77\mathrm{K}$

$$R_0 = \frac{f_{848}}{f_{804}}$$

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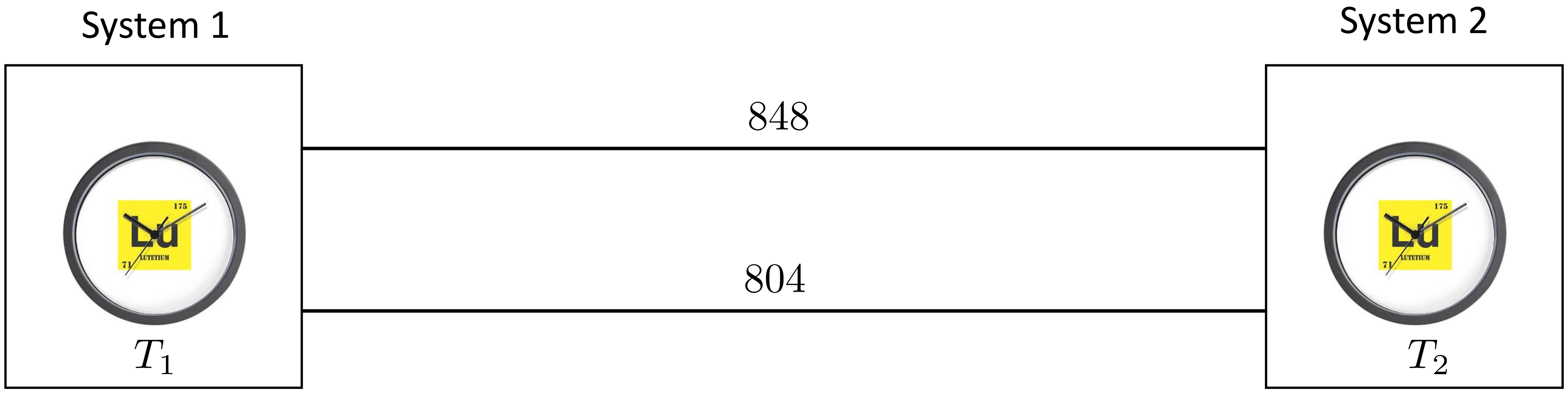
A local frequency reference



$$R_1 = \frac{f_{848}}{f_{804}}$$

$$R_2 = \frac{f_{848}}{f_{804}}$$

- In situ ratios determine system temperatures
- Comparison on the primary determines the redshift
- Comparison on the secondary confirms the temperature and redshift

The transitions are sensitive to α variations, the ratio is not.

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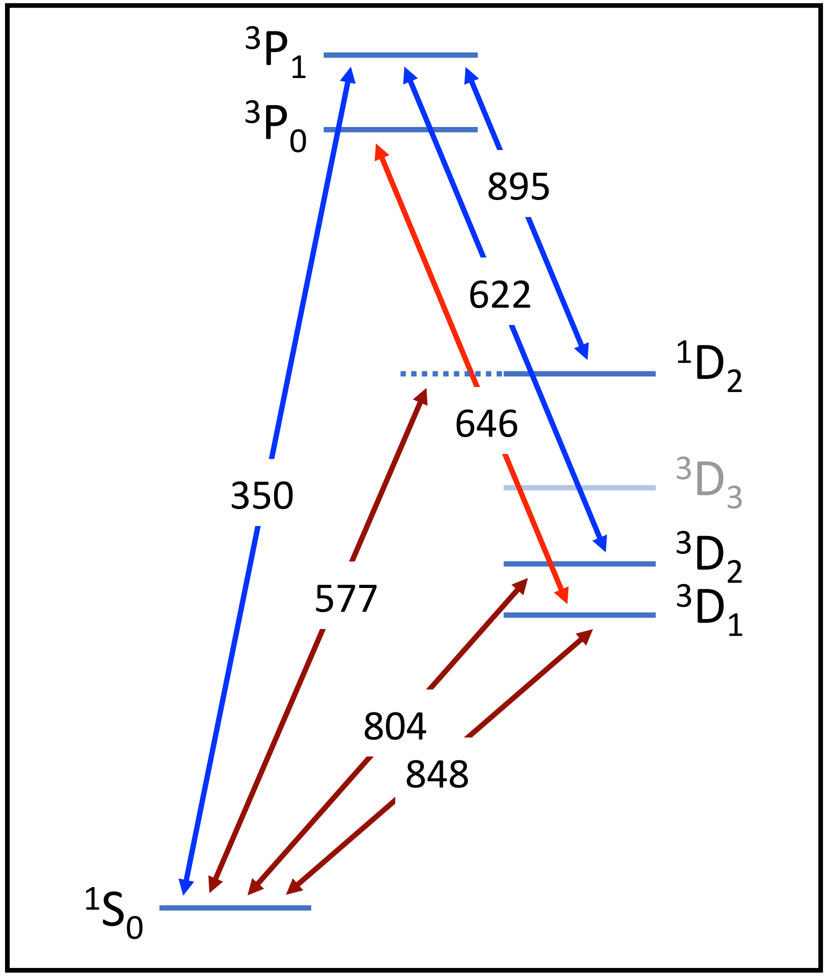
What lutetium has to offer…

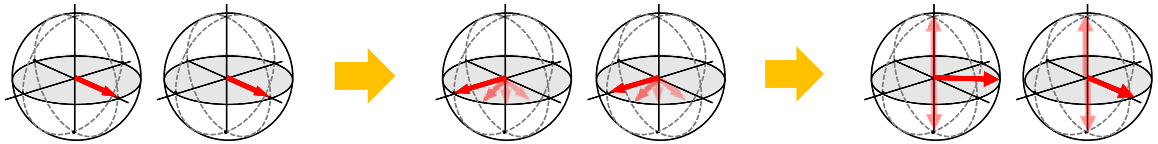
- A technically easy path to 10^{-18} and beyond
    - Long lifetimes
    - Low systematics for robust < 10^{-18} operation.
    - Suitable isotope for magnetic field insensitivity
    - Easily accessible laser systems
    - State sensitive detection channel
- A pathway to ``many’’ ions.
- Two clock transitions to verify clock performance.
    - Similar sensitivities to alpha variation.



The group

(Left to Right)
Rattakorn Keawuam (NIMT),
Scott Bustabad (NMC),
Nakarin Jayjong,
Kyle Arnold
Me
Zhang Zhao
Zhao Qi
Qin Qichen
Michael Kang

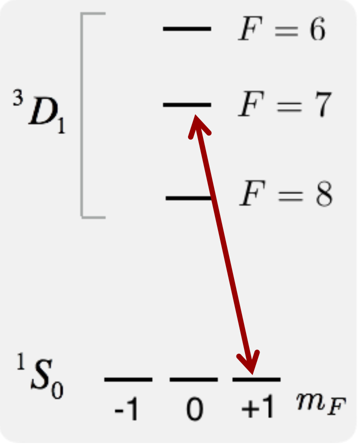
Multiple ions

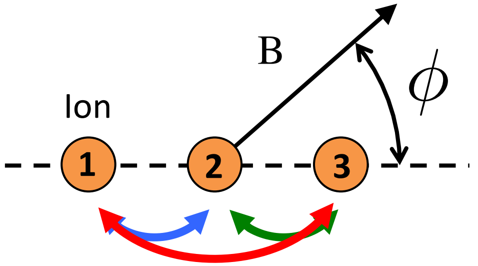


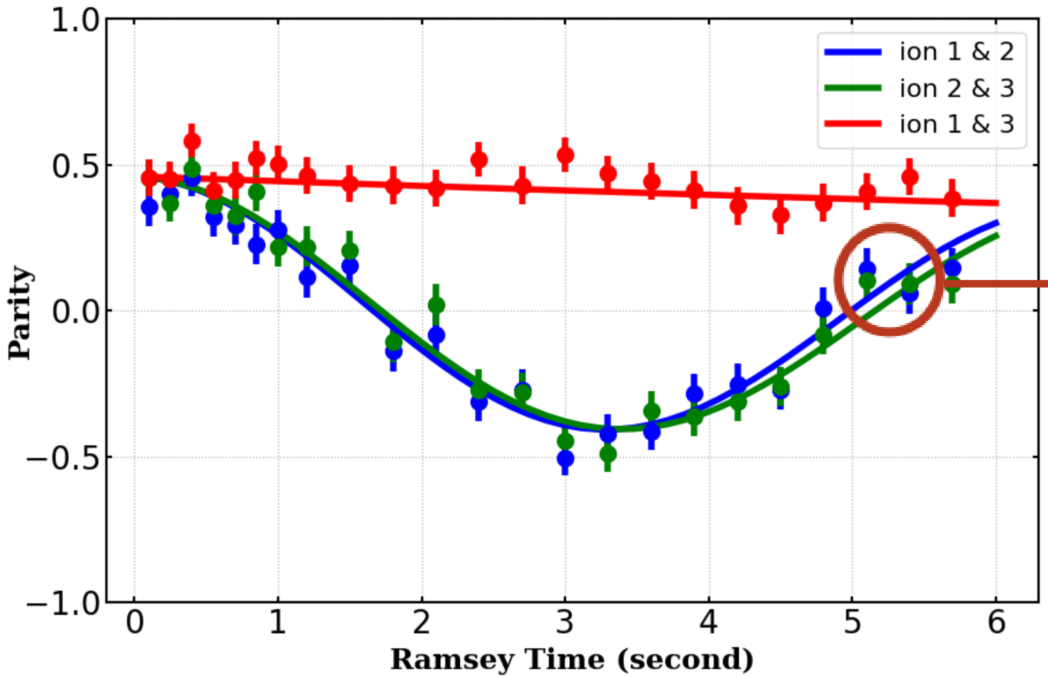
1st Ramsey Pulse

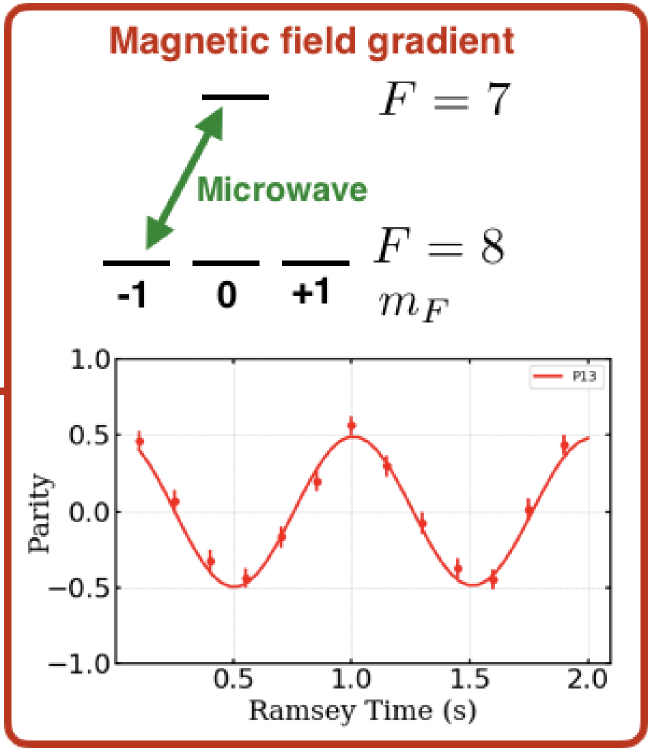
Free evolution beyond laser coherence time

2nd Ramsey Pulse followed by detection and compute $\sigma_z^{(i)}\sigma_z^{(j)} / \cos(' _{ij})$



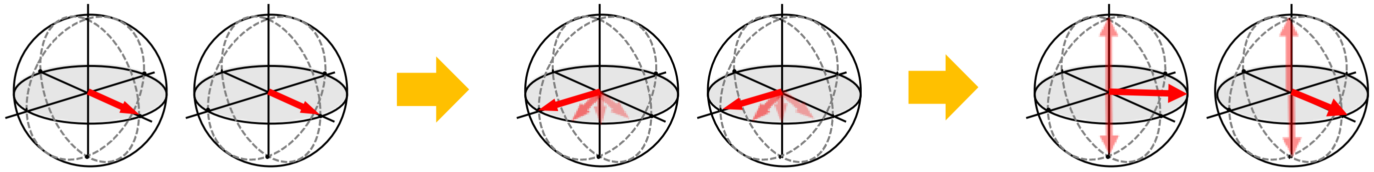






Magnetic field gradient

Correlation spectroscopy

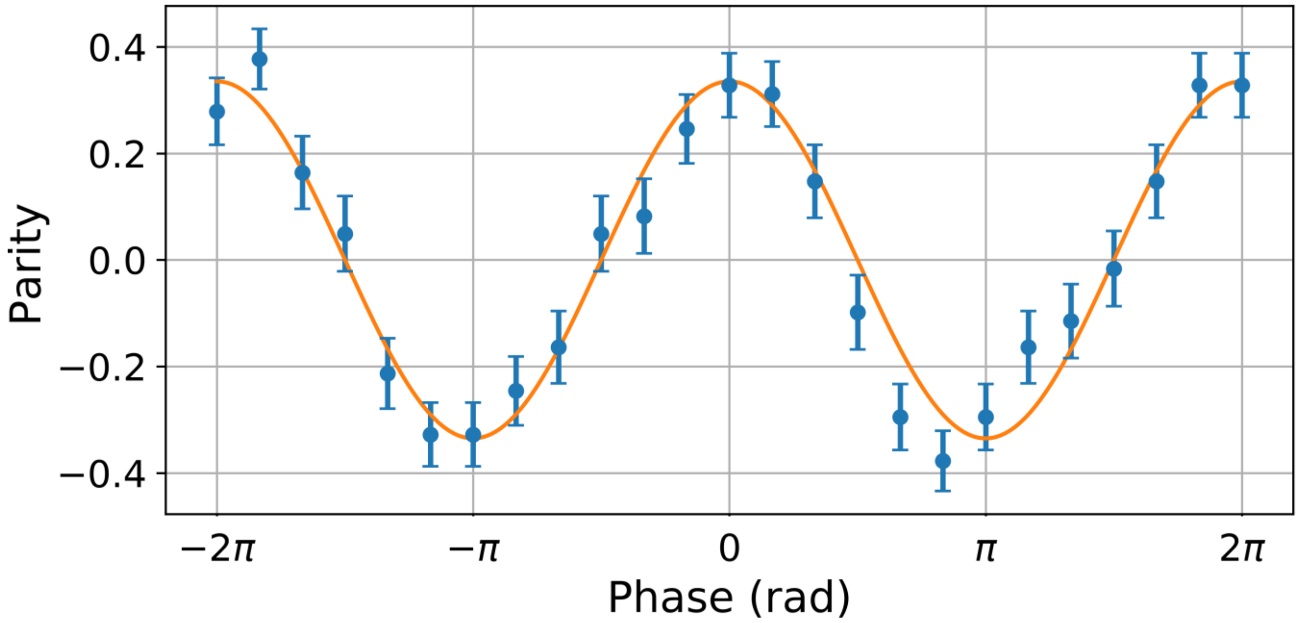


1st Ramsey Pulse

Free evolution beyond laser coherence time

2nd Ramsey Pulse followed by detection and compute $\sigma_z^{(i)}\sigma_z^{(j)}$ / cos(' $_{ij}$)

$$p_{12} = \langle \sigma_{z,1}, \sigma_{z,2} \rangle = \frac{p_c}{2} \cos(2\pi T_R \Delta f)$$



$T_R = 744\ ms$

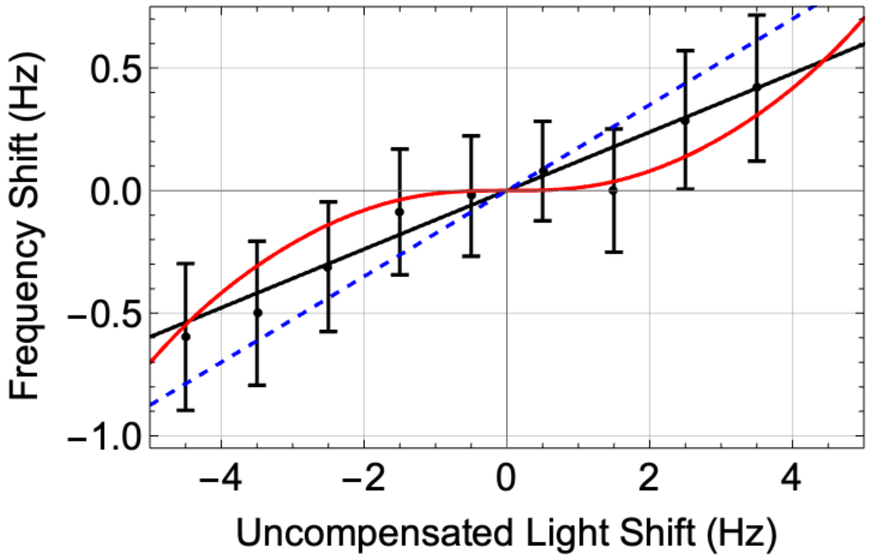
E. R. Clements, et al, PRL **125**, 243602 (2020)

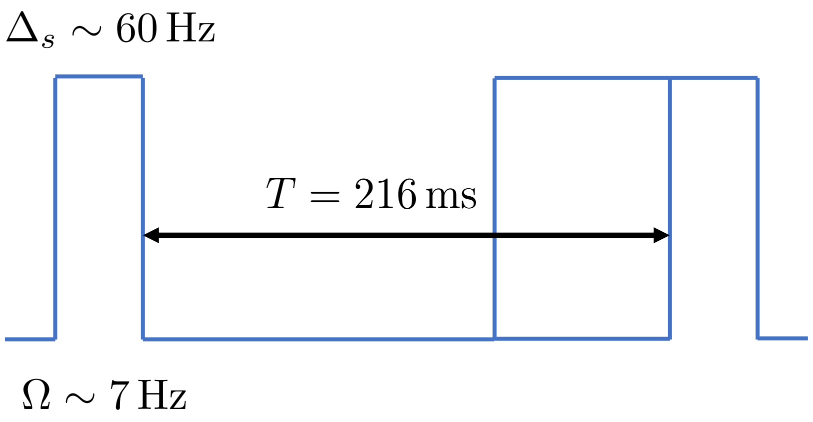
T. R. Tan, et al. PRL, **123**, 063201 (2019)

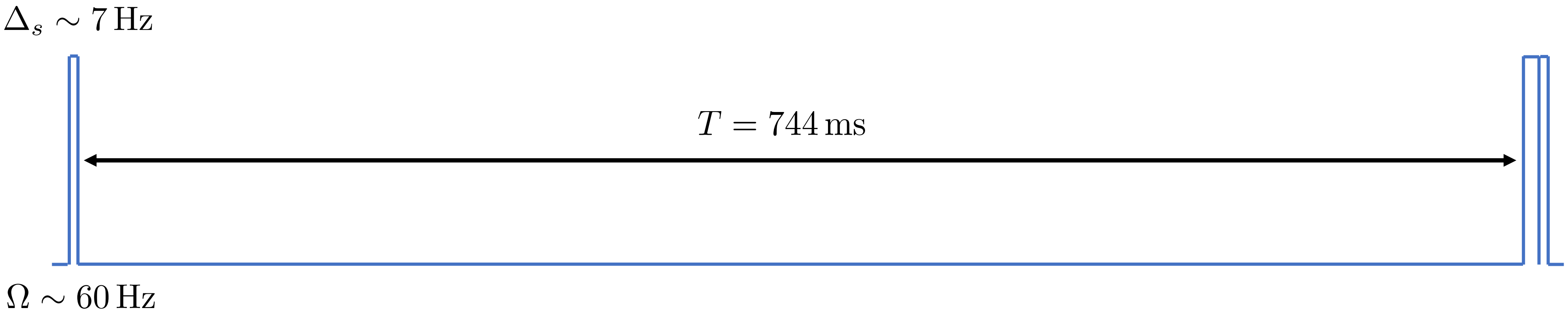
Hyper-Ramsey Suppression

PRL **109,** 213002 (2012) PHYSICAL REVIEW LETTERS week ending 21 NOVEMBER 2012

Generalized Ramsey Excitation Scheme with Suppressed Light Shift







Segal’s Adage

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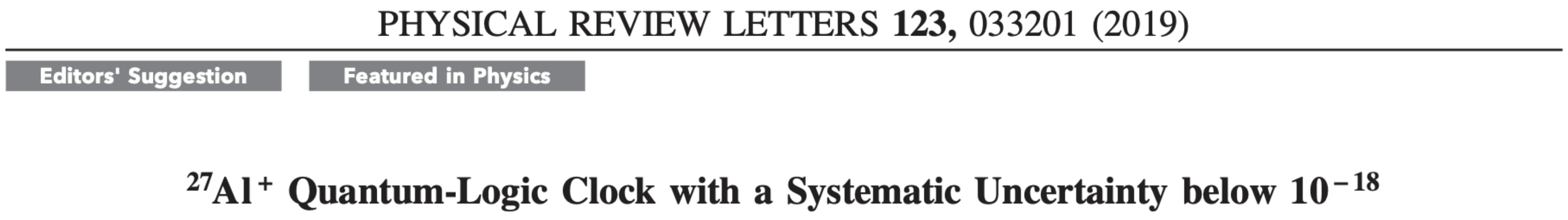
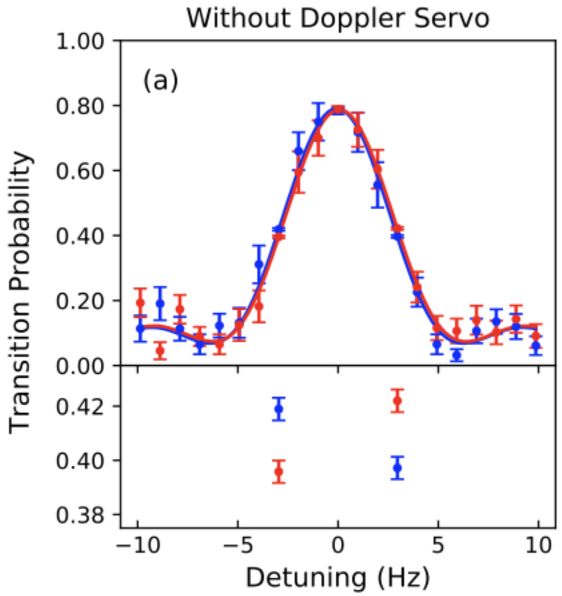


PHYSICAL REVIEW LETTERS **123,** 033201 (2019)

Editors' Suggestion **Featured in Physics**

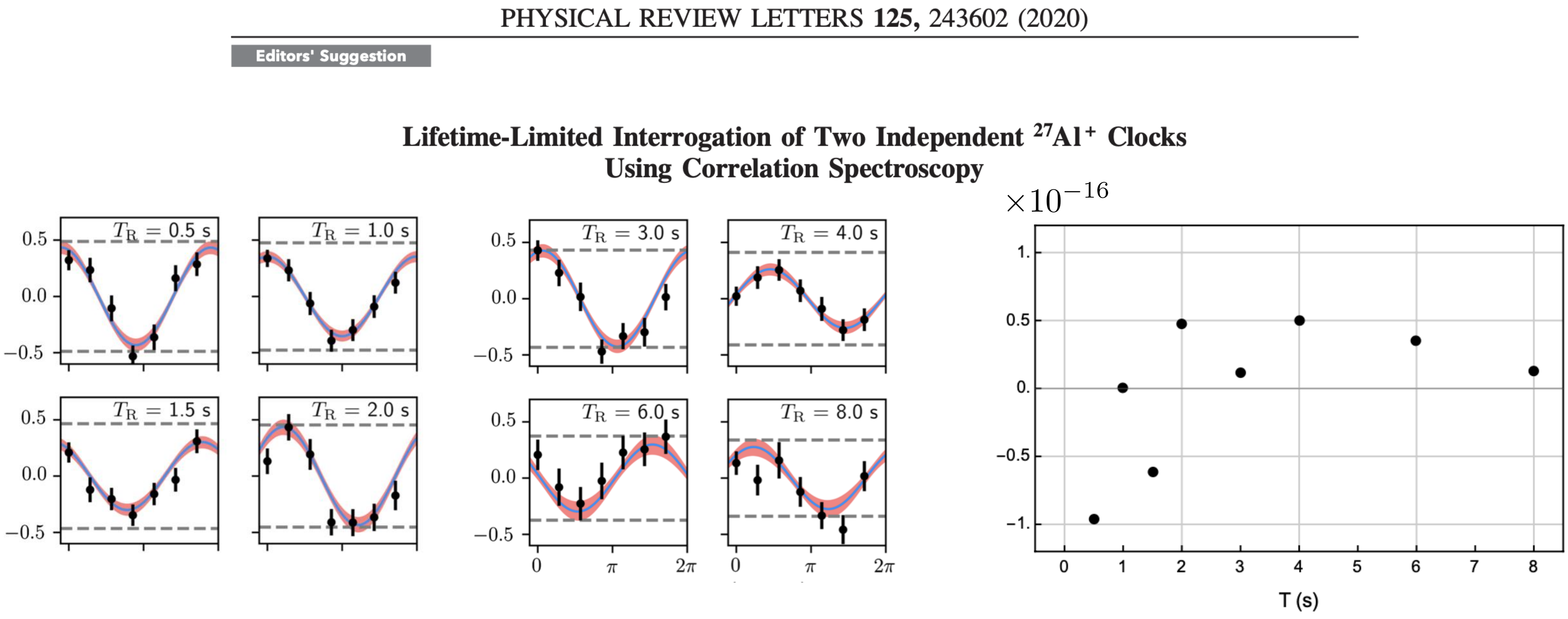
^{27}Al^{+} Quantum-Logic Clock with a Systematic Uncertainty below 10^{-18}



10^{-16} difference from counter propagating beams

Segal’s Adage

A man with one clock knows exactly what time it is, the man with two clocks is never sure.



PHYSICAL REVIEW LETTERS **125,** 243602 (2020)

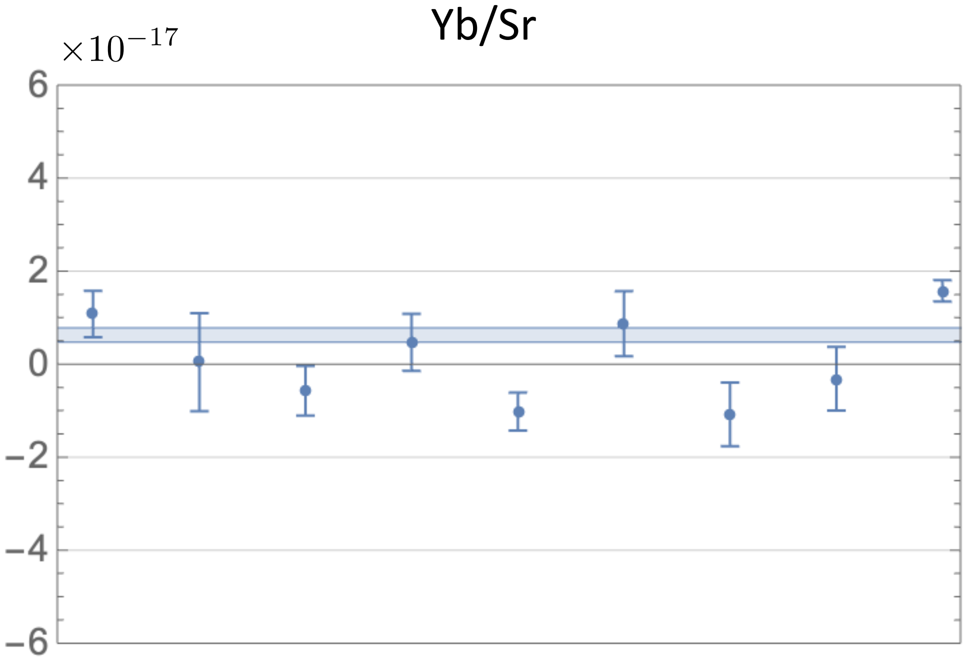
Editors' Suggestion

Lifetime-Limited Interrogation of Two Independent $^{27}Al^+$ Clocks Using Correlation Spectroscopy

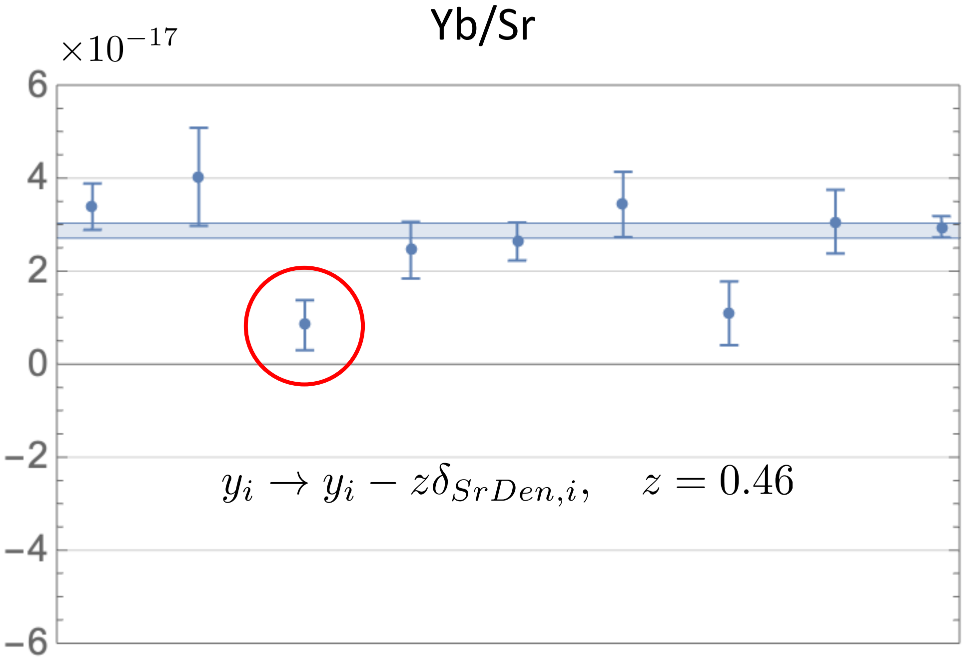
10^{-16} differences between two clocks

Centre for
Quantum
Technologies

The BACON Collaboration



$$\chi^2_\nu = 6.0, \quad P(\chi^2_\nu > 6.0) = 10^{-7}$$



$$y_i \to y_i - z\delta_{SrDen,i}, \quad z = 0.46$$

$$\chi^2_\nu = 1.5, \quad P(\chi^2_\nu > 1.5) = 0.16$$

- Underspread in the Al⁺/Sr ratio: $\chi^2_\nu = 0.2, \quad P(\chi^2_\nu < 0.2) = 0.014$
- Overspread in the Yb/Sr ratio: $\chi^2_\nu = 6.0, \quad P(\chi^2_\nu > 6.0) = 10^{-7}$
- Correlation in the Yb/Sr ratio measurements with the Sr density shift