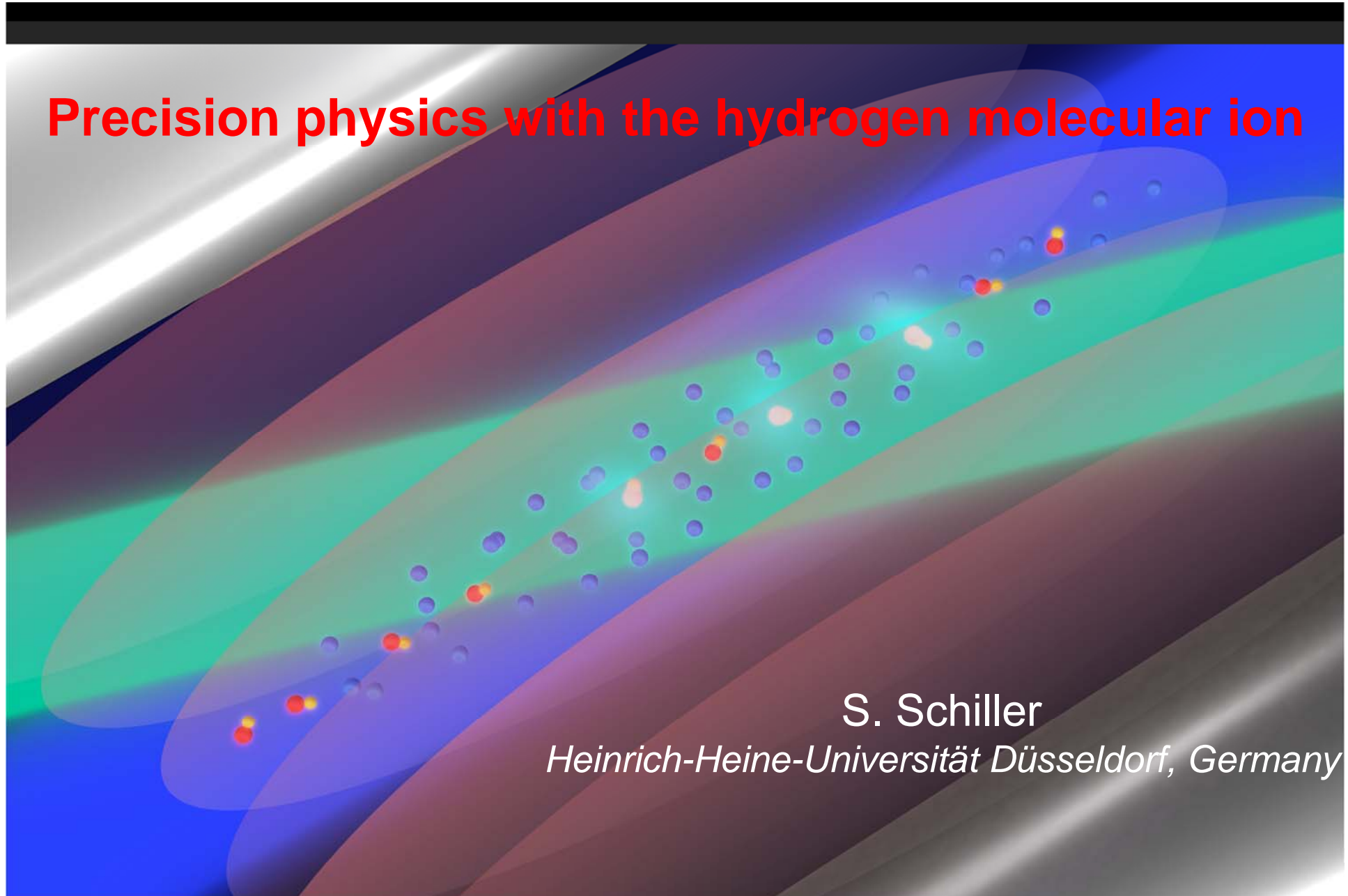


Precision physics with the hydrogen molecular ion



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Topics in this Seminar

- Test of QED
- Parity violation
- Time-dependence of fundamental constants (oscillations, drift)
- CPT Symmetry
- Lorentz Invariance
- Precision mass measurements
- Fifth force
- Proton Radius Puzzle

Motivation

Precision physics with „simple“ quantum systems

- Test QED, CPT
- Determine fundamental constants

$$R_\infty, \alpha, m_e/m_p, m_p/m_d, \\ r_p, r_d, g_e, g_p, g_{p^-} \dots \\ m_{e^+}/m_e, m_{p^-}/m_p, \dots$$

Single particles

Penning traps

Atom interferometry

$$m_e/u, m_p/u, m_d/u, \dots \\ g_e, g_p, g_{p^-}, \dots \\ m_{e^+}/m_e, m_{p^-}/m_p, \dots \\ \alpha$$

Two-body systems

H, D
Muonic hydrogen
Anti-hydrogen
positronium
muonium
H-like ions

$$R_\infty, m_e/u \\ r_p, r_d, g_e, \dots \\ R^+_\infty, \dots$$

Three-body systems

He, Li⁺
Antiprotonic He
Molecular hydrogen ions
He-like ions

$$R_\infty, \alpha \\ m_e/m_p, m_p/m_d \\ r_p, r_d, r_{He}, \dots \\ m_{p^-}/m_e$$

Precision theory is essential

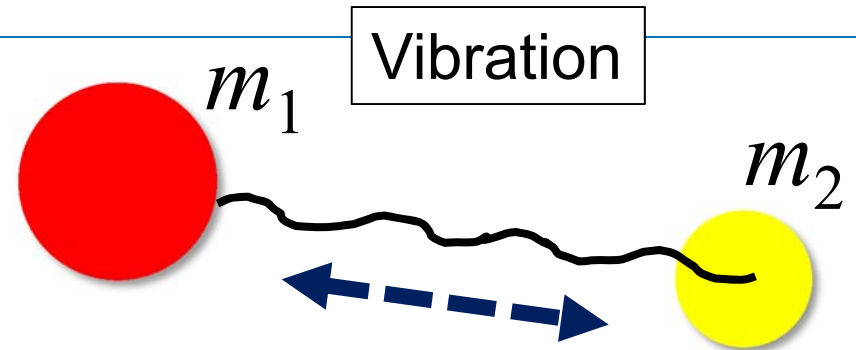
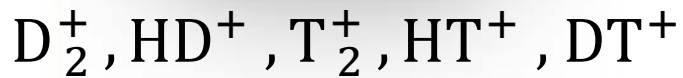
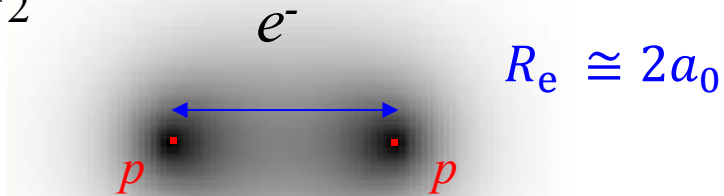
The interaction between a proton and a deuteron



$$F = F_{Coulomb} + F_{dip-dip} + F_{charge-induced\ dip} + \dots$$

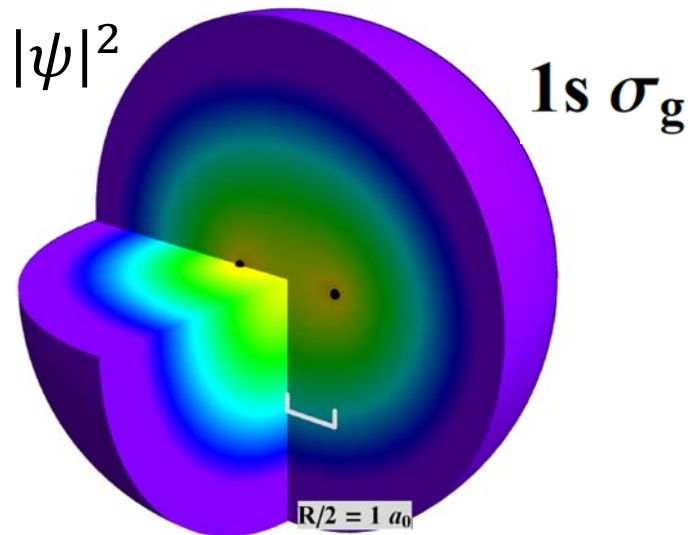
- A probe for new physics
- Cf. Experiments on short-range gravity
- Approach: precision measurement on a *bound* state containing p and d
- Bound state must contain at least 1 negatively charged particle $\rightarrow e^-$
- The interaction of p with e^- (and d with e^-) has been tested via H spectroscopy, with uncertainty 2×10^{-12} (CODATA 2018)

Molecular hydrogen ions: basics

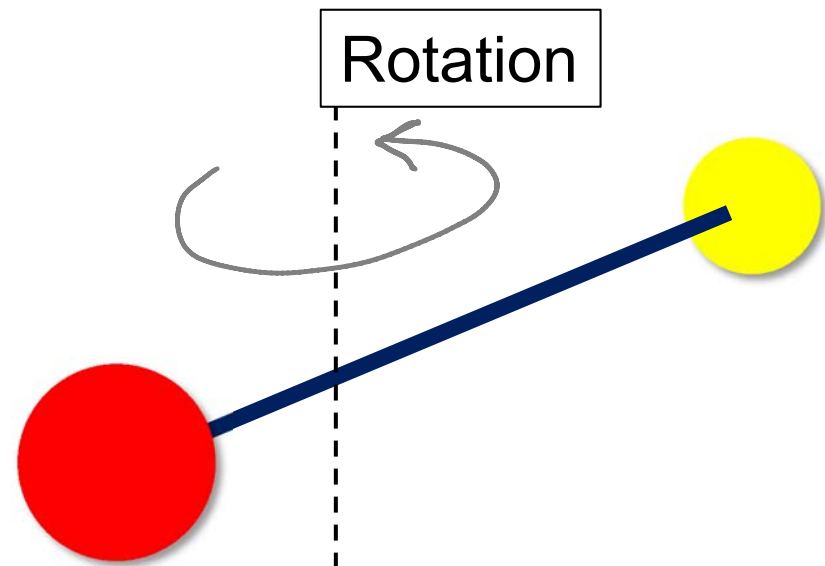


$$f_{\text{vib}} \approx cR_\infty \sqrt{m_e (1/m_1 + 1/m_2)}$$

+ QED + nuclear charge radii

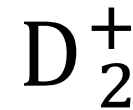
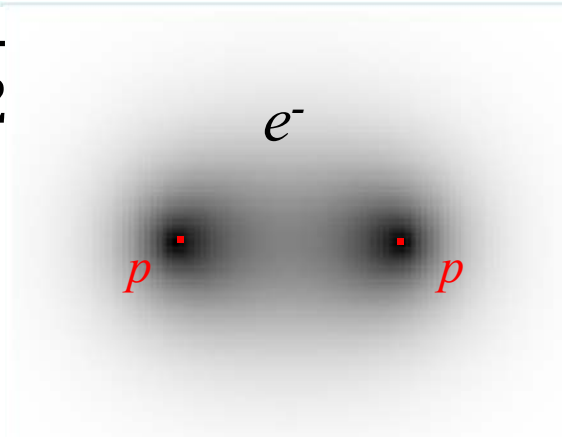
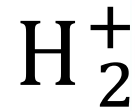


+ QED + nuclear charge radii



$$f_{\text{rot}} \approx cR_\infty m_e (1/m_1 + 1/m_2)$$

Molecular Hydrogen Ions: fundamental quantum systems



- Since 1927, a fundamental system of quantum physics [1]
- Test of QED in molecular systems [2]
- Determination of fundamental constants [3,4,8]
- Search for Physics beyond the Standard Model [5,9]
- Molecular clock: candidate for tests of time-independence of fundamental constants [6]
- „Test molecule“ for novel manipulation methods [3,7]
- Mirror image to anti-H₂⁺ [10]

[1] *Reviews:*

Leach and Moss, *Annu. Rev. Phys. Chem.* 46, 55 (1995)

Roth et al, *Springer Lect. Notes in Phys.* 745, 205 (2007)

Zhong et al. *Chin. Phys. B* 24, 053102 (2015)

[2] Koelemeij et al. *PRL* 98, 173002 (2007);

[3] Bressel et al. *PRL* 108, 183003 (2012),

[4] Biesheuvel et al. *Nat. Comm.* 7, 10385 (2015) .

[5] Salumbides et al. *PRD* 87, 112008 (2013)

[6] Bakalov and Schiller, *Appl. Phys. B.* 114, 213 (2014);

Schiller et al. *PRL* 113, 023004 (2014)

Karr, *J. Mol. Spec.* 300, 37 (2014)

[7] Schneider et al. *Nat. Phys.* 6, 275 (2010),

[8] Alighanbari et al. *Nat. Phys.* 14, 555 (2018)

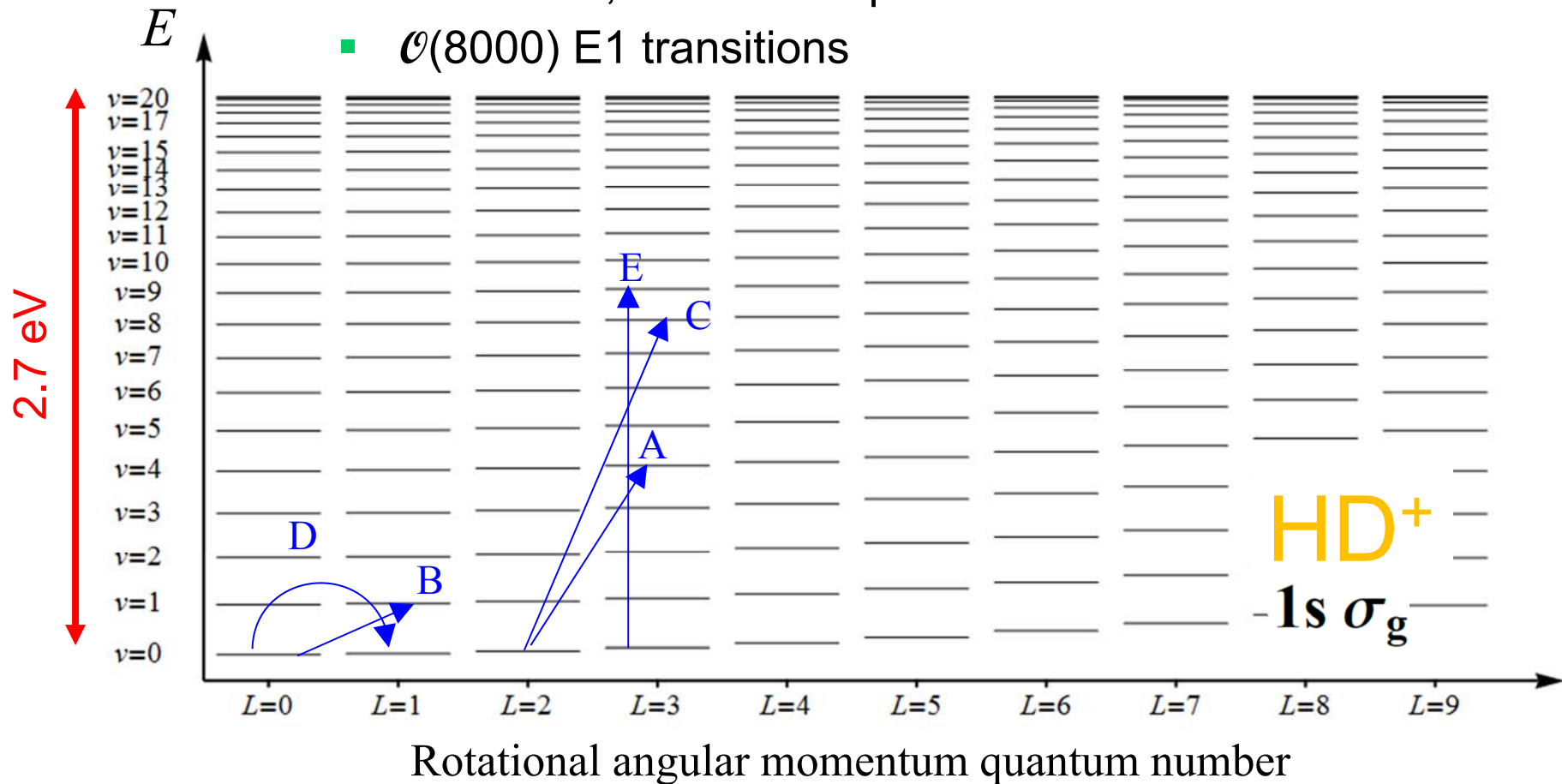
[9] Salumbides et al. *New J. Phys.* 17 (2015) 033015

[10] Dehmelt, *Phy. Scr.* T59, 432 (1995);

Myers *PRA* 98, 010101 (2018)

The richness of the rotation-vibration level structure

- 637 levels, 80% have spontaneous lifetimes ≥ 10 ms
- $\mathcal{O}(8000)$ E1 transitions



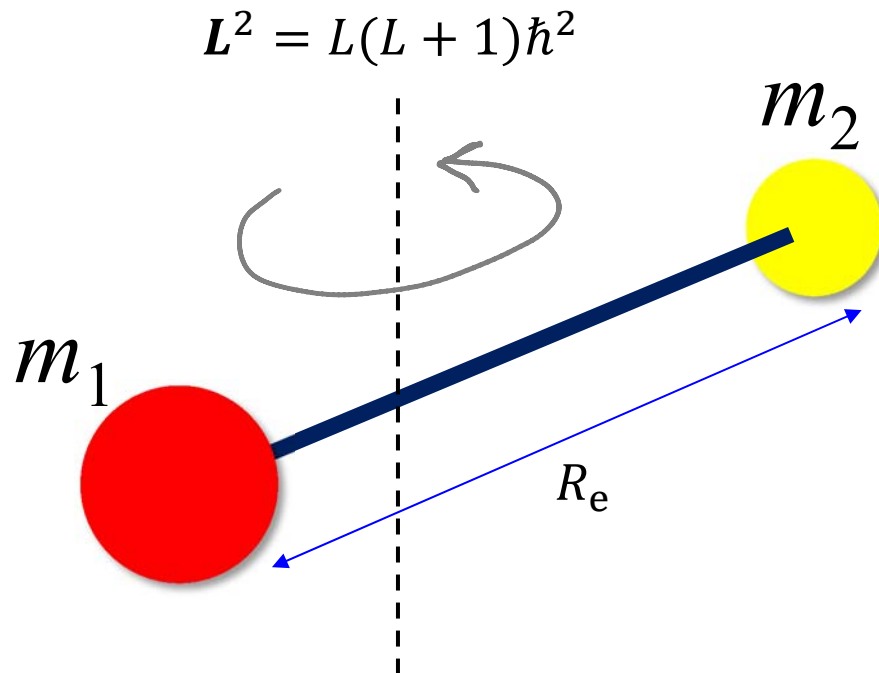
Trapped-ion measurements since year 2000:

A, B, D: Düsseldorf (2006, 2007, 2012, 2018, 2019)

C, E: Amsterdam (2012, 2016, 2019)

Rotating molecular hydrogen ions

... one of the simplest motions of a quantum system



$$E_{\text{rot}} = \frac{L^2}{2I}$$

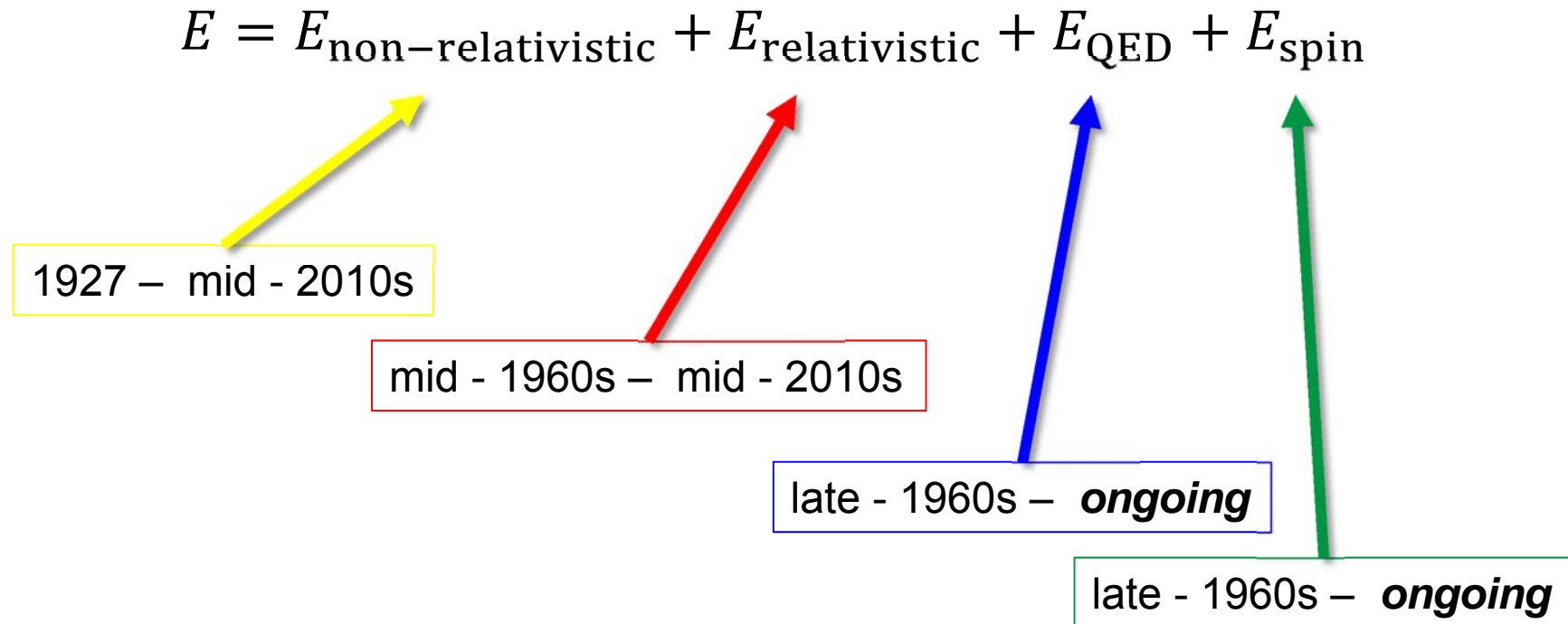
$$E_{\text{rot}}(L) \approx L(L + 1) \left(\frac{R_e}{a_0}\right)^{-2} \frac{m_e}{\mu_{12}} E_R$$

$$f_{\text{rot}}(L = 0 \rightarrow L' = 1) \approx 2 \left(\frac{R_e}{a_0}\right)^{-2} \frac{m_e}{\mu_{pd}} cR_{\infty} \approx 1.3 \text{ THz}$$

- HD⁺: rotational transitions are E1-allowed (Shen et al. PRA 85, 032519 (2012))

Ab-initio theory of molecular hydrogen ions

Theory of MHIs: approaching a century of efforts



Progress in the theoretical solution of the nonrelativistic problem

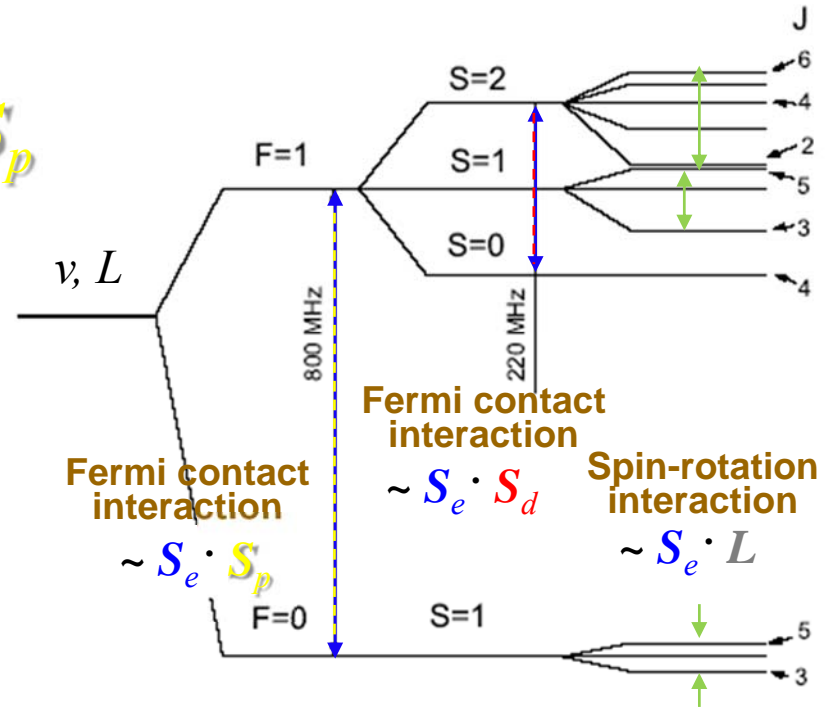
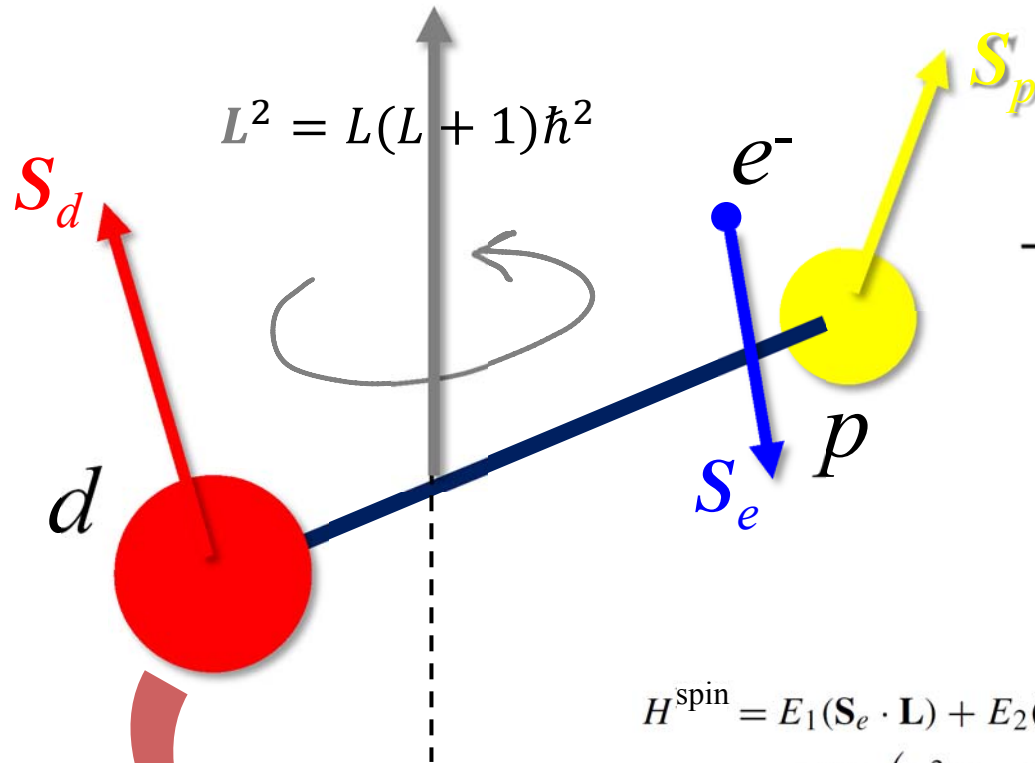
$$\hat{H} \psi(R, r_a, r_b) = E \psi(R, r_a, r_b)$$

Year	Method	f_{rot} (10^{-4} atomic units)	Reference
1967	Non-adiabatic (partial)	1.9975	Hunter and Pritchard
1977	Adiabatic	1.998508	Bishop and Cheung
1977	Variational (partial)	1.99839	Bishop and Cheung
1989	Adiabatic	1.9985	Moss and Sadler
1989	Transformed H, partially non-adiabatic	1.9985	Moss and Sadler
1989	Transformed H, variational, Σ	1.9985	Moss and Sadler
1989	Transformed H, variational, Σ, Π	1.9984	Moss and Sadler
1990	Transformed H, scattering	1.99841	Balint-Kurti et al.
1999	Transformed H, variational	1.998404167	Moss
1999	Transformed H, scattering	1.998404167	Moss
2005	Variational	1.99840416720	Schiller and Korobov
2006	Variational	1.9984041668	Korobov
2006	Variational	1.9984041668	Karr and Hilico
2014	Variational	1.9984041668069(1)	Tang et al.
2018	Variational	1.998404166825285	Alighanbari et al.

Experimental accuracy (2020) 

All rescaled to CODATA2014

Spin structure



Electric quadrupole moment

$$\begin{aligned}
 H^{\text{spin}} = & E_1(\mathbf{S}_e \cdot \mathbf{L}) + E_2(\mathbf{S}_p \cdot \mathbf{L}) + E_3(\mathbf{S}_d \cdot \mathbf{L}) + E_4(\mathbf{S}_e \cdot \mathbf{S}_p) + E_5(\mathbf{S}_e \cdot \mathbf{S}_d) \\
 & + 2E_6 \left(\mathbf{L}^2(\mathbf{S}_e \cdot \mathbf{S}_p) - 3(\mathbf{S}_p \cdot \mathbf{L})(\mathbf{S}_e \cdot \mathbf{L}) \right) \\
 & + 2E_7 \left(\mathbf{L}^2(\mathbf{S}_e \cdot \mathbf{S}_d) - 3(\mathbf{S}_d \cdot \mathbf{L})(\mathbf{S}_e \cdot \mathbf{L}) \right) \\
 & + 2E_8 \left(\mathbf{L}^2(\mathbf{S}_p \cdot \mathbf{S}_d) - 3(\mathbf{S}_p \cdot \mathbf{L})(\mathbf{S}_d \cdot \mathbf{L}) \right) \\
 & + E_9 \left(\mathbf{L}^2 \mathbf{S}_d^2 - \frac{3}{2}(\mathbf{S}_d \cdot \mathbf{L}) - 3(\mathbf{S}_d \cdot \mathbf{L})^2 \right)
 \end{aligned}$$

Ab initio theory of rotational frequency

- Non-relativistic Schrödinger equation $f_{\text{rot,non-rel}} = 1\,314\,886\,776.526$ kHz
(zero-point vibration,... exact)
- Relativistic corrections $\propto \alpha^2$ $\approx 4 \times 10^{-5}$
 Contributions from finite nuclear size: deuteron: $\approx -3 \times 10^{-9}$
 proton: $\approx -5 \times 10^{-10}$
- QED corrections and relativistic corr. ($\propto \alpha^3, \alpha^4, \dots$) $\approx -7 \times 10^{-6}$
- Total: $f_{\text{rot,spin-avg}} = 1\,314\,925\,752.896(18)$ kHz

Theoretical uncertainty: $u_r(f_{\text{rot,spin-avg}}) \approx 1.4 \times 10^{-11}$
 [excl. CODATA uncertainties]

$$f_{\text{rot,spin-avg}} \propto R_\infty m_e (1/m_p + 1/m_d)$$

- Spin structure $f_{\text{spin}} = \mathcal{O}(10 \text{ MHz}) \approx 1 \times 10^{-5}$

Korobov, Hilico, Karr, *Phys. Rev. Lett.* 118, 233001 (2017)

Alighanbari et al. *Nature Phys.* 14, 555 (2018)


Alighanbari et al., *Nature* (2020) 10.1038/s41586-020-2261-5


Using CODATA 2018

Sensitivity to the fundamental constants

$$f_{\text{spin-avg}}^{(\text{theor})} \approx 1,314,925,752,896(18)_{\text{theory}} \text{ Hz}$$

$$+ (2.5 \Delta R_{\infty,r} - 80 \Delta \mu_{p,r} - 24 \Delta M_{d,r} - 2.9 \Delta r_{p,r} - 2.9 \Delta r_{d,r}) \text{ Hz}$$


$$\mu_p = m_p/m_e$$


$$M_d = m_d/m_p.$$

with the normalized deviations of the fundamental constants from their nominal values:

$$\Delta X_r = \frac{X - X(\text{CODATA 2018})}{u(X(\text{CODATA 2018}))}$$

Precision spectroscopy of MHs: a half-century of efforts

1960s – end 80s: magnetic resonance and spin resonance in traps

Dehmelt,...
Jefferts,
Werth,...

~ 1969 – 70: classical optical and photoelectron spectroscopy

Herzberg
Takezawa
Asbrink,...

mid - 1970s – end 1990s: ion beams

Wing,...
Carrington, ...
McNab,...

late 1980s – **ongoing**: Rydberg states of *neutral* molecular hydrogen

Lundeen
Merkt,...

Early 2000s – **ongoing**: Trapped and sympathetically laser-cooled

Düsseldorf,
Amsterdam,
Paris,...

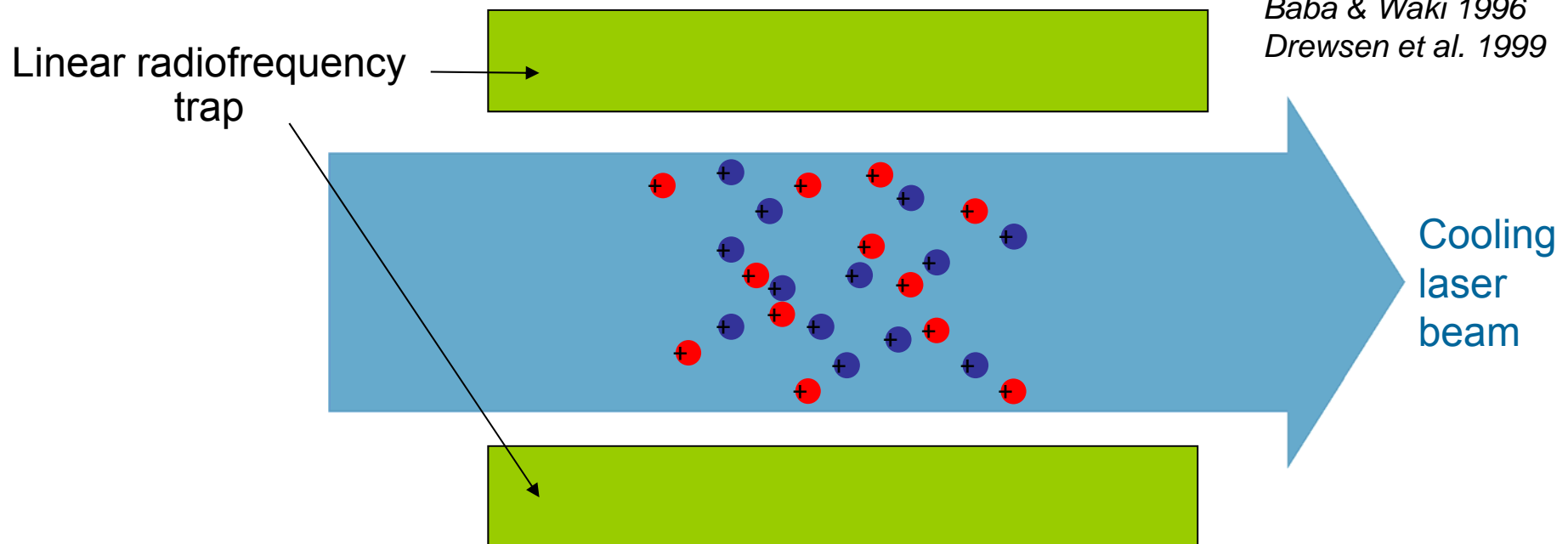
Sympathetic cooling of charged particles

- Efficient, since Coulomb force is long-range
- General method: independent of nature of particle (only charge and mass relevant)
- Final state: Coulomb „crystal“ (cluster) (particles on sites, well-separated)

First experiments:

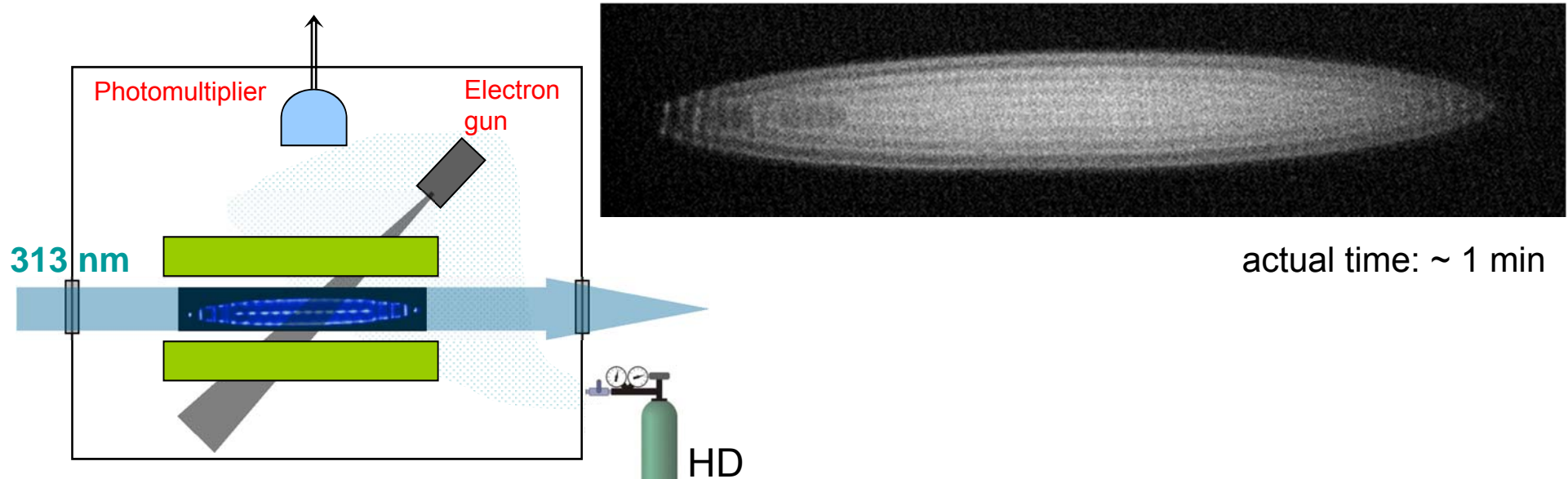
Penning trap: Drullinger et al. 1980
Larson et al. 1986

Paul trap: Diedrich et al. 1987
Waki et al 1992
Raizen et al 1992
Baba & Waki 1996
Drewsen et al. 1999



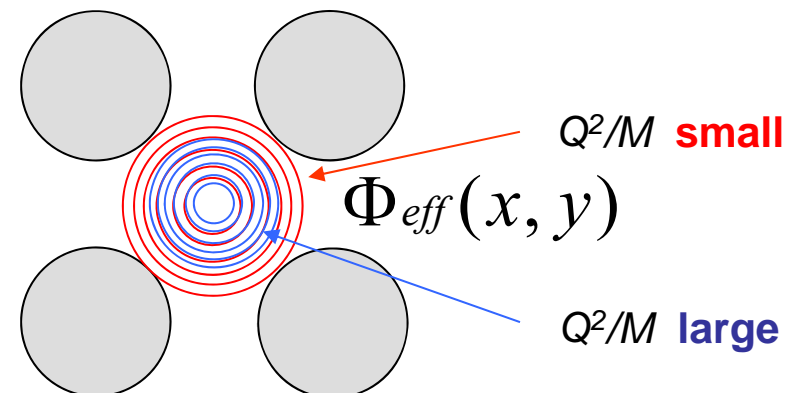
Sympathetic cooling of HD⁺

Blythe et al., PRL 95,183002 (2005)



- Load Be⁺ ; laser-cool
- Neutral gas inlet via leak valve at $\sim 3 \cdot 10^{-10}$ mbar
- Ionised by a 200 eV electron beam
- Loading for 1 - 5 s

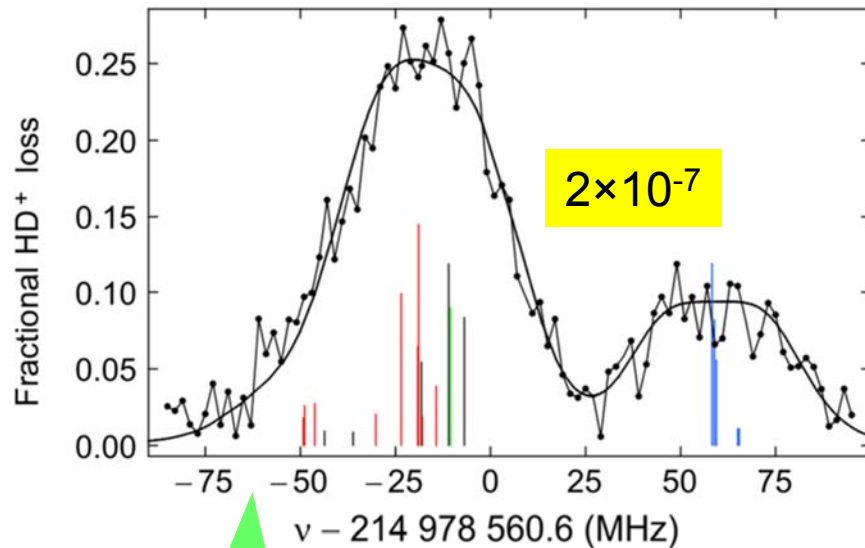
- Lighter particles are closer to axis:



Rovibrational spectroscopy of trapped and sympathetically cooled molecular ions

2007

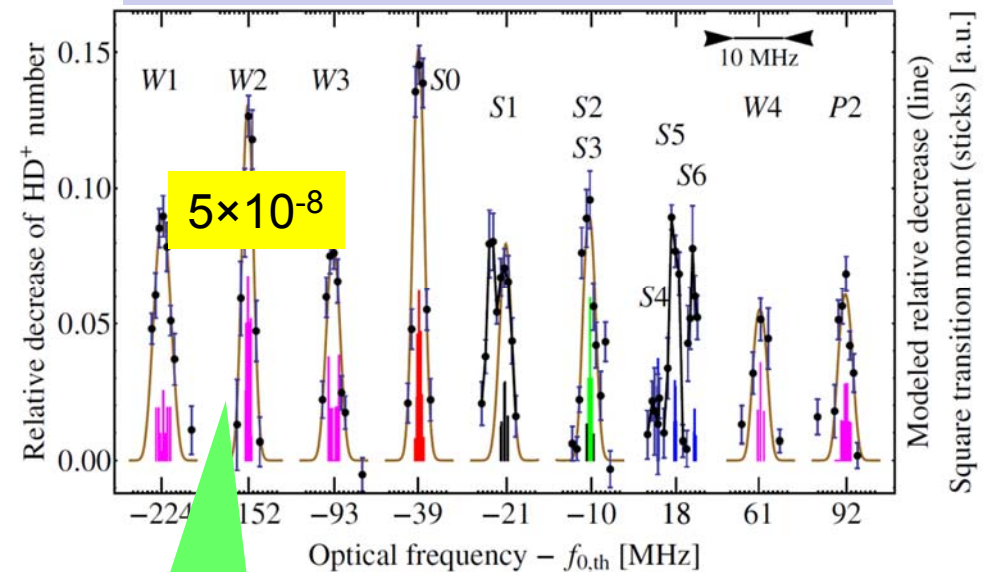
Rovibrational, overtone (1.4 μm)



Doppler-limited

2011

Rovibrational, fundamental (5 μm)



Doppler-limited

Koelmeij, et al., Phys. Rev. Lett. 98 (2007) 173002

Bressel, et al., Phys. Rev. Lett. 108 (2012) 183003

High-resolution rotational spectroscopy of HD⁺

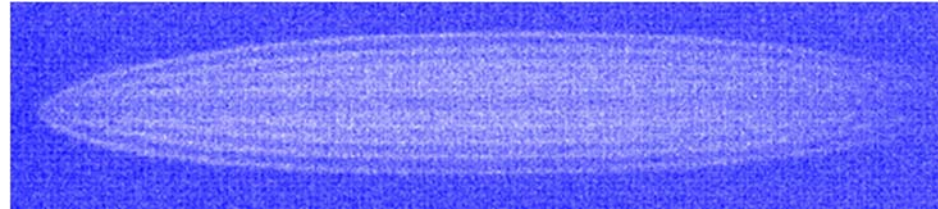
Why rotational spectroscopy?

- ✓ Doable
- ✓ Allows precision measurement of hyperfine structure
- ✓ Allows characterization of systematic shifts
- ✓ Allows improving the mass ratio values

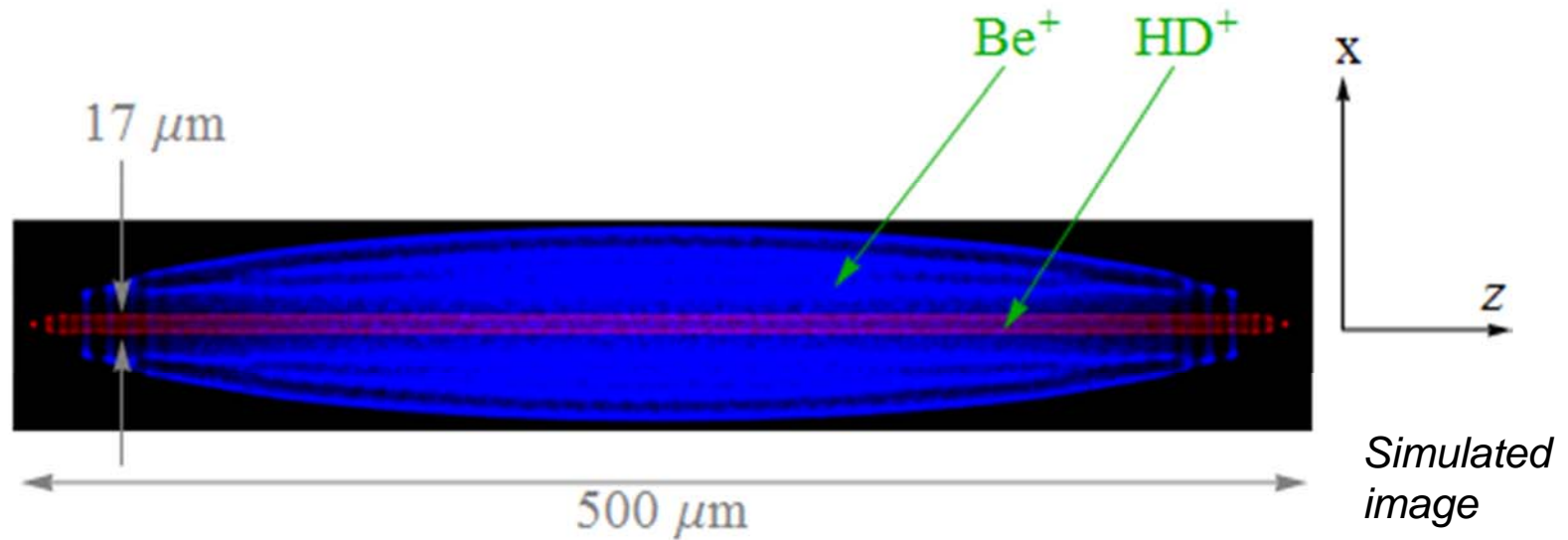
.... little substantial progress in spectroscopic resolution of rotational spectroscopy resolution in ~ 40 years

TICTES: Trapped ion cluster transverse excitation spectroscopy

S. Alighanbari et al, *Nat. Phys.* 14, 555 (2018)



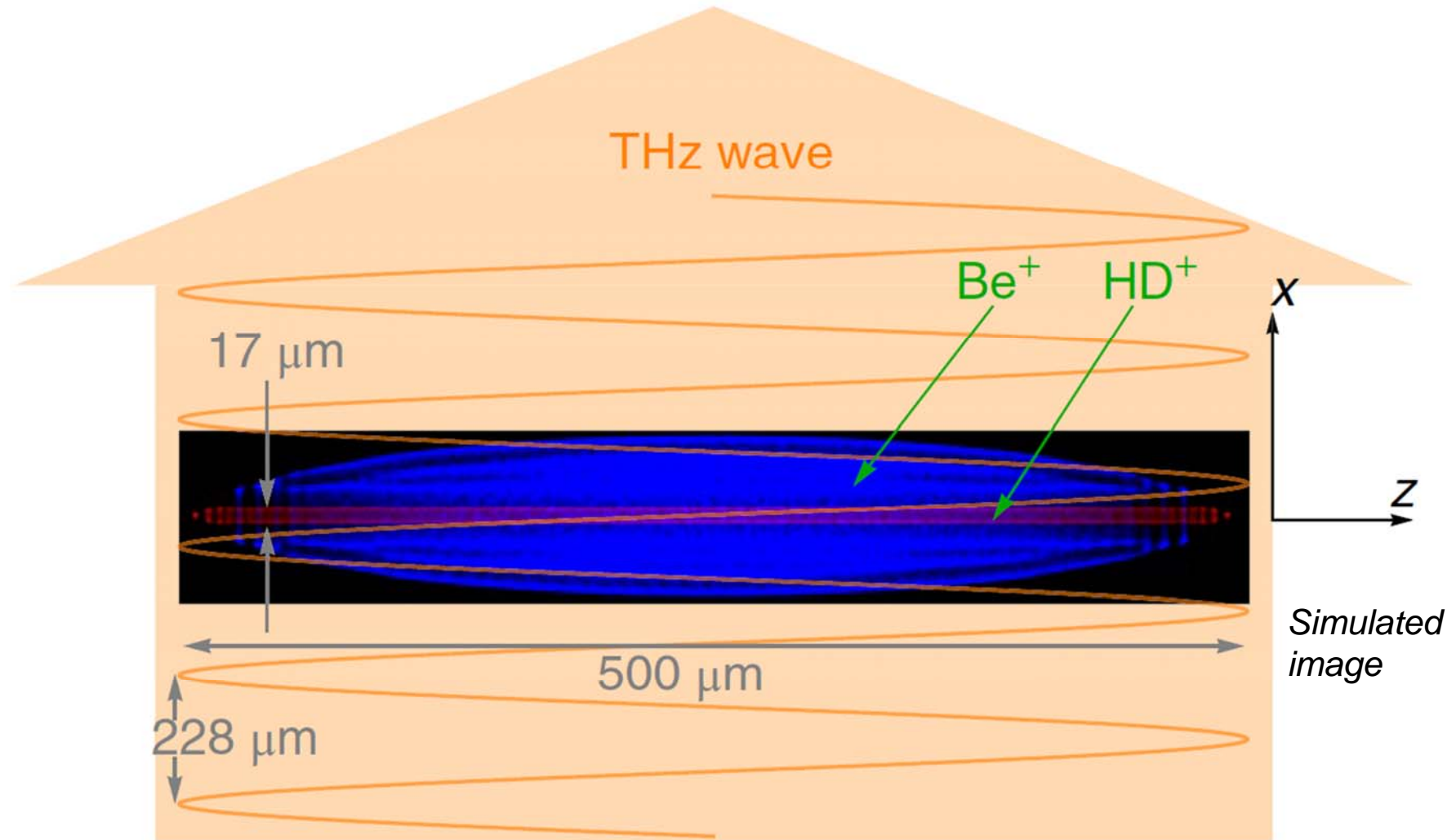
Coulomb cluster;
sympathetically
cooled HD^+ [1]



[1] Blythe et al., *PRL* 95, 183002 (2005)

TICTES: Trapped ion cluster transverse excitation spectroscopy

S. Alighanbari et al, *Nature Phys.* 14, 555 (2018)

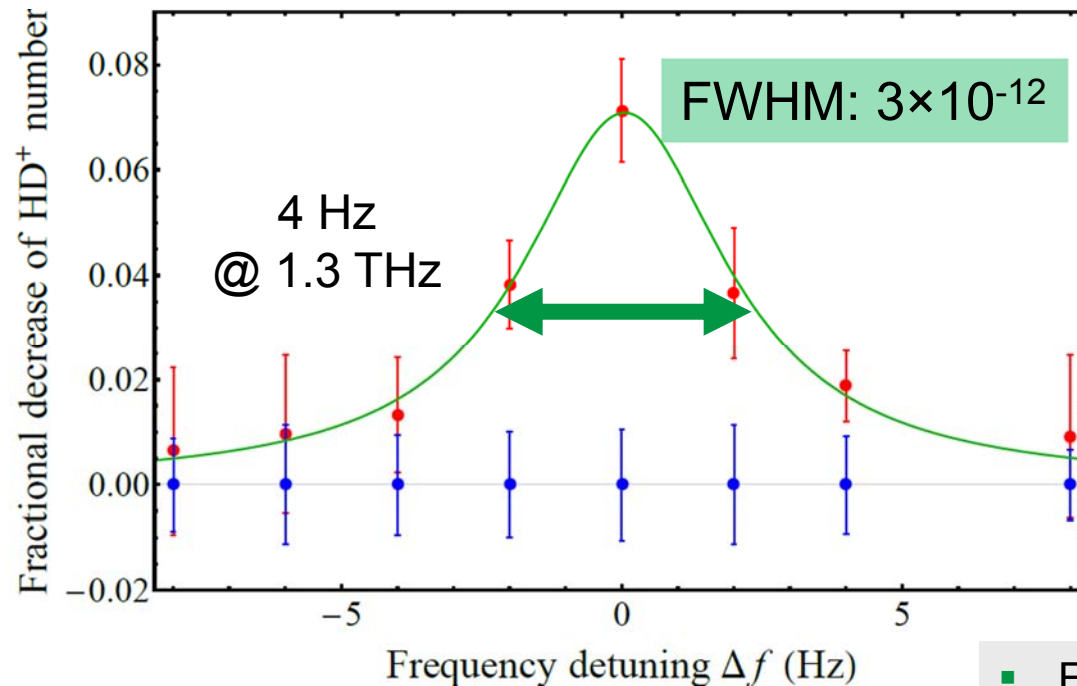


$$\Delta x \approx 8 \mu\text{m} @ 12 \text{ mK}$$
$$\rightarrow \Delta x \ll \lambda_{\text{rot}}/2\pi$$

Spectroscopy in the Lamb-Dicke regime

Spectroscopy: Doppler-free

Alighanbari et al. *Nature Phys.* 14, 555 (2018)
Alighanbari et al. *Nature* (2020)
10.1038/s41586-020-2261-5

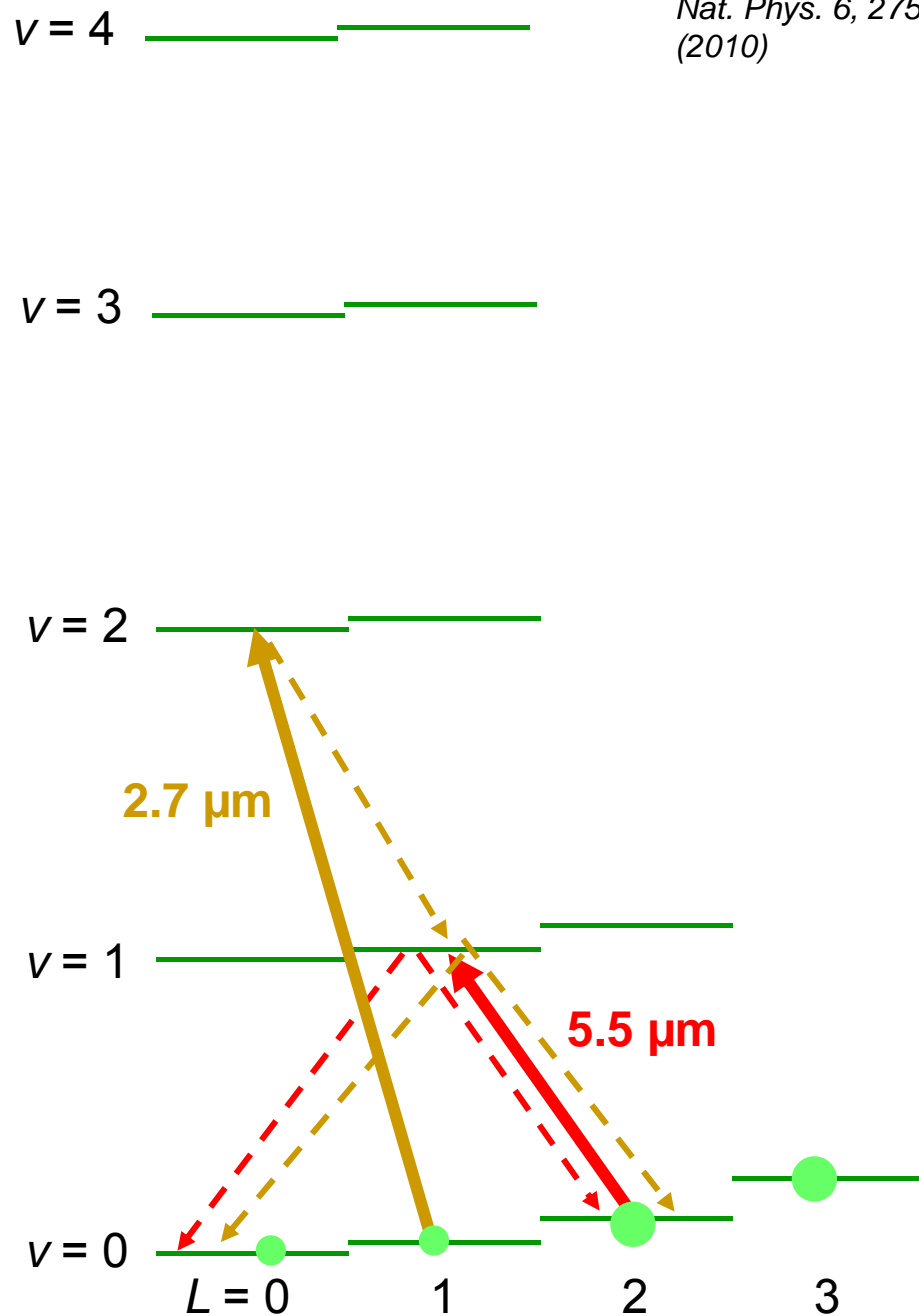


- Spectroscopy source:
frequency multiplier,
referenced to H-maser
Schiller et al., Appl. Phys. B 95, 55 (2009)

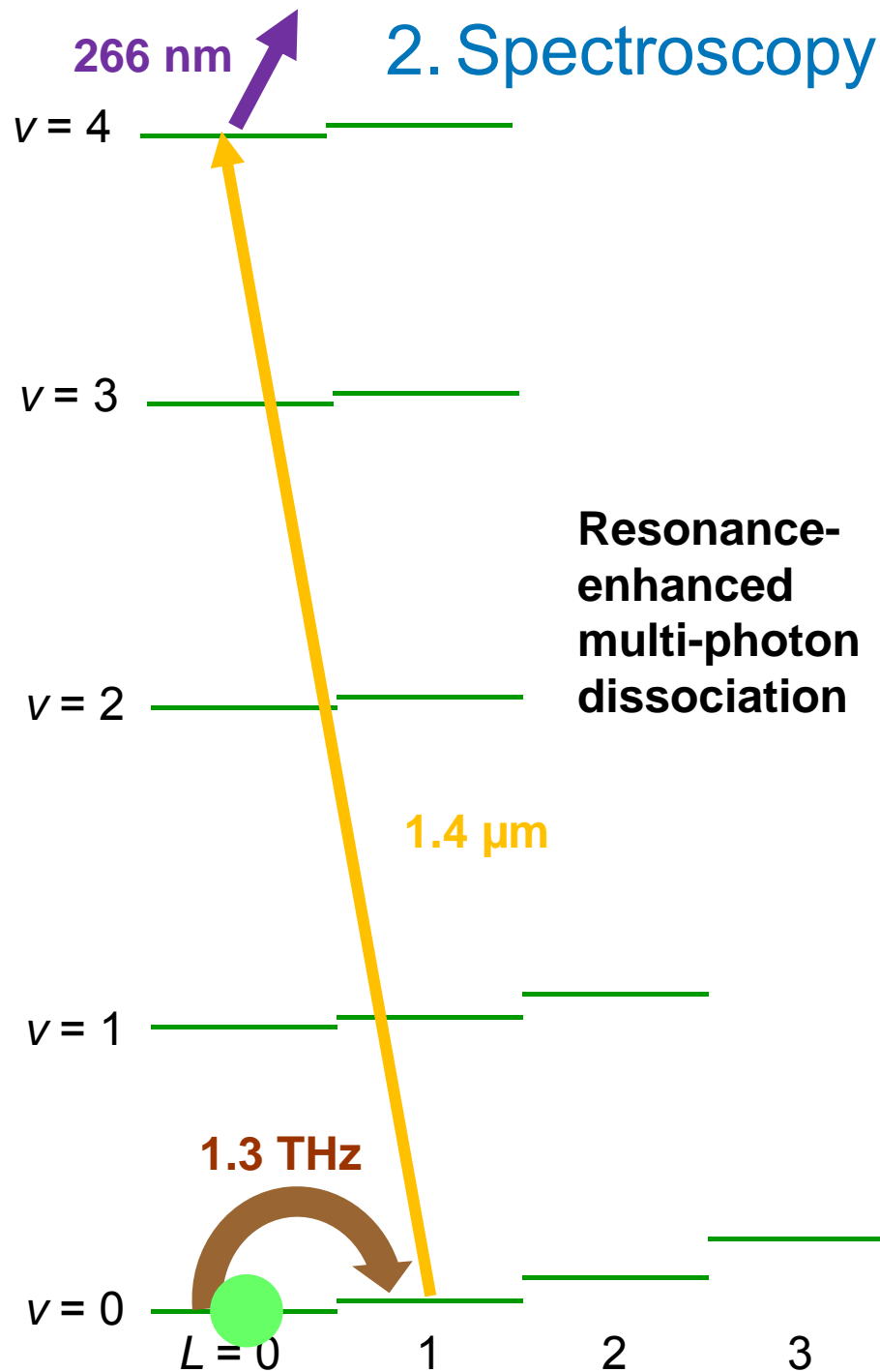
- Experimental resolution better than theory uncertainty (1.4×10^{-11}) of the spin-averaged frequency
- $> 100 \times$ better than theory uncertainty of spin frequency
- Can resolve Zeeman splittings

1. Rotational cooling

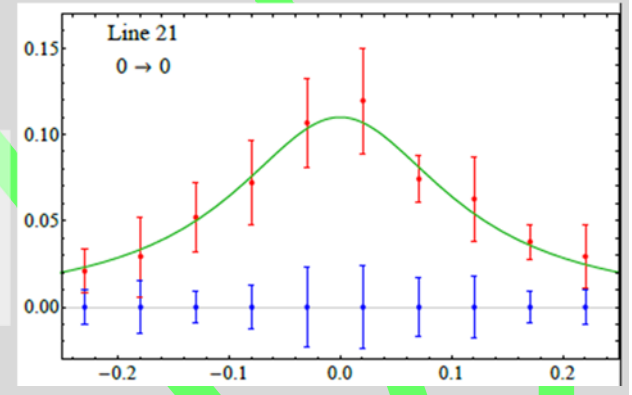
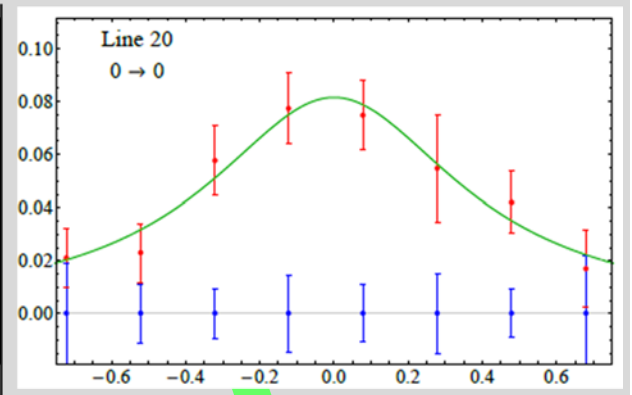
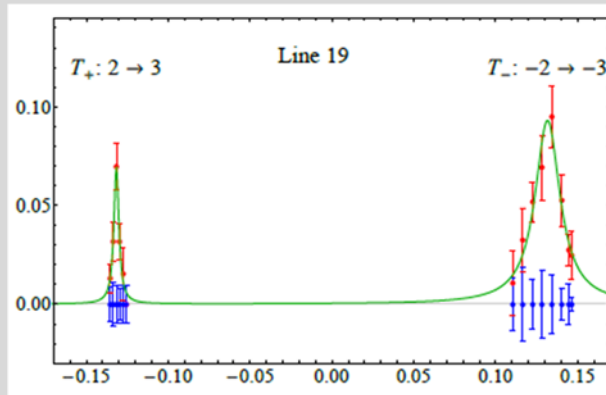
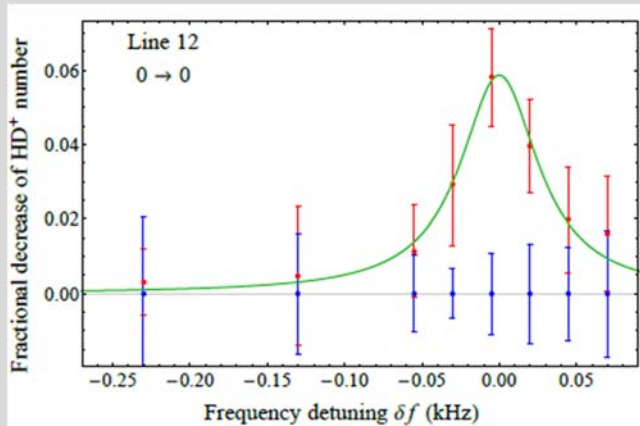
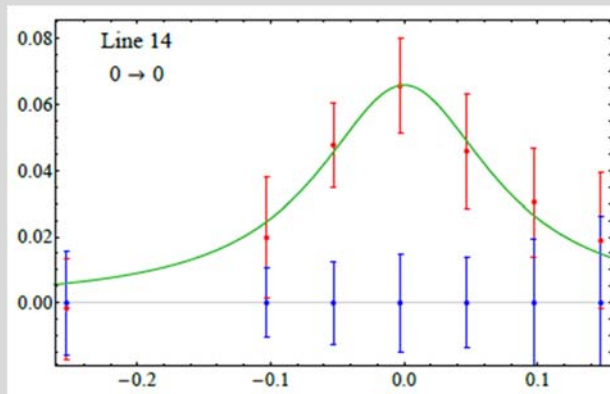
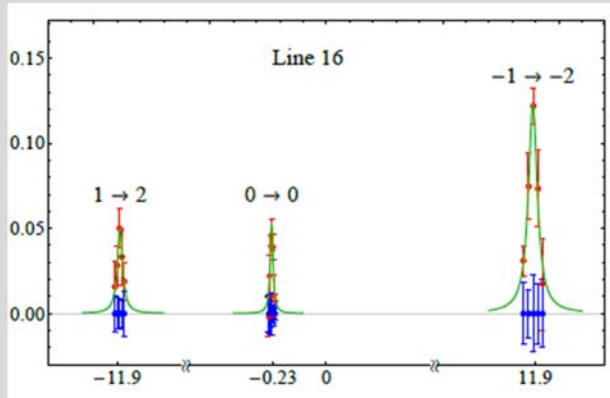
Schneider, et al.
Nat. Phys. 6, 275
(2010)



2. Spectroscopy

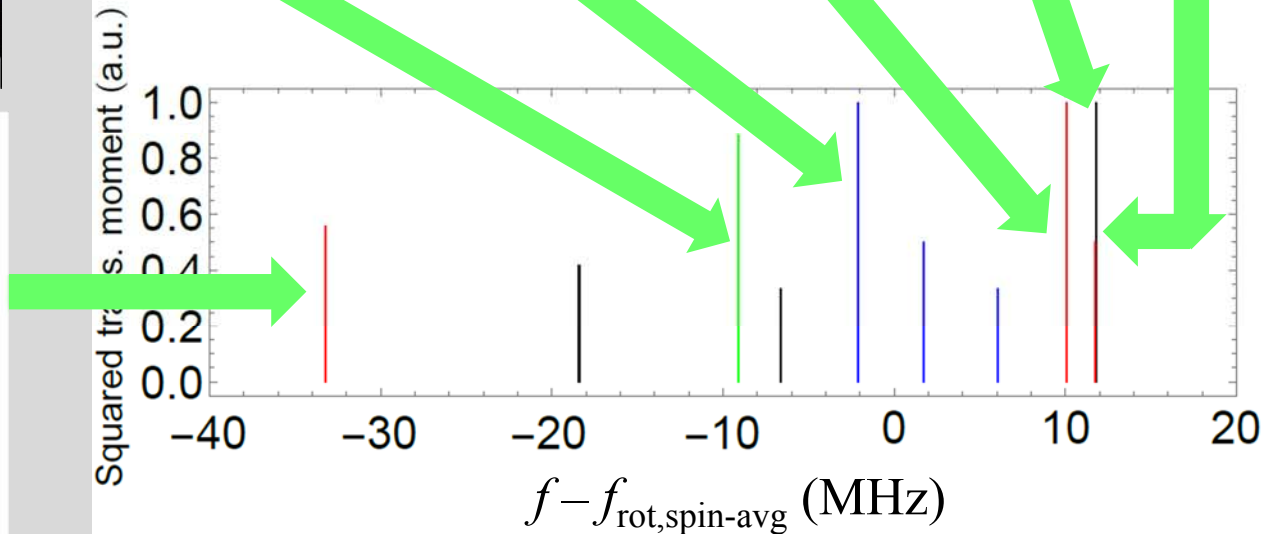


Spin structure



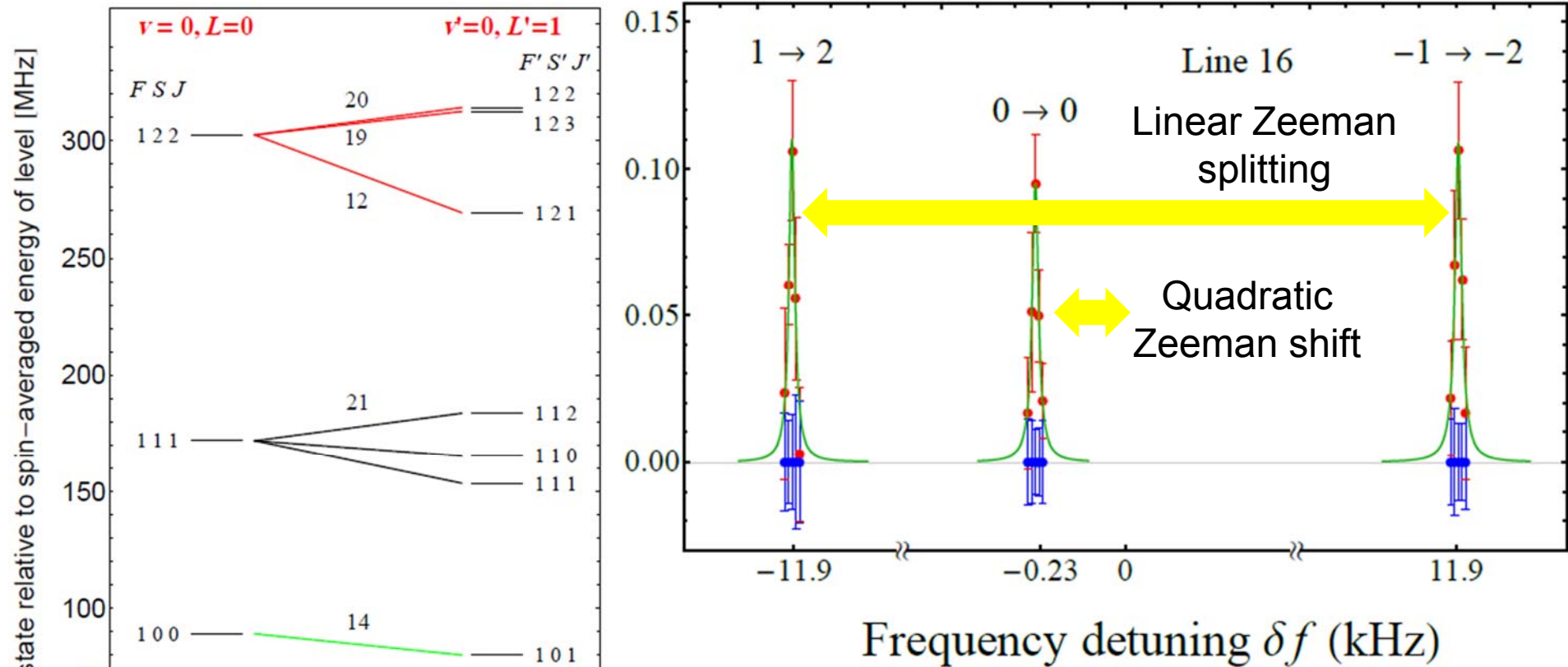
10 favoured transitions

6 measured



Theory – experiment comparison

Alighanbari et al. Nature (2020)
10.1038/s41586-020-2261-5

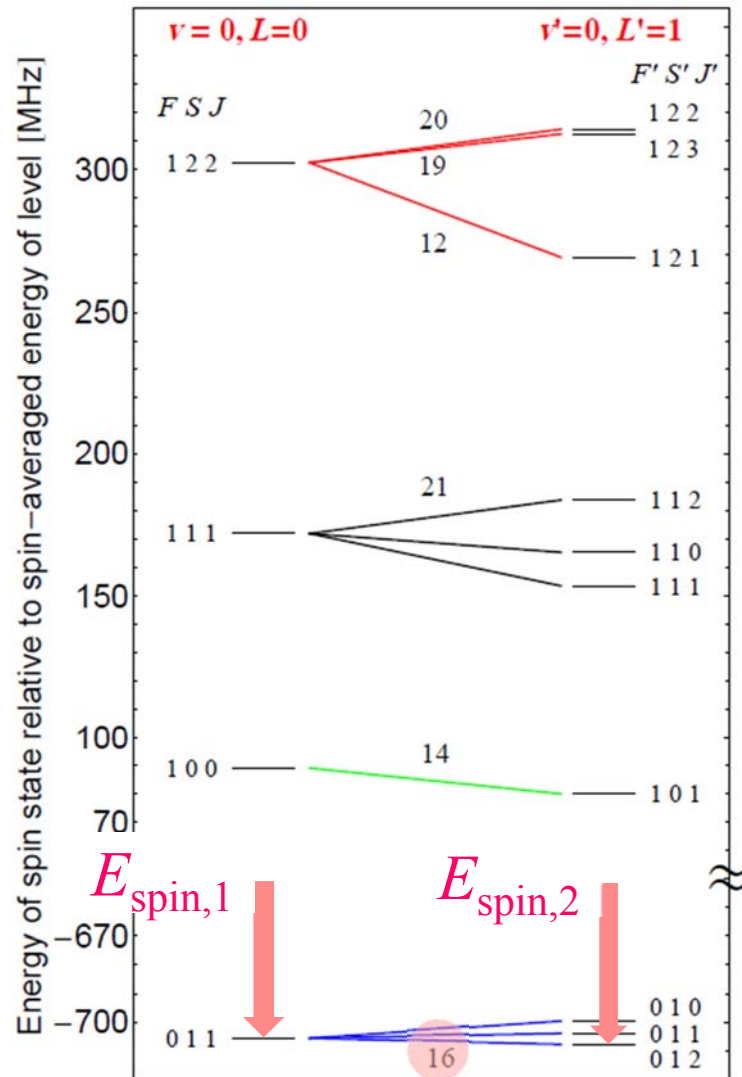


Careful characterization of the systematic shifts and corrections

Zeeman shift theory: Bakalov, Korobov, Schiller J. Phys. B 44 (2011) 025003

Theory – experiment comparison

Alighanbari et al. Nature (2020)
10.1038/s41586-020-2261-5



$$u_r(\text{exp.}) = 1.3 \times 10^{-11}$$

$$f_{16}^{(\text{exp})} = 1\,314\,923\,618.028(17) \text{ kHz}$$

$$f_{16}^{(\text{theory})} = 1\,314\,923\,617.93(20) \text{ kHz}$$

Ab initio theory
uncertainties:

$$1.5 \times 10^{-10}$$

$$f_{16}^{(\text{theor})} = f_{\text{spin-avg}} + f_{\text{spin},16}$$

$$u_r(\text{QED}) = 1.4 \times 10^{-11}$$

$$u_r(\text{CODATA}) = 4.6 \times 10^{-11}$$

Theory – experiment comparison via a composite frequency

Alighanbari et al. Nature (2020)
10.1038/s41586-020-2261-5

Form an approximation to the spin-averaged frequency:

$$f_{\text{spin-avg}}^{(\text{exp})} = \sum_{i=12,14,16,19,20,21} b_i (f_i^{(\text{exp})} - f_{\text{spin},i}^{(\text{theory})})$$

with $\sum_i b_i = 1$

Minimize spin theory uncertainty w.r.t. $\{b_i\}$:

$$u_r(\text{spin theory}) = 1 \times 10^{-12}$$

$$u_r(\text{exp.}) = 1.3 \times 10^{-11}$$

$$f_{\text{spin-avg}}^{(\text{exp})} = 1\,314\,925\,752.910(17) \text{ kHz}$$

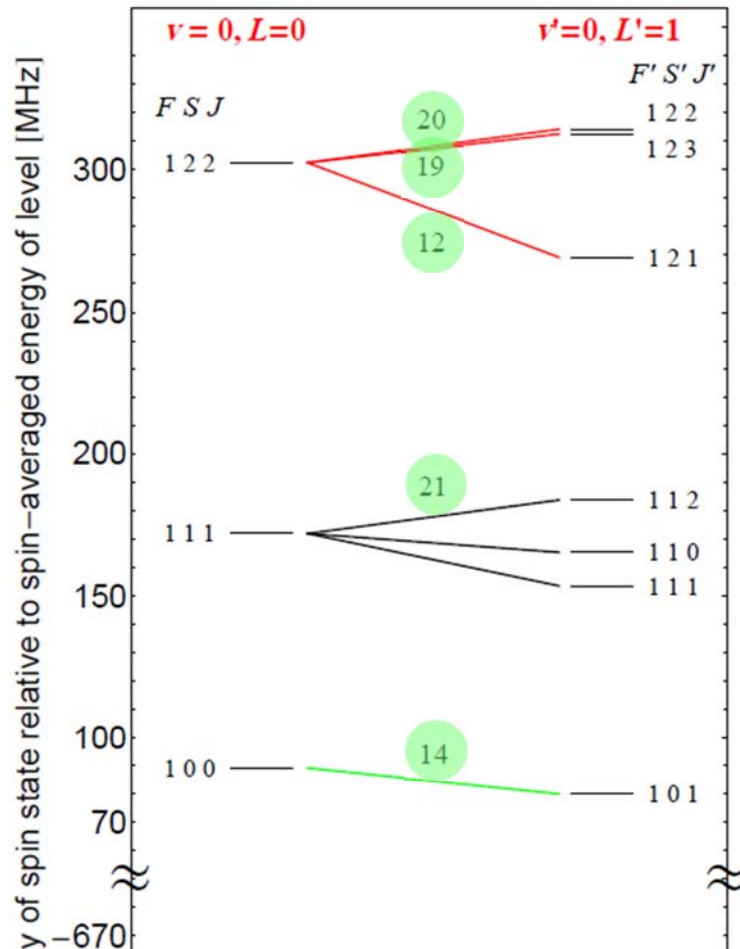
$$f_{\text{spin-avg}}^{(\text{theory})} = 1\,314\,925\,752.896(18) \text{ kHz}$$

$$u_r(\text{QED}) = 1.4 \times 10^{-11}$$

But must add uncertainty of fundamental constants in the prediction:

$$u_r(\text{CODATA}) = 4.6 \times 10^{-11}$$

$$0.061 \text{ kHz}$$



Test of three-body physics successful at 5×10^{-11} level, limited by uncertainty of fundamental constants

Interpretation

$$u(\text{exp.}) = 0.017 \text{ kHz} \\ (1.3 \times 10^{-11})$$

$$u(\text{CODATA}) = 0.061 \text{ kHz} \\ (4.6 \times 10^{-11})$$

$$u(\text{QED}) = 0.018 \text{ kHz} \\ (1.4 \times 10^{-11})$$

Deuteron's r_d^2 is confirmed at 1.5% level.
 CODATA₂₀₁₄: $u_r(r_d^2)=0.24\%$
 CODATA₂₀₁₈: $u_r(r_d^2)=0.07\%$

Proton's r_p^2 is confirmed at 10% level.
 CODATA₂₀₁₄: $u_r(r_p^2)=1.4\%$
 CODATA₂₀₁₈: $u_r(r_p^2)=0.45\%$

Term	Relative order	Contribution (kHz)	Origin
$f^{(0)}$	1	1 314 886 776.526	Solution of 3-body Schrödinger eq.
$f^{(2)}$	α^2	48 416.268 Deuteron: - 4.120(3) Proton: - 0.644(3)	Relativistic corrections in Breit-Pauli approximation; nuclear radii
$f^{(3)}$	α^3	-9 378.119	Leading-order radiative corrections (e.g. leading-order Lamb shift, anomalous magnetic moment)
$f^{(4)}$			Higher-order radiative corrections;
$f^{(5)}$	α^5	3.923(3)	Radiative corrections up to 3-loop diagrams; Wichman-Kroll contrib.
$f^{(6)}$	α^6	-0.070(18)	Higher-order radiative corrections
Total: $f_{\text{spin-avg}}^{(\text{theor})}$		1 314 925 752.896(18)	

Electric quadrupole moment of deuteron:
 Value confirmed with 1.5% frac. uncertainty

Determination of a fundamental constants combination

$$f^{(\text{theor})} = 2 c R_{\infty} \bar{f}^{(\text{theor})}(m_e/m_p, m_e/m_d, \alpha, r_p, r_d)$$

$$\bar{F}^{(\text{theor})} = \frac{\bar{f}^{(\text{theor})}(m_e/m_p, m_e/m_d, \alpha, r_p, r_d)}{m_e/m_p + m_e/m_d}$$

$$f^{(\text{theor})} = 2 c R_{\infty} \left(\frac{m_e}{m_p} + \frac{m_e}{m_d} \right) \bar{F}^{(\text{theor})}$$

True
values

Ab initio dimensionless
number;
not affected by the
CODATA2018
uncertainties of
 $\alpha, m_e/m_p, m_p/m_d, r_p, r_d, \dots$
at a relevant level

Therefore, comparison experiment – theory determines

$$\left(\frac{m_e}{m_p} + \frac{m_e}{m_d} \right) = \frac{f^{(\text{exp})}}{2 c \bar{F}^{(\text{theor})}}$$

True
values

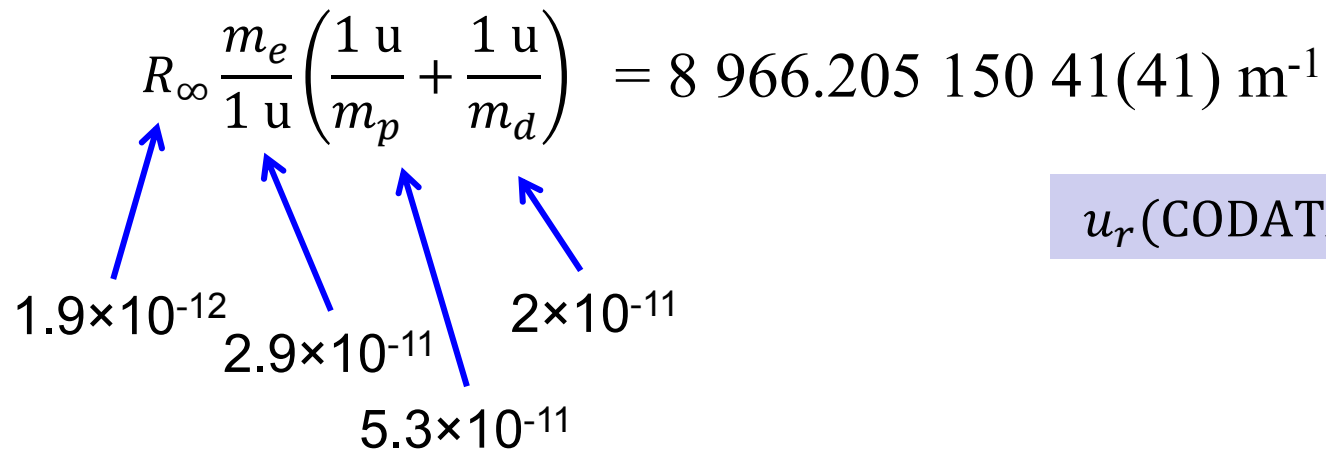
Determination of a fundamental constants combination

Alighanbari et al. Nature (2020)
10.1038/s41586-020-2261-5

$$R_\infty \left(\frac{m_e}{m_p} + \frac{m_e}{m_d} \right) = 8\,966.205\,150\,50(12)_{\text{exp}}(12)_{\text{theor}}(4)_{\text{nucl}} \text{ m}^{-1}$$

$$u_r = (1.3_{\text{exp}}, 1.3_{\text{theor}}) \times 10^{-11}$$

CODATA2018

$$R_\infty \frac{m_e}{1 \text{ u}} \left(\frac{1 \text{ u}}{m_p} + \frac{1 \text{ u}}{m_d} \right) = 8\,966.205\,150\,41(41) \text{ m}^{-1}$$


1.9 × 10⁻¹²
2.9 × 10⁻¹¹
5.3 × 10⁻¹¹
2 × 10⁻¹¹

$$u_r(\text{CODATA2018}) = 4.6 \times 10^{-11}$$

Our uncertainty is 2.4 times lower than CODATA2018

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Determination of proton mass

Alighanbari et al. Nature (2020)
10.1038/s41586-020-2261-5

From rotational spectroscopy of HD⁺

$$m_p/u = 1.007\,276\,466\,605\,(20)_{\text{exp}}(21)_{\text{theor}}(45)_{\text{CODATA2018}}$$

$$u_r = 5.3 \times 10^{-11}$$

Direct measurement with single proton in a Penning trap [1]

$$m_p/u = 1.007\,276\,466\,598\,(16)_{\text{stat}}(29)_{\text{syst}}$$

$$u_r = 3.3 \times 10^{-11}$$

[1] *F. Heiße et al., PRA 100 (2019) 022518*

Determination of proton-electron mass ratio

Alighanbari et al. Nature (2020)
10.1038/s41586-020-2261-5

From:

- rotational spectroscopy of HD^+
- atomic hydrogen spectroscopy (CODATA2018)
- single-ion Penning trap mass spectrometry of H_2^+ vs. D^+ ($\rightarrow m_d/m_p$) [3]

$$m_p/m_e = 1\,836.152\,673\,449\ (24)_{\text{exp}}(25)_{\text{theor}}(13)_{\text{CODATA2018,Fink\&Myers}}$$

$$u_r = 2.0 \times 10^{-11}$$

From:

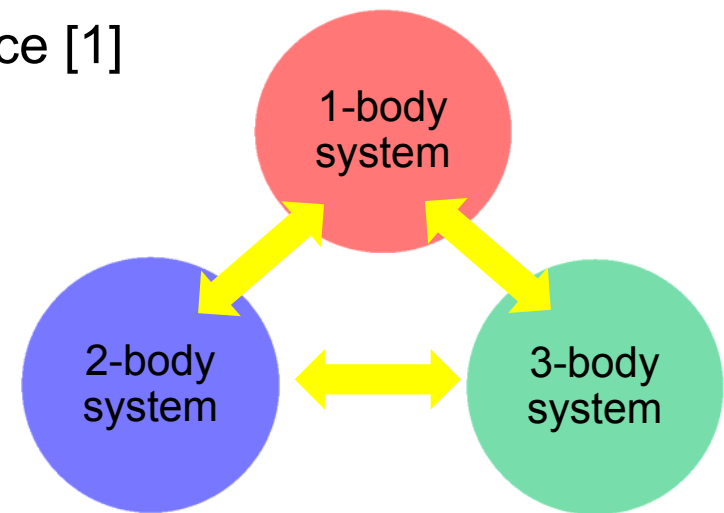
- m_e : $^{12}\text{C}^{5+}$ hydrogen-like ion spin resonance [1]
- m_p : single proton in a Penning trap [2]

$$m_p/m_e = 1\,836.152\,673\,374\ (78)_{\text{exp}}$$

$$u_r = 4.2 \times 10^{-11}$$

CODATA2018: $1\,836.152\,673\,43(11)$

$$u_r = 6.1 \times 10^{-11}$$

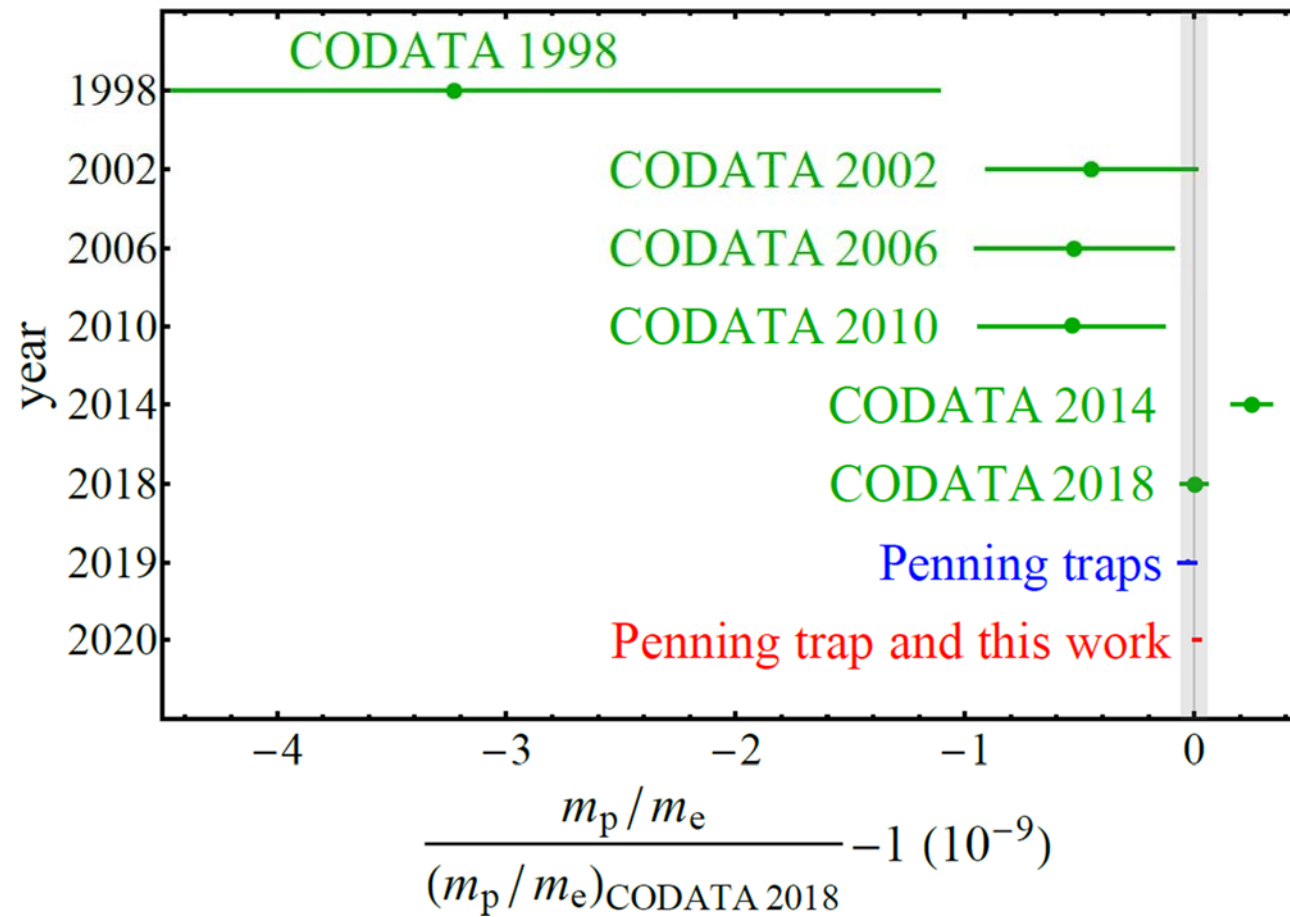


[1] S. Sturm et al. Nature 506 (2014) 467

[2] F. Heiße et al., PRA 100 (2019) 022518

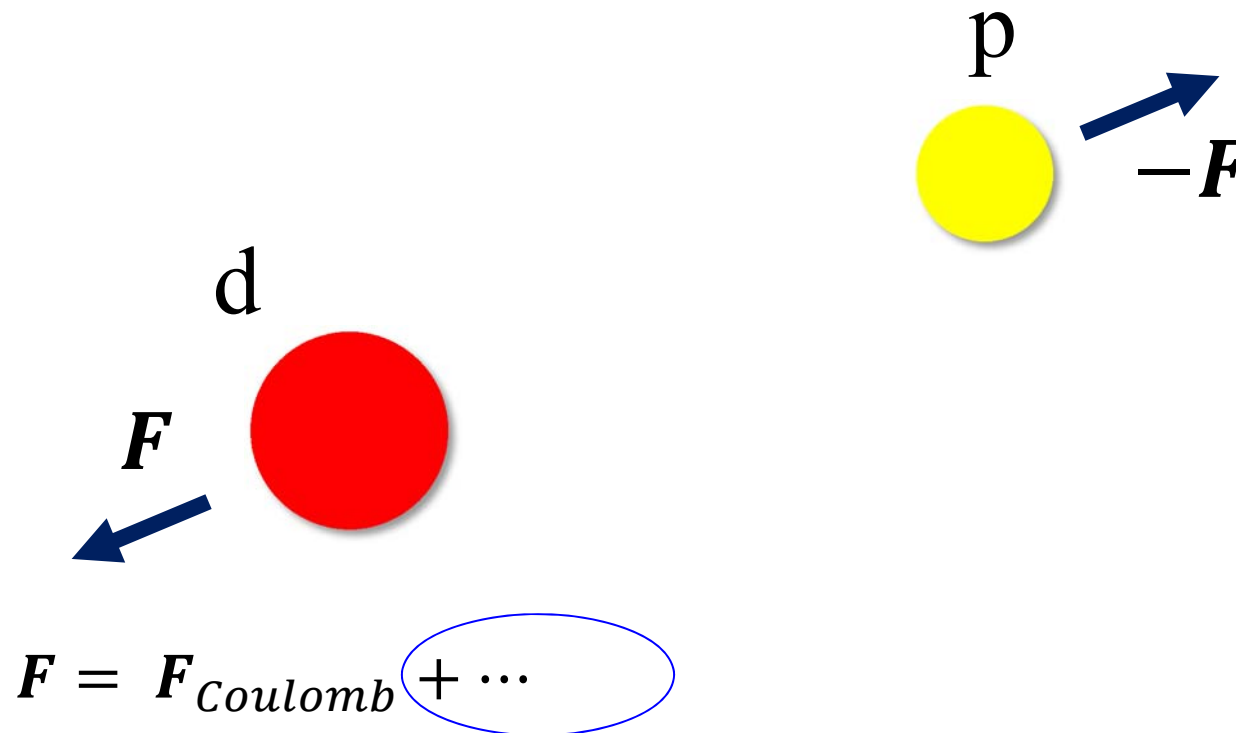
[3] D. Fink and E. Myers, PRL 124 (2020) 013001

Comparison with CODATA evolution



Bound on a 5th force on the Angstrom scale

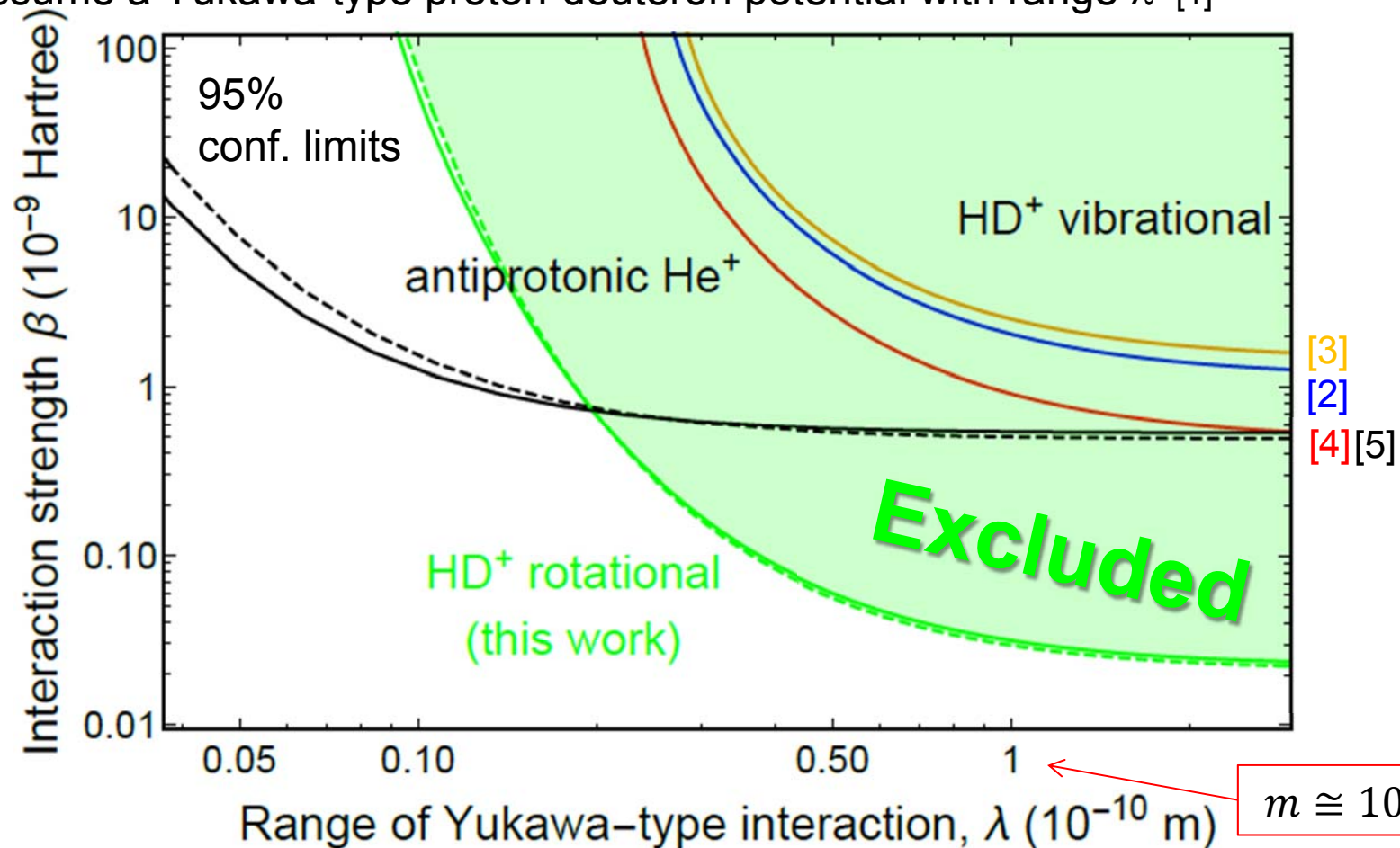
- An additional interaction between the nuclei changes the bond length R_e of the molecule \rightarrow change in rotational frequency
- Assume a Yukawa-type proton-deuteron potential with range λ [1]



[1] Salumbides et al. PRD 87, 112008 (2013)

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[1] Salumbides et al. PRD 87, 112008 (2013)

[3] Bressel et al, PRL 108, 183003 (2012)

[5] M. Hori et al., Nature 475, 484 (2011)

[2] Koelemeij et al, PRL 98, 173002 (2007)

[4] Biesheuvel et al., Nat. Comm. 7, 10385 (2016)

Summary

- High-accuracy, Doppler-free spectroscopy of a rotational transition in a molecular ion
- Most precise comparison theory – experiment in a 3-body system: 5×10^{-11} limited by uncertainty of fundamental constants
- Comparison of ab-initio theory and experiment allows determining

$$R_\infty (m_e/m_p + m_e/m_d)$$

with uncertainty 2×10^{-11}

$$m_p/m_e$$

with uncertainty 2×10^{-11}

- 20-fold improved bound on fifth force between proton and deuteron for range $\lambda = 1 \text{ \AA}$
- Progress in 10 years: $\sim 100 \times$ improved experimental accuracy & theory

*Excellent prospects to reduce experimental uncertainty further
→ More accurate QED theory highly desirable!*

Perspectives

- Improve experimental precision on rotational & vibrational frequencies of HD⁺
- Apply new techniques of quantum logic or optical forces (work of PTB, NIST, U. Basel, MPI-K, ...) → nondestructive spectroscopy
- Extend work to H₂⁺
- Push accuracy → test of time-independence of m_e/m_p ?
- Future: apply to anti-H₂⁺ → CPT test of extremely high accuracy?

The team



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