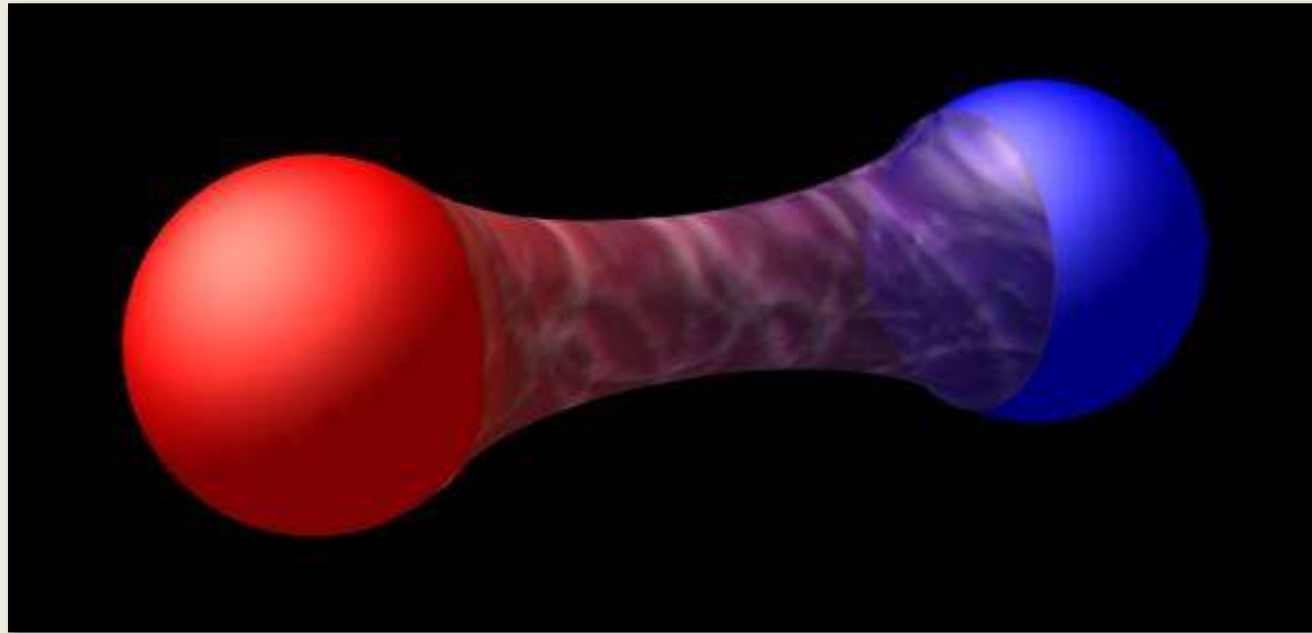


Quantum Logic Spectroscopy of Trapped Ions



P. O. Schmidt

QUEST Institute for Experimental Quantum Metrology
PTB Braunschweig and Leibniz Universität Hannover

Quantum Science Seminar, 17.09.2020

Physikalisch-Technische Bundesanstalt



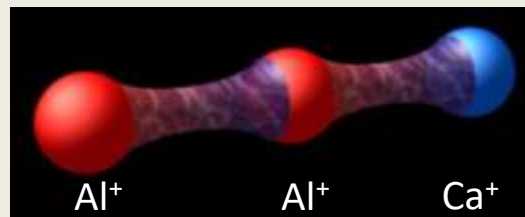
Hermann v. Helmholtz



Location Braunschweig: 1 km², approx. 1500 employees

- National Metrology Institute, founded 1887
- **Tasks:** determination of fundamental constants, dissemination of SI units, development of measurement techniques,...
- ca. **1800 employees**, of which are **>200 PhD candidates**
- 60% research: >600 publications per year

Quantum Logic Spectroscopy Group



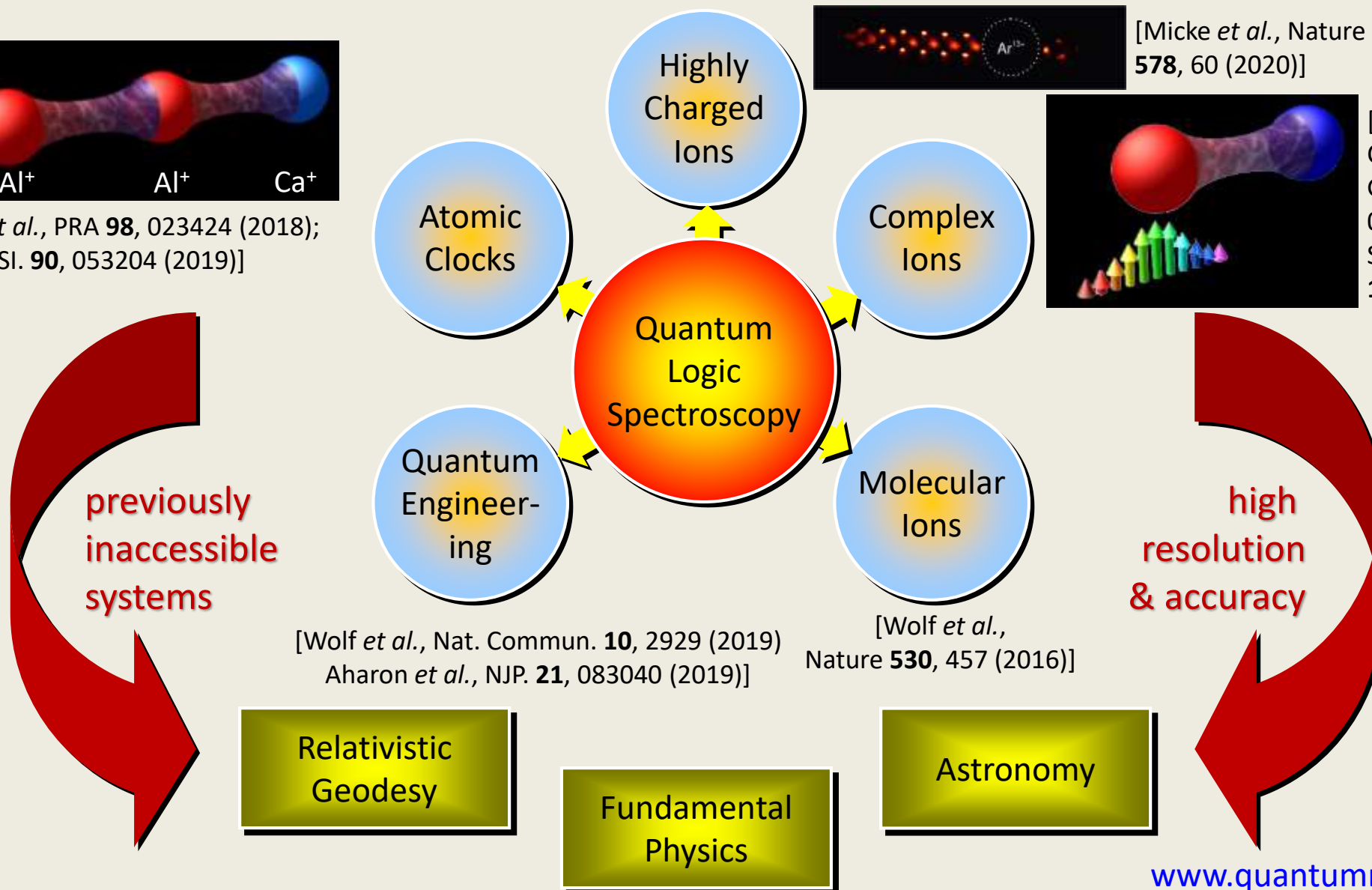
[Scharnhorst *et al.*, PRA **98**, 023424 (2018);
Hannig *et al.*, RSI. **90**, 053204 (2019)]



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



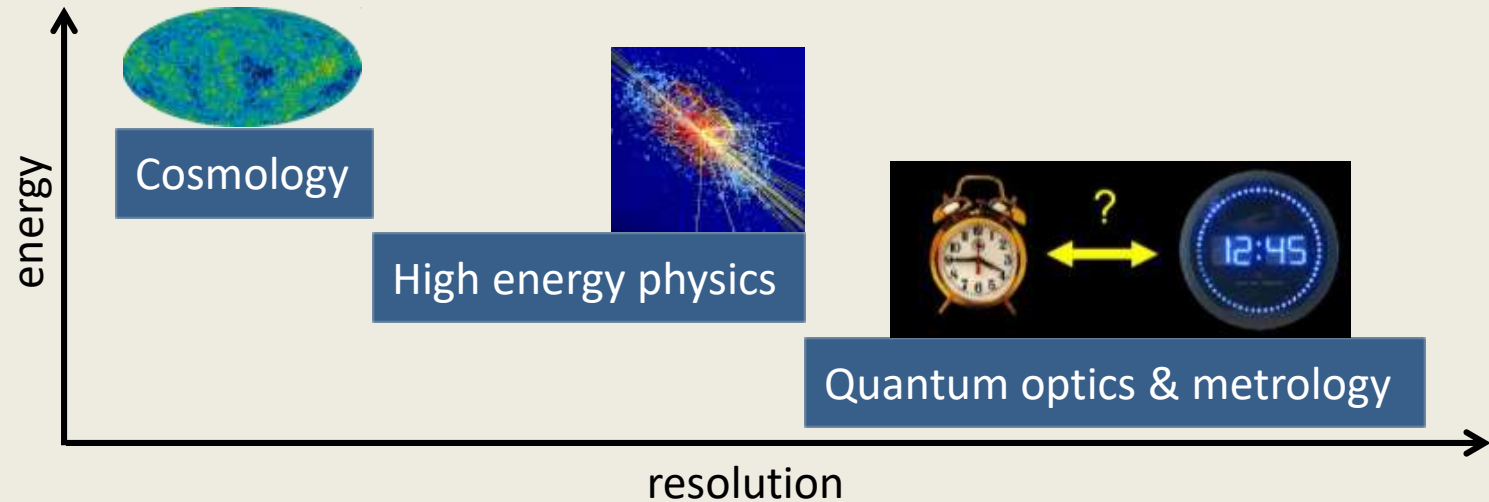
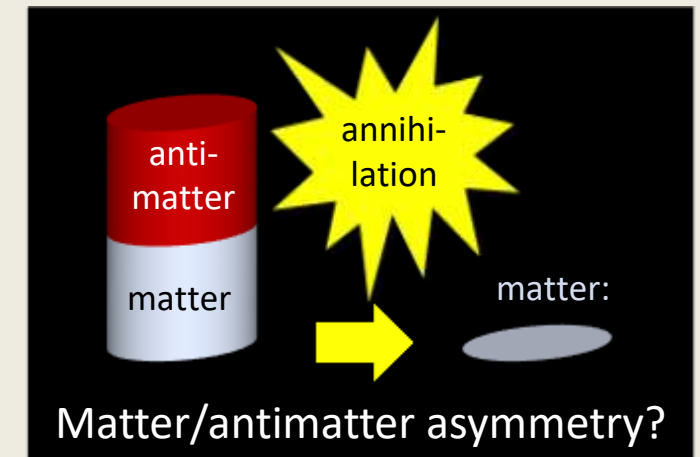
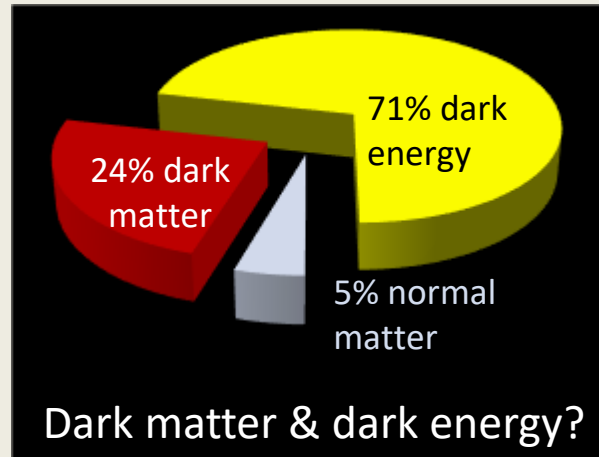
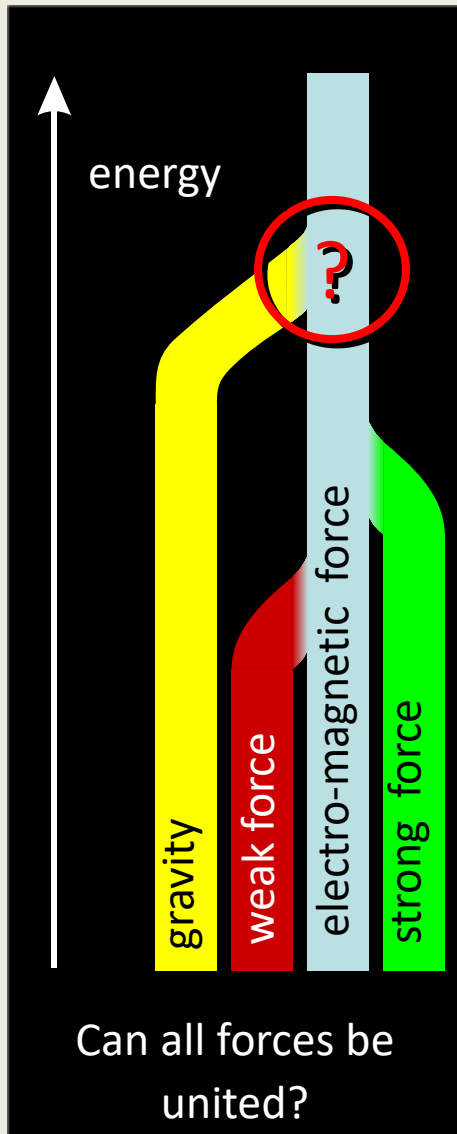
[Wan *et al.*, Nat. Commun. **5**, 4096 (2014);
Gebert *et al.*, PRL **115**, 053003 (2015);
Shi *et al.*, Appl. Phys. B **123**, 2 (2017)]



[Wolf *et al.*, Nat. Commun. **10**, 2929 (2019)
Aharon *et al.*, NJP. **21**, 083040 (2019)]

[Wolf *et al.*,
Nature **530**, 457 (2016)]

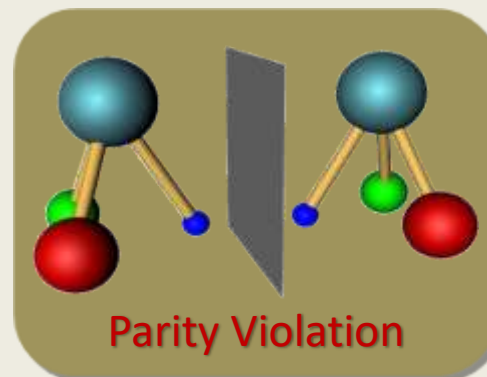
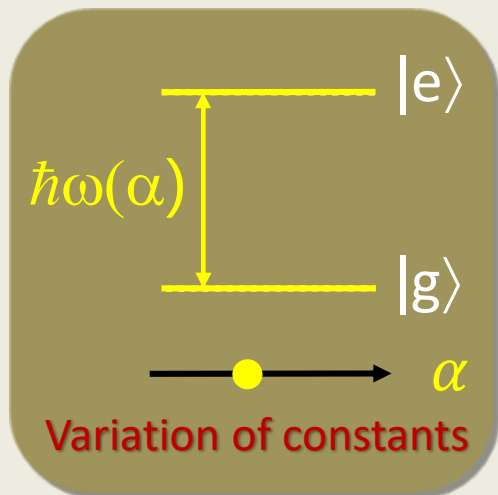
Open questions in physics



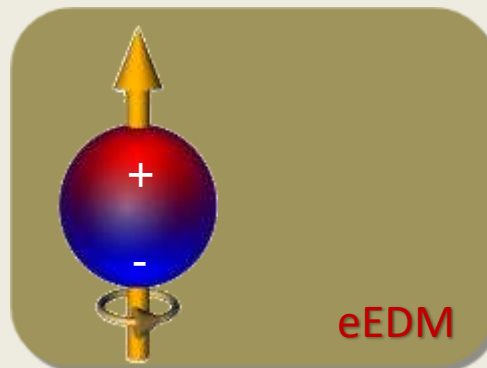
→ low energy, high resolution

atomic & molecular systems
are sensitive probes

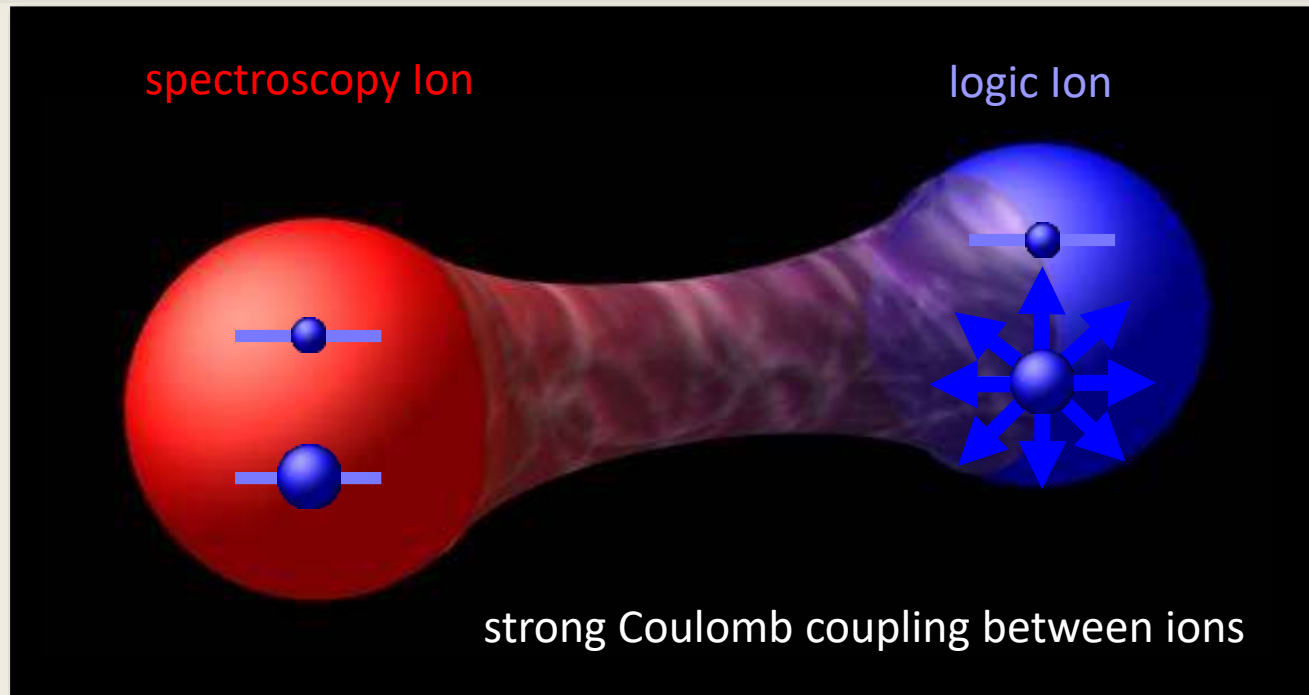
Spectroscopy probes fundamental physics



Need transition for
laser cooling and
detection!



Quantum Logic Spectroscopy



Many applications demonstrated,
e.g.

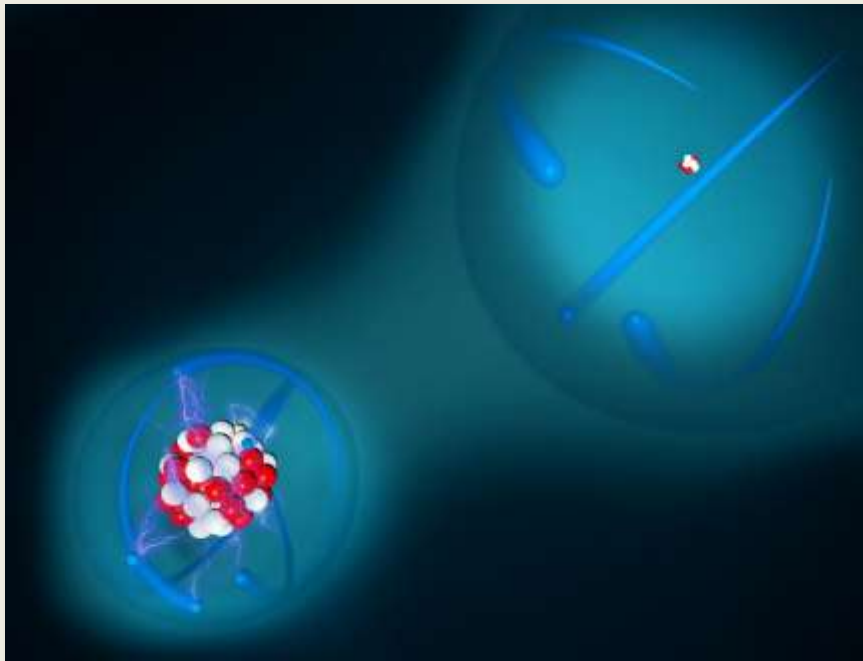
- Most accurate clock: Al^+
[Brewer *et al.*, PRL **123**, 033201 (2019)]
→ Talk by D. Leibrandt
<https://indico.cern.ch/event/942276>

- ions in linear Paul trap → high accuracy achievable
 - logic ion provides sympathetic cooling & signal readout
 - strong Coulomb interaction couples motional modes
 - composite system: combine advantages of both species
- investigation of previously inaccessible species

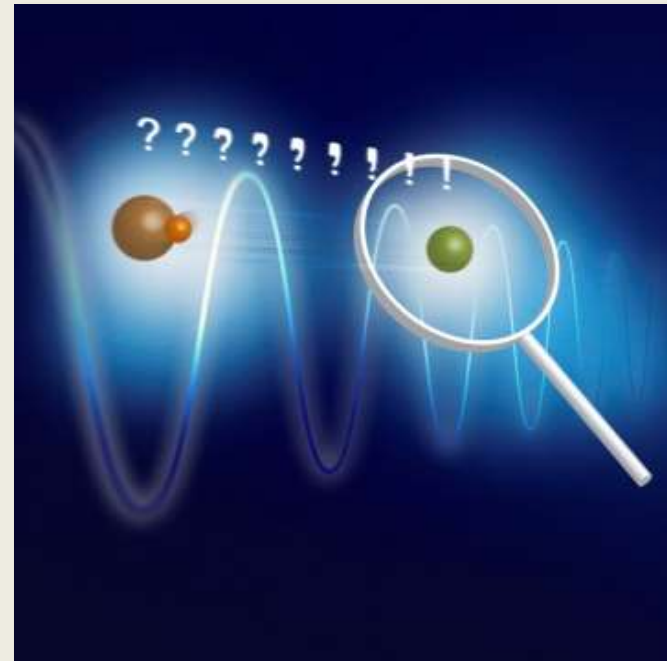
Overview

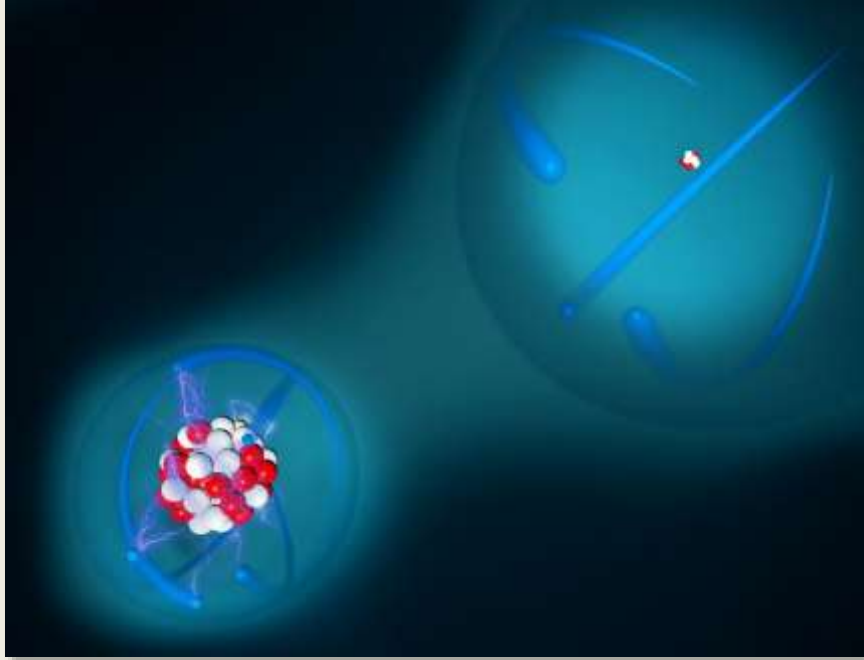
- Introduction & motivation for quantum logic spectroscopy (QLS)

QLS of highly charged ions



QLS of molecular ions



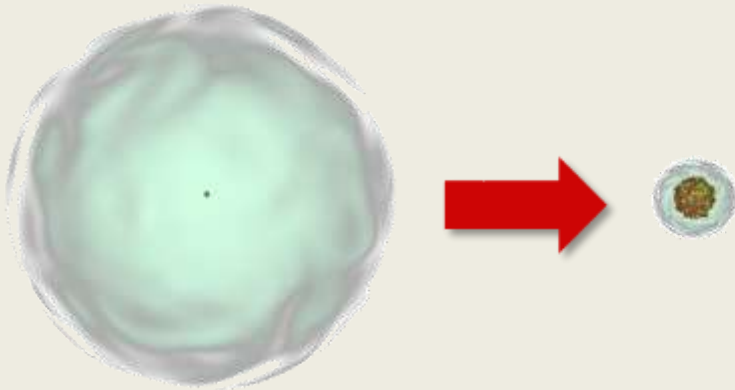


QUANTUM LOGIC SPECTROSCOPY OF HIGHLY CHARGED IONS

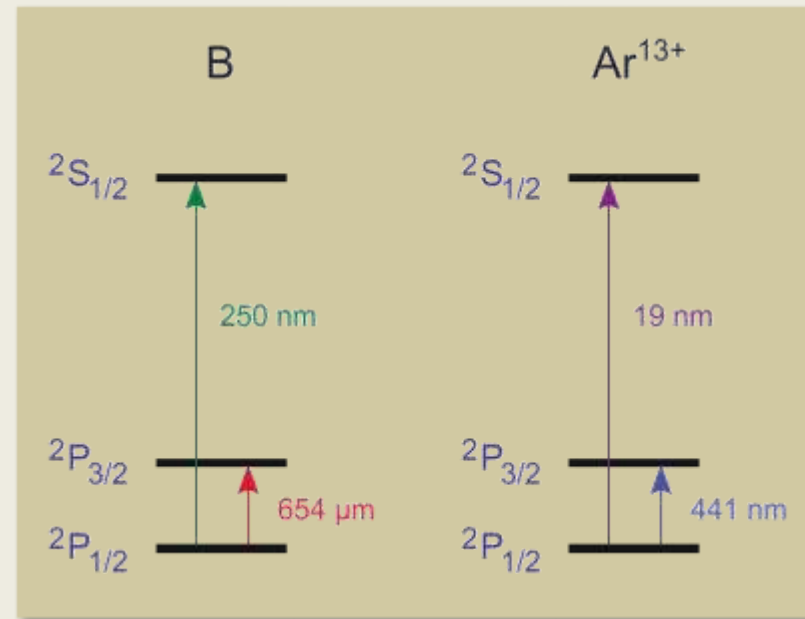
Highly Charged Ions

Charge state dependence:

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



H	$\rightarrow$	$U^{91+} (H\text{-like})$
10 eV	$\rightarrow$	140 keV
μeV	$\rightarrow$	eV
μeV	$\rightarrow$	300 eV

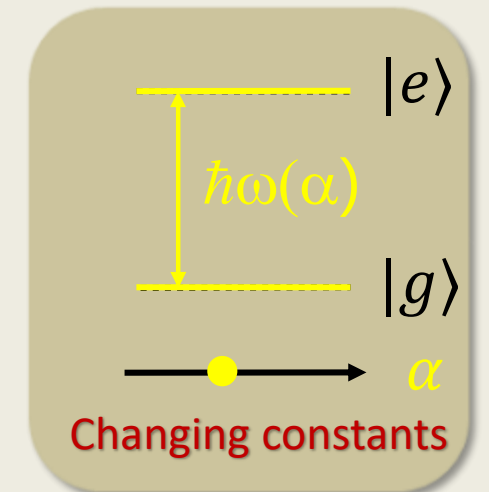
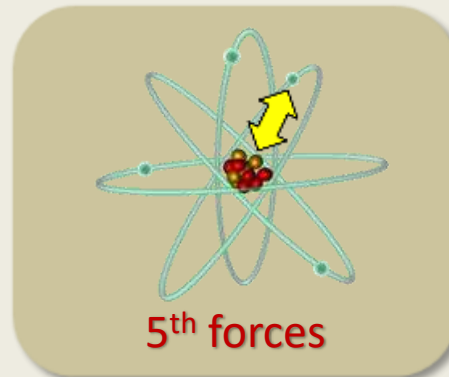
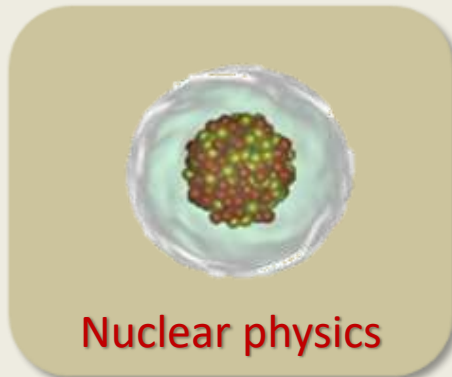
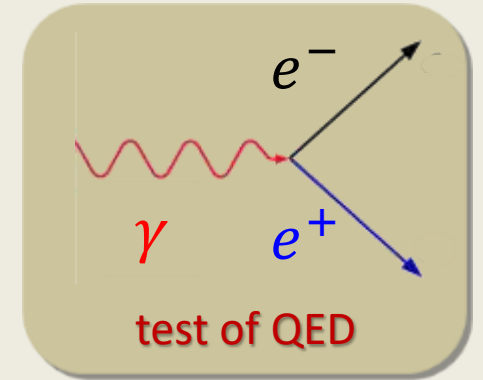


- optical transitions: fs, hfs, **level crossings**

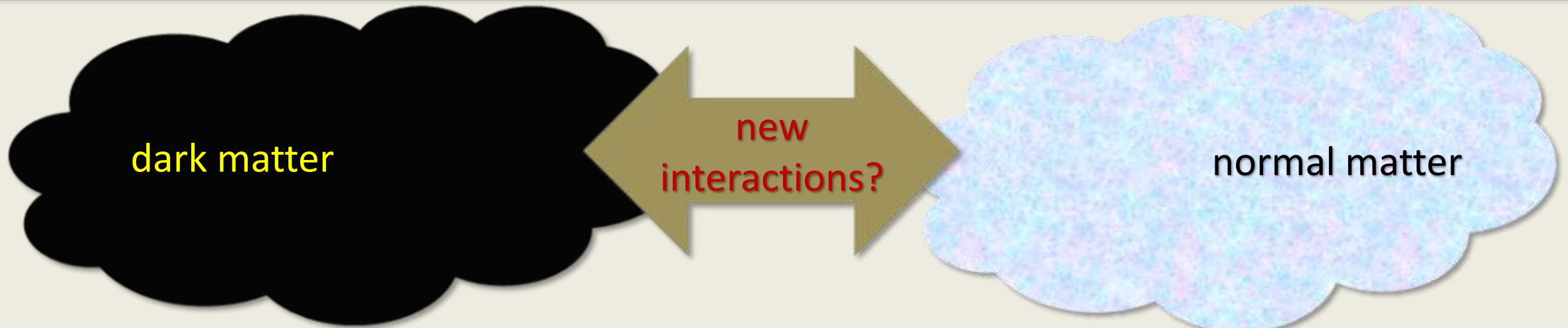
[Kozlov *et al.* Rev. Mod. Phys **90**, 045005 (2018)]

Testing fundamental physics with HCl

- simple electronic structure → testbed for atomic structure theory
- QED test: g-factor → QED in strong fields
- sensitive to
 - α → highest sensitivity of all atomic species
 - violation of local Lorentz invariance
 - isotope shifts (5th forces)
 - parity violation in XUV transitions
 - nuclear physics
 - ...

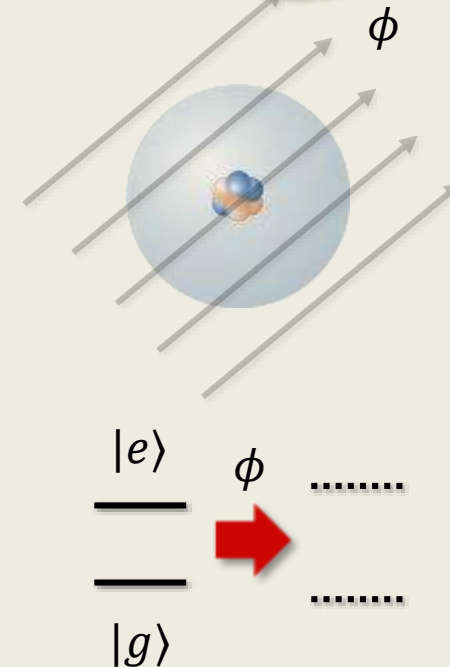


Effects of dark matter on normal matter



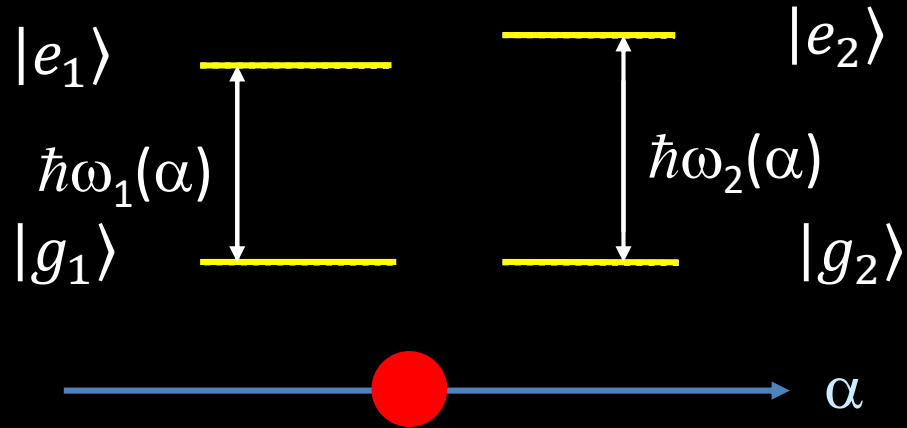
- dark matter candidate: scalar field ϕ
 - oscillating field
 - topological field (forming „clumps“)
 - ...
 - weak (non-gravitational) coupling to matter changes energy levels in atoms/molecules
- **apparent variation of fundamental constants**

[review: Safronova *et al.*, RMP **90**, 025008 (2018)]



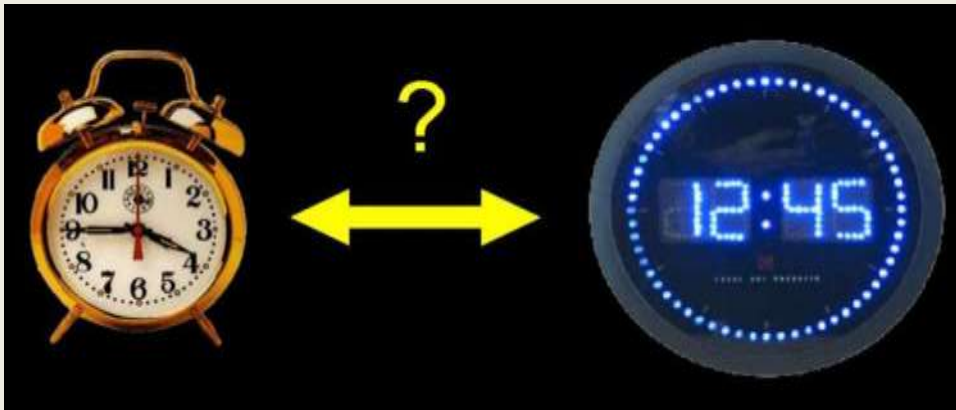
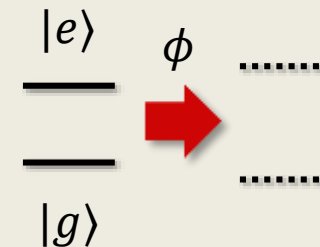
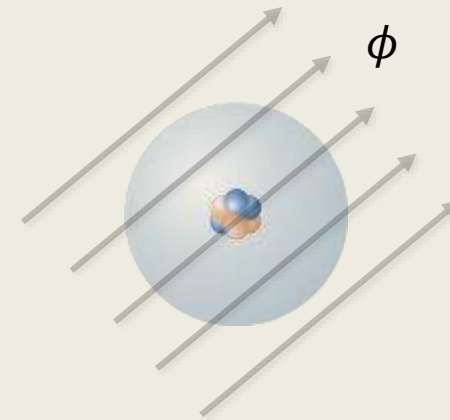
Variation of Fundamental Constants

fine-structure constant α

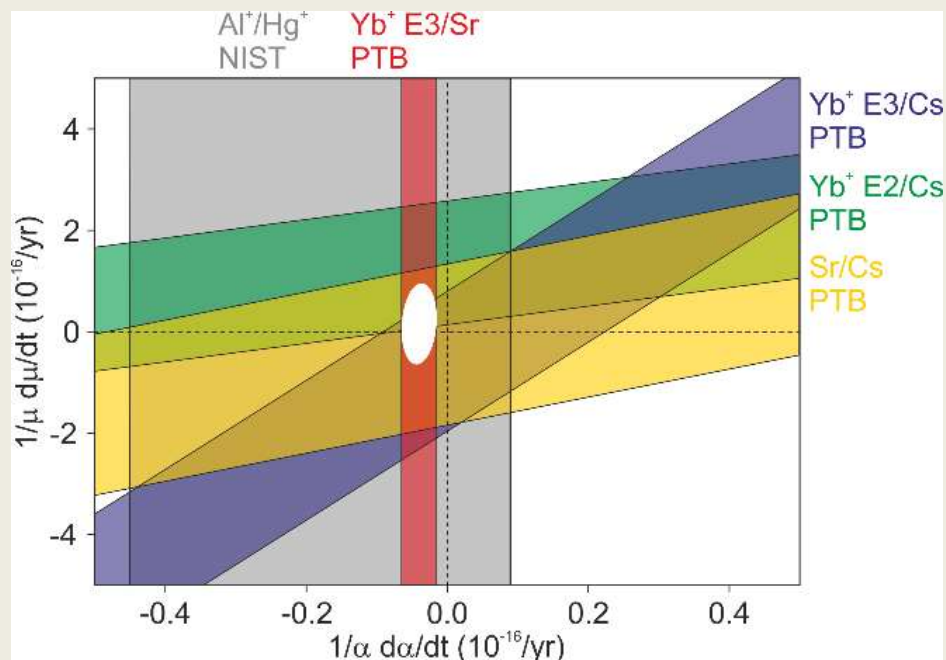


$$\frac{\Delta\omega}{\omega} = K \frac{\Delta\alpha}{\alpha}$$

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



Combined data from clocks



$$\dot{\alpha}/\alpha = -4.1(2.5) \times 10^{-18}/\text{year}$$

$$\dot{\mu}/\mu = -1.3(8) \times 10^{-17}/\text{year}$$

[Peik/Lisdat (PTB), preliminary]

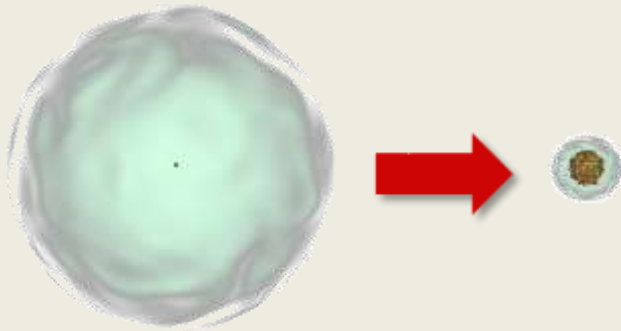
$$\frac{\Delta\omega}{\omega} = K \frac{\Delta\alpha}{\alpha}$$

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
Ir ¹⁷⁺ T1	-109	ca. 1416
Ir ¹⁷⁺ T2	145	ca. 2000
Cf ^{15+*}	57	ca. 618
Cf ^{17+*}	-44	ca. 485
Th* nuclear	8000	ca. 150

highest sensitivity of
all known atomic
systems

Highly charged ions as optical clocks?

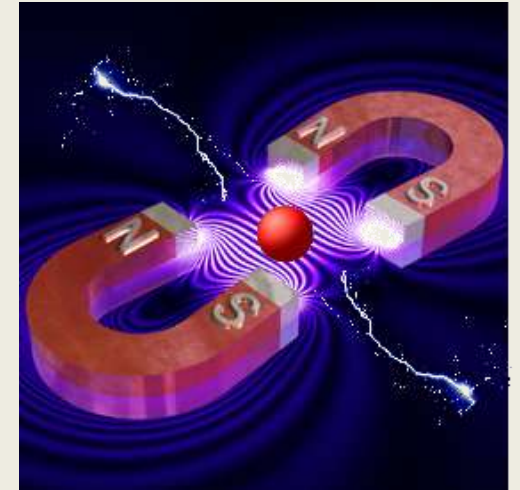
- **High accuracy**
 - ➔ low sensitivity to resonance shifts
- **HCI advantage: suppressed shifts**



Hydrogen-like HCI:

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

[Berengut *et al.*, EPJ Web of Conferences **57**, 02001 (2013)]

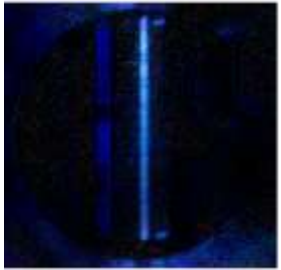


electric & magnetic fields

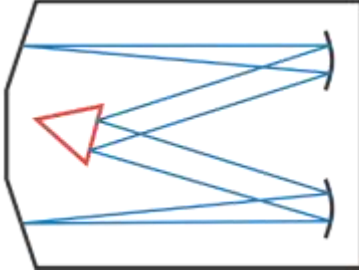
Other clock species requirements can be fulfilled

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

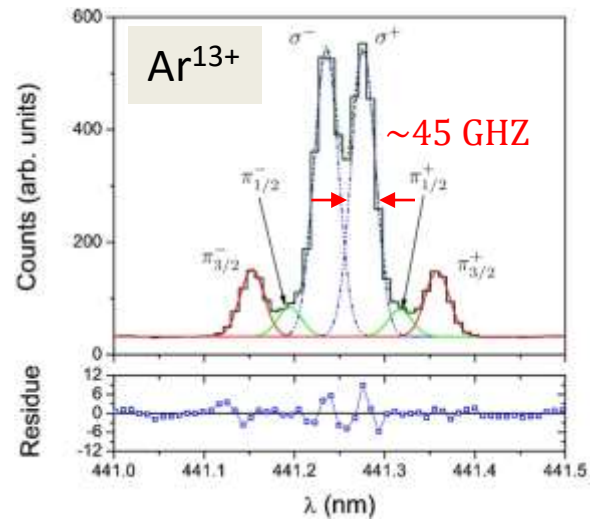
State-of-the-art HCl spectroscopy



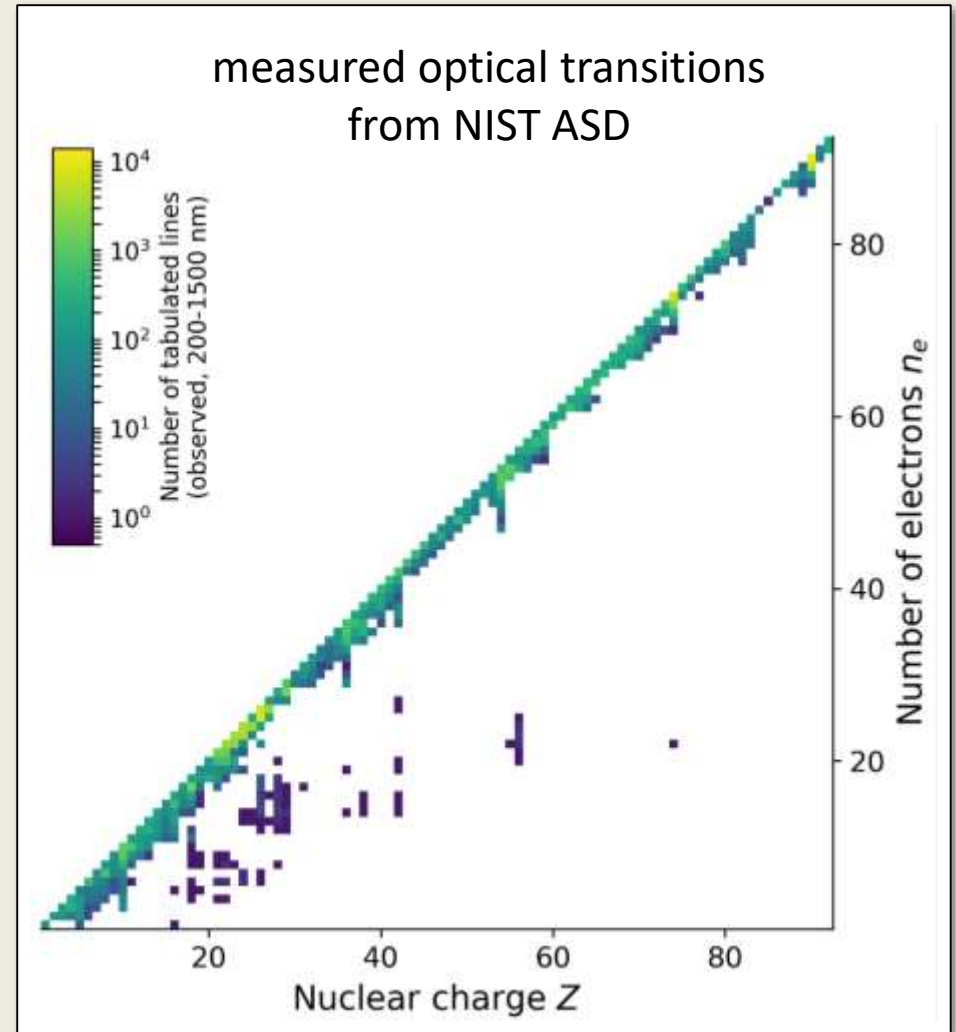
Plasma (EBIT)



Grating spectrometer



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

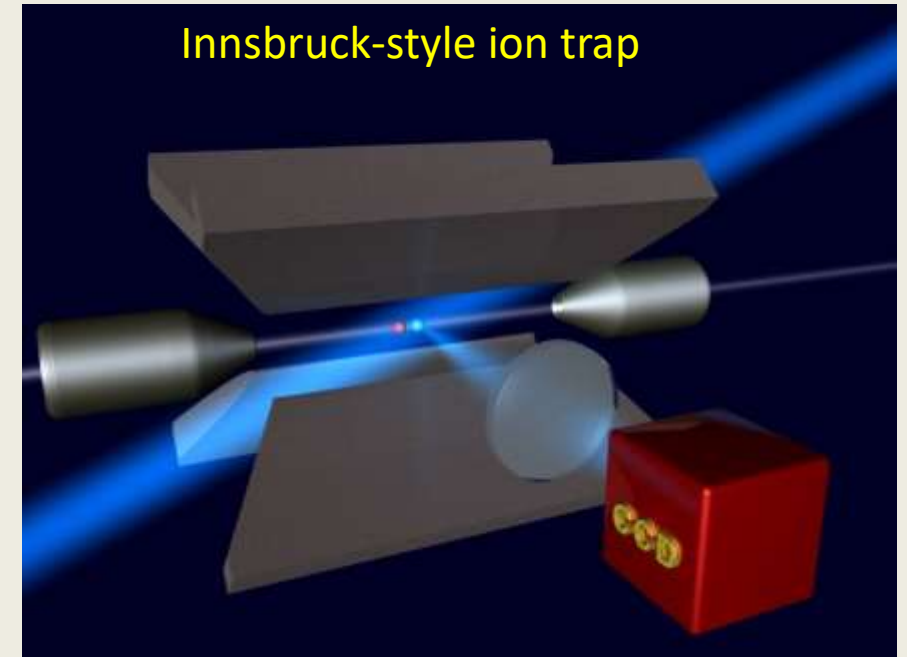


Doppler-limited resolution of ~ 150 MHz

Laser spectroscopy of single trapped ions

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
→ high accuracy



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

Yb^+ single-ion clock: 3.2×10^{-18}

[Huntemann *et al.*, PRL **116**, 063001 (2016)]

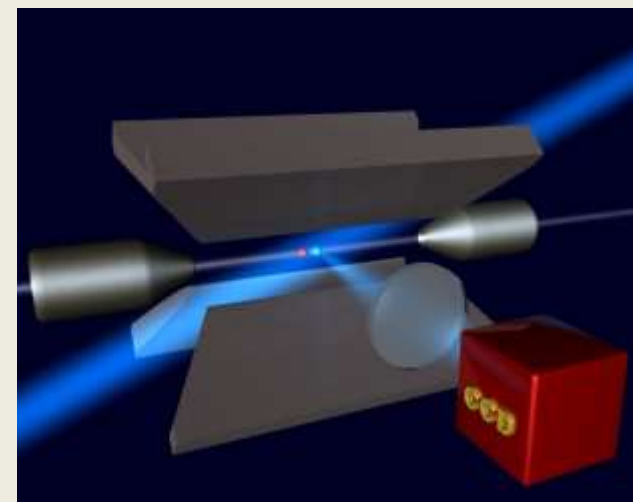
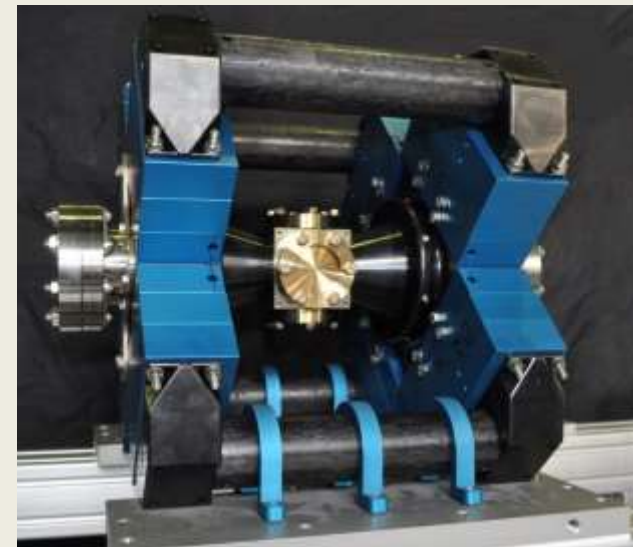
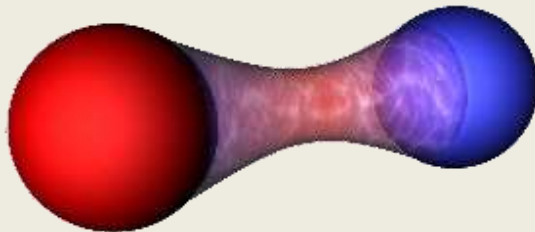
High resolution spectroscopy of HCl?

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



PTB approach to precision HCl spectroscopy

Machine room

1 m

Laser Laboratory

30 cm

cryogenic linear Paul trap

cryogenic
linear Paul trap

55 mm

[Leopold *et al.*, Rev. Sci. Instr. **90**, 073201 (2019)]

mini EBIT

[Micke *et al.*, RSI **89**, 063109 (2018)]

[Micke *et al.*, Rev. Sci. Instr. **90**, 065104 (2019)]

Specs vacuum system:

- Vacuum: $\sim 10^{-14}$ mbar
- Temperature: < 5 K
- Vibrations: < 20 nm
- Magnetic field: < 0.5 nT

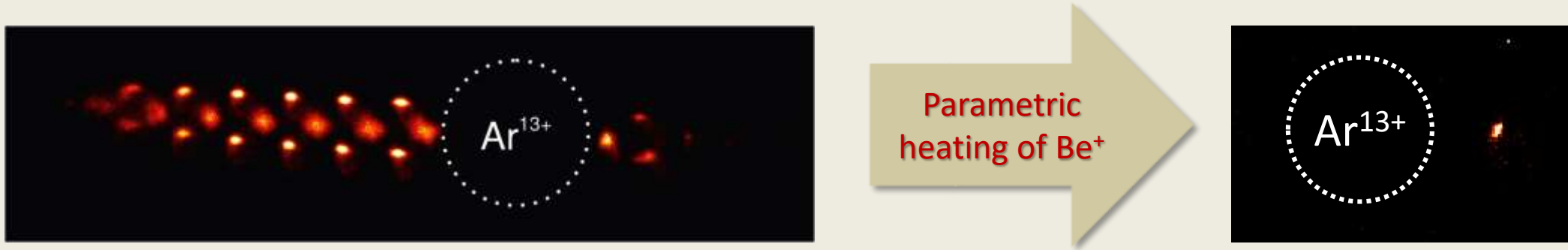
Specs EBIT:

- 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
- $f/\# \sim 1$ imaging with bi-aspheric lens

Preparation & Lifetime of a 2-Ion Crystal



- total preparation time of $\text{Be}^+/\text{Ar}^{13+}$ crystal: $\sim$ few min
- Ar^{13+} lifetime: $\tau = (38.4 \pm 3.8)$ min
→ residual pressure: $< 1.5 \times 10^{-14}$ mbar
(assuming Langevin collisions)
- Sideband cooling to the motional ground state ($T < 3 \mu\text{K}$)

Quantum Logic with Trapped Ions

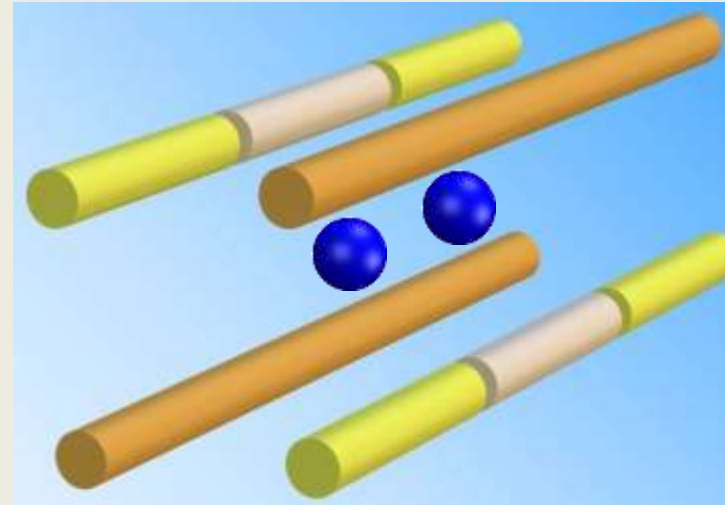
- Idea by:

J. I. Cirac



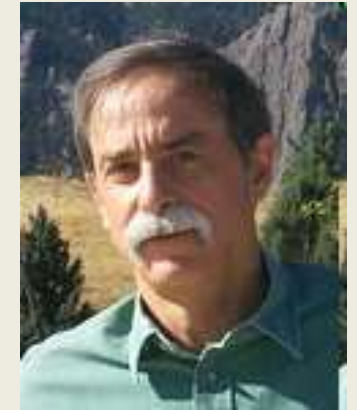
PRL **74**, 4091 (1995)

P. Zoller



Collective motion of ions
described by normal modes

D. Wineland



NP 2012

VOLUME 74, NUMBER 20

PHYSICAL REVIEW LETTERS

15 MAY 1995

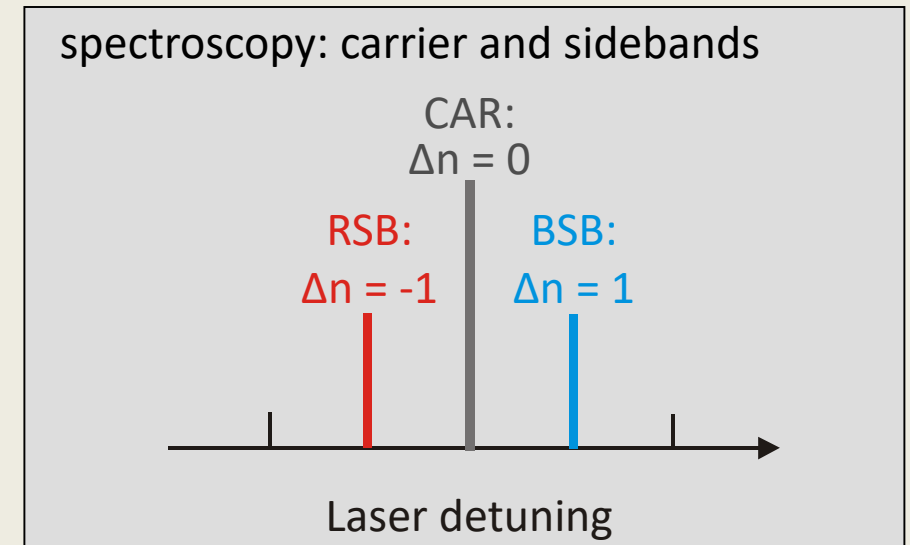
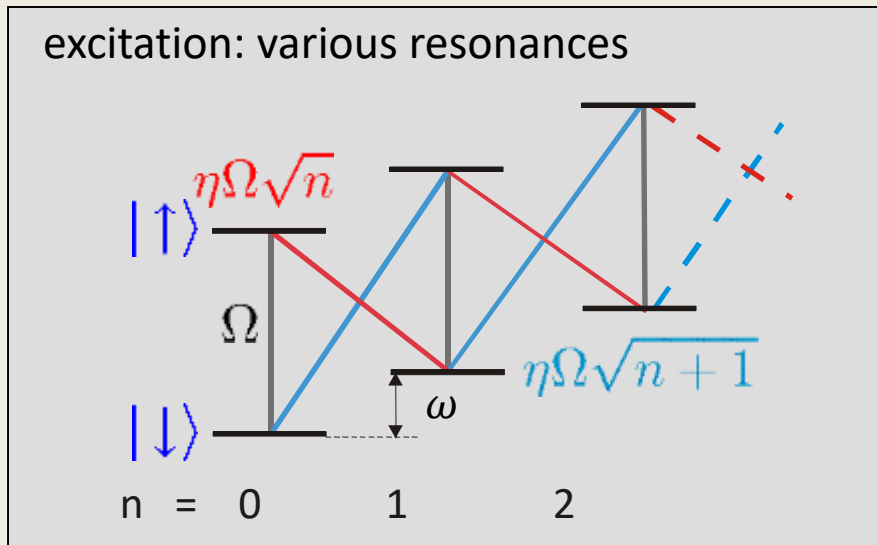
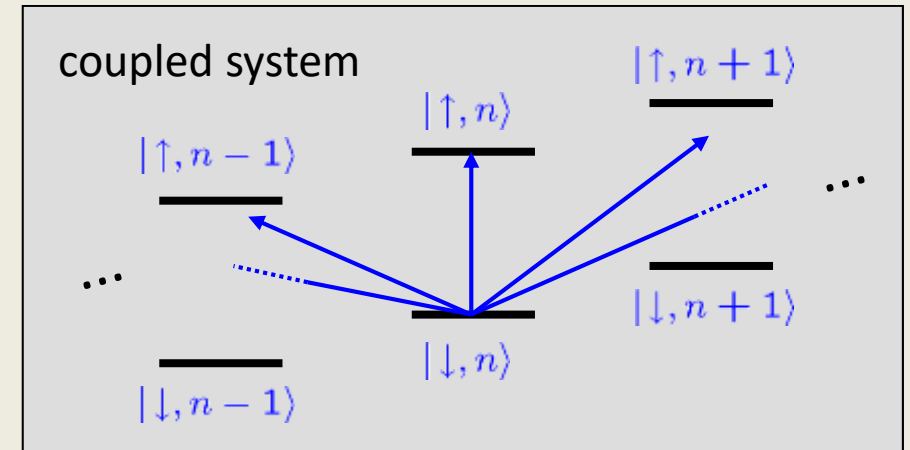
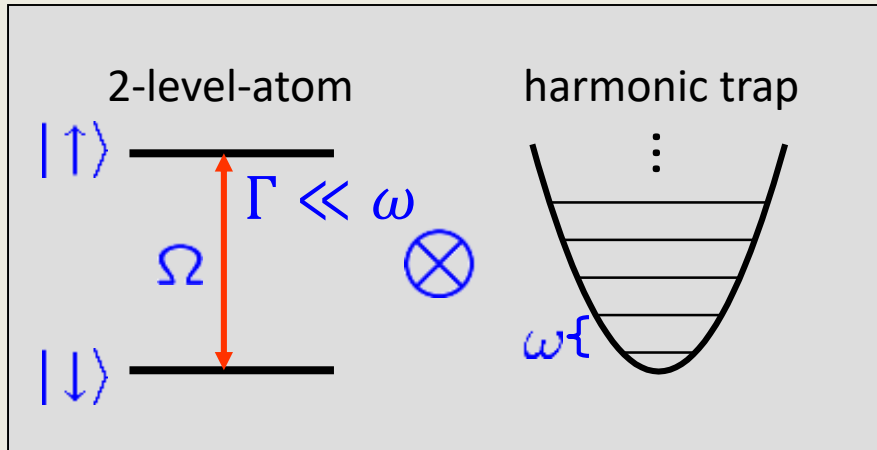
Quantum Computations with Cold Trapped Ions

J. I. Cirac and P. Zoller*

Institut für Theoretische Physik, Universität Innsbruck, Technikerstrasse 25, A-6020 Innsbruck, Austria
(Received 30 November 1994)

A quantum computer can be implemented with cold ions confined in a linear trap and interacting with laser beams. Quantum gates involving any pair, triplet, or subset of ions can be realized by coupling the ions through the collective quantized motion. In this system decoherence is negligible, and the measurement (readout of the quantum register) can be carried out with a high efficiency.

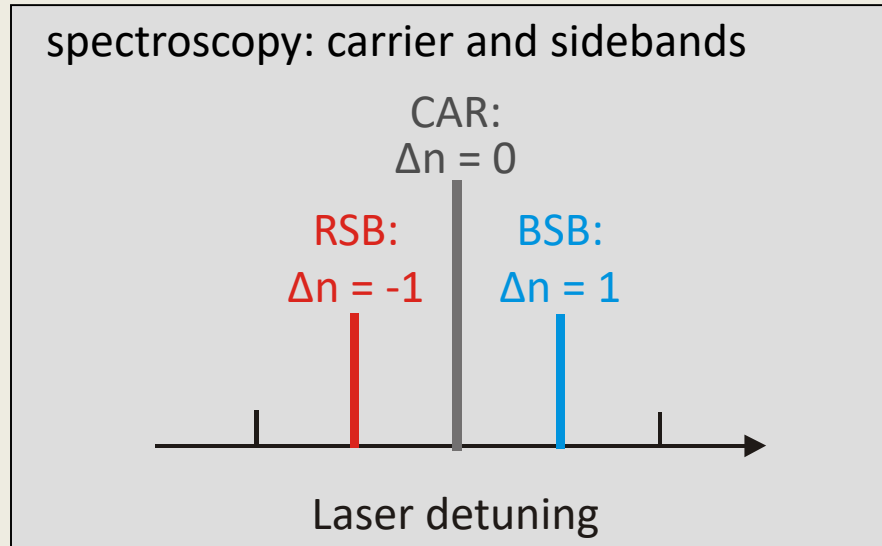
Quantum Logic with Trapped Ions



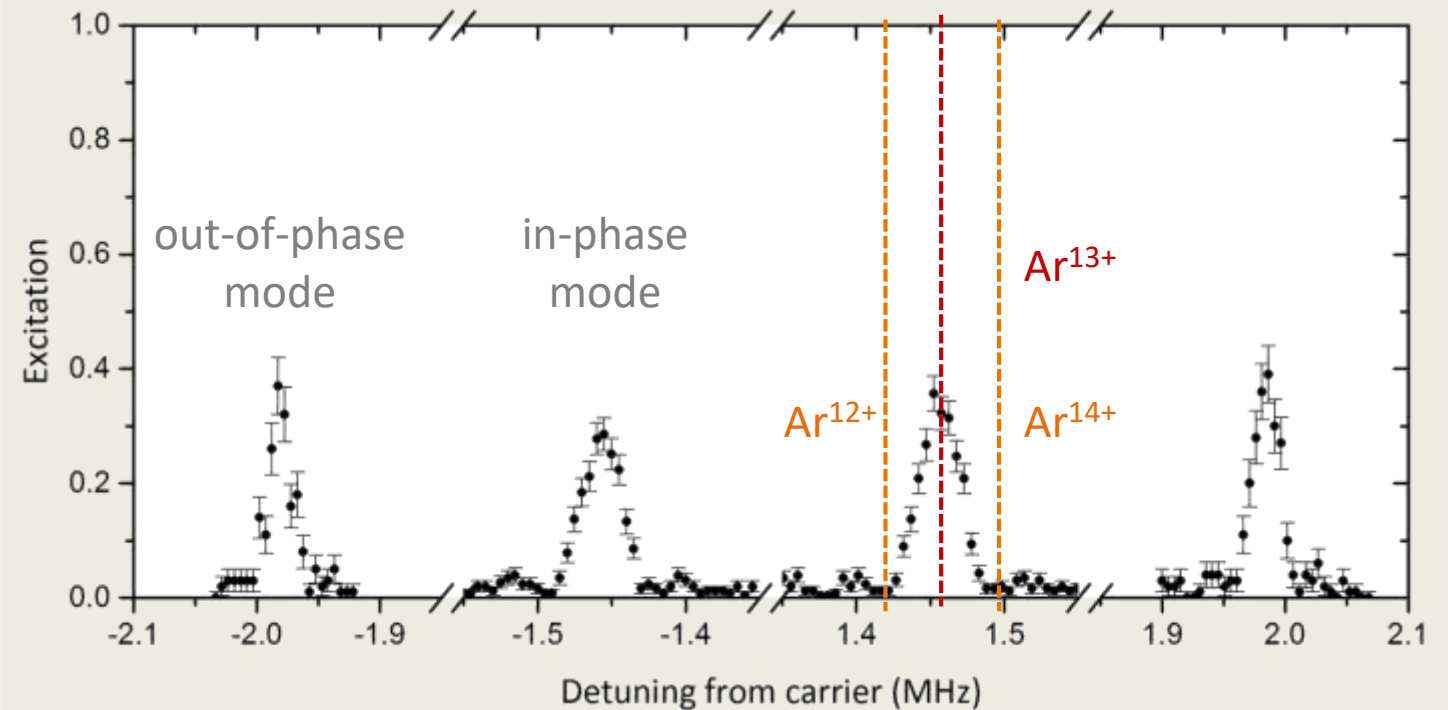
Ω : Carrier Rabi frequency; $\eta = kz_0$: Lamb-Dicke factor



Doppler cooling & charge state identification



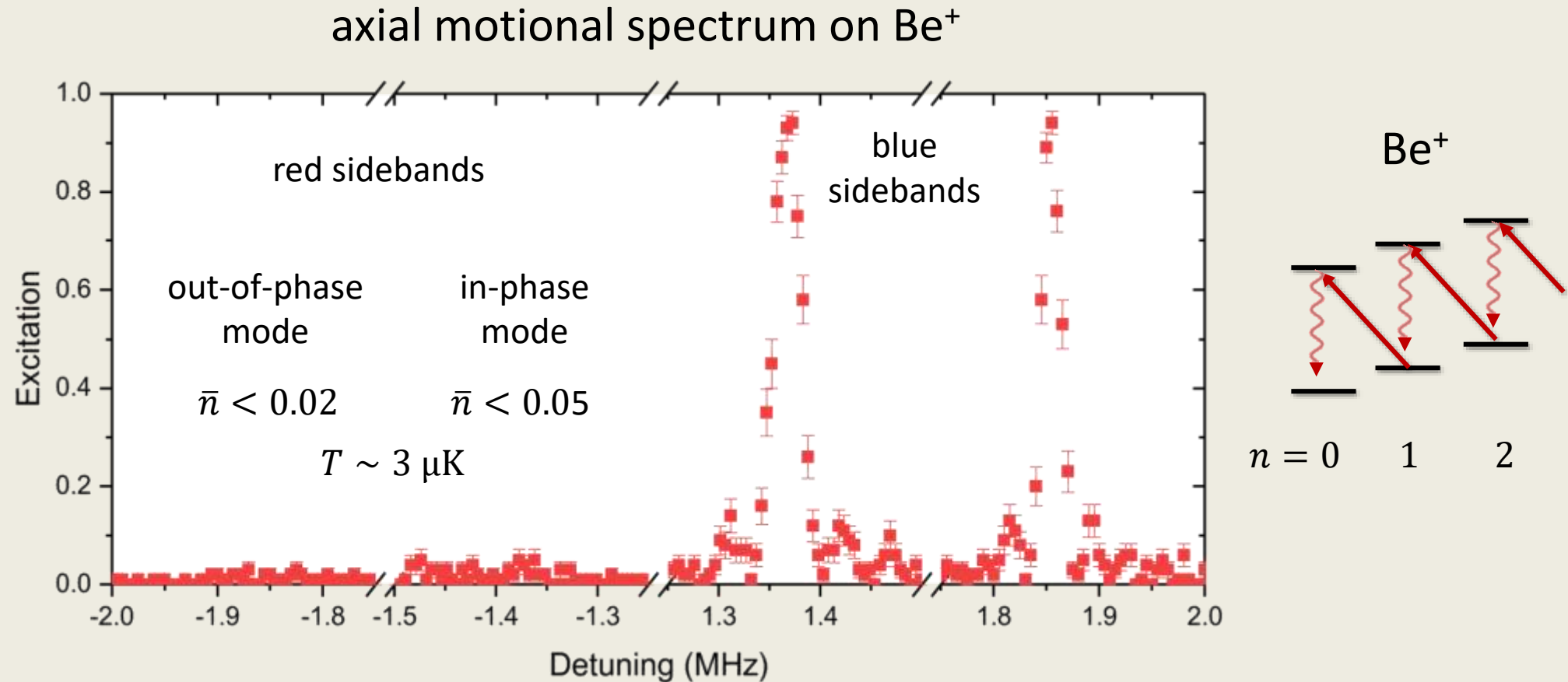
axial motional spectrum on Be^+



- single Be^+ axial frequency: 0.995 MHz
→ $\text{Be}^+/\text{Ar}^{13+}$ axial frequencies:
1.47 MHz and 1.99 MHz

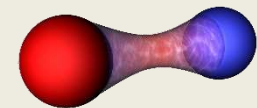


Sympatetic ground state cooling of Ar^{13+}

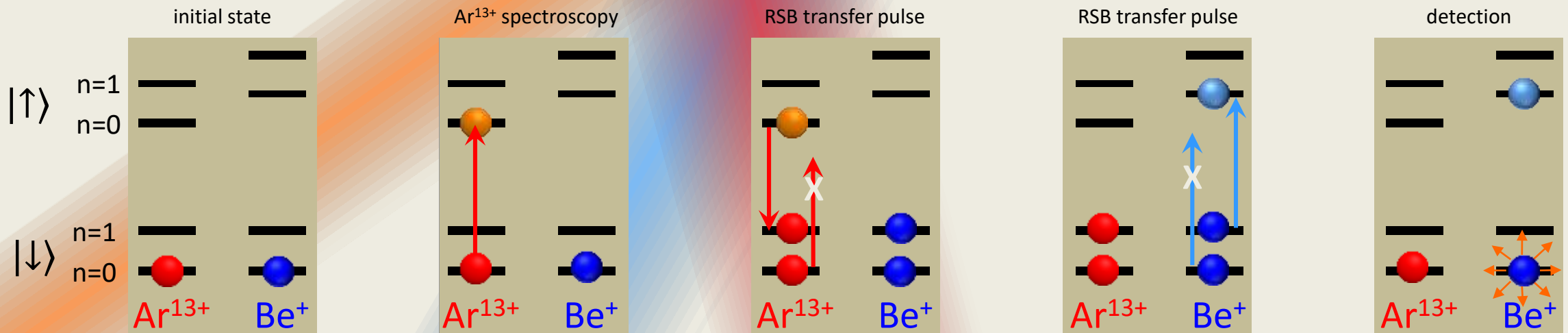
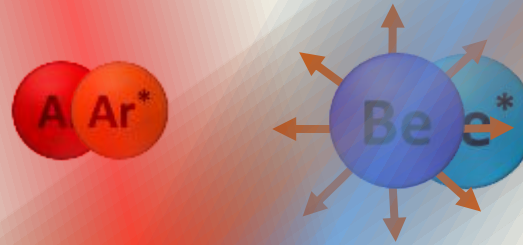


- resolved Raman sideband cooling on Be^+
- Lamb-Dicke parameter: $\eta_z = 0.82\sqrt{\text{MHz}/\nu_z}$

[King *et al.*, in preparation]

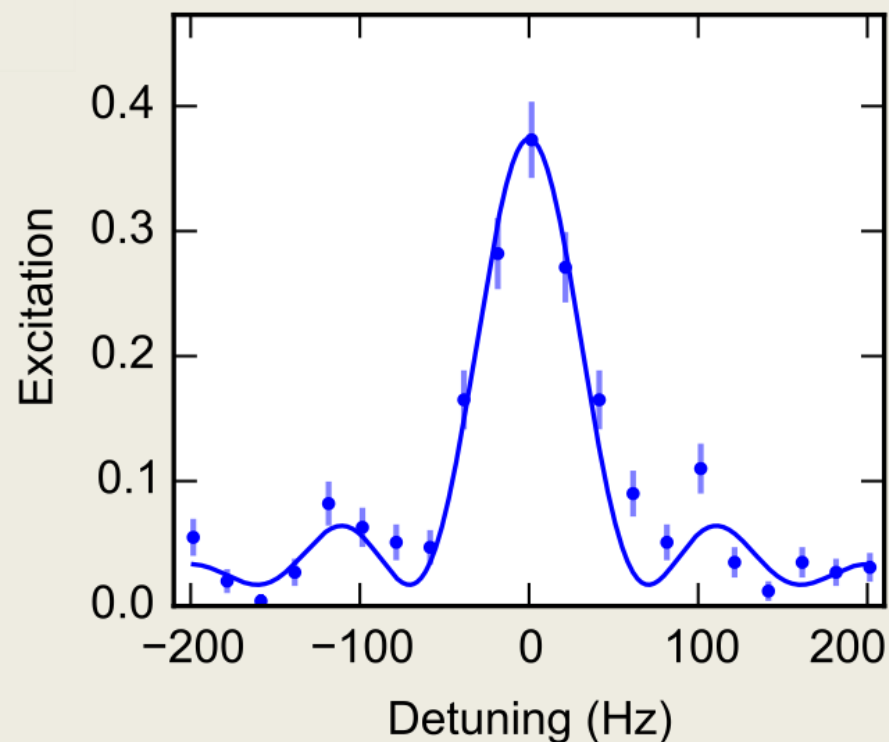


Quantum Logic State Transfer

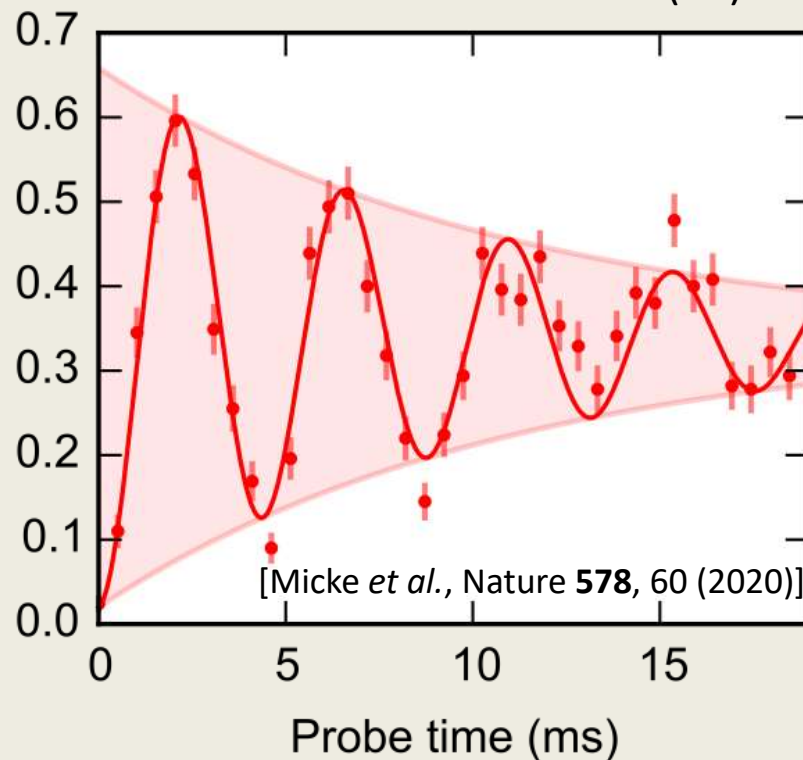


Quantum Logic Spectroscopy of Ar^{13+}

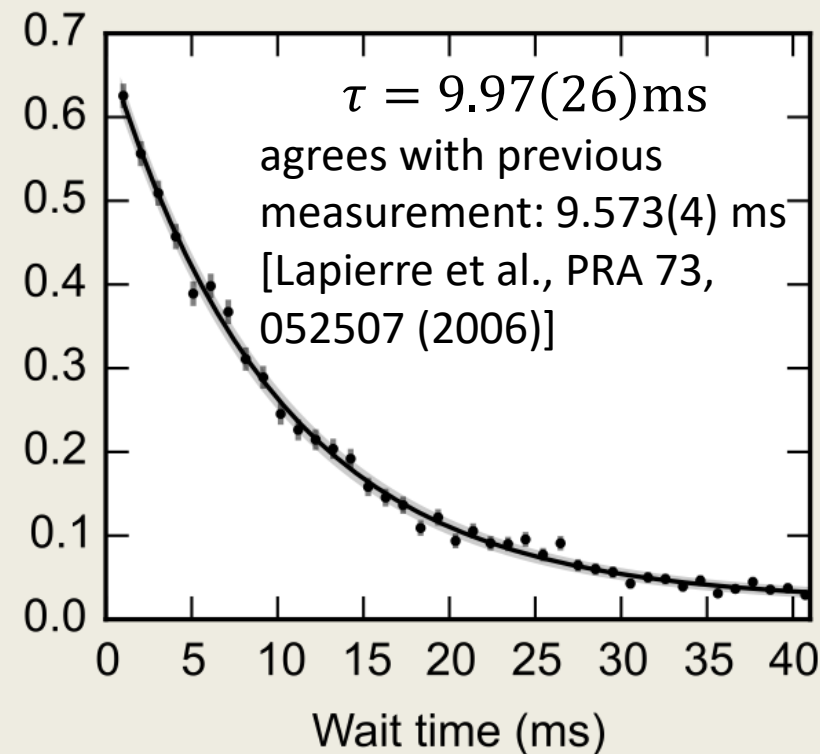
Fourier-limited linewidth: 65 Hz
(12 ms probe time) resolution: ~ 5 Hz



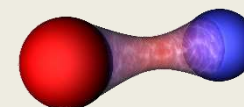
dephasing dominated by
excited state lifetime of 9.97(26)ms



dedicated lifetime measurement

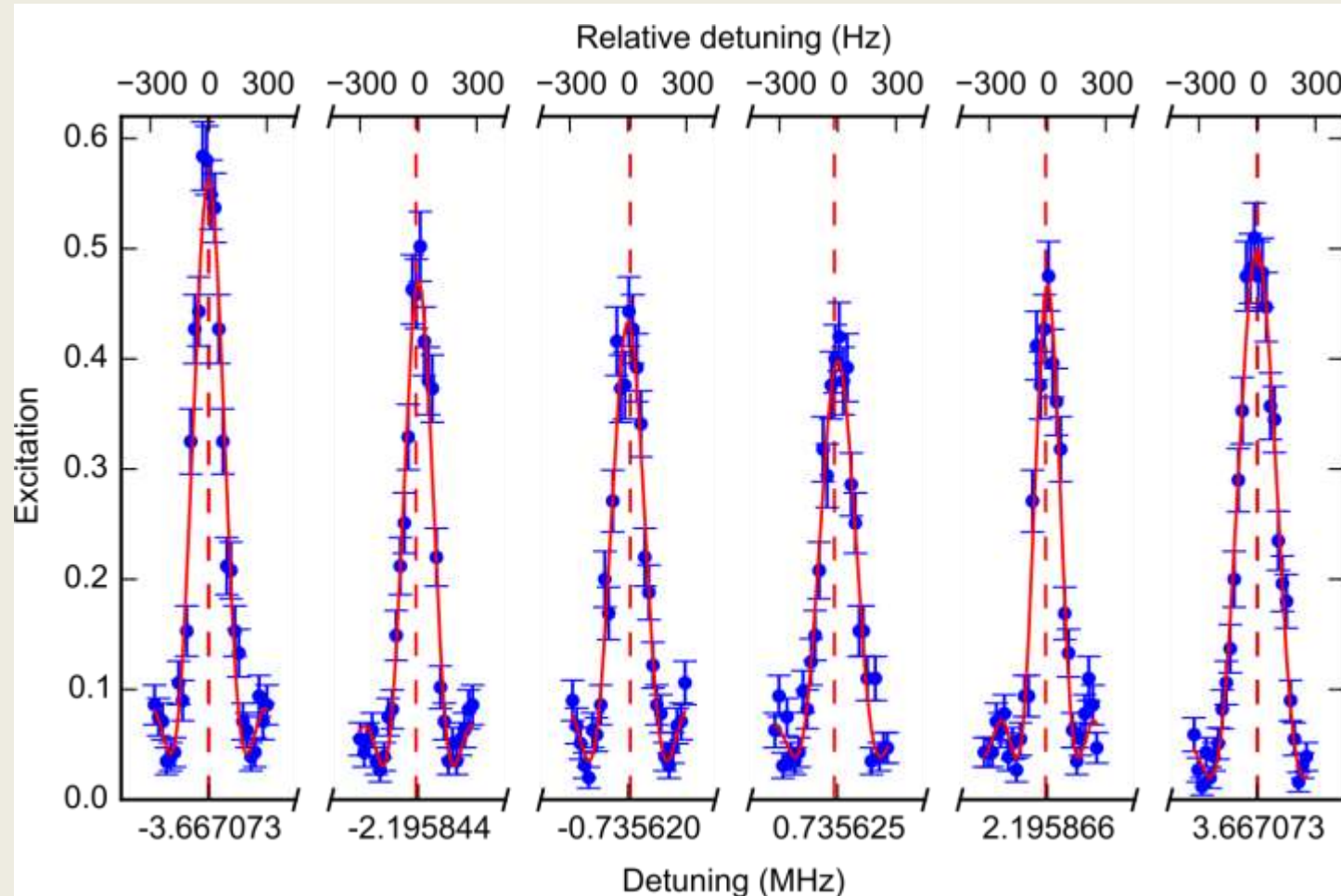


- spectroscopy laser transfer locked of Ar^{13+} to Si cavity-stabilized laser
[Sterr & Benkler @ PTB: D. G. Matei *et al.*, Phys. Rev. Lett. **118**, 263202 (2017)]



Ar¹³⁺ Zeeman structure

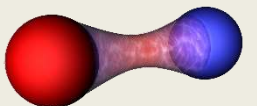
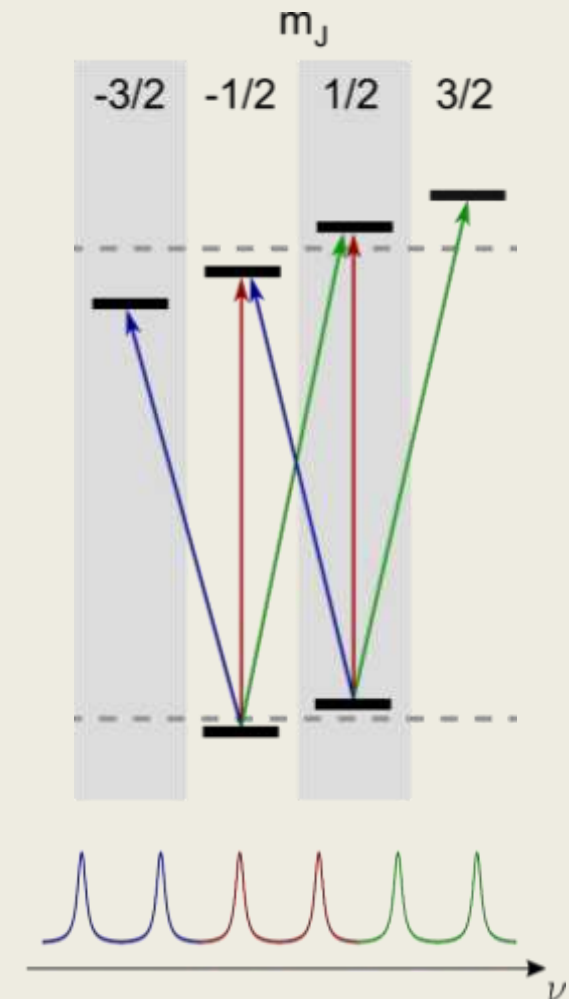
--- Direct + faint interactions + QED



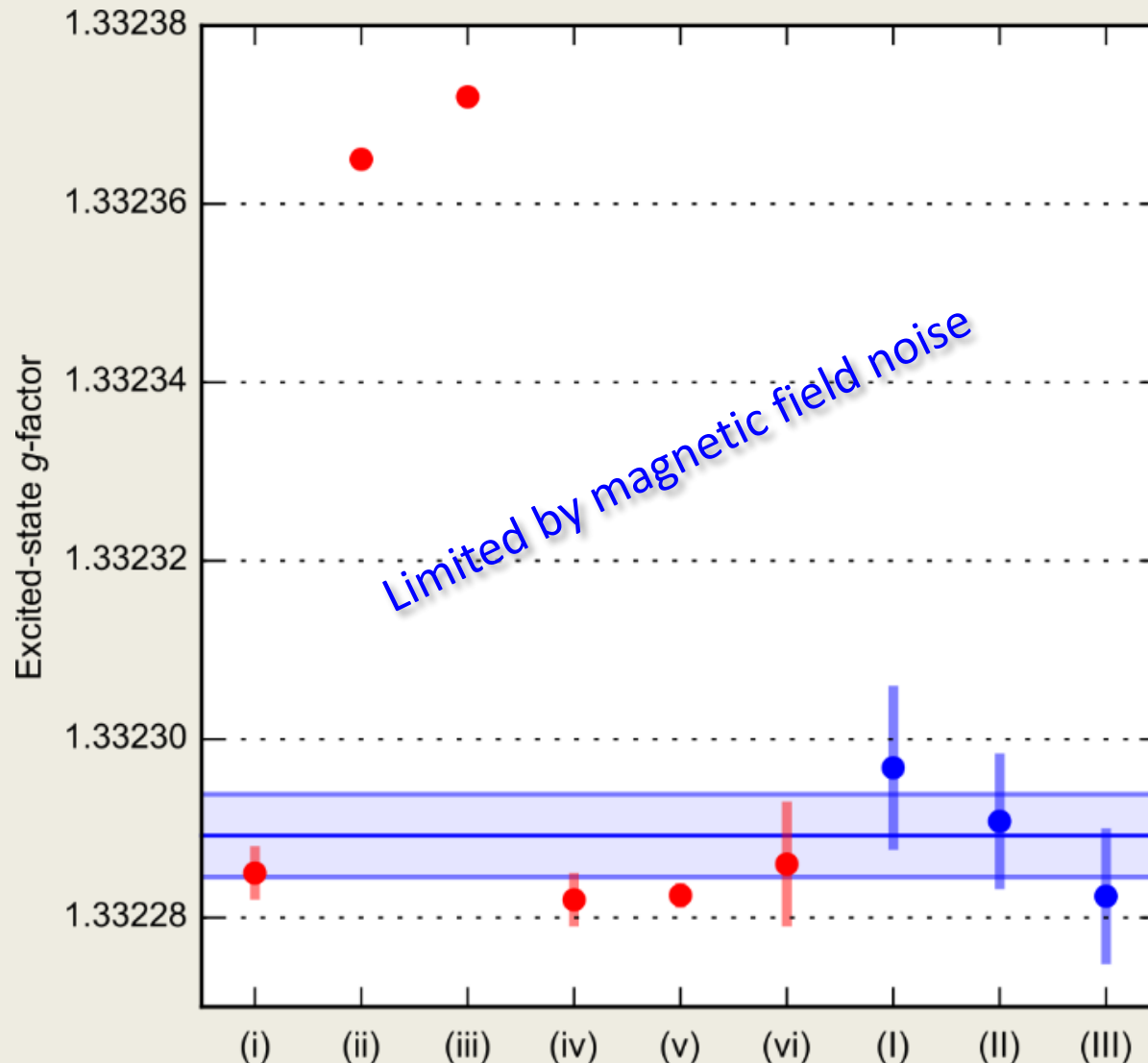
[Micke *et al.*, Nature **578**, 60 (2020)]

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

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



Excited state g -factor



Theory:

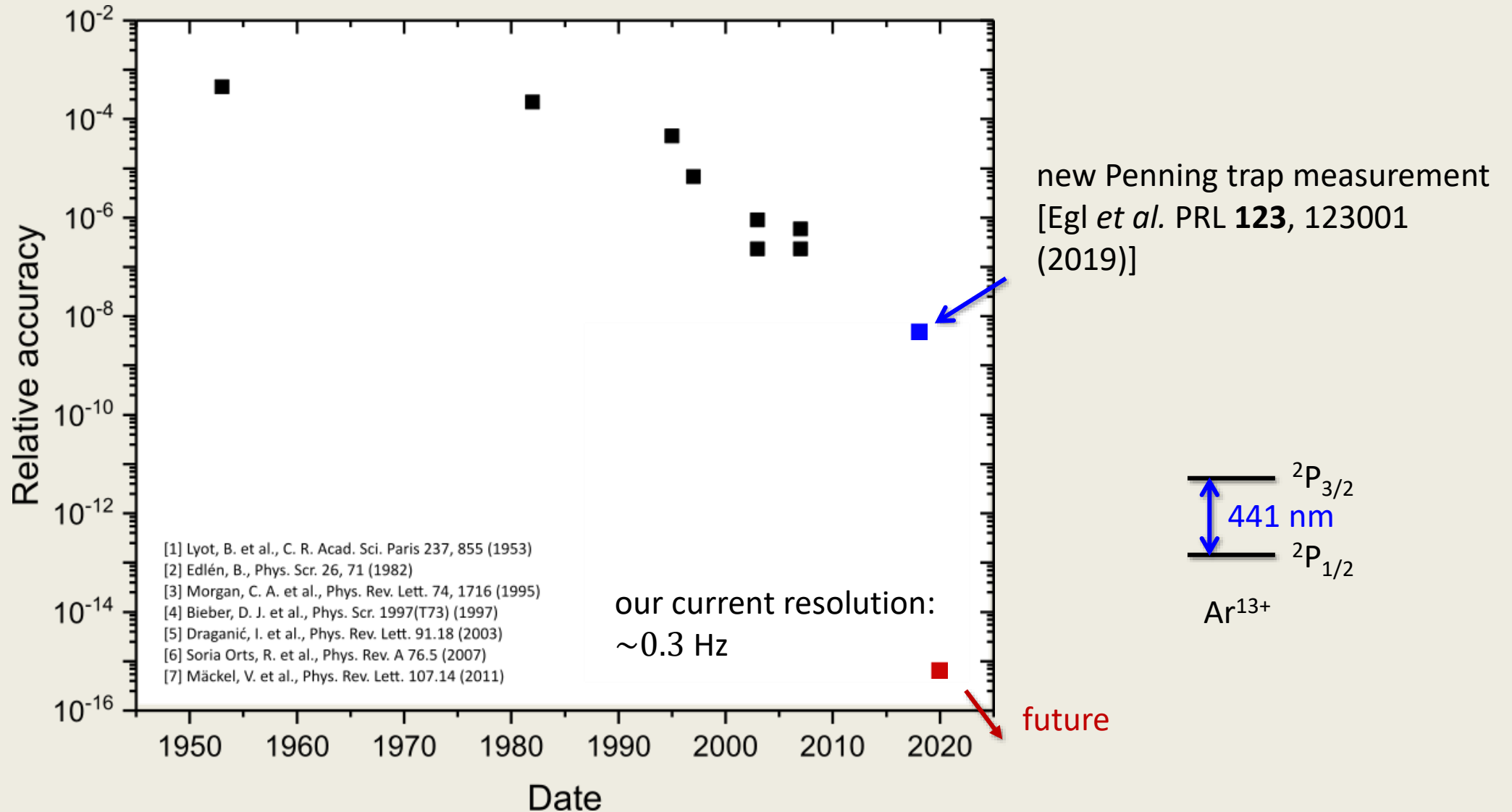
- (i) Glazov *et al.*, Phys. Scr. **T156**, 014014 (2013)
- (ii) Verdebout *et al.*, At. Data Nucl. Data Tables **100**, 1111 (2014)
- (iii) Marques *et al.*, Phys. Rev. A **94**, 042504 (2016)
- (iv) Shchepetnov *et al.*, J. Phys. Conf. Ser. **583**, 012001 (2015)
- (v) Agababaev *et al.*, arXiv:1812.06431 (2018)
- (vi) Maison *et al.*, Phys. Rev. A **99**, 042506 (2019)

Experiment:

(I)-(III) This work

QED test of excited state g -factor

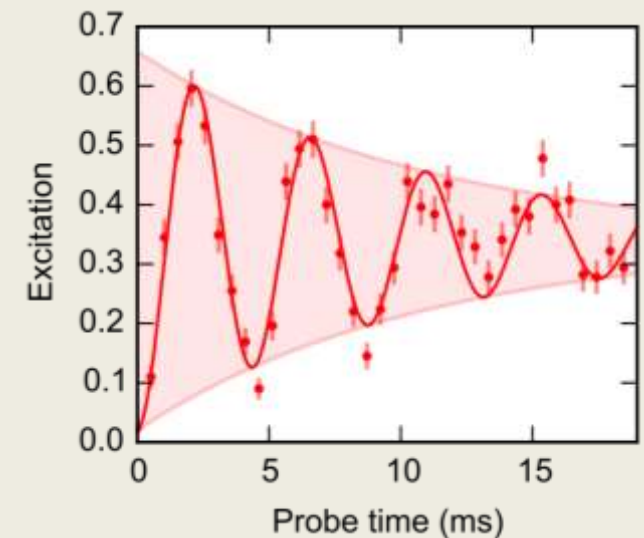
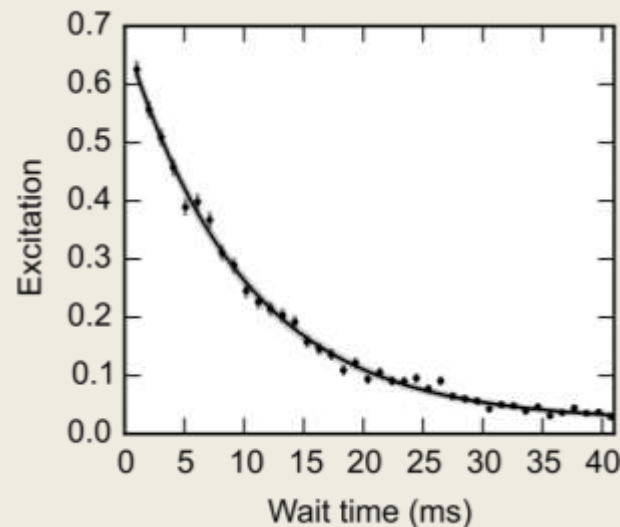
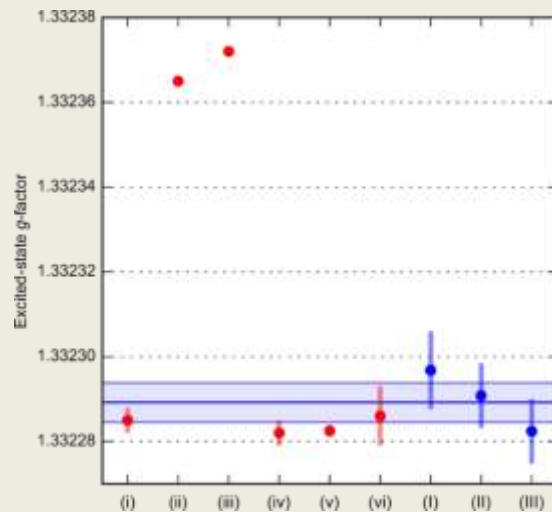
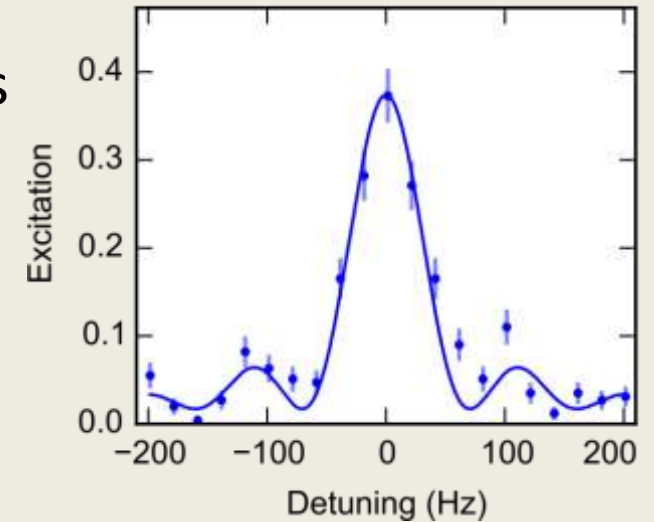
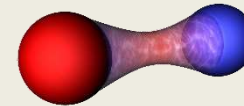
History of Ar^{13+} frequency measurements



HCI Summary

Summary

- precision spectroscopy of HCI addresses fundamental physics
- full quantum optical control over HCI achieved
- first coherent spectroscopy of HCI
- measured excited state g-factor & lifetime
- “universal” spectroscopy scheme

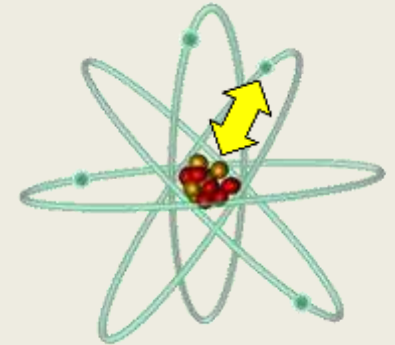


For more details see: <https://indico.cern.ch/event/901588/>

What's next?

First optical HCI „clock“

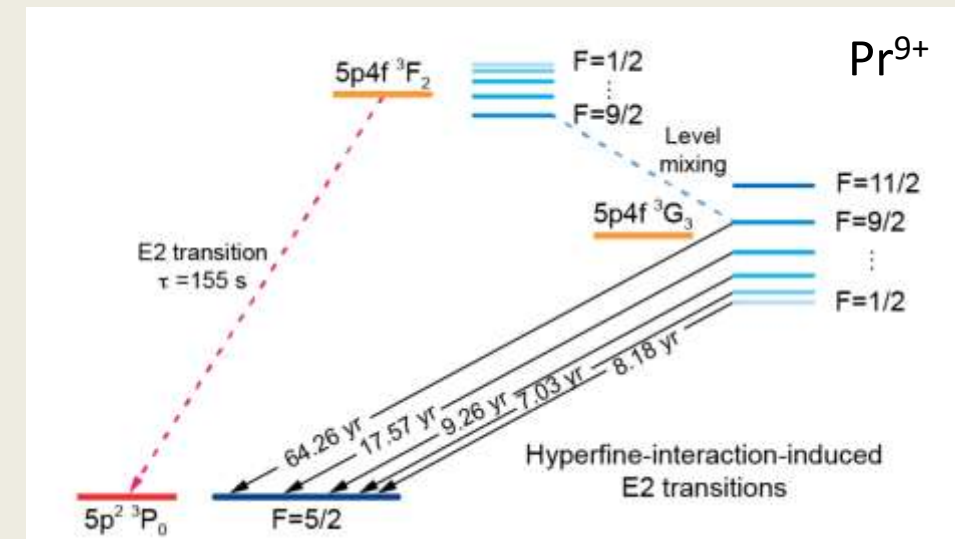
- $^{36,40}\text{Ar}^{13+}$ $P_{1/2}$ - $P_{3/2}$ lines: full evaluation of systematic uncertainties
- verify isotope shift atomic structure calculations
[Yerokhin *et al.*, Phys. Rev. A **101**, 012502 (2020)]
- Isotope shift spectroscopy of $\text{Ca}^{14+/15+}$ to search for 5th forces
[Berengut *et al.*, PRL **120**, 091801 (2018)]

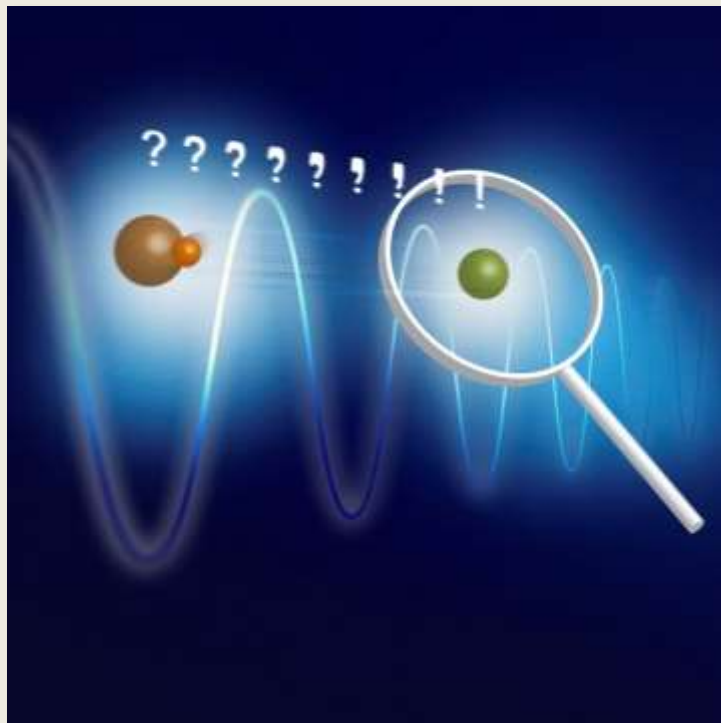


Future

- Clock candidate: $^{58}\text{Ni}^{12+}$
[Yu & Sahoo, Phys. Rev. A **97**, 041403 (2018)]
- α -sensitive level-crossings: Pr^{9+} , Ir^{17+} , $\text{Cf}^{15+/17+}$
[Bekker *et al.*, Nat. Commun. **10**, 5651 (2019)]
[Windberger *et al.*, PRL **114**, 150801 (2015)]
[Porsev *et al.*, PRA, **102**, 012802 (2020)]

**goal: optical clock-like spectroscopy of
HCI to test fundamental physics**





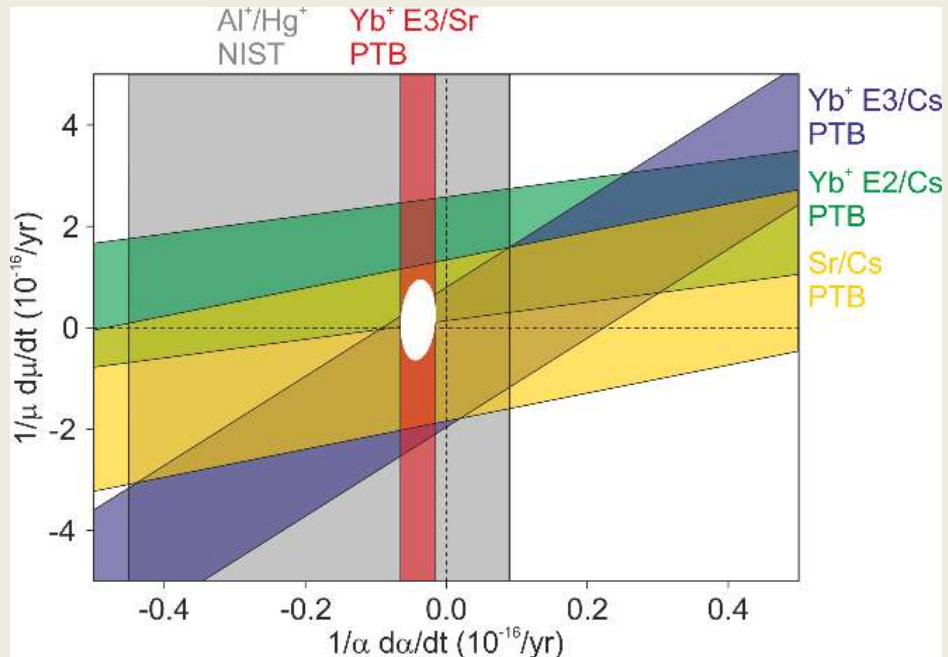
QUANTUM LOGIC WITH MOLECULAR IONS

Electron-to-proton mass ratio



$$\frac{\dot{\mu}}{\mu} \sim 40 \frac{\dot{\alpha}}{\alpha}$$

[Calmet & Fritzsch, Phys. Lett. B **540**, 173 (2002)]

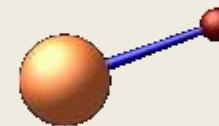


$$\begin{aligned}\dot{\alpha}/\alpha &= -4.1(2.5) \times 10^{-18} / \text{year} \\ \dot{\mu}/\mu &= -1.3(8) \times 10^{-17} / \text{year}\end{aligned}$$

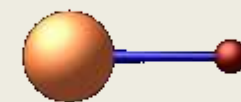
[Peik/Lisdat (PTB), preliminary]

- change in μ from clocks is model dependent
- limited uncertainty from microwave transition

ro-vibrational optical transition
in molecules provides high
sensitivity for $\dot{\mu}/\mu$



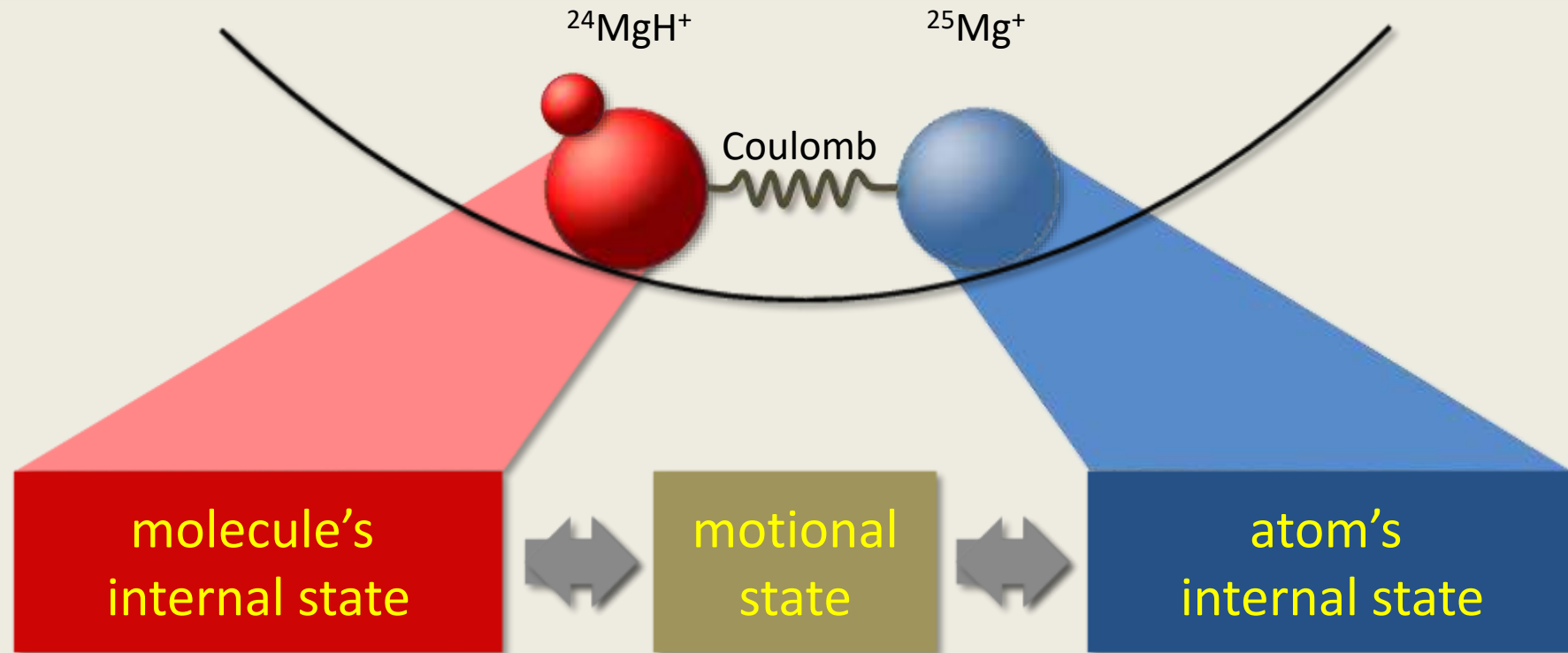
$$E_{vib} \sim \sqrt{\mu}$$



$$E_{rot} \sim \mu$$

[Schiller & Korobov, Phys. Rev. A **71**, 032505 (2005)]

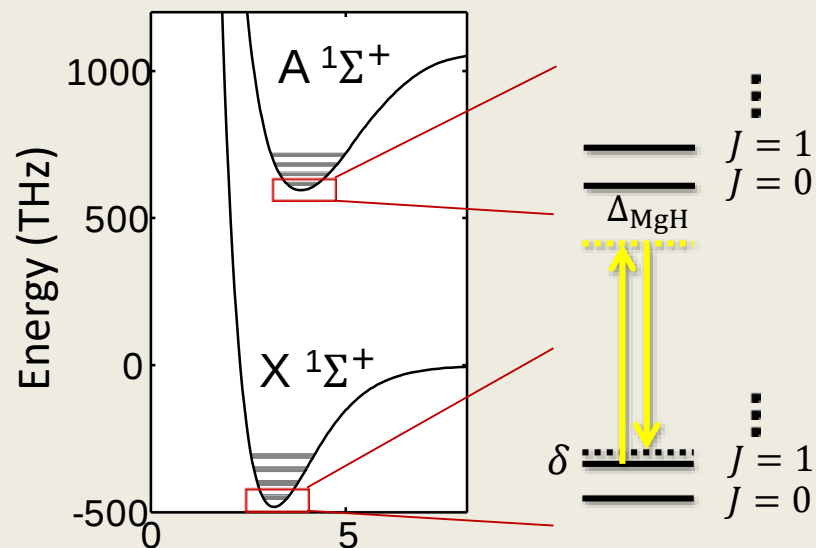
State detection of a molecular ion



- atomic ion is a sensor for molecular ion
 - molecular ion can be controlled through atomic ion
 - composite system: combine advantages of both species
- ➔ make single molecular ions accessible for spectroscopy

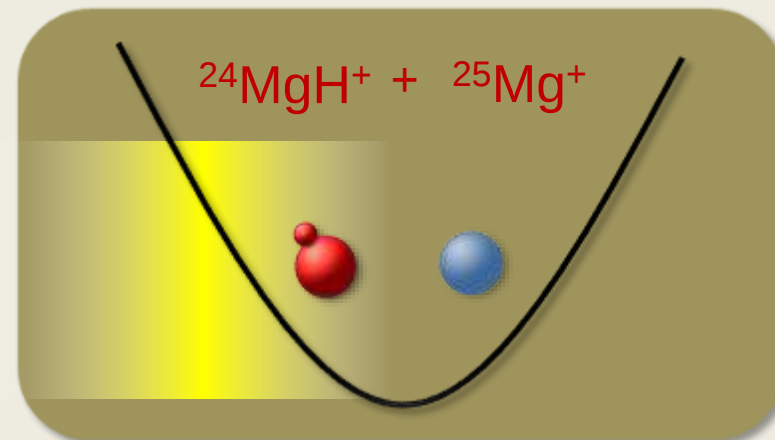
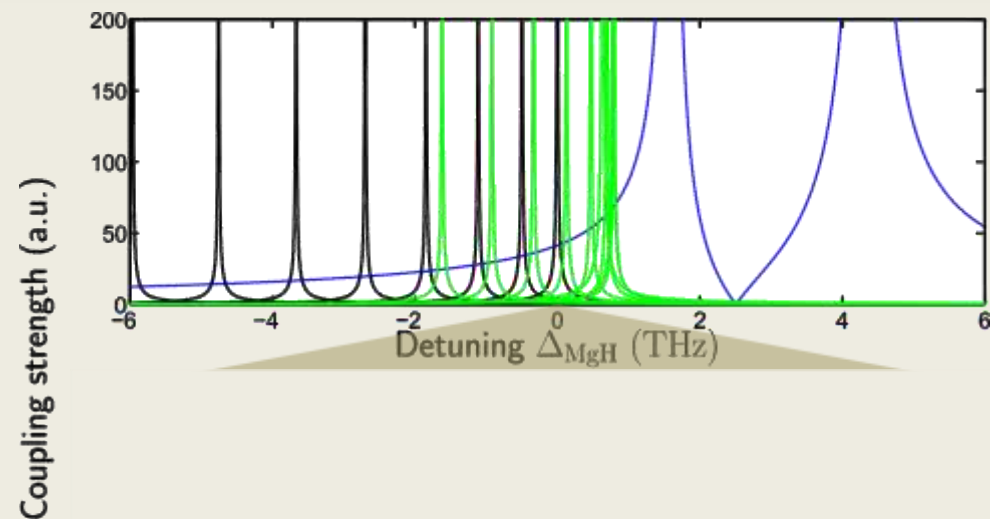
[similar proposals by: Drewsen, Keller, Koelemeji; demonstrated with atoms: Hume @ NIST]

Dipole force on MgH^+/Mg^+ system



- $J \rightarrow J - 1$
- $J \rightarrow J + 1$
- $^{25}\text{Mg}^+$ D1 & D2 lines
→ excitation offset

rotational state selectivity
through laser detuning



BBR-induced quantum jumps

Detection sequence:
(30x per data point)

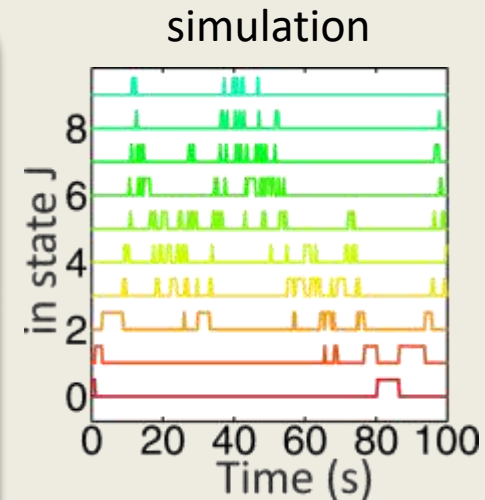
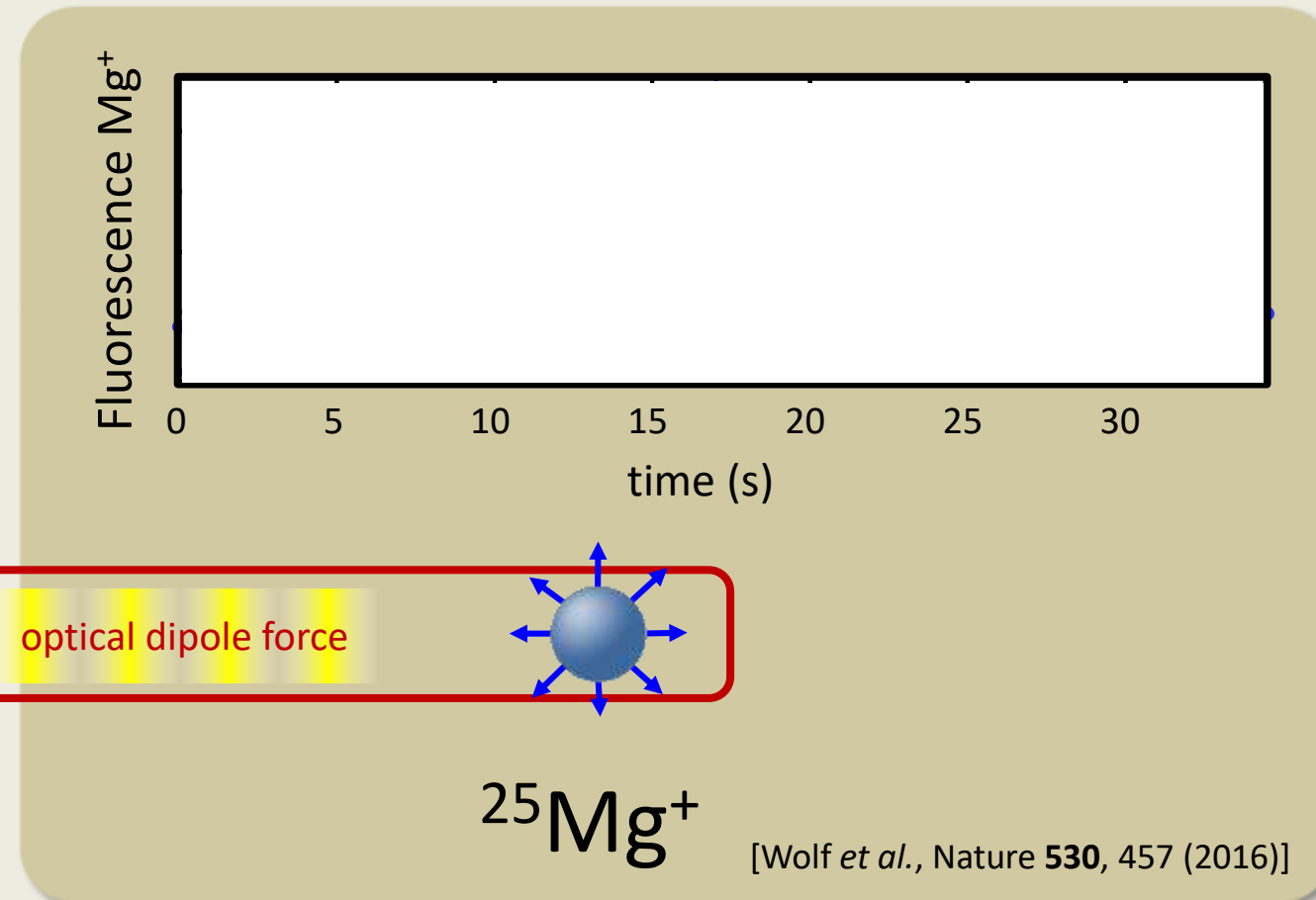
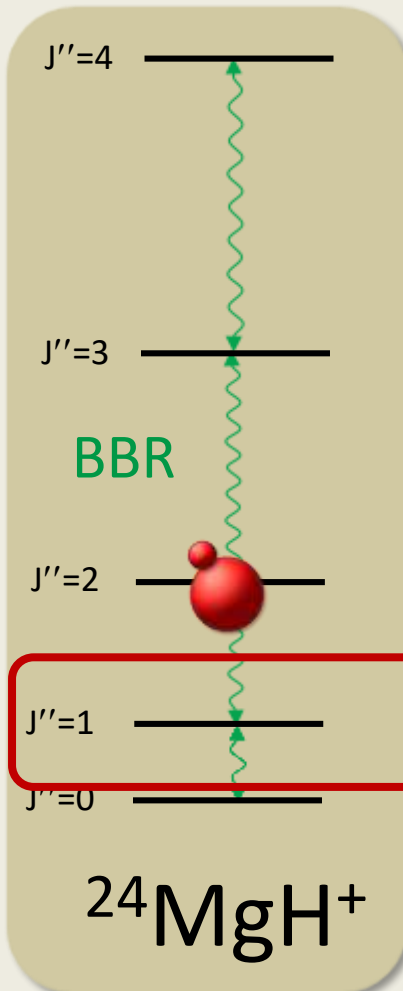
ground
state
cooling

spin-
motion
mapping

optical
dipole
force

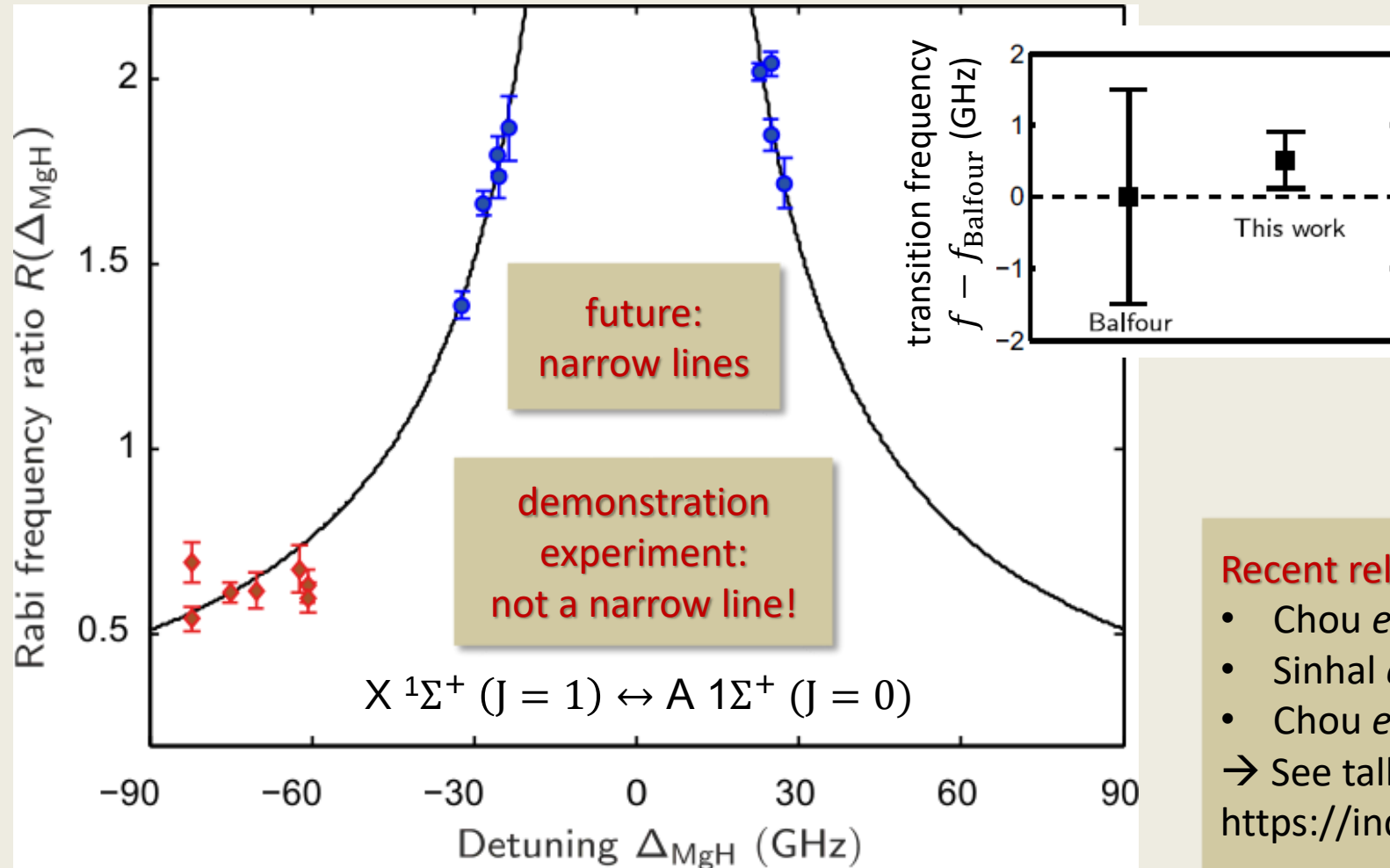
motion-
spin
mapping

detect



[Wolf *et al.*, Nature **530**, 457 (2016)]

Quantum Logic Spectroscopy of MgH^+



Recent related work:

- Chou *et al.*, Nature **545**, 203 (2017)
- Sinhal *et al.*, Science **367**, 1213 (2020)
- Chou *et al.*, Science **367**, 1458 (2020)

→ See talk by J. Chou

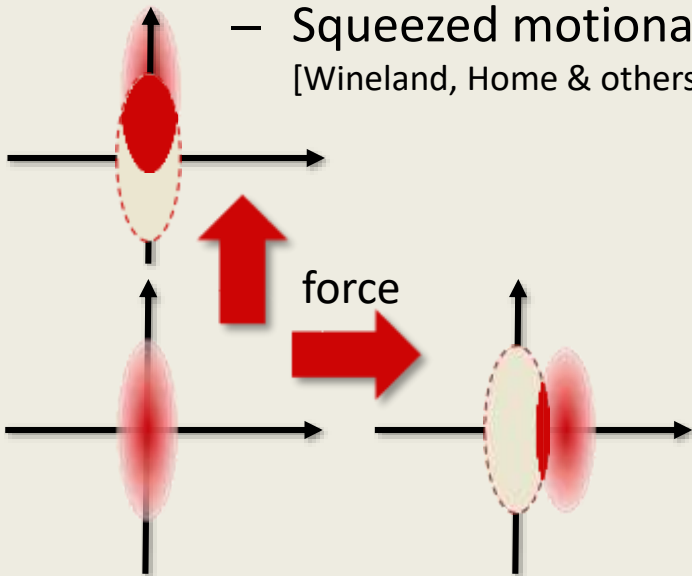
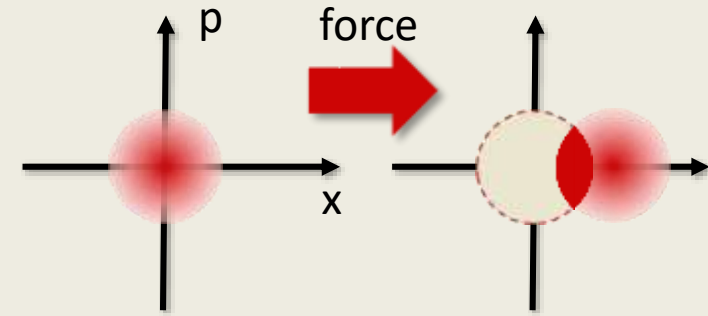
<https://indico.cern.ch/event/930162/>

$$f_0 = 1067.74789(40) \text{ THz}$$

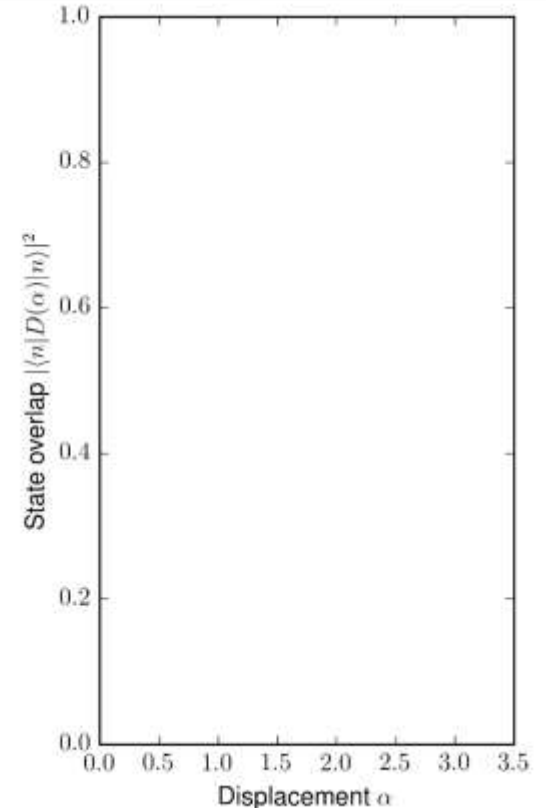
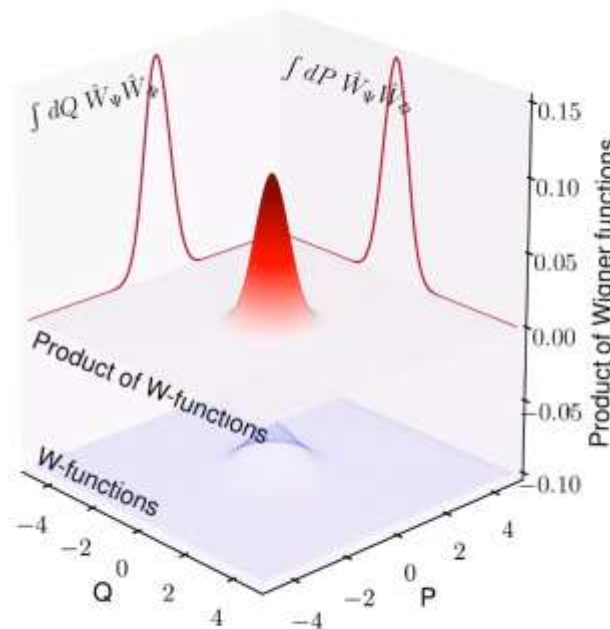
[in agreement with $f_0 = 1067.74730(150) \text{ THz}$
from Balfour *et al.*, Can. J. Phys. **50**, 1082 (1972)]

Classical detection scheme

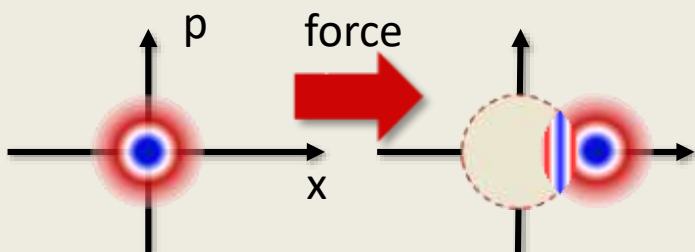
- classical scheme:
 - prepare $|0\rangle$
 - apply displacement $D(\alpha)|0\rangle$
 - overlap with $|0\rangle$: $|\langle 0|D(\alpha)|0\rangle|^2 = e^{-|\alpha|^2}$
- nonclassical schemes:
 - Schrödinger cat states
[Hempel *et al.*, Nat Photon **7**, 630 (2013)]
 - Squeezed motional states
[Wineland, Home & others]



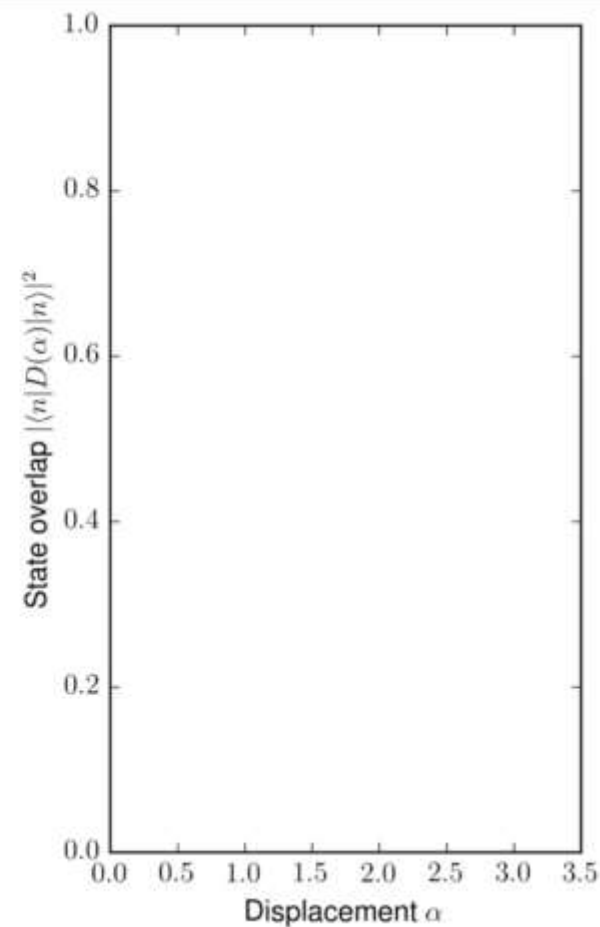
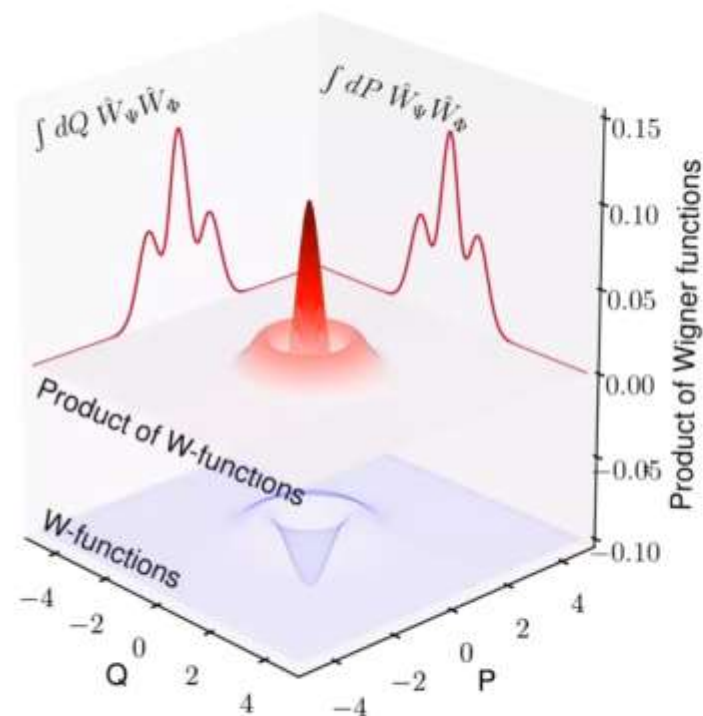
$$|\langle \Phi | \Psi \rangle|^2 = \int d\beta W_\Phi(\beta) W_\Psi(\beta)$$



Fock state interferometry

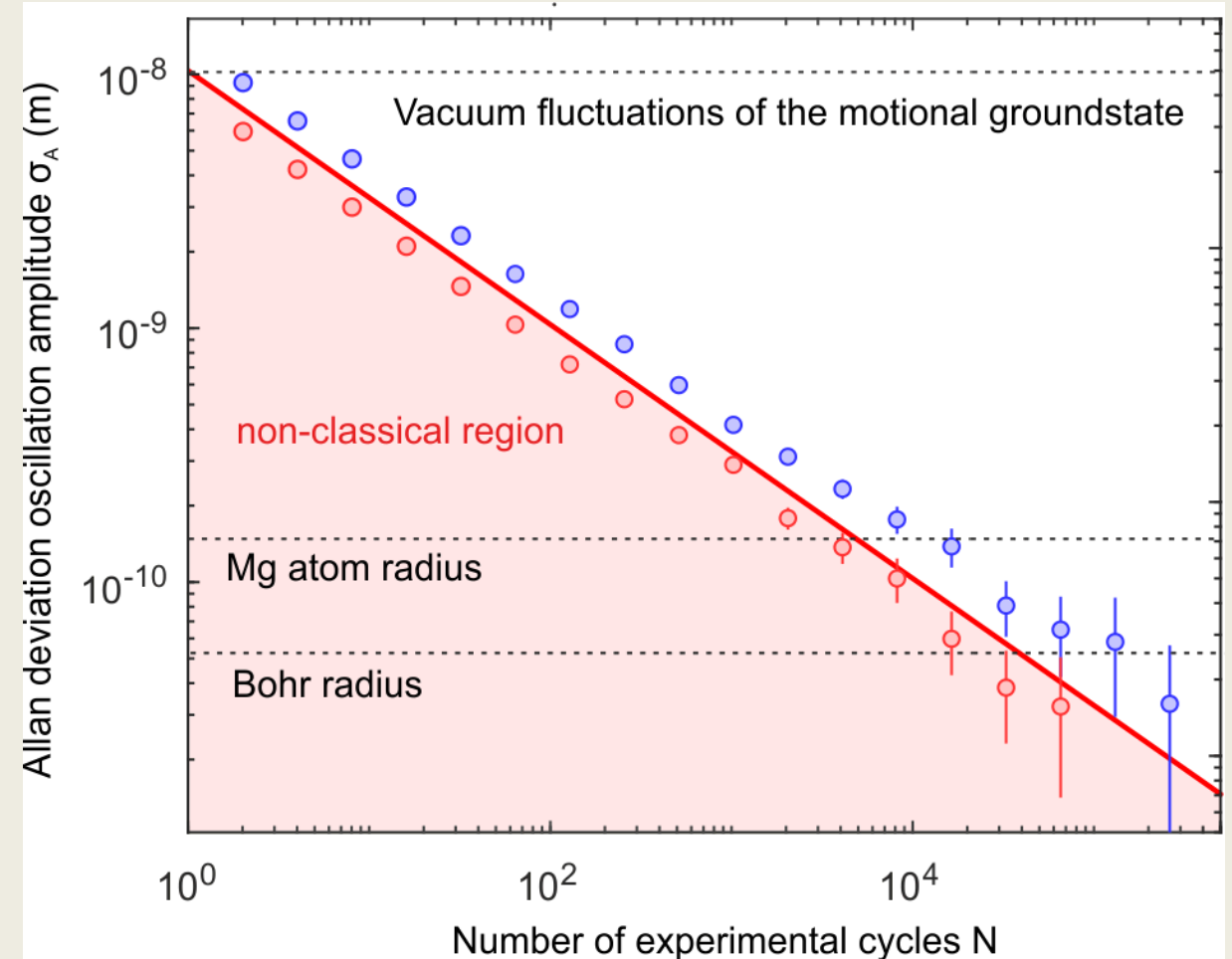


$$|\langle \Phi | \Psi \rangle|^2 = \int d\beta W_{\Phi}(\beta) W_{\Psi}(\beta)$$



Displacement/amplitude measurement

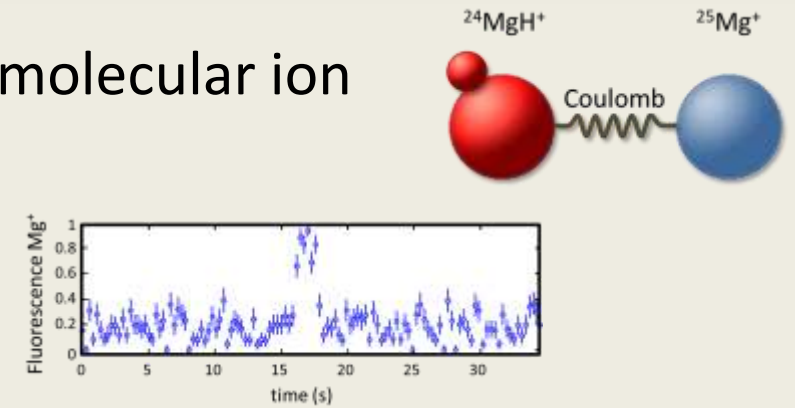
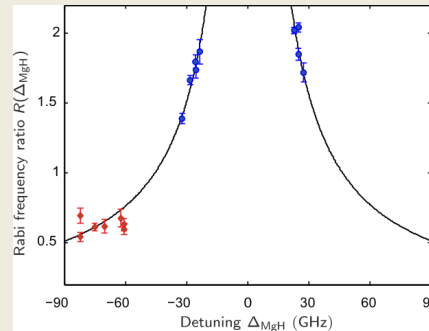
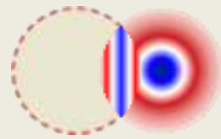
- resolution limited by measurement time (QPN)
- $n = 1$ Fock state
 - 1.3 dB sensitivity below theoretical SQL
 - 3.6 dB sensitivity below experimental SQL
- ➔ reduce averaging time by x2
- sensitivity of force measurement:
 $\sim 112 \text{ yN}/\sqrt{\text{Hz}}$



related work using classical states of motion:
[Gilmore et al. PRL **118**, 263602 (2017),
Shaniv et al., Nat. Commun. **8**, 14157 (2017),
Biercuk et al., Nat Nano **5**, 646 (2010)]

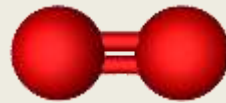
Summary & Future Molecules

- first step towards extending quantum optics control to a molecular ion
- demonstrated non-destructive state detection & simple spectroscopy
- Demonstrated sub-SQL Fock state metrology

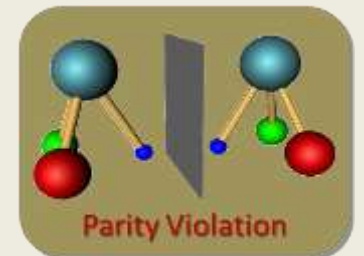


Future:

- O_2^+ spectroscopy
[F. Wolf et al., arXiv:2002.05584]
- deterministic state preparation
 - ➔ full control over molecular state
 - ➔ high-precision spectroscopy
 - ➔ towards applications in chemistry, molecular & fundamental physics



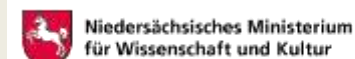
Dream: probe for parity violation in chiral molecules



Quantum Logic Spectroscopy Group

M. Schwarz, L. Schmöger & J. Crespo

PhD/PostDoc
openings



Collaborators:

- J. Crespo López-Urrutia (MPIK, Heidelberg)
- J. Berengut (U. of New South Wales)
- K. Hammerer (LUH, Hannover)
- A. Smerzi (LENS, Florence)
- A. Retzker (U. of Jerusalem)
- A. Surzhykov (PTB & TU Braunschweig)
- M. Safronova (U. of Delaware)

HCI: P. Micke, L. Spieß, S. King, T. Leopold

Molecules: J.C. Heip, F. Wolf, M. Zawierucha

THE END