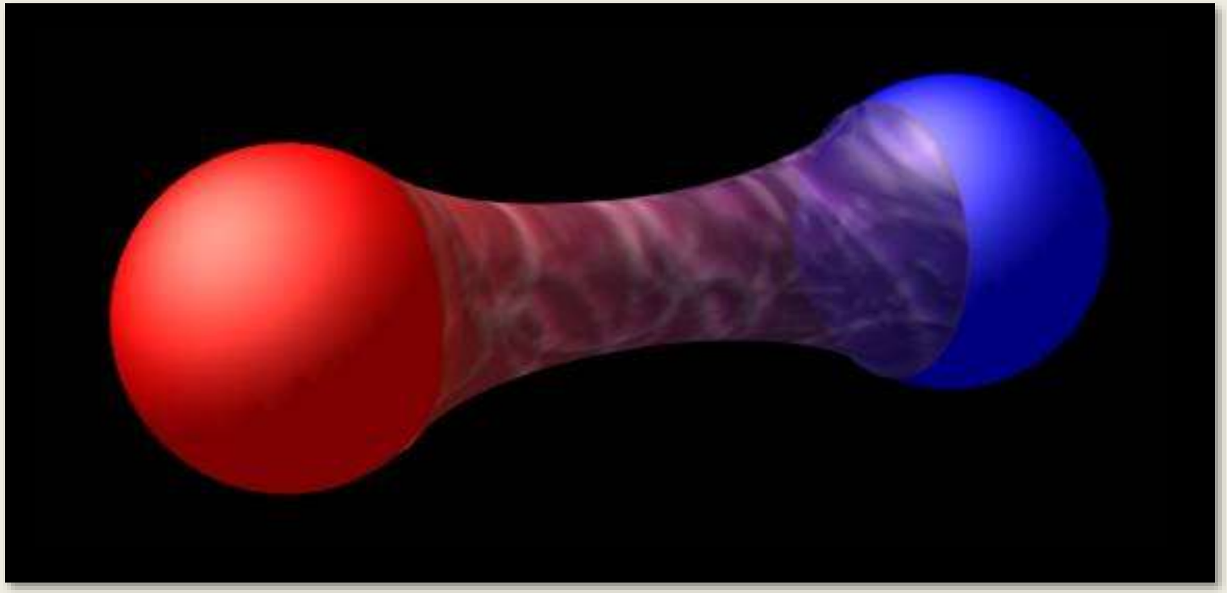
Quantum Logic Spectroscopy & Clocks II



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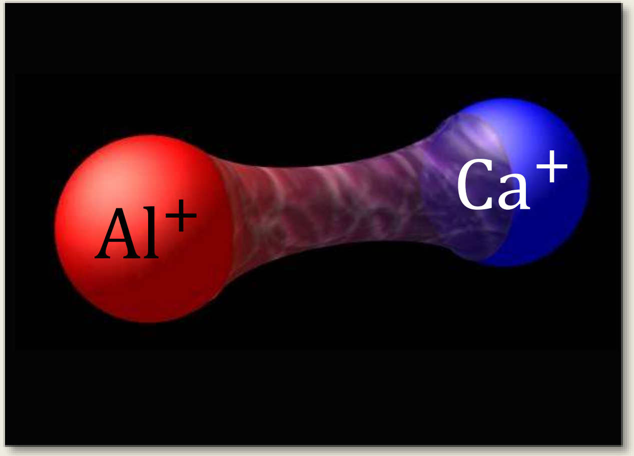
Frontiers of Quantum Metrology for New Physics Searches

Bad Honnef Physics School, Physikzentrum Bad Honnef, May 11 - 16, 2025

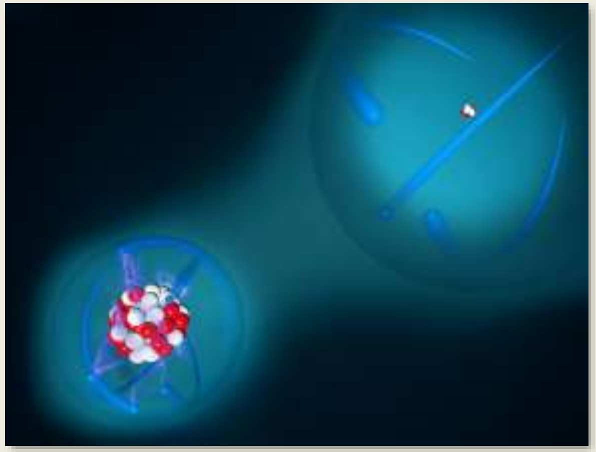
Overview

- Complete introduction to quantum logic with trapped ions
- Applications:

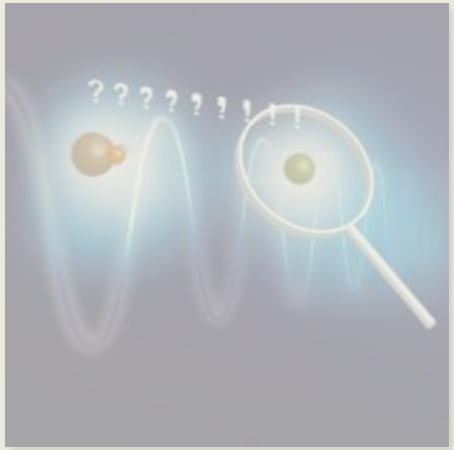
Al^+ quantum logic clock

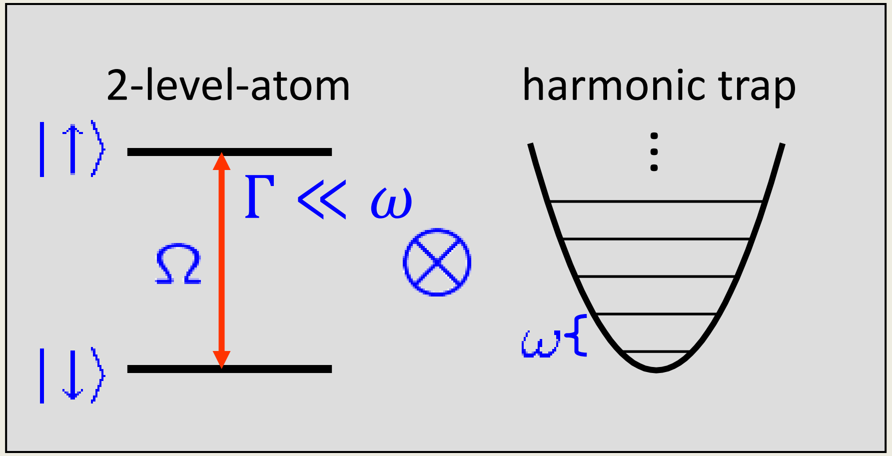


QLS of highly charged ions



QLS of molecules





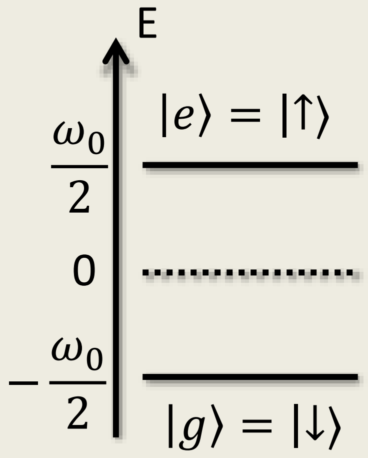
QUANTUM LOGIC WITH TRAPPED IONS

Atom-light interaction

- Hamiltonian: $H = H_a + H_m + H_i$
- Atom: $H_a = \frac{\hbar\omega_o}{2}(|e\rangle\langle e| - |g\rangle\langle g|) = \frac{\hbar\omega_0}{2}\sigma_z$
- Motion: $H_m = \hbar\omega_z a^\dagger a$
- Atom-Light-Interaction: $H_i = -\hat{\vec{d}}\vec{E} = e\hat{\vec{r}}\vec{E}_0 \cos(k\hat{z} - \omega t + \phi)$

$$\Rightarrow \mathrm{H_i} = \frac{\hbar\Omega}{2}(\sigma_+ + \sigma_-)\left(e^{i(k\hat{z}-\omega t+\phi)} + e^{-i(k\hat{z}-\omega t+\phi)}\right)$$

with $\Omega = \Omega_{ge} = \frac{e\vec{E}_0}{\hbar}\langle e|\hat{\vec{r}}|g\rangle$ and $\sigma_+ = |e\rangle\langle g|$, $\sigma_- = |g\rangle\langle e|$, Basis: $\{|g,n\rangle, |e,n\rangle\}$

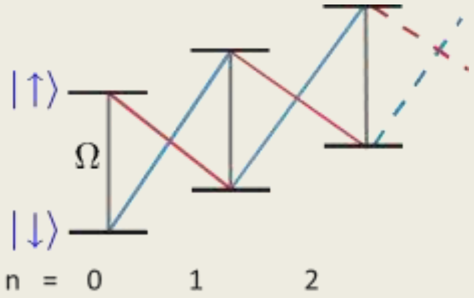


→ quantum dynamics simulations using QuTiP

[D. J. Wineland *et al.*, J. Res. Natl. Inst. Stand. Technol. **103**, 259–328 (1998)]

Quantized atom-light interaction including motion

- Interaction in Lamb-Dicke regime:

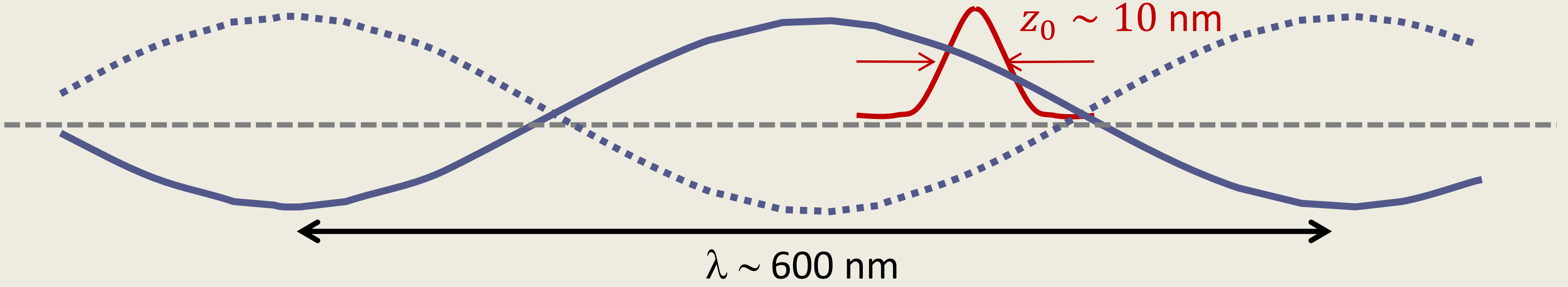


$$H_i' \approx \left(\hbar\Omega\hat{\sigma}_+ e^{-i\Delta t+i\phi}\right)/2 + h.c.$$ carrier (Δn=0)

$$+\left(i\hbar\eta\Omega\hat{\sigma}_+\hat{a}e^{-i(\Delta+\omega_z)t+i\phi}\right)/2 + h.c.$$ red sideband

$$+\left(i\hbar\eta\Omega\hat{\sigma}_+\hat{a}^\dagger e^{-i(\Delta-\omega_z)t+i\phi}\right)/2 + h.c.$$ blue sideband

with $\eta = kz_0$ and $z_0 = \sqrt{\hbar/2m\omega_z}$

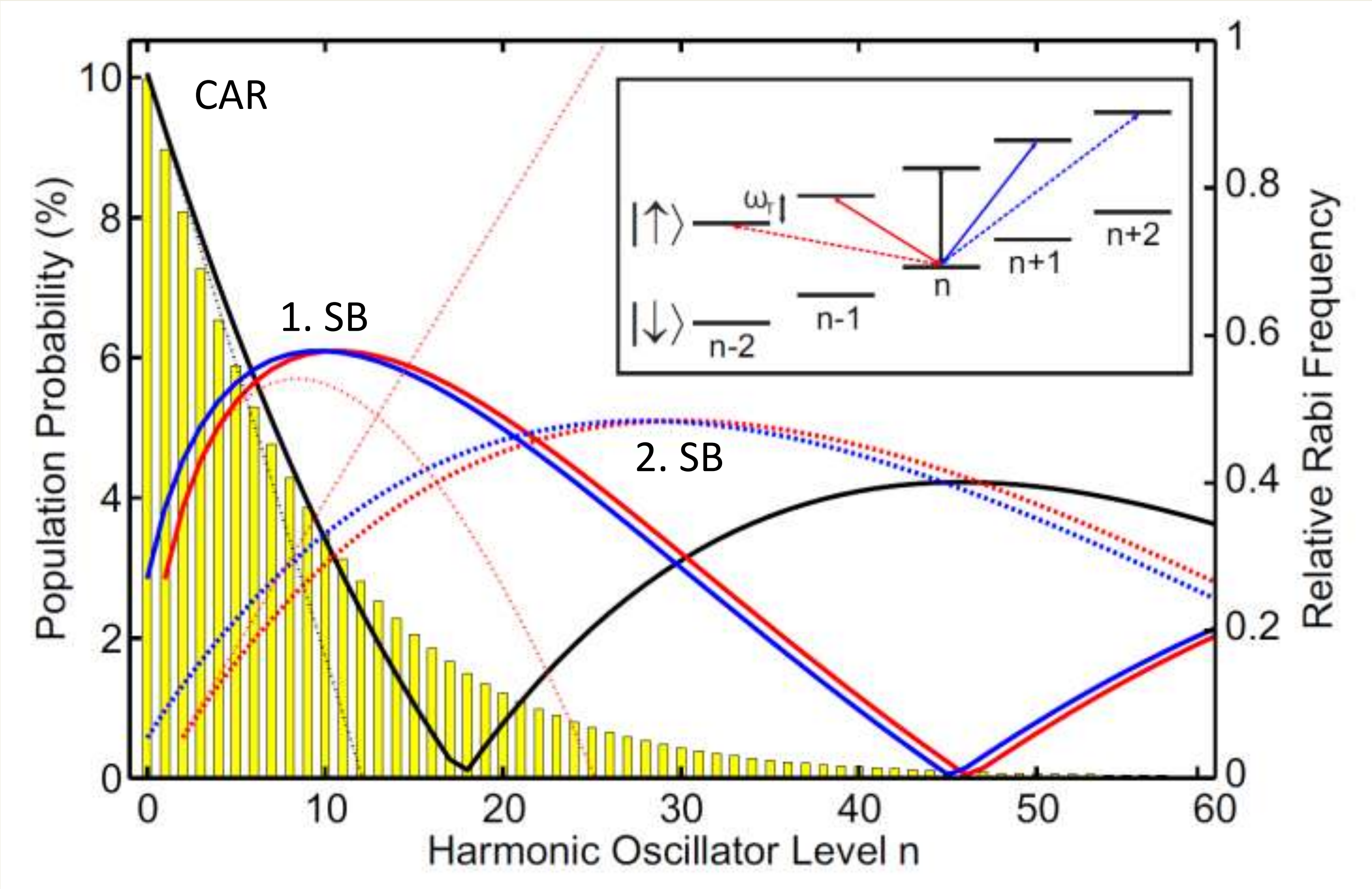


- $E = const$ drives carrier
- $\vec{\nabla}E$ drives sidebands (smaller by η)

What happens for
$(k\langle\hat{z}^2\rangle)^{1/2} = \eta\sqrt{\langle\Psi|(\hat{a}+\hat{a}^\dagger)^2|\Psi\rangle} = \eta_c > 1$?

[D. J. Wineland *et al.*, J. Res. Natl. Inst. Stand. Technol. **103**, 259–328 (1998)]

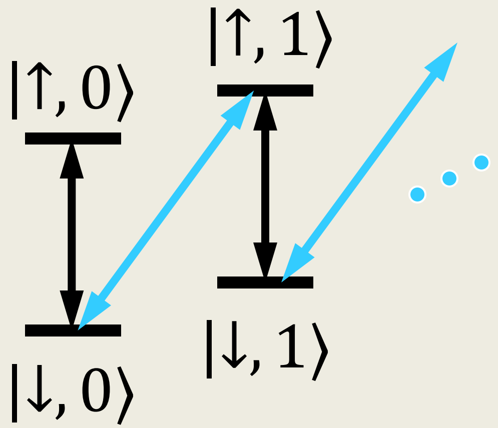
Higher-order sidebands

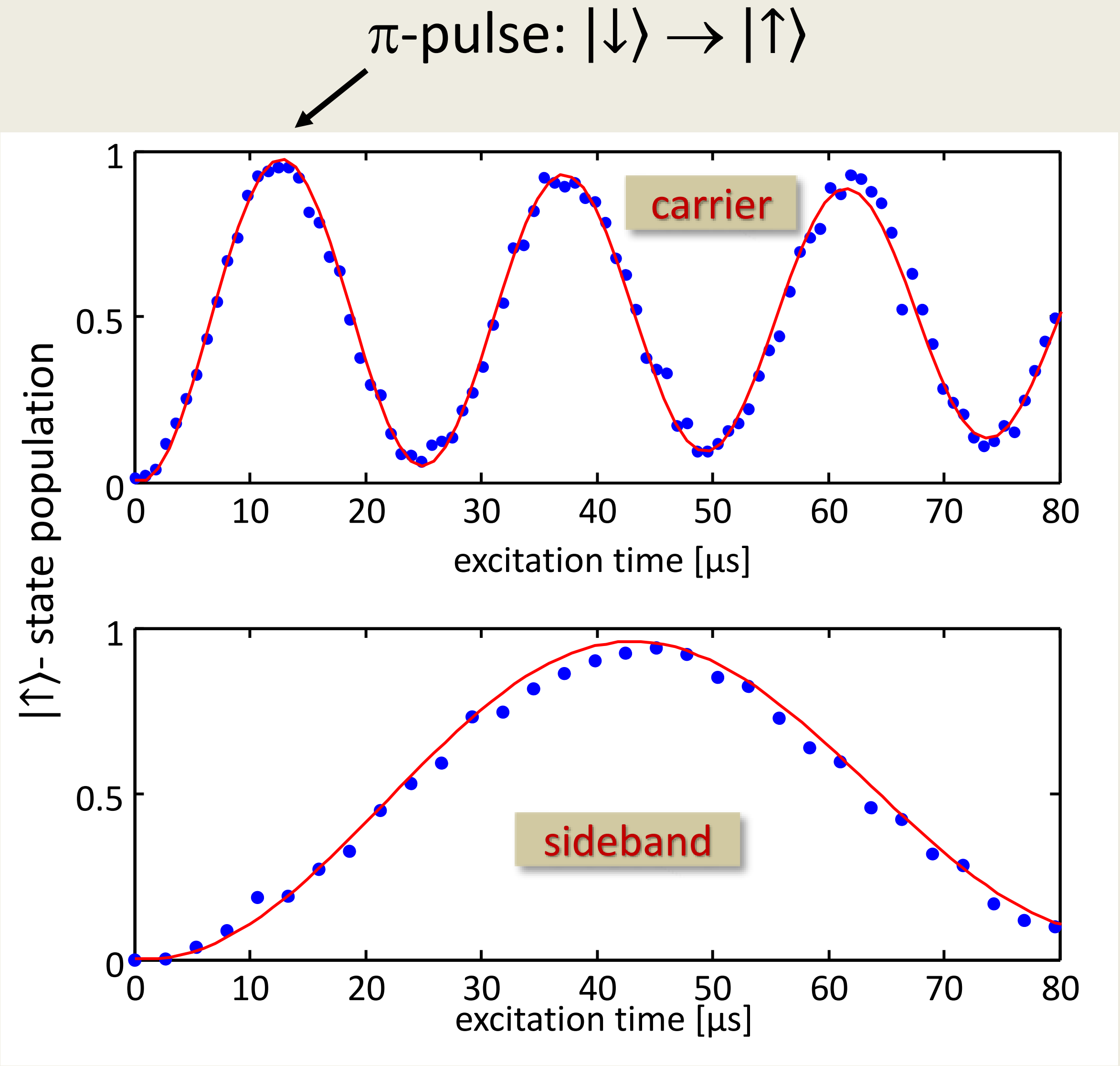


$\eta = 0.28$, $\omega_z = 2\pi\times2.2$ MHz, $T_D \approx 1$ mK, $\bar{n} \approx 10$

[B. Hemmerling *et al.*, Appl. Phys. B **104**, 583 (2011)]

Coherent state manipulation



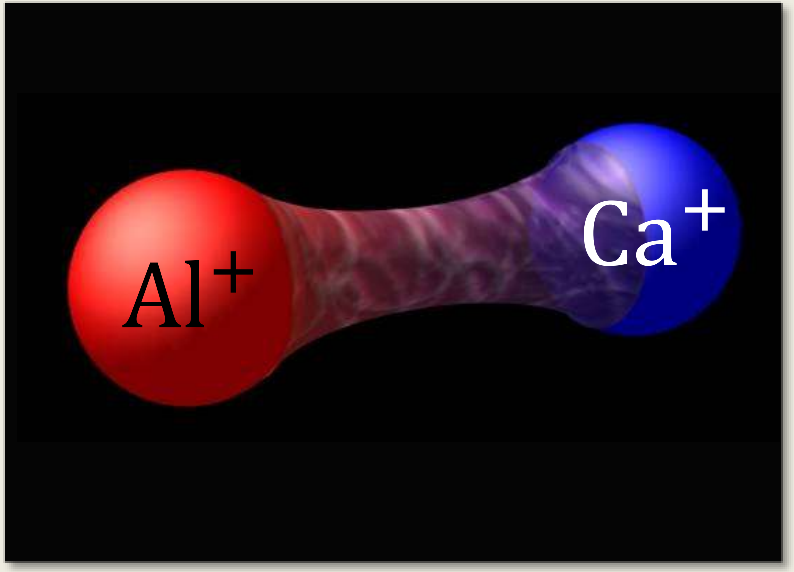


Rabi frequencies:

- CAR: Ω
- BSB: $\eta\Omega\sqrt{n+1}$
- RSB: $\eta\Omega\sqrt{n}$

(BSB and RSB: Jaynes-Cummings physics)

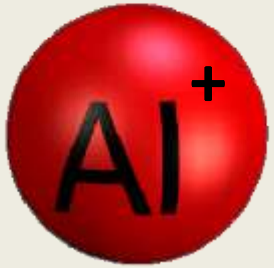
- Lamb-Dicke parameter: $\eta = kz_0$

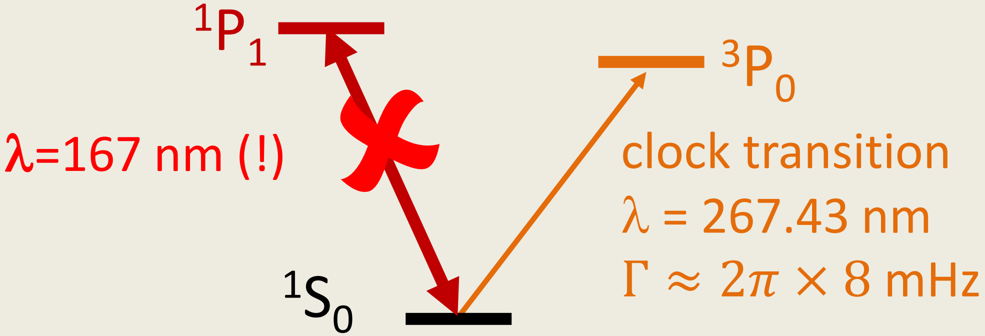


AL^{+} QUANTUM LOGIC CLOCK

Aluminum as Optical Clock Atom

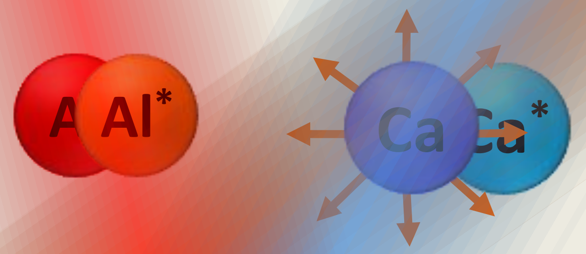
- Hans Dehmelt 1992 (NP 1989)
- Al^+ Features:
  - narrow optical transition
  - no electric quadrupole shift
  - small black-body shift
  - But: no accessible cooling transition

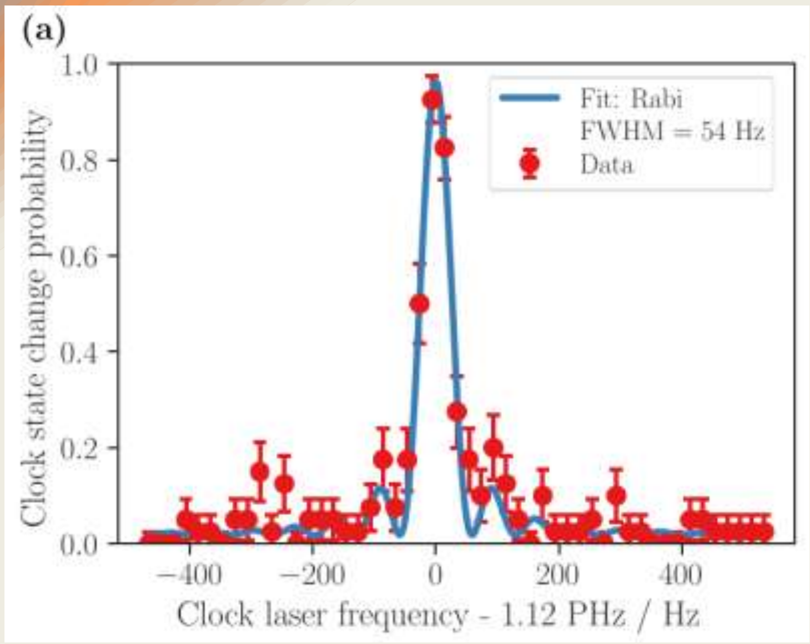


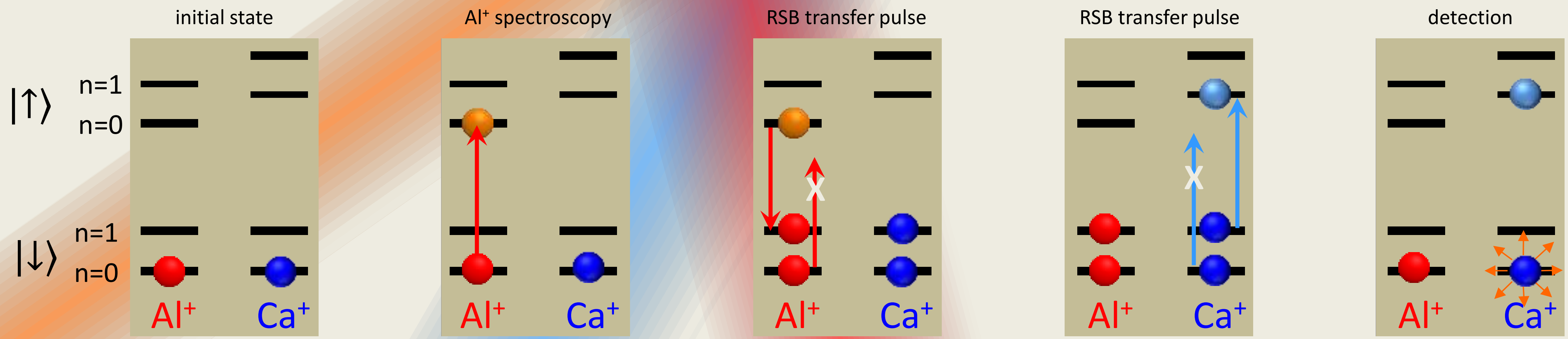


$^{27}Al^+$ (I=5/2) partial level scheme

Quantum Logic State Transfer





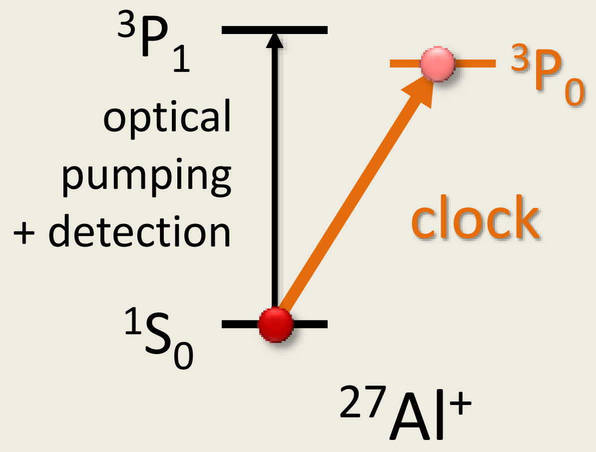


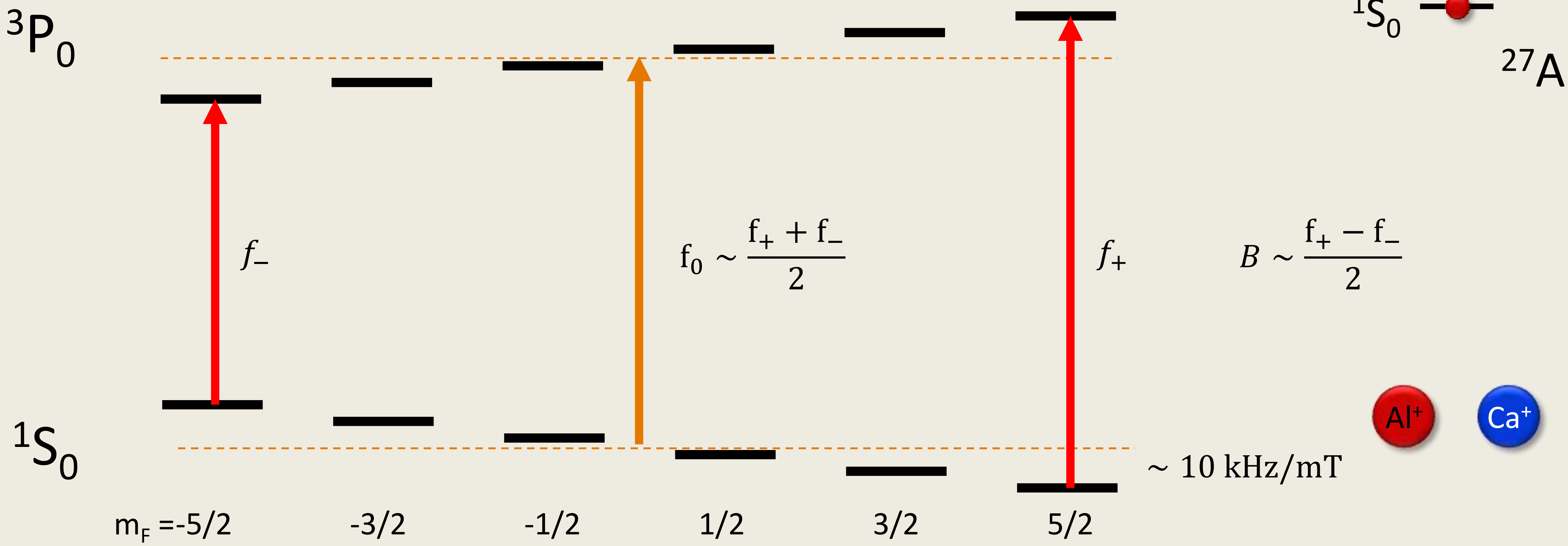
[D.J. Wineland *et. al.*, Proc. 6th Symposium on Frequency Standards and Metrology, 361 (2001); P.O. Schmidt *et al.*, *Science*, **309**, 749 (2005)]

Clock Interrogation Sequence

Laser cooling 15 ms	State prep. <1 ms	Probe clock transition 100 – 300 ms	Detect Al+ state 10 ms

+ Optical pumping
+ Interleaved measurements → 60% duty cycle



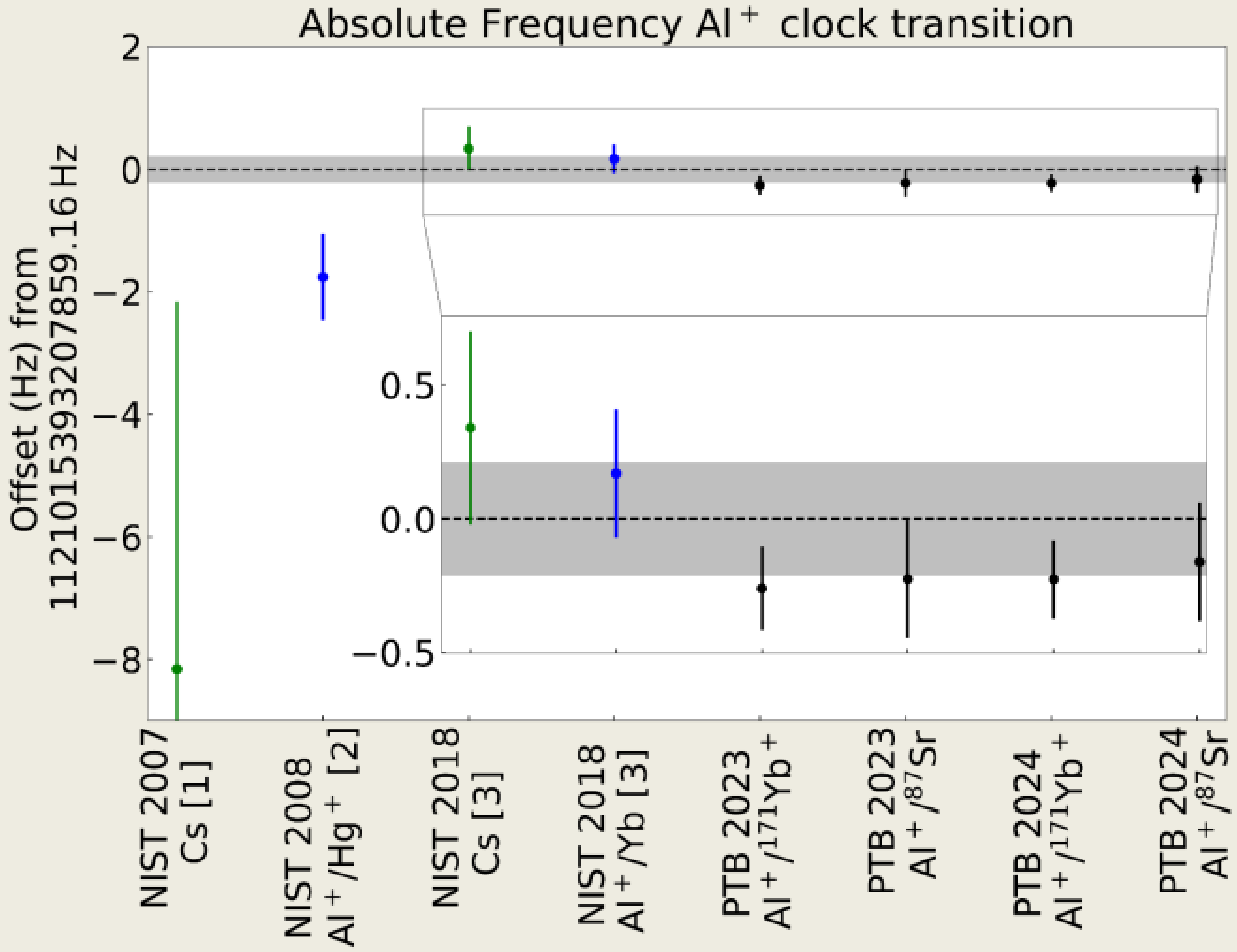


PTB Al^+ clock error budget its absolute frequency

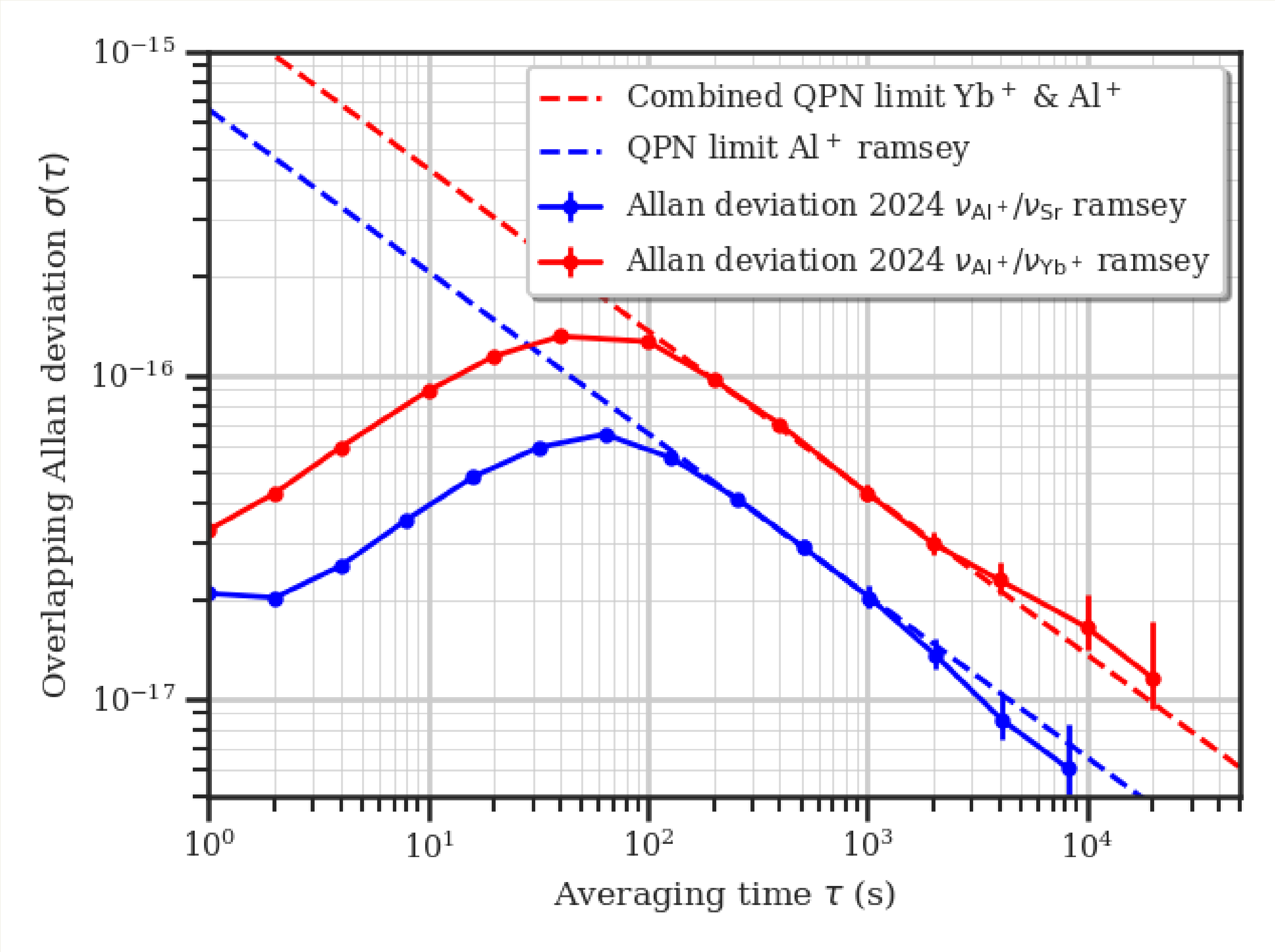
Name	Total shift (e-18)	Uncertainty (e-18)
Excess micromotion	**-1.00**	Re-Evaluation: $< 5 \times 10^{-19}$
Cooling laser Stark	**-6.0**	
Time-dilation shift	-1.79	0.40
Quadratic Zeeman – dc	-1486.88	0.49
Quadratic Zeeman – ac	-15.72	0.25
BBR	-3.34	0.30
Clock laser Stark	0.00	0.3
Other shifts	0.72	1.3
Total	**-1521.6**	**2.6**

NIST Al^+ clocks:

- 9.4×10^{-19} [Brewer *et al.*, PRL **123**, 033201 (2019)]
- 5.5×10^{-19} [Marshall *et al.*, arXiv:2504.13071]



PTB Al^+/Sr frequency ratio



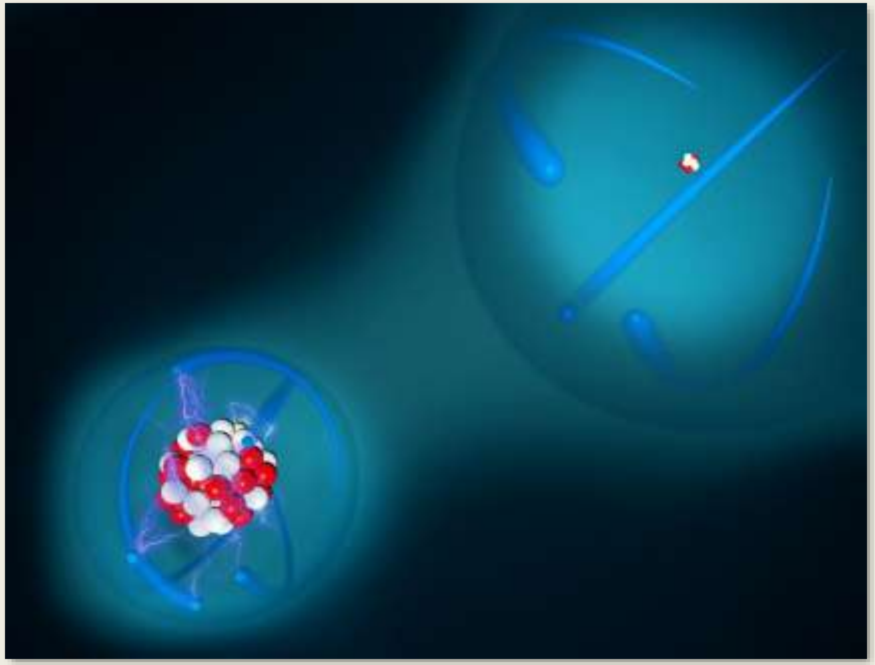
- Ramsey interrogation with 250 ms (50 ms Ramsey pulses)
- Duty cycle: 60% (live monitoring of magnetic field & micromotion)
- Small instability thanks to Si-stabilized laser: $6.6 \times 10^{-16}/\sqrt{\tau/s}$
- First clock with EIT cooling while probing → long probe times [F. Dawel *et el.*, in preparation]

Al^+/Sr frequency ratios

Discrepancies currently unresolved

Summary Al^+ clock

- NIST Al^+ clock currently most accurate clock:
  5.5×10^{-19} [Marshall *et al.*, arXiv:2504.13071]
- Clear path towards 1×10^{-19}
- Discrepancies between frequency ratios measured by different groups
  → more frequency ratio measurements required
- Al^+ clock transition has a very small sensitivity to new physics
  → anchor transition

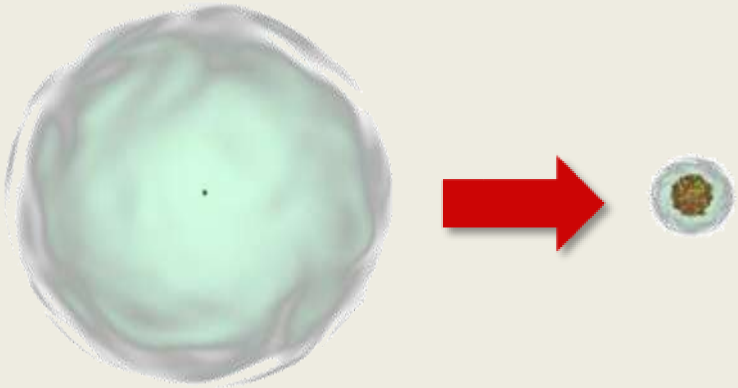


QUANTUM LOGIC SPECTROSCOPY OF HIGHLY CHARGED IONS

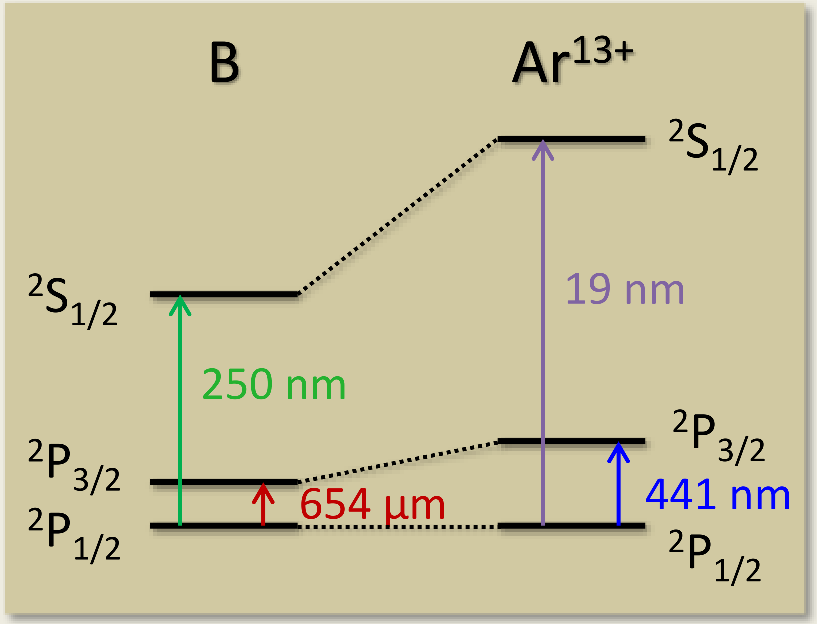
Highly Charged Ions

Charge state dependence:

		H	→	***U^{91+} (H-like)***
• Binding energy	~ Z^2	10 eV	→	140 keV
• Hyperfine splitting	~ Z^3	µeV	→	eV
• QED effects	~ Z^4	µeV	→	300 eV
• Stark shifts	~ Z^{-6}			



strongly relativistic systems with large QED effects



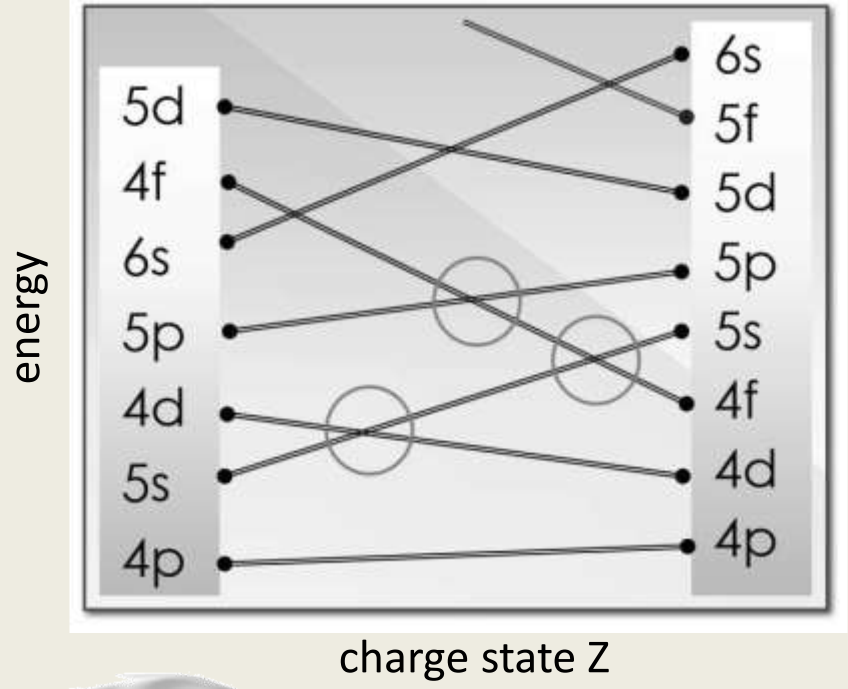
- optical transitions: fs, hfs, level crossings

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

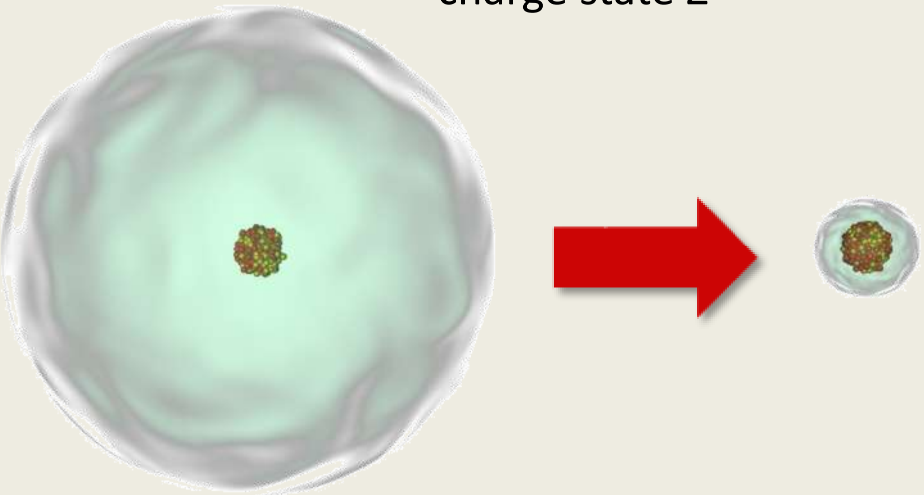
Optical level crossing transitions in HCI

Madelung ordering
(neutral)

Coulomb ordering
(H-like)

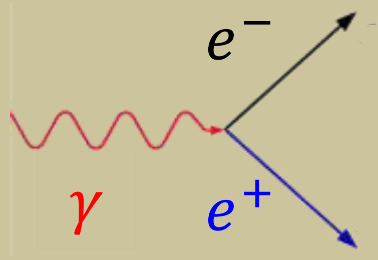


most interesting candidates for many tests of fundamental physics



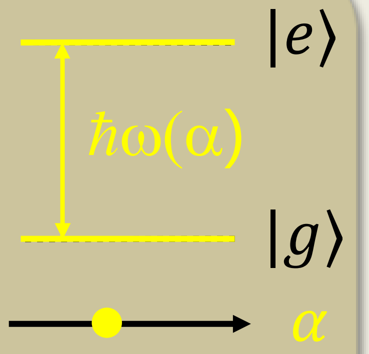
[Berengut *et al.* Phys. Rev. Lett. **105**, 120801 (2010)
Berengut *et al.* Phys. Rev. Lett. **106**, 210802 (2011)
Berengut *et al.* Phys. Rev. A **86**, 022517 (2012)]

Testing fundamental physics with HCI

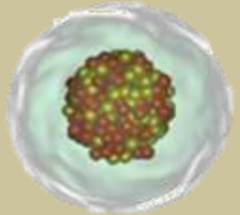


test of QED

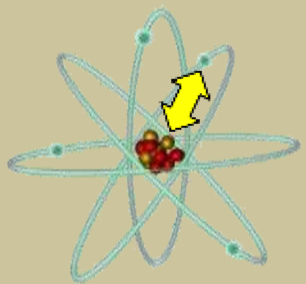
Lorentz invariance



Changing constants



Nuclear physics



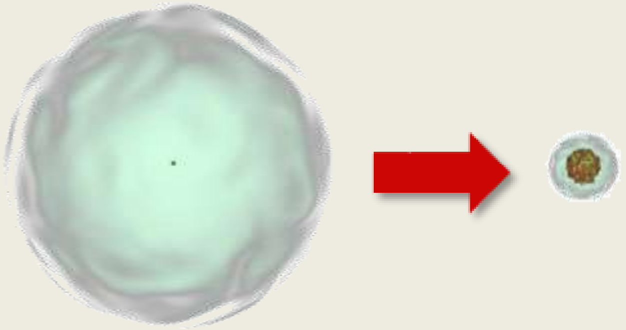
5th forces

Dark matter

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

Highly charged ions as optical clocks?

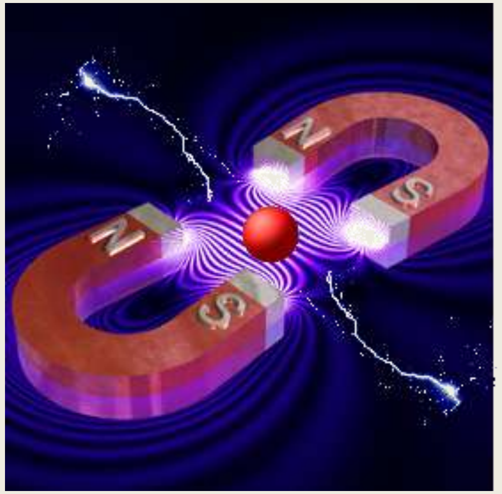
- **High accuracy**
  → low sensitivity to resonance shifts
- **HCI advantage: suppressed shifts**



Hydrogen-like HCI:

Linear Stark shift	Z^{-1}
Second order Stark shift	Z^{-4}
Linear Zeeman shift	Z^{0}
Second order Zeeman shift	$Z^{-3\ldots-4}$
Electric quadrupole shift	Z^{-2}

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

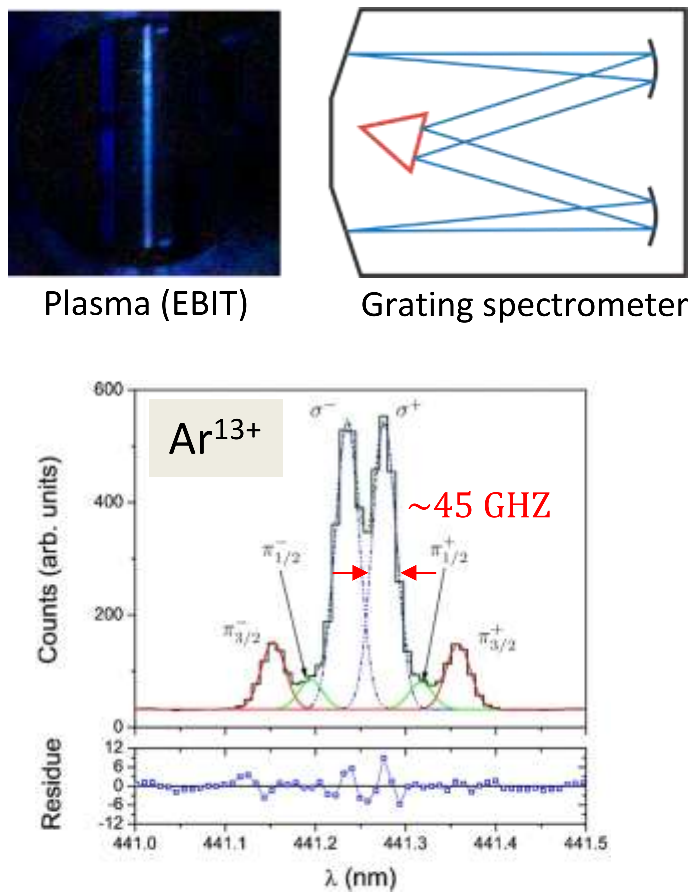


electric & magnetic fields

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)]

State-of-the-art HCI spectroscopy

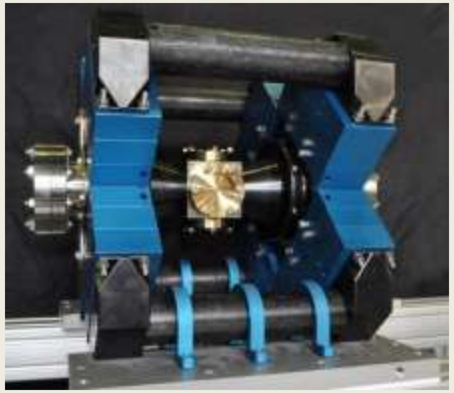


[Soria Orts et al., PRA **76**, 052501 (2007)]

Doppler-limited resolution of ~ 150 MHz

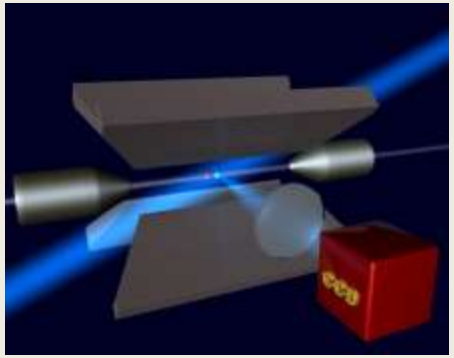
Problem:

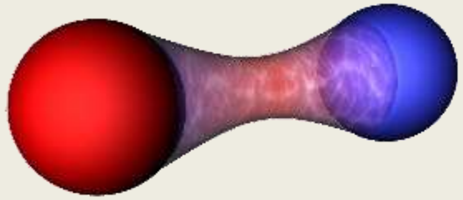
- Electron beam ion trap (EBIT) is a noisy environment
- No cycling transition for cooling & state detection



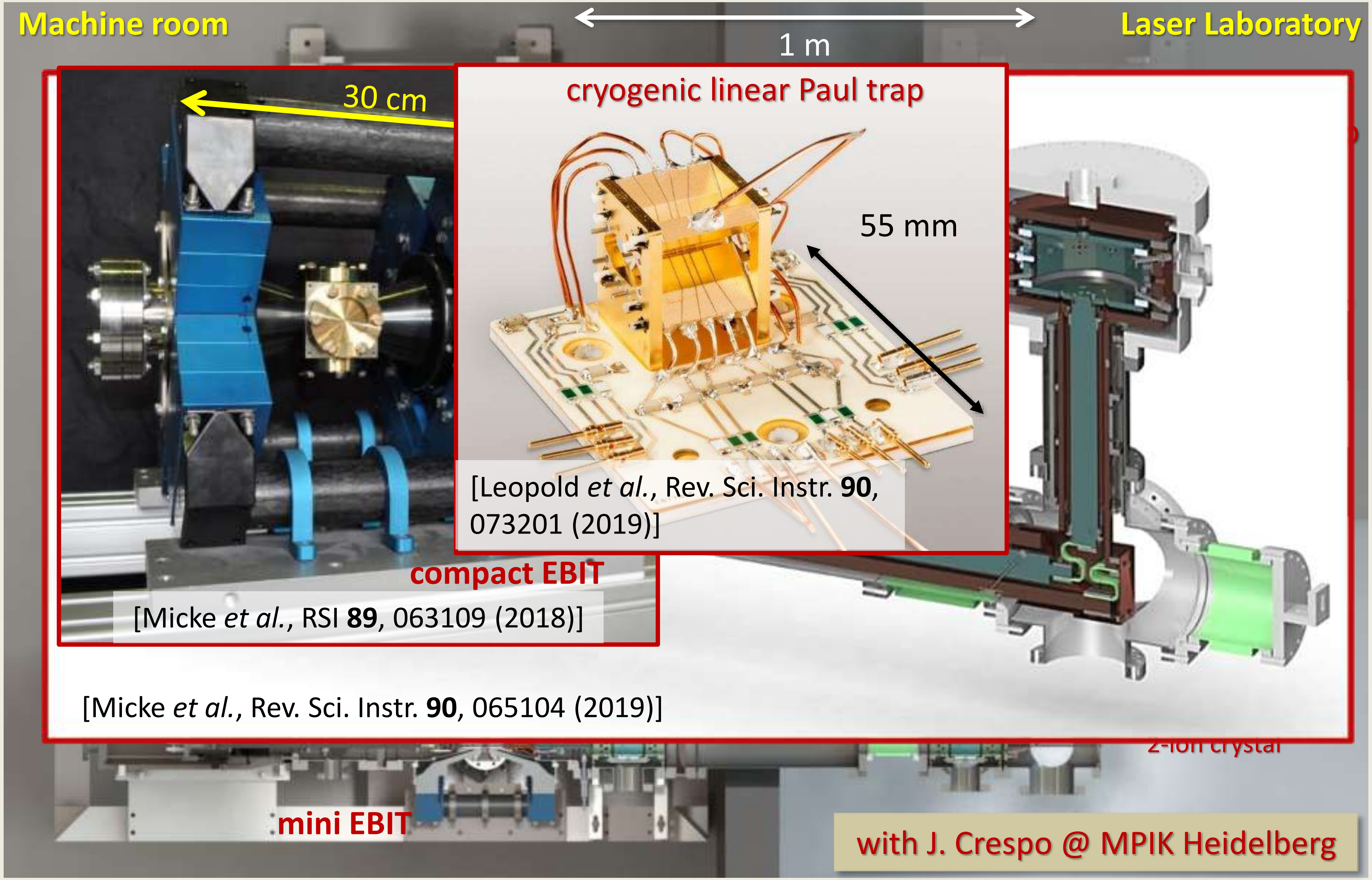
Solution:

- Paul trap environment
- cooling & detection
  ➔ Quantum Logic Spectroscopy





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

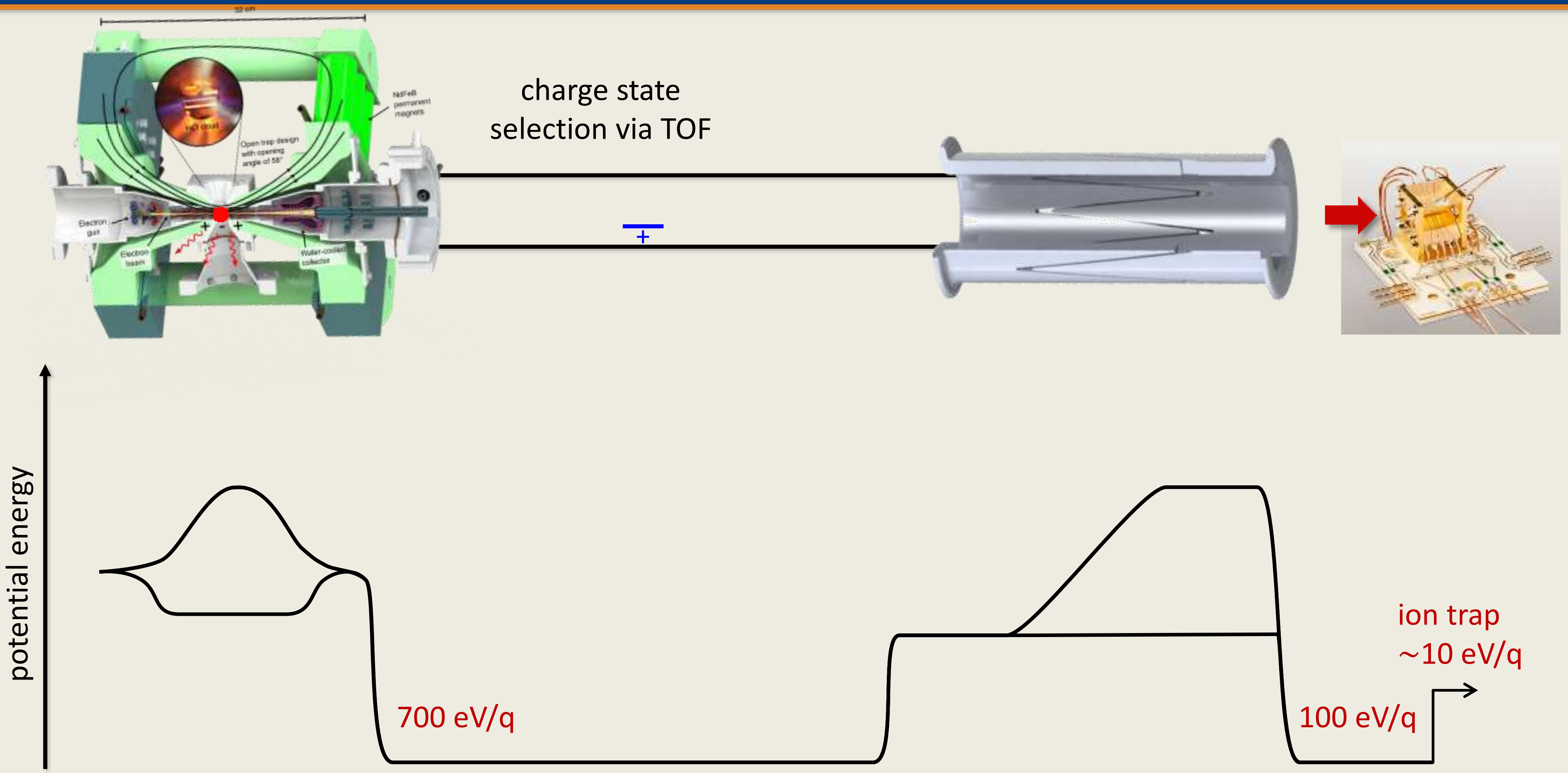
Specs EBIT:

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

Specs ion trap:

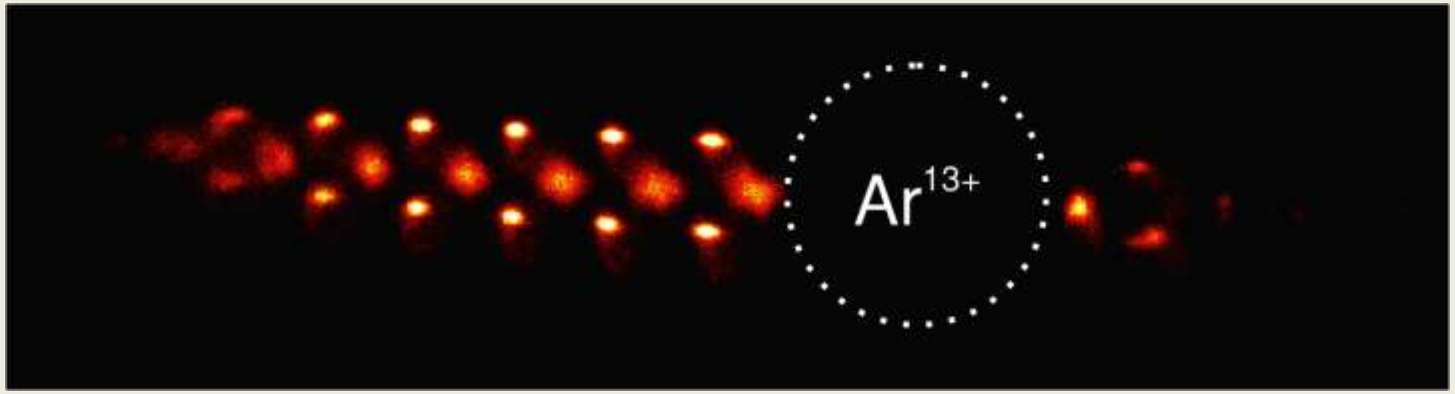
- 5 segments, Au-coated Al_2O_3, 0.7 mm ion-electrode distance
- Trapping frequencies: > 1 MHz
- Heating rates: ~ 1 1/s
- f/# ~ 1 imaging with bi-aspheric lens

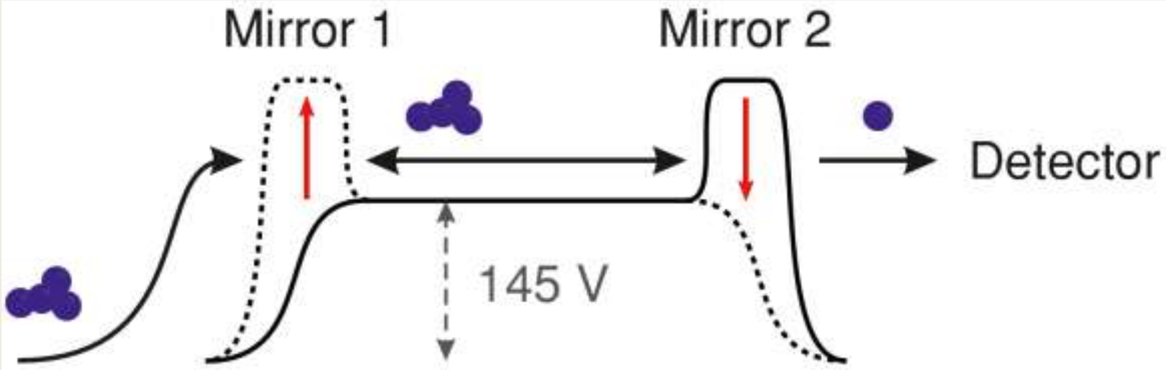
Slowing & Cooling



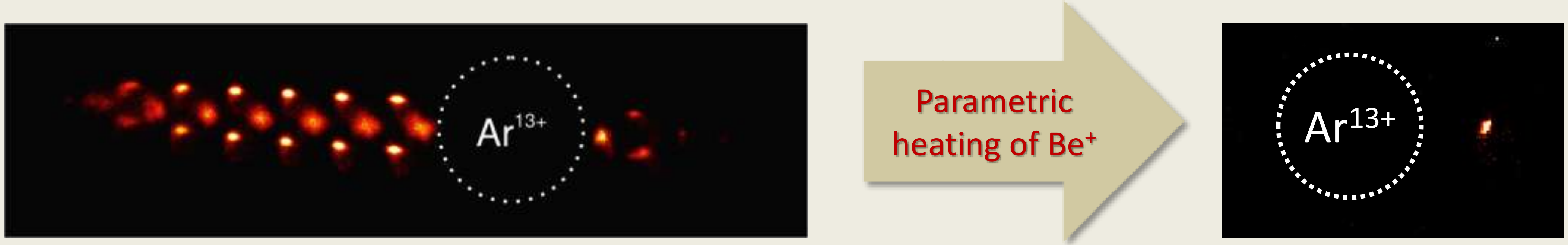
[Schmöger *et al.,* Rev. Sci. Instrum. **86**, 103111 (2015)]

Sympathetic Doppler cooling of a HCI



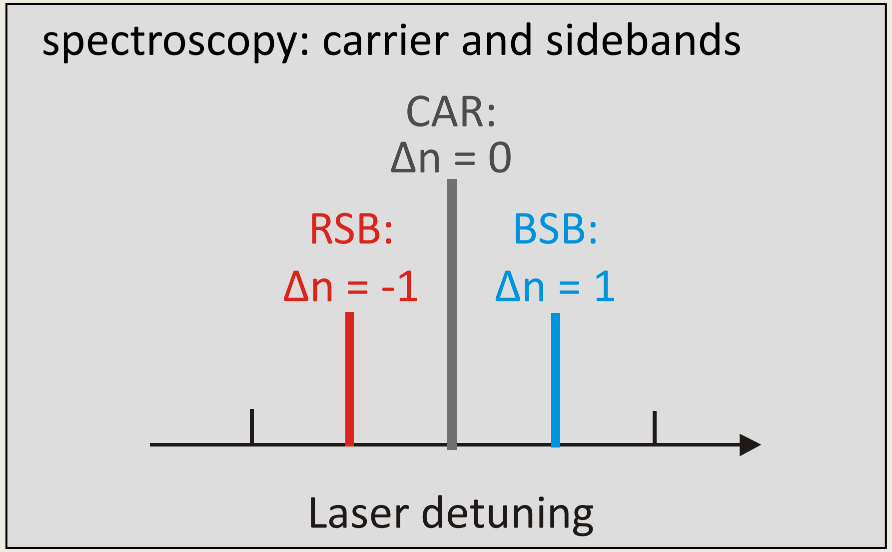


Preparation & Lifetime of a 2-Ion Crystal

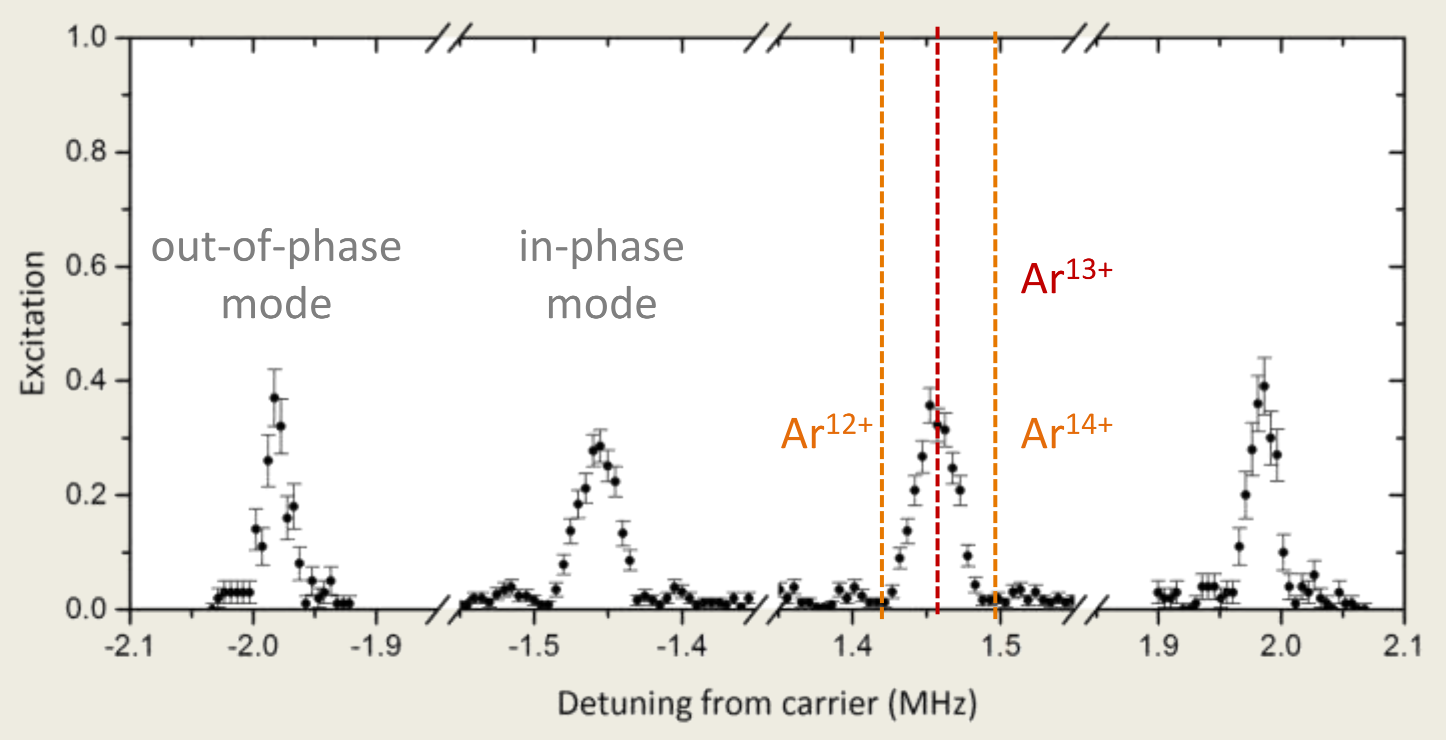


- total preparation time of Be^{+}/Ar^{13+} crystal: $\sim$ few min
- Ar^{13+} lifetime: $\tau \sim 100$ min
  ➔ residual pressure: $< 10^{-14}$ mbar
  (assuming Langevin collisions)
- Sideband cooling to the motional ground state ($T < 3$ μK)

Doppler cooling & charge state identification



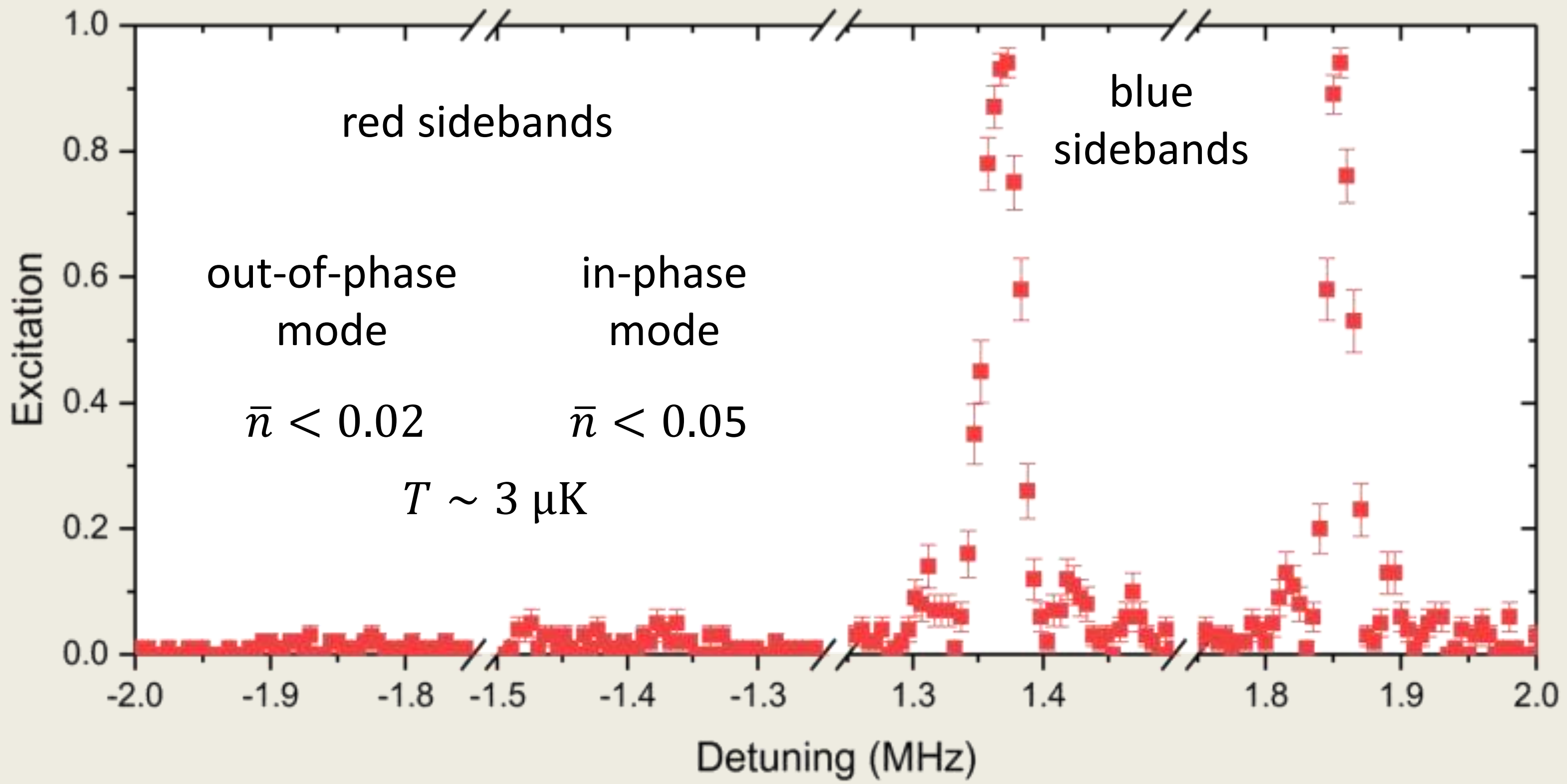
axial motional spectrum on Be^{+}

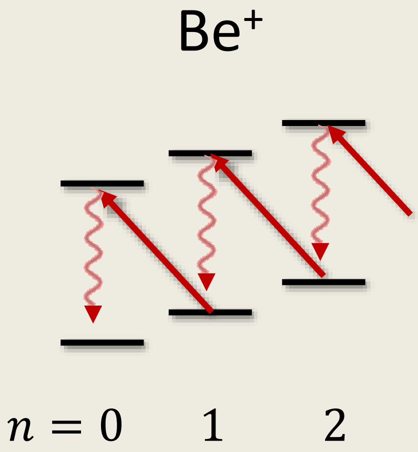


- single Be^{+} axial frequency: 0.995 MHz
  ➔ Be^{+}/Ar^{13+} axial frequencies:
  1.47 MHz and 1.99 MHz

Sympatetic ground state cooling of Ar^{13+}

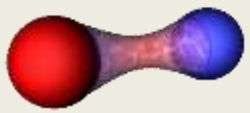
axial motional spectrum on Be^+



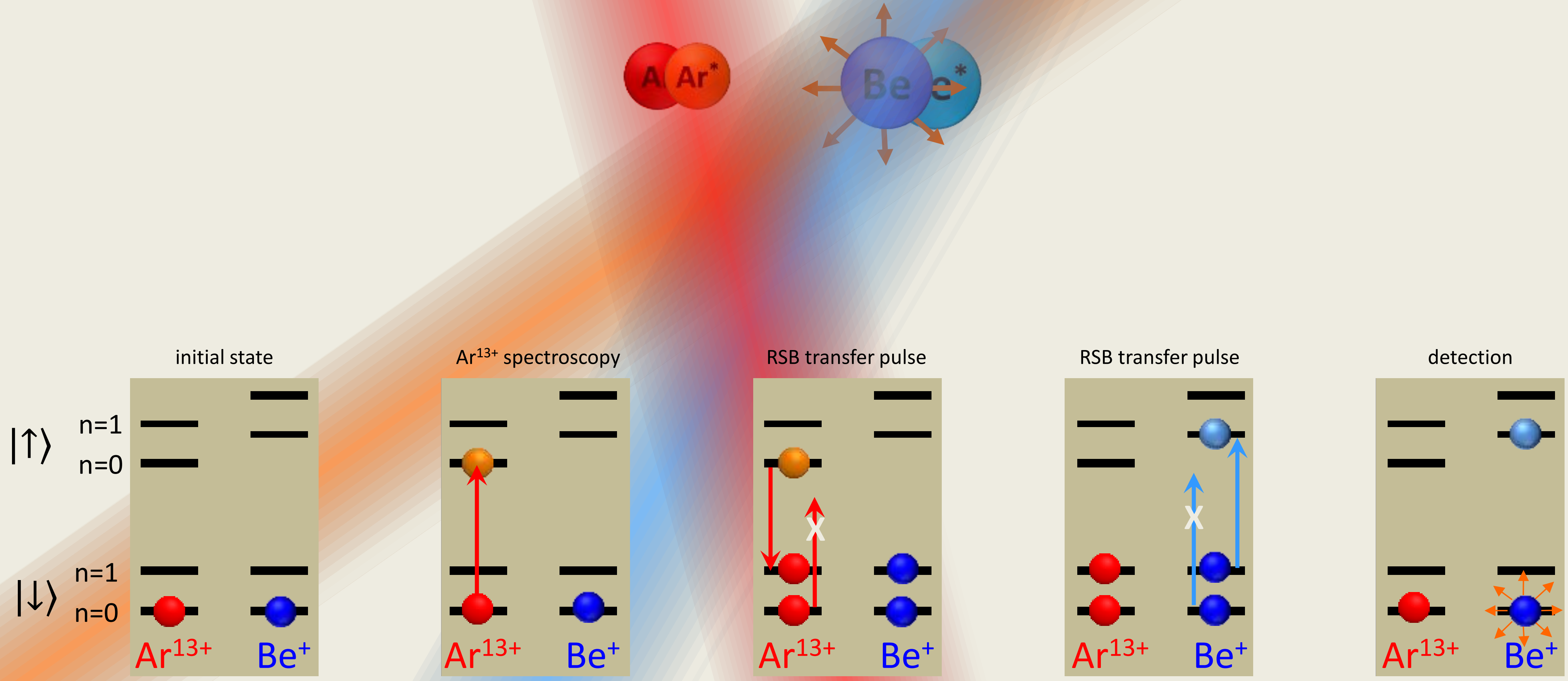


- resolved Raman sideband cooling on Be^+
- Lamb-Dicke parameter: $\eta_z = 0.82\sqrt{\mathrm{MHz}/\nu_z}$

[King *et al.*, in preparation]



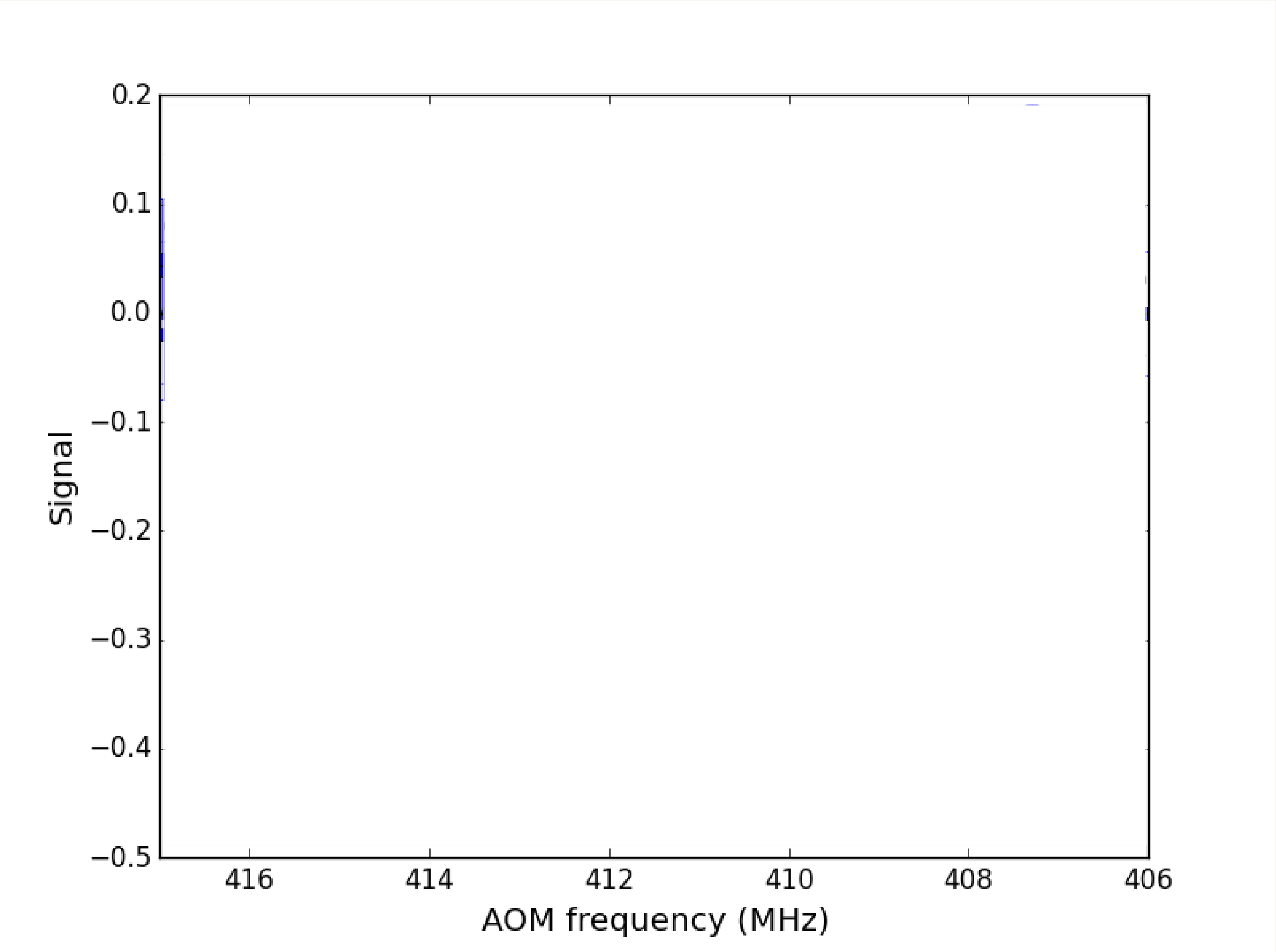
Quantum Logic State Transfer



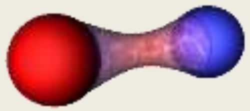
[D.J. Wineland *et. al.*, Proc. 6th Symposium on Frequency Standards and Metrology, 361 (2001); P.O. Schmidt *et al.*, *Science*, **309**, 749 (2005)]

First Ar^{13+} signal

19.9.2018

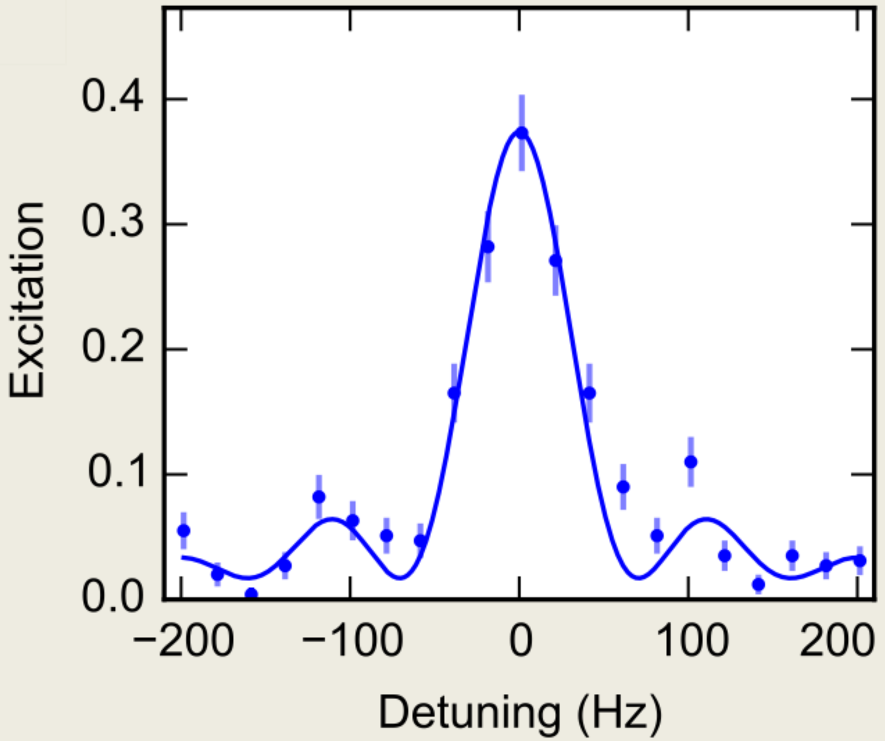


- blue sideband (BSB) heating signal on Ar^{13+} detected via red sideband (RSB) excitation on Be^{+}

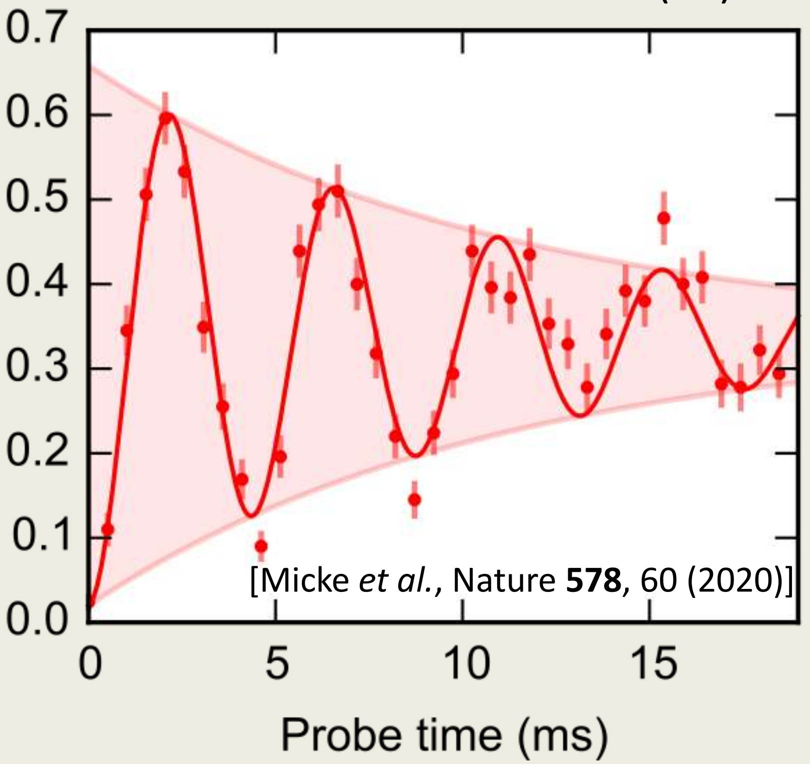


Quantum Logic Spectroscopy of Ar^{13+}

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

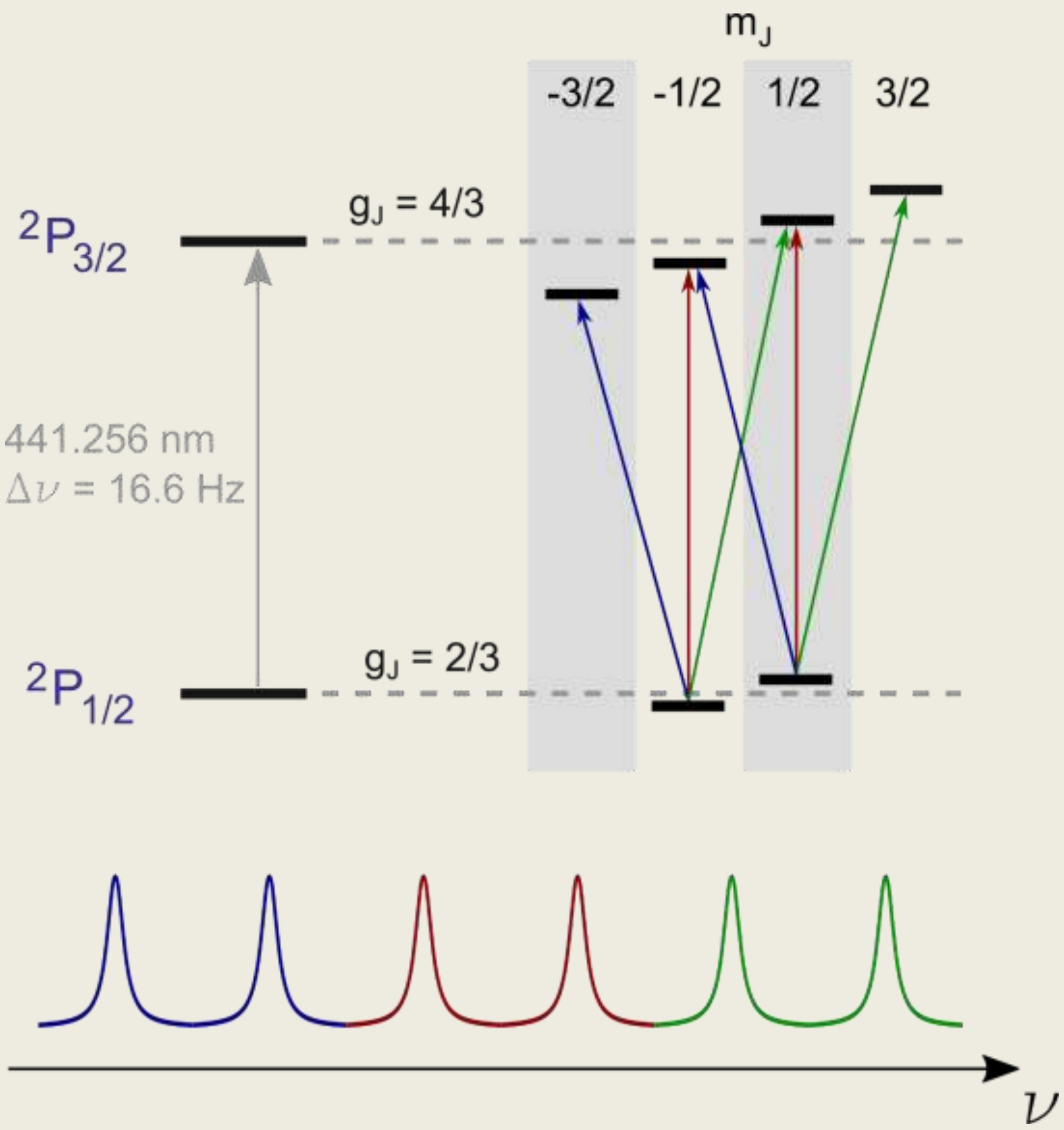


dephasing dominated by
excited state lifetime of 9.97(26)ms

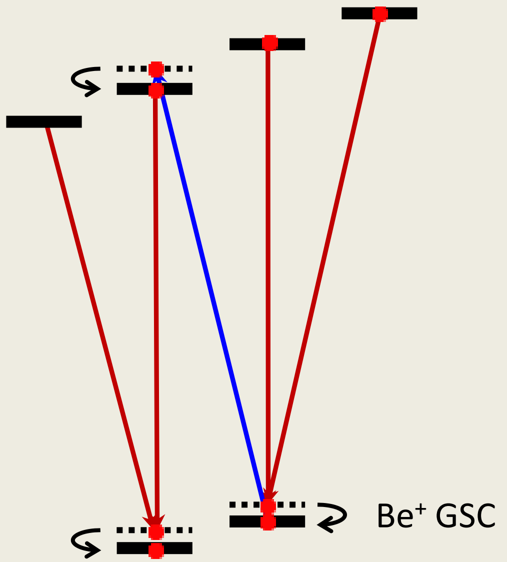


- spectroscopy laser transfer locked of Ar^{13+} to Si cavity-stabilized laser
  [Sterr & Benkler @ PTB: D. G. Matei *et al.*, Phys. Rev. Lett. **118**, 263202 (2017)]

Ar^{13+} levels, spectrum & preparation



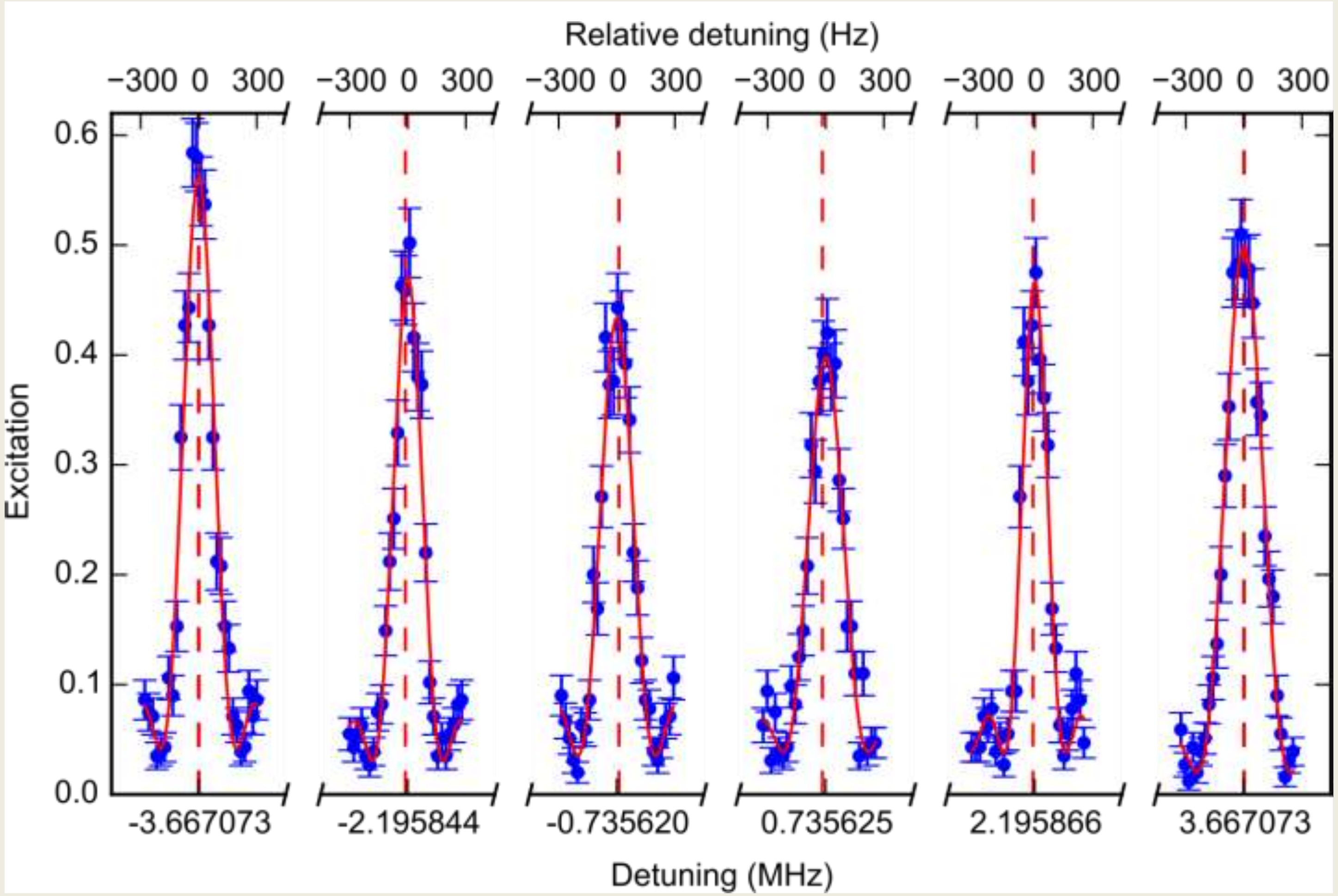
QL state preparation sequence

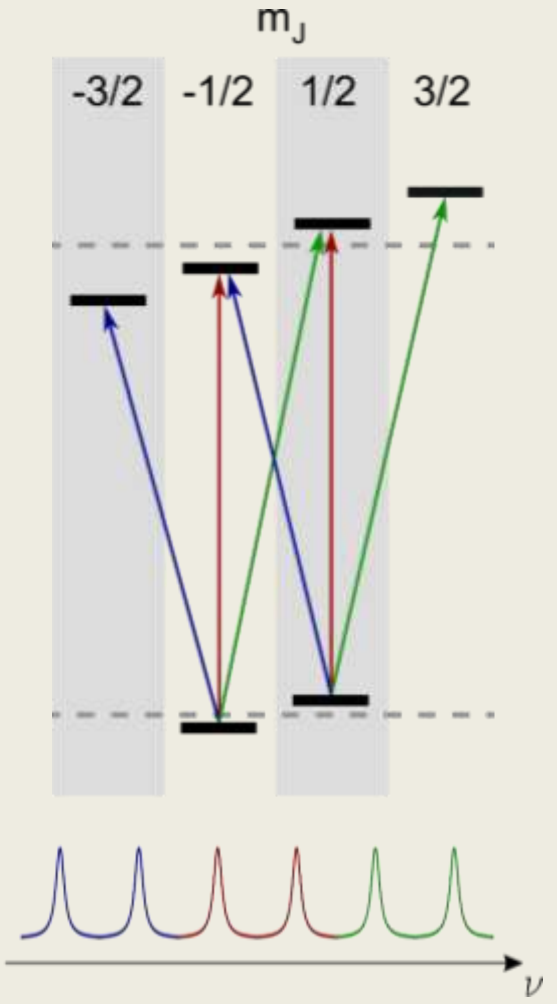


- Coherent manipulation on HCI
- Dissipation via ground state cooling on Be^+

Ar^{13+} Zeeman structure

- - - - - Dirac g-factors / Landé + interactions + QED





[Micke *et al.*, Nature **578**, 60 (2020)]

g-factors: [Agababaev *et al.* X-Ray Spectrom. 1-6 (2019)]

➔ measurement of ground- and **excited** state g-factors with <10 ppm

Systematic shifts for Ar^{13+}

Shift source	Mitigation	Shift (10^{-18})	Uncertainty (10^{-18})
Micromotion	Real-time measurement	-443	22
AC Zeeman shift	Calibration at much higher powers and extrapolation	0	2
First-order Doppler	Counter-propagating beams	0	< 1
Electric quadrupole	Small coefficient, averaging over multiple Zeeman components	0	< 1
Linear Zeeman	Averaging over multiple Zeeman components	0	< 1
Quadratic Zeeman	Small coefficient, small field	< 1	$\ll 1$
2nd order Doppler	Algorithmic cooling [King *et al.*, PRX **11**, 041049 (2021)]	-1	< 1

} no fundamental limitations

$^{40}Ar^{13+}$ clock with 2.2×10^{-17} estimated systematic uncertainty

8 orders of magnitude improvement

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

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

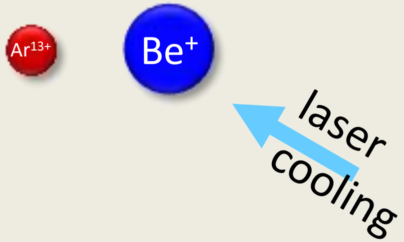
Cooling challenges....

Large q/m mismatch between ions

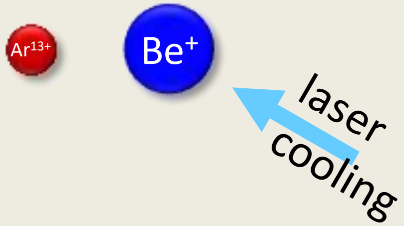
→ large difference in radial amplitudes

→ inefficient cooling of radial modes

- radial mode x_1:
  → strong cooling

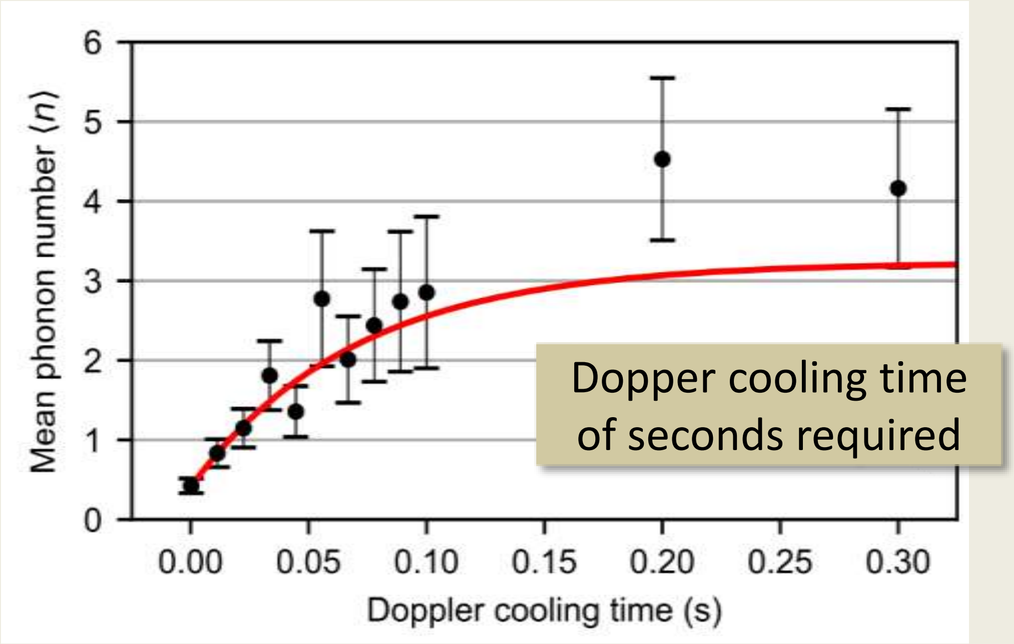


- radial mode x_2:
  → weak cooling



[Kozlov *et al.*, Rev. Mod. Phy. **90**, 045005 (2018)]

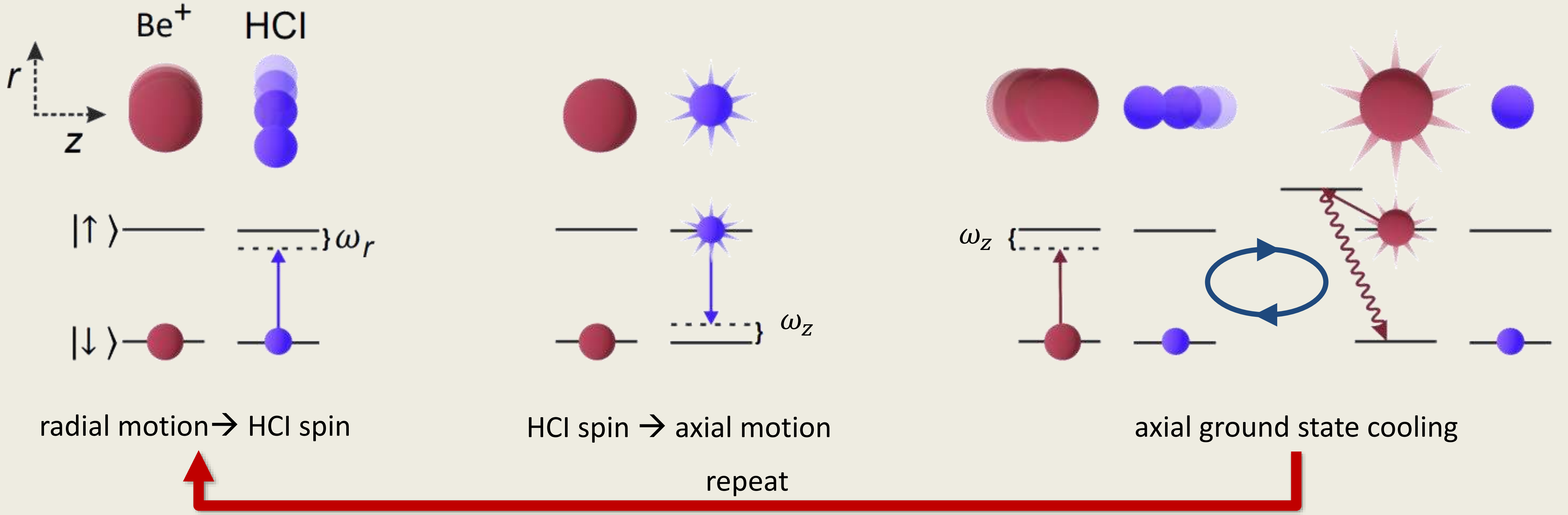
Doppler „heating“



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

Ground-state cooling using quantum logic

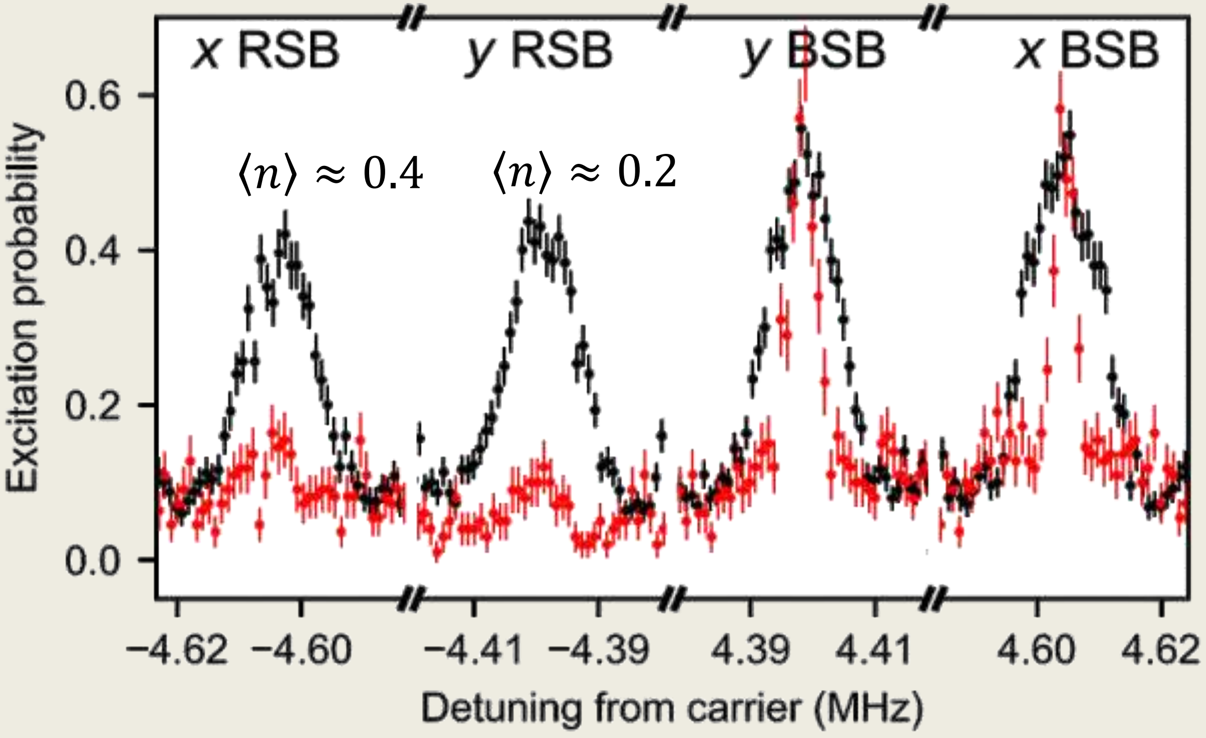
Coherently transfer radial mode phonons to efficiently-cooled axial modes
„Algorithmic cooling“

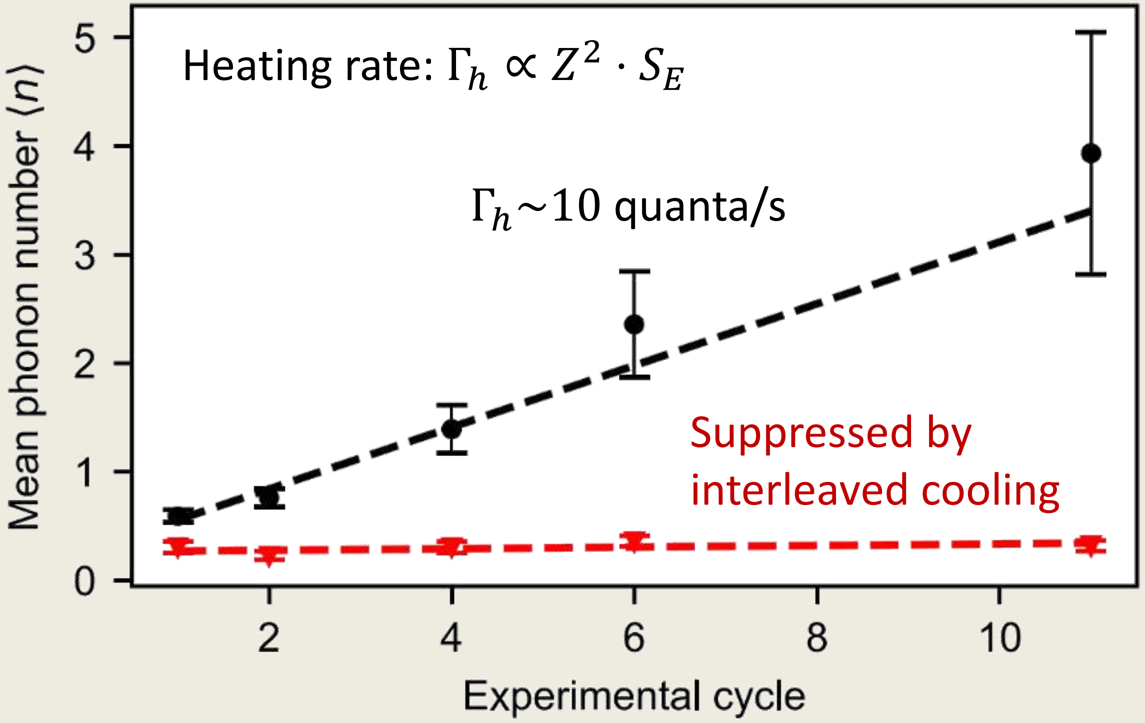


Generic scheme applicable to other systems:
large molecules, nanoparticles, …

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

Algorithmic cooling: results





Coldest HCI of all time!
T < 200 µK in all modes

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

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

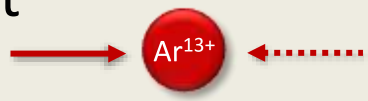
Clock operation

Stabilisation to 4 Zeeman components

- eliminate linear Zeeman shift
- eliminate quadrupole shift
- measure quadrupole moment & g-factor

Interrogation from two possible directions

- eliminate linear Doppler shift

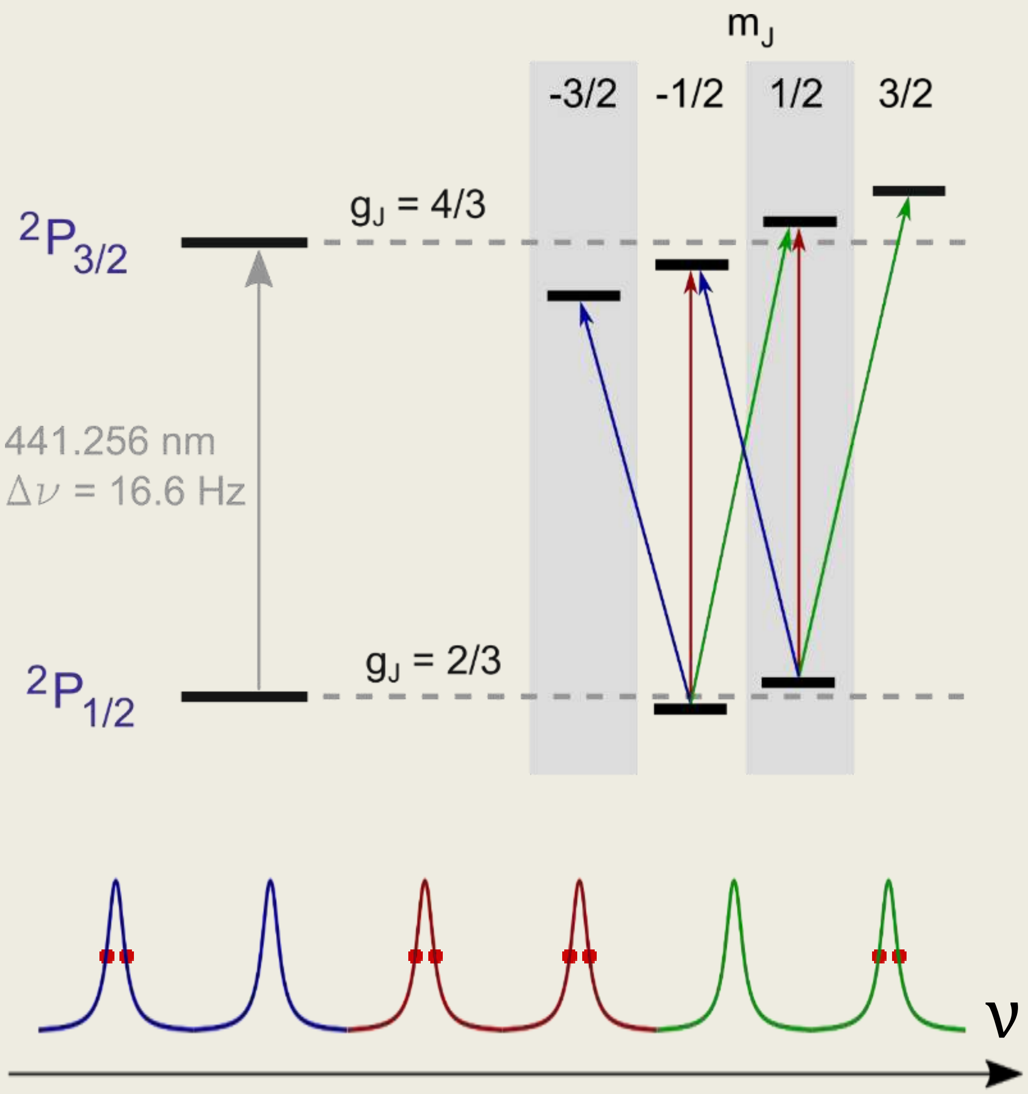


Pseudorandomisation of component & direction

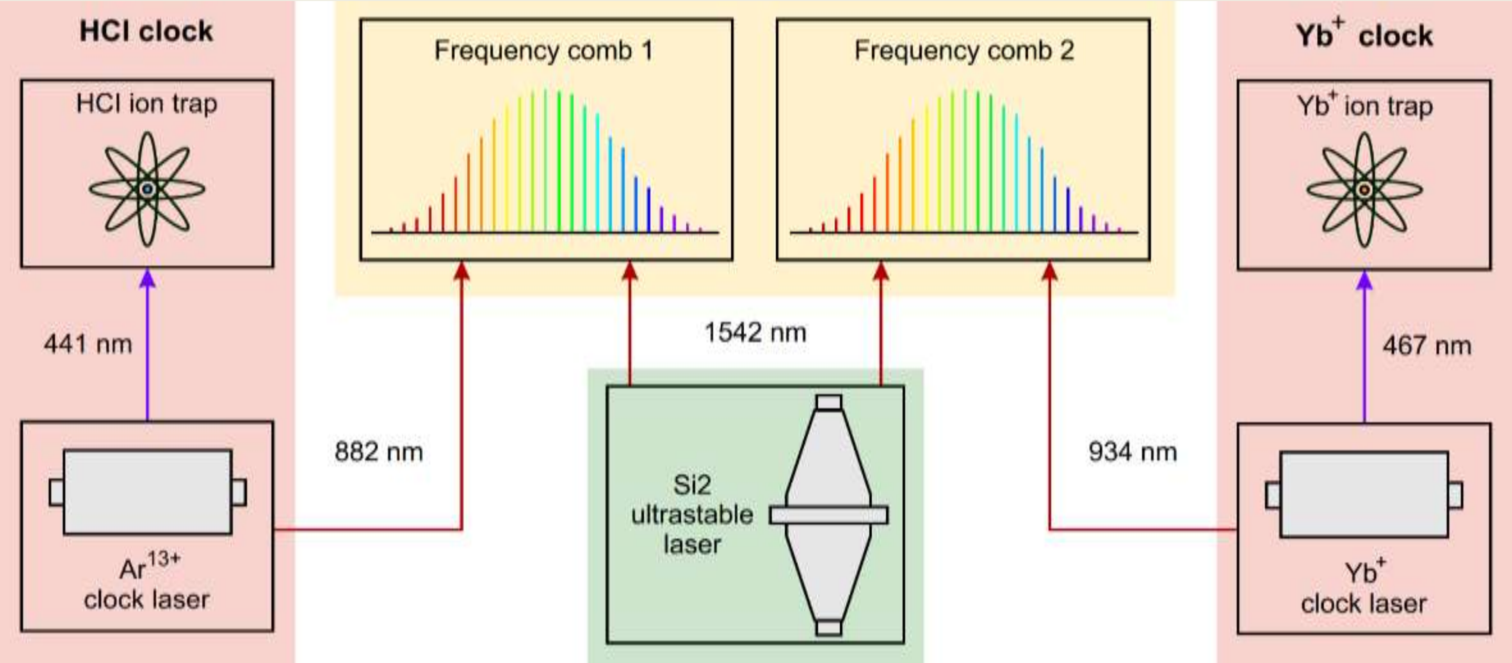
- sensitivity to drifts suppressed

Interleaved measurements:

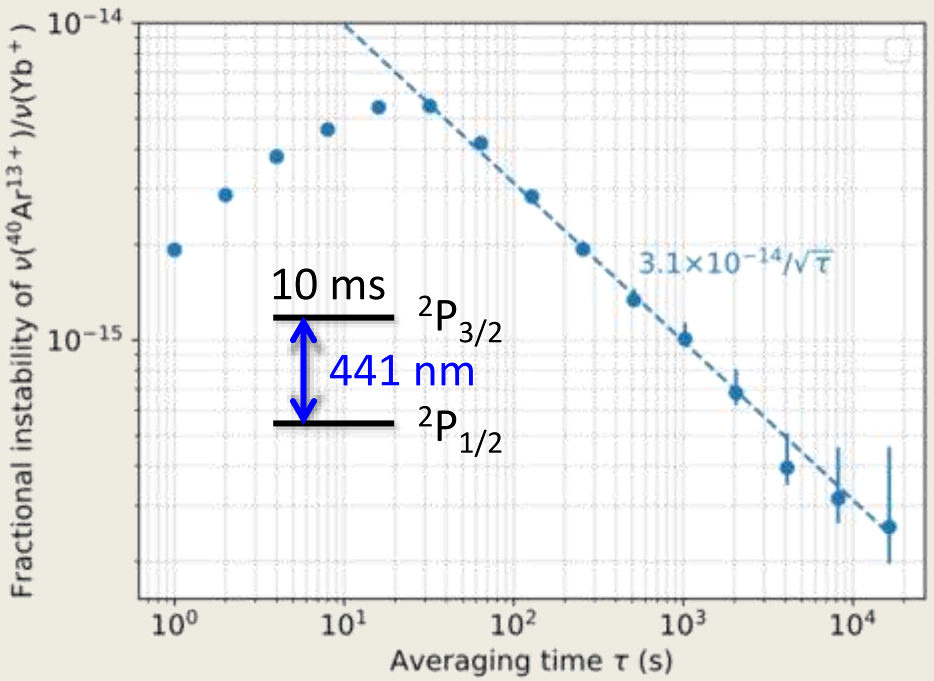
- Micromotion
- Secular temperature



Frequency ratio measurement Ar^{13+}/Yb^{+} E3



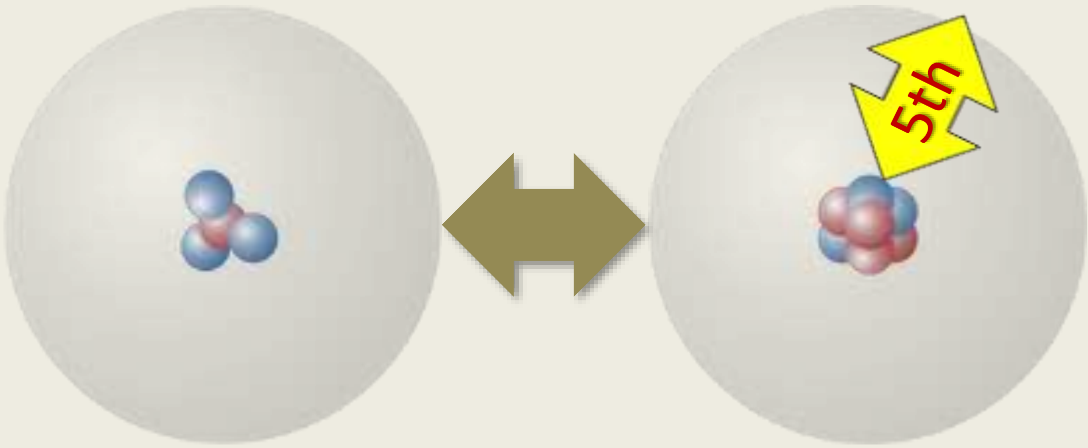
Comparison: E. Benkler, Yb^{+} E3 clock: R. Lange, N. Huntemann

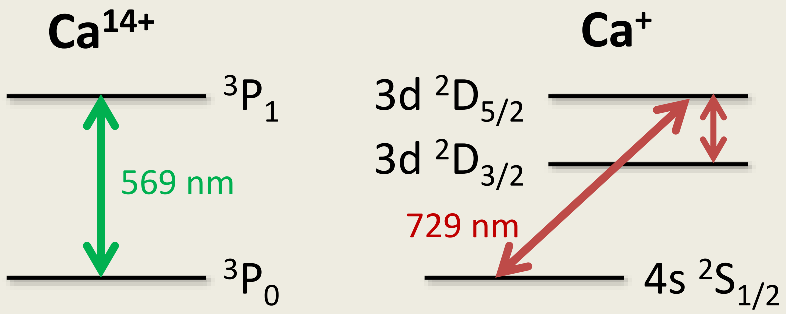


R = 1.05776938758748093(11)

- Frequency ratio uncertainty limited by Ar^{13+} excited state lifetime to $\sim 3 \times 10^{-14}/\sqrt{\tau}$
- Measurements to $\sim 1 \times 10^{-16}$ statistical uncertainty for $^{40}Ar^{13+}$ and $^{36}Ar^{13+}$
- Yb^{+} E3 absolute frequency known with 1.3×10^{-16} fractional uncertainty

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



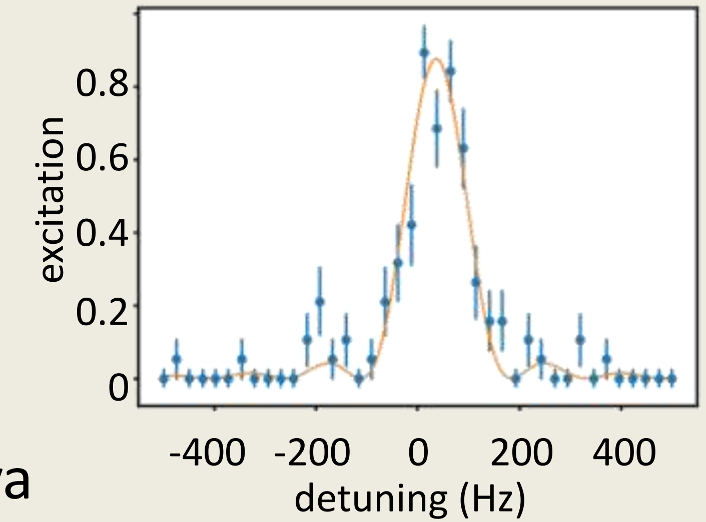


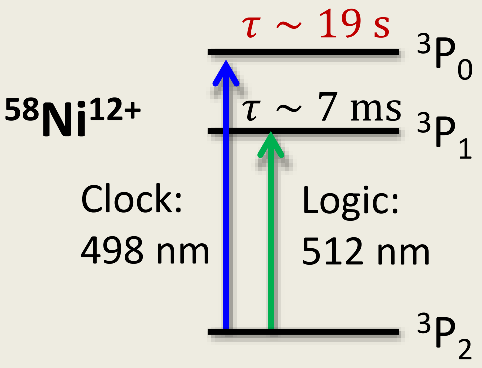
→ See Elina's talk

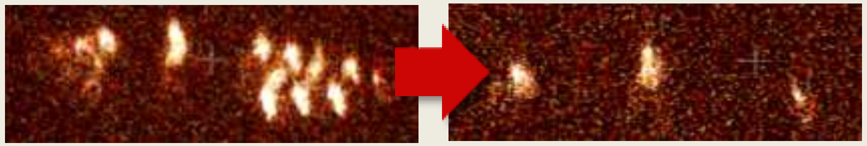
ISOTOPE SHIFT SPECTROSCOPY & SEARCH FOR 5TH FORCES/DARK MATTER

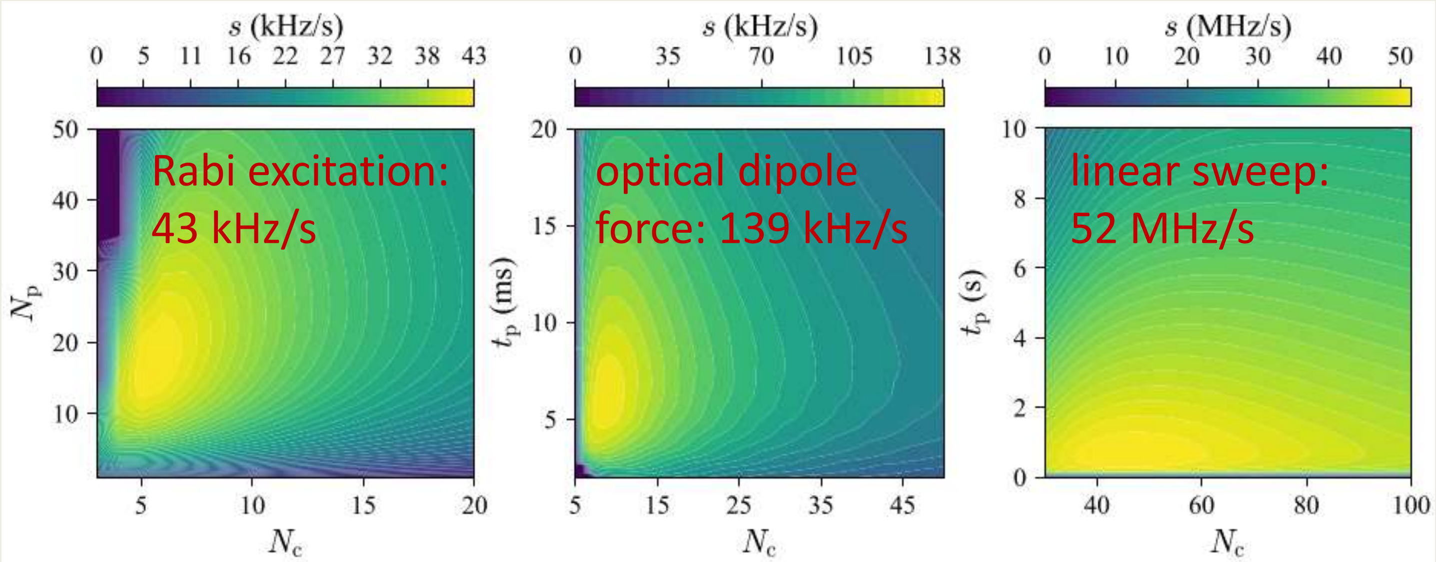
Current work

- Clock candidate: $^{58}Ni^{12+}$
  [Yu & Sahoo, Phys. Rev. A **97**, 041403 (2018);
  Chen *et al.*, Phys. Rev. Res. **6**, 013030 (2024)]
- observed logic transition at 512 nm
- Challenge: clock transition energy uncertainty of several THz (few nm)
  → improved calculations by M. Safronova
  → efficient search strategies
  [Chen *et al.*, Phys. Rev. Appl. **22**, 054059 (2024);
  Cheung *et al.*, arXiv:2502.05386]
- future prospects: multi-ion HCI
  [Zawierucha *et al.*, PRA **110**, 013107 (2024);
  Pelzer *et al.*, PRL **133**, 033203 (2024)]







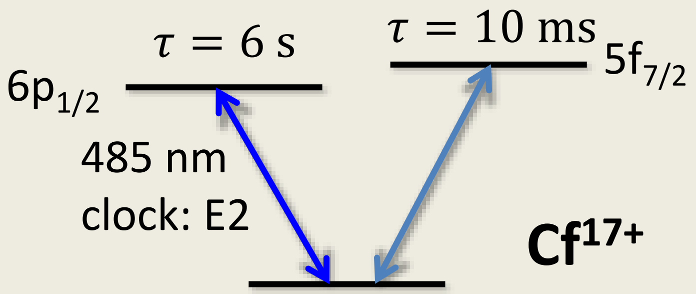


Outlook

Future: further tests of fundamental physics

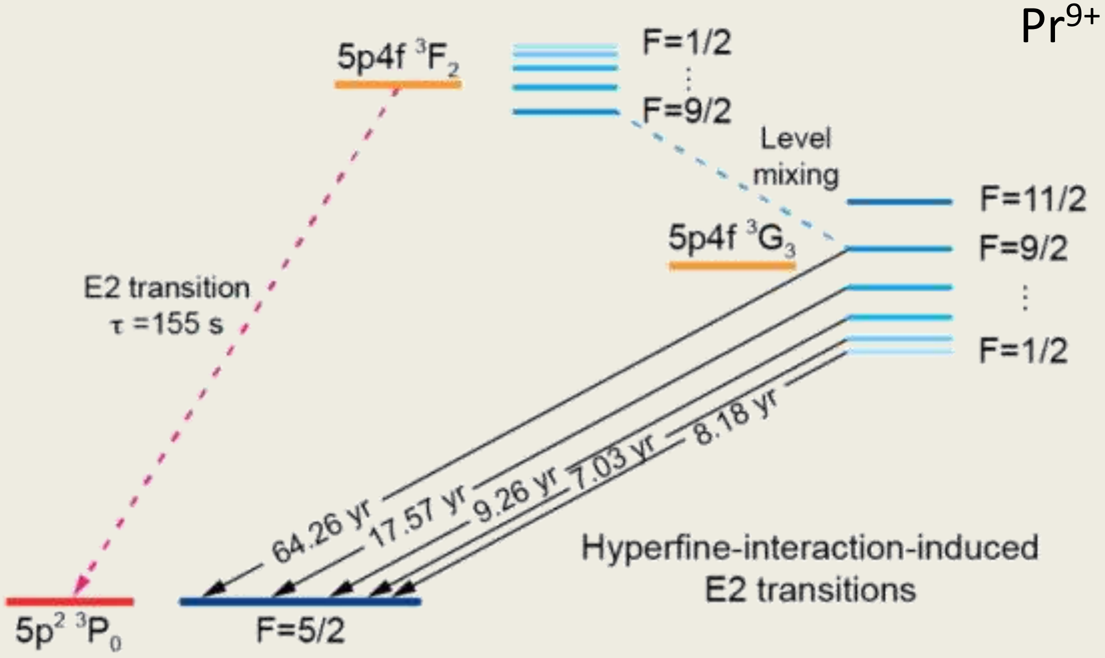
- dark matter & α-sensitive level-crossings:
  - Pr^{9+} [Bekker *et al.*, Nat. Commun. **10**, 5651 (2019)]
  - Ir^{17+} [Windberger *et al.*, PRL **114**, 150801 (2015)]
  - Cf^{15+} & Cf^{17+} [Porsev *et al.,* PRA **102**, 012802 (2020)]

    also: V. Schäfer (MPIK), G. Barontini (Birmingham)



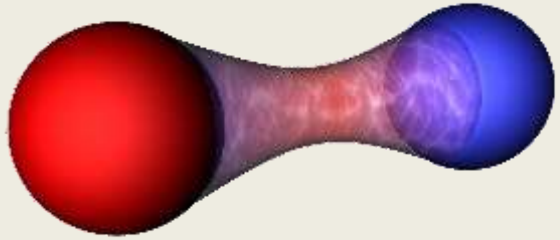
Other ideas

- Ba^{4+}, Pr^{10+}: S. Brewer (Colorado State)
- XUV clocks: J. Crespo (MPIK)
- few-electron HCI: P. Micke (GSI/Jena)

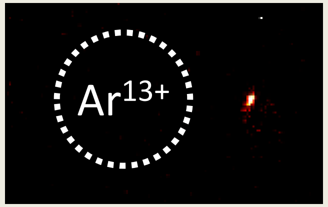


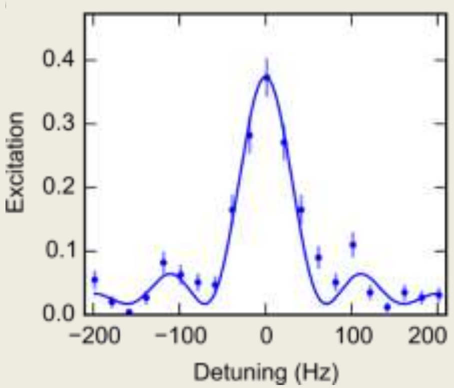
Emerging field with many new ideas & applications!

Summary



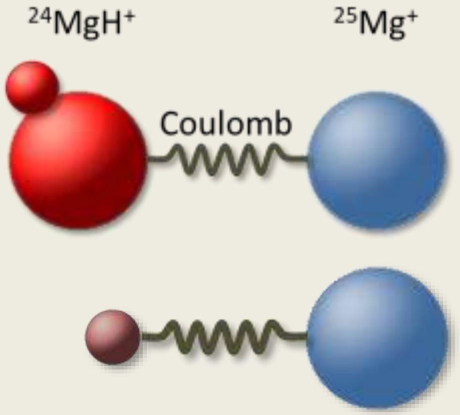
- Quantum logic spectroscopy is a powerful & versatile technique
  - full coherent control over internal & external degrees of freedom
  - dissipation for cooling & state-preparation on logic ion
  - enables readout and manipulation of „complicated“ spectroscopy species
- Al^+ clock with 5.5×10^{-19} systematic uncertainty
- Highly charged ions





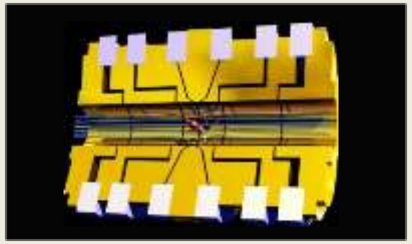
- Molecular ions

  [Wolf *et al.*, Nature **530**, 457 (2016)
  Chou *et al.*, Nature **545**, 203 (2017)
  Sinhal *et al.*, Science **367**, 1213 (2020)
  Chou *et al.*, Science **367**, 1458 (2020)]



- (Anti-)Protons are next

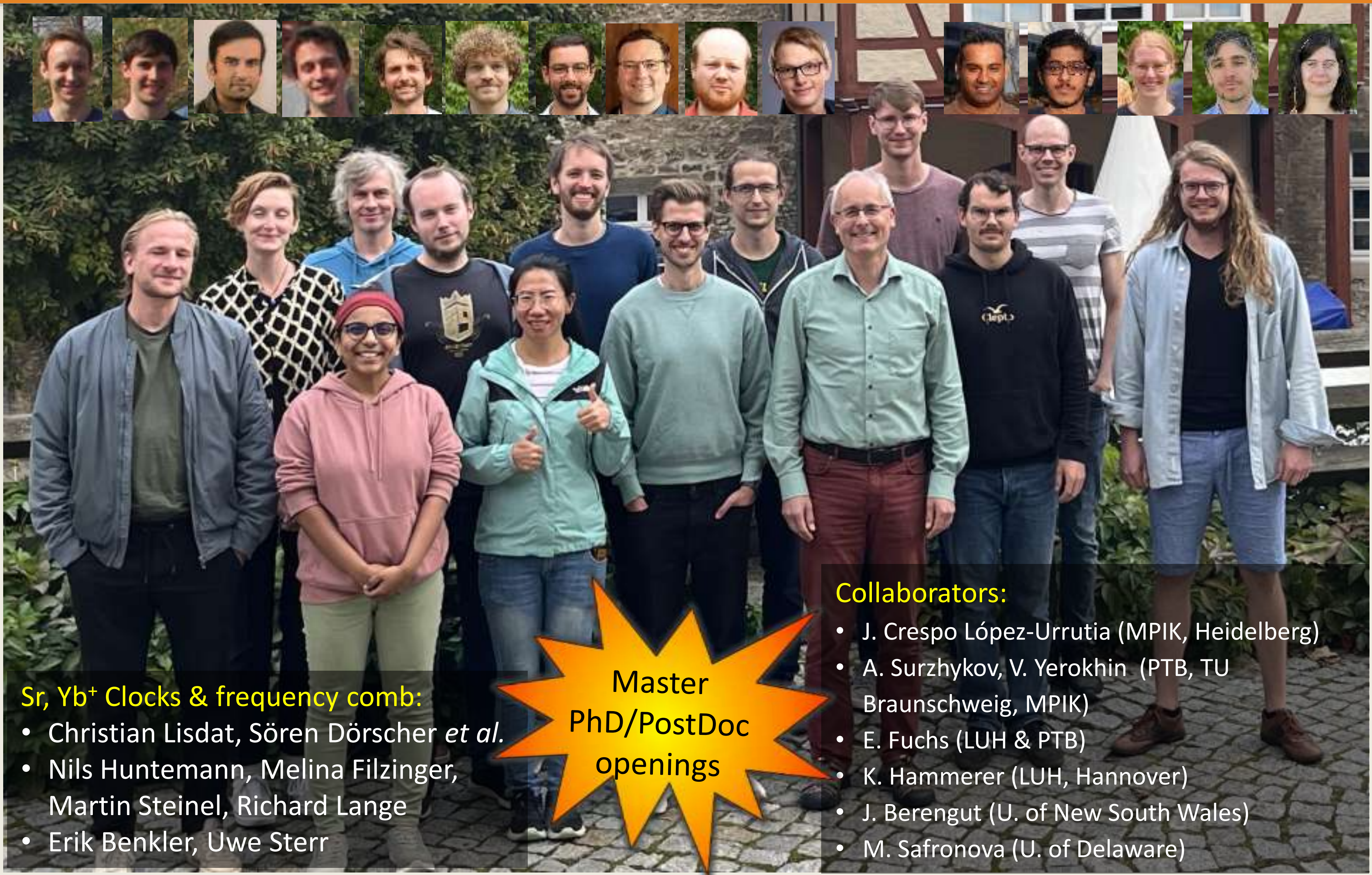
  [Nitzschke *et al.*, Adv. Quant. Techn. **3**, 1900133 (2020)]



Bright future for quantum logic spectroscopy & applications

Quantum Logic Spectroscopy Group

€



Sr, Yb⁺ Clocks & frequency comb:

- Christian Lisdat, Sören Dörscher *et al.*
- Nils Huntemann, Melina Filzinger, Martin Steinel, Richard Lange
- Erik Benkler, Uwe Sterr

Master PhD/PostDoc openings

Collaborators:

- J. Crespo López-Urrutia (MPIK, Heidelberg)
- A. Surzhykov, V. Yerokhin (PTB, TU Braunschweig, MPIK)
- E. Fuchs (LUH & PTB)
- K. Hammerer (LUH, Hannover)
- J. Berengut (U. of New South Wales)
- M. Safronova (U. of Delaware)

ERC Adv. Grant „FunClocks"

Quantum Valley Lower Saxony

EMPIR

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