Hydrogen molecular ions and fundamental constants

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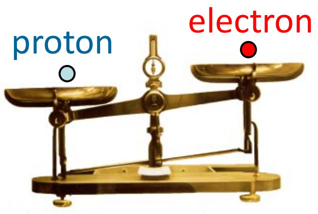
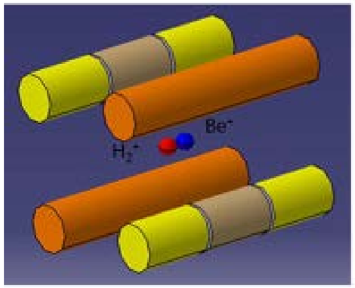
Outline

I. **Determining Fundamental Constants: how to exploit the spectroscopy of Hydrogen Molecular Ions**



II. **Probing the *time variation* of FC: an optical clock based on HMI**



Hydrogen molecular ions

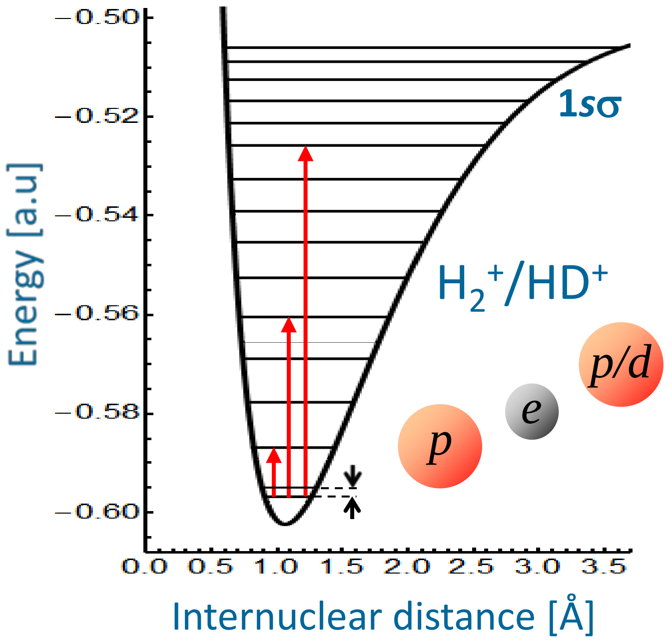
Dependence of ro-vibrational transition frequencies on fundamental constants :

$$\nu = cR_\infty \left[\underbrace{\varepsilon_{nr}(\mu_{ne})}_{\text{Schrödinger}} + \underbrace{\alpha^2 F_{QED}(\alpha)}_{\text{Relativistic and QED corrections}} + \sum_n \underbrace{A_n^{fs}(r_n / a_0)^2}_{\text{Nuclear finite size correction}} \right]$$

Vibrational: $\varepsilon_{nr} \propto \sqrt{m_e / m_r}$

Rotational: $\varepsilon_{nr} \propto m_e / m_r$

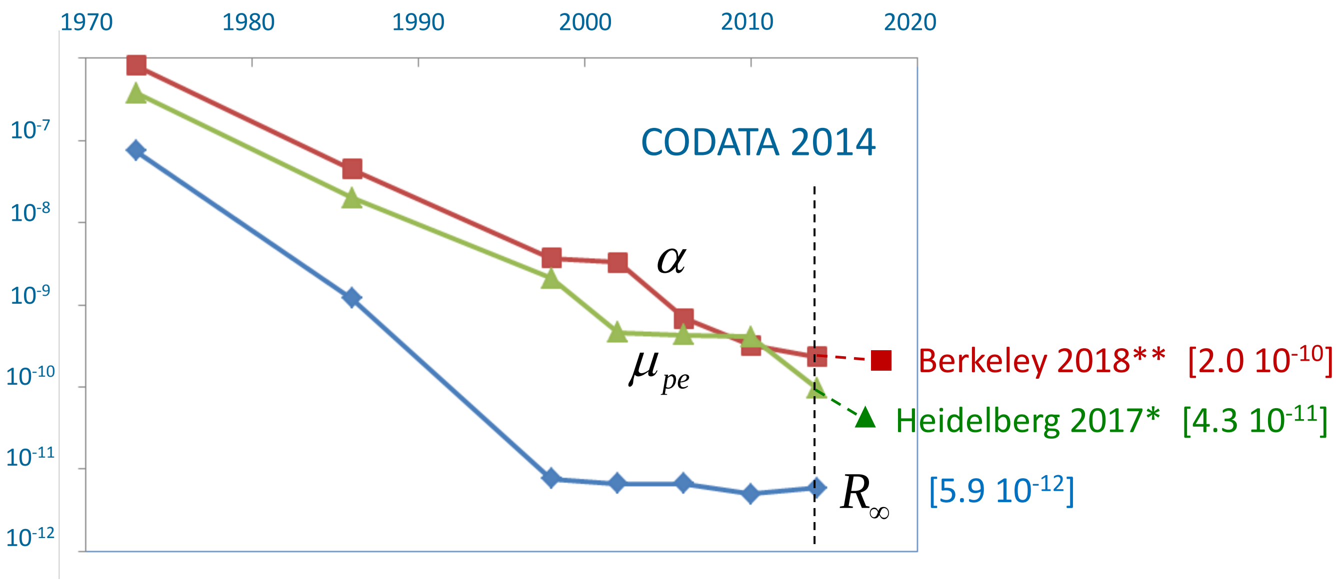
New feature w.r.t. atomic transitions



$\Rightarrow$ Spectroscopy of H_2^+/HD^+ can be used to determine μ_{pe}, μ_{de}.

W.H. Wing, G.A. Ruff, W.E. Lamb Jr., J.J. Spezeski, PRL **36**, 1488 (1976)

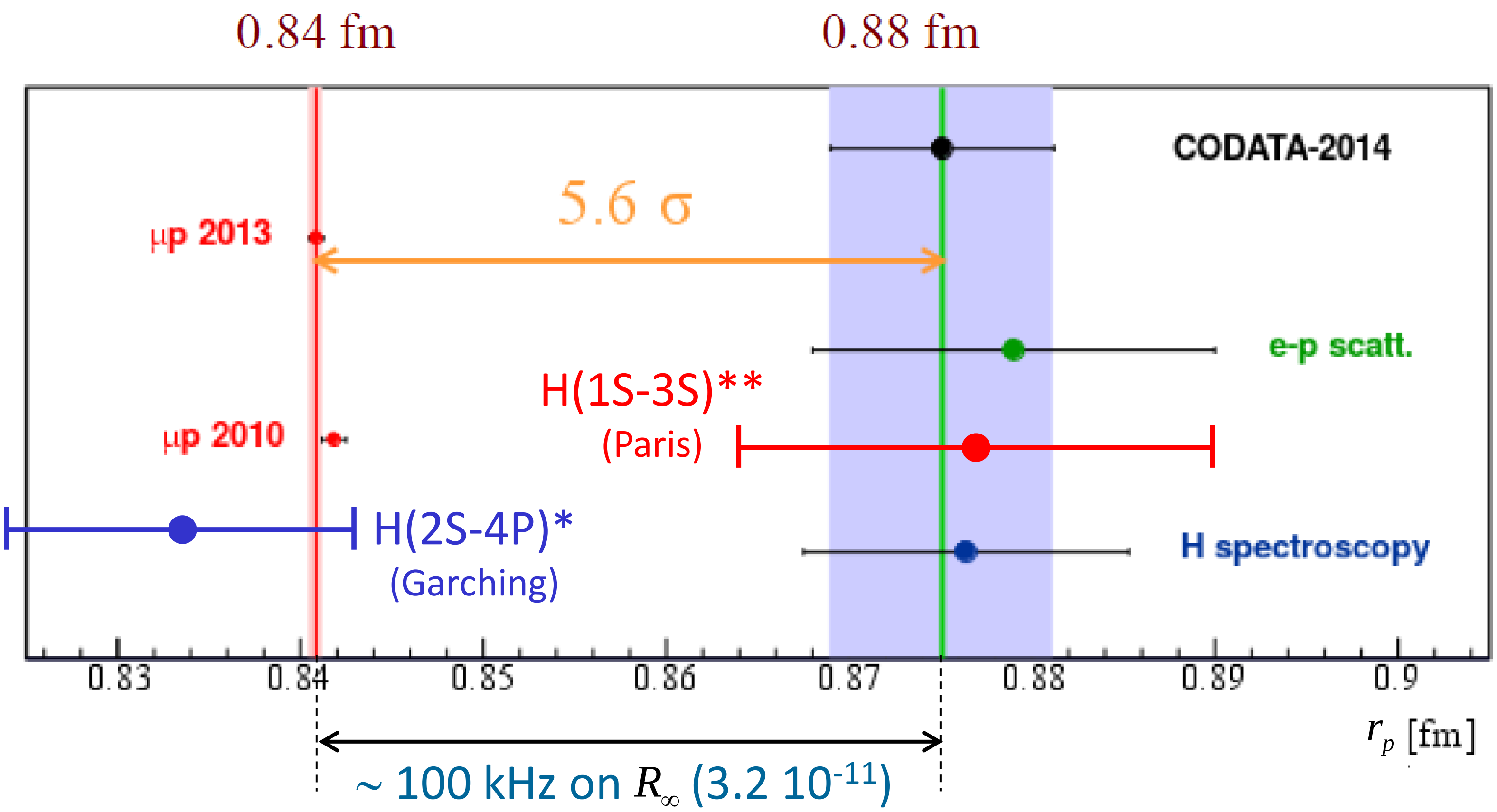
Fundamental constants: progress in accuracy



* F. Heiße et al., Phys. Rev. Lett. **119**, 033001 (2017)

** R.H Parker et al., Science **360**, 191 (2018)

The proton-radius puzzle (updated)



~ 100 kHz on R_∞ (3.2 10^{-11})

(r_p and R_∞ strongly correlated by H(1S-2S) measurement)

- Similar discrepancy on r_d

* A. Beyer et al., Science **358**, 79 (2017)

** H. Fleurbaey et al., PRL **120**, 183001 (2018)

Hydrogen molecular ions: status of theory

H_2^+ (v = 0, L=0) → (v'=1, L'=0) interval in kHz

ν_{nr}	$65\,687\,511\,047.0$
ν_{α^2}	$1091\,040.5$
ν_{α^3}	$-276\,545.1$
ν_{α^4}	$-1952.0(1)$
ν_{α^5}	$121.8(1)$
ν_{α^6}	$-2.3(5)$
ν_{tot}	$65\,688\,323\,710.1(5)$**(29)(11)**

V.I. Korobov, L. Hilico, J.-Ph. Karr,
PRL **118**, 233001 (2017)

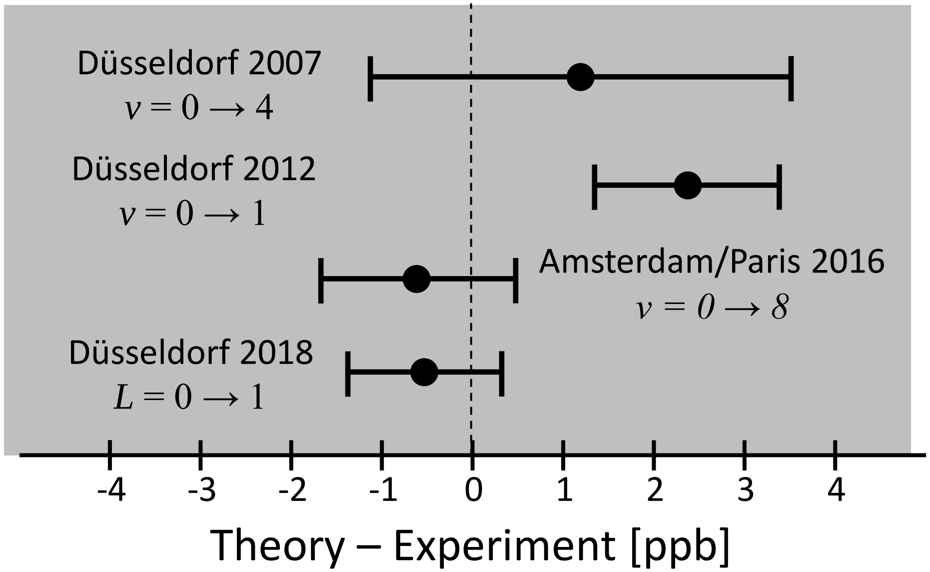
See next talk !

1. Theoretical uncertainty = **7.6 10^{-12}**
2. CODATA 2014 uncertainty on μ_{pe}
3. Discrepancies on R_∞ and r_p between CODATA 2014 and μp + H(1S-2S)

- From a **single** high-precision measurement one could get:
  - **a determination of** μ_{pe} accurate to
    - ~ **1.6 10^{-11}** if r_p-puzzle resolved
    - ~ **3.6 10^{-11}** if not.
  - **or, assuming** a big improvement in μ_{pe} ,
    - R_∞, r_p discrimination at ~ 2.2σ level

Experimental state of art

One-photon transitions In HD$^+$



- ~ppb accuracy, limited by Doppler broadening (exc. for rotational transition)
- Good agreement with theory
- Determinations of μ_{pe}

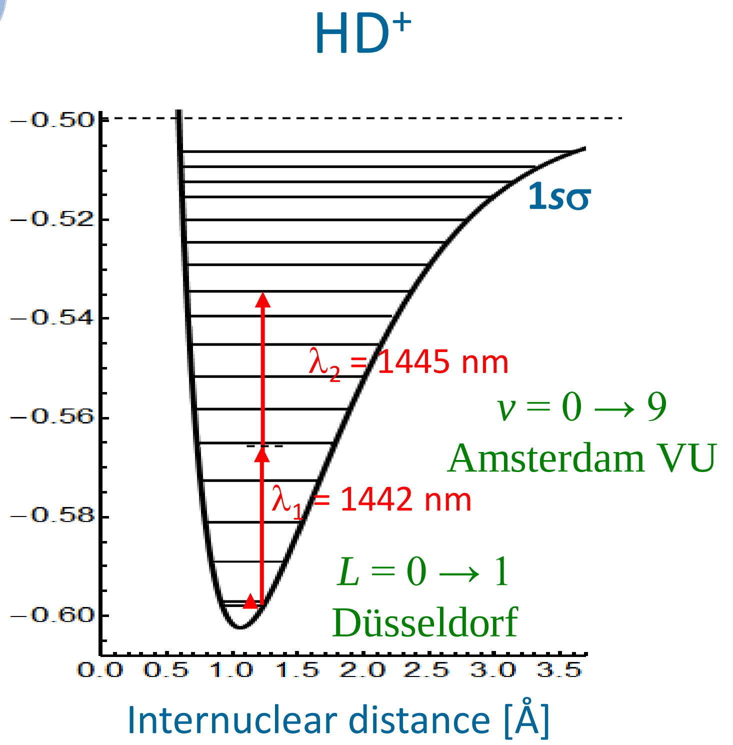
from 3 vibrational transitions: $\mu_{pe} = 1836.152\,682\,4(46)$ 2.5 10^{-9}

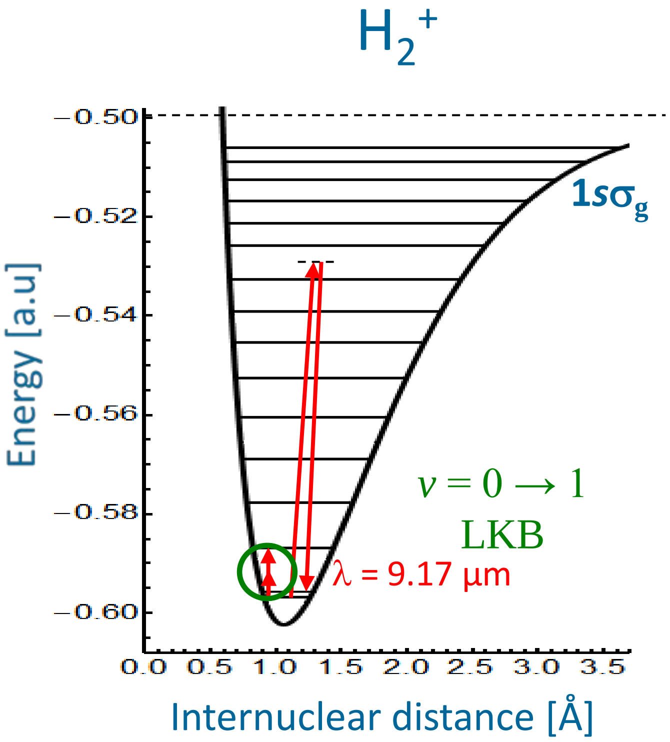
S. Patra et al., J. Phys. B **51**, 024003 (2018)

from rotational transition: $\mu_{pe} = 1836.152\,6739(24)$ 1.3 10^{-9}

S. Alighanbari et al., Nat. Phys. (2018)

Prospects: Doppler-free spectroscopy





- Decay to *v* = 0 within hundreds of ms
- Rotational transitions (THz or Raman)
- **Quasi-degenerate** two-photon transitions

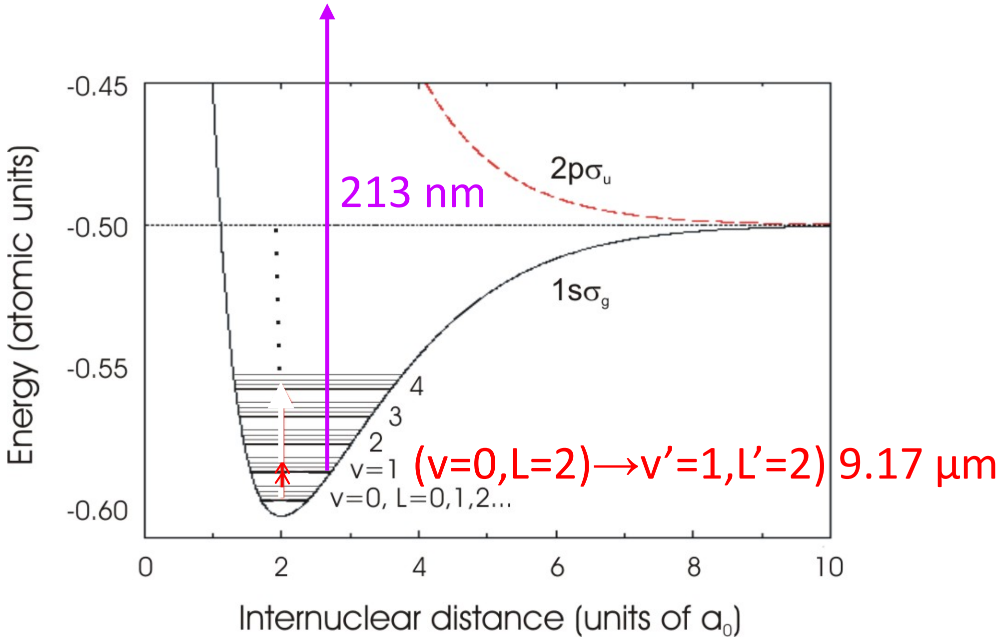
$$\lambda_{eff} = \frac{2\pi}{\left|\vec{\mathbf{k}}_1 + \vec{\mathbf{k}}_2\right|} >> \Delta r$$

V.Q. Tran et al., Phys. Rev. A **88**, 033421 (2013)

- All ro-vibrational states metastable
- $\Delta v = 1$ Doppler-free two-photon transitions
- Rotational Raman transitions

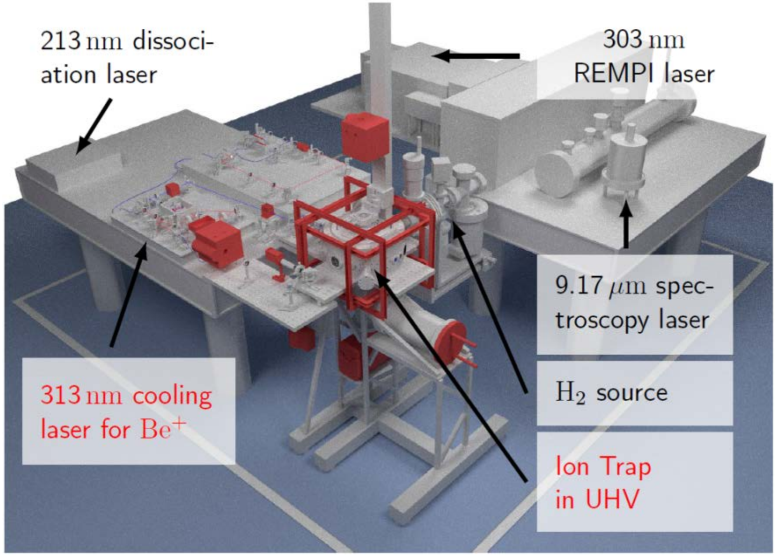
Experimental accuracies of 10^{-12} or better are achievable !

The H_2^+ experiment in Paris

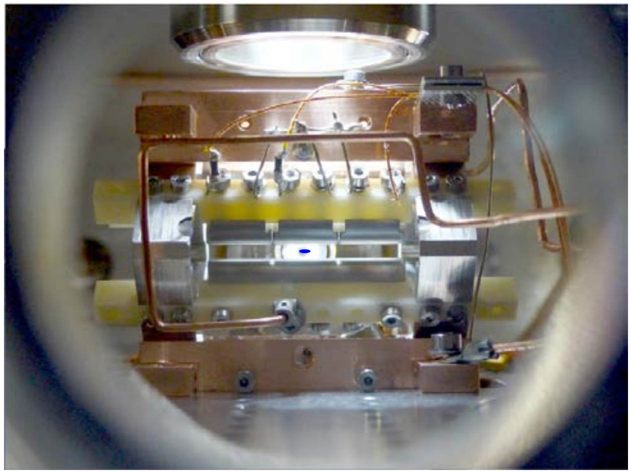


- Linear rf ion trap, sympathetic cooling by laser-cooled Be^+ (λ = 313 nm)
- State-selected H_2^+ production by Resonance-Enhanced Multi Photon Ionization (REMPI) of H_2 at λ = 303 nm
- In-vacuum enhancement cavity for the two-photon transition
- Detection via photodissociation of v=1: Resonance-Enhanced Multi Photon Dissociation (REMPD). Signal = ion loss

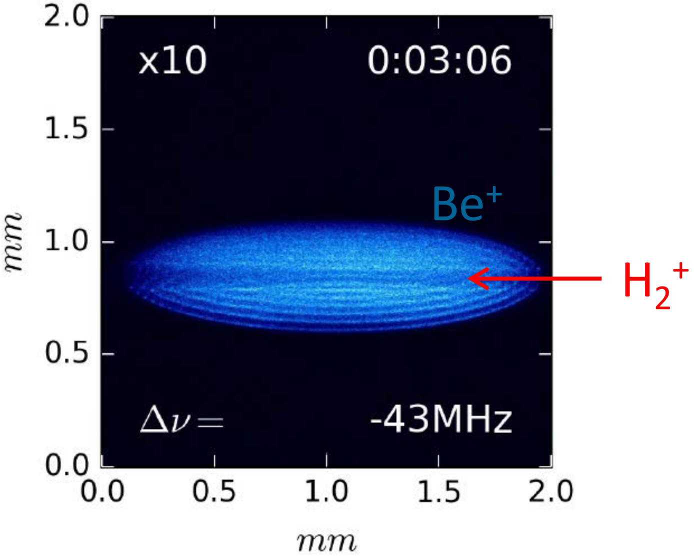
H_2^+ two-photon spectroscopy setup



Linear ion trap



Be^+/H_2^+ ion crystal



Prospects for fundamental constants

- Simulate the extraction of FC from (hypothetical!) experimental data, using a CODATA-like least-squares adjustment procedure.
- Assumptions:
  - All transitions frequencies calculated with accuracy of 3 10^{-12}
  - Experimental uncertainties of 1 10^{-12} for all transitions
- Input data:
  - 1S-2S transitions in H and D
  - three transitions in HMI: **HD^+** $L = 0 \rightarrow 1$; **HD^+** $v = 0 \rightarrow 9$; **H_2^+** $v = 0 \rightarrow 1$

FC	$\Delta y / y$	Δy	**Improv.**
μ_{pe}	1.5 10^{-11}		2.7
μ_{dp}	1.4 10^{-11}		2.9
R_∞	1.2 10^{-11}	39 kHz	2.7
r_p	1.3 10^{-2}	0.011 fm	3.0
r_d	2.2 10^{-3}	4.7 10^{-3} fm	3.3

J.-Ph. Karr et al., PRA **94**, 050501(R) (2016)

Prospects for fundamental constants

- Which would be the best new transition to investigate ?

- **H_2^+** $L = 0 \longrightarrow 2$ (Raman)
- **H_2^+** $v = 6 \longrightarrow 7$
- **HD^+** $v = 0 \longrightarrow 2$

$\Rightarrow \sim \times 5$ improvement

- ➢ HMI spectroscopy can shed light on $\mathrm{r_p} / r_d / R_\infty$ puzzle at $\sim 5\sigma$ significance level ;
- ➢ Key advantage: measure only very narrow lines

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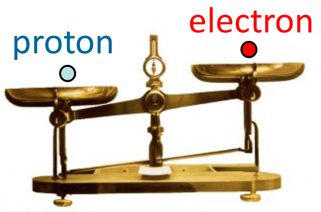

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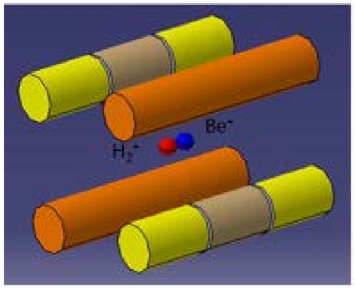
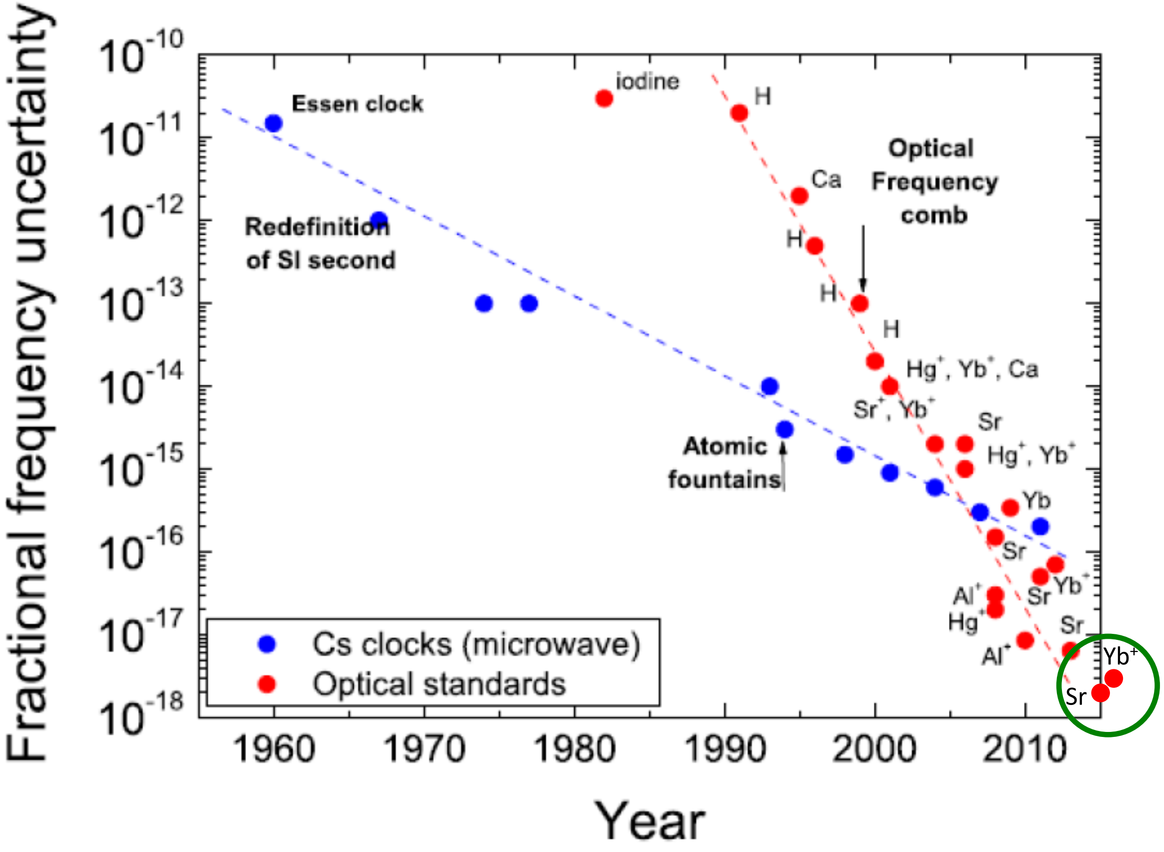


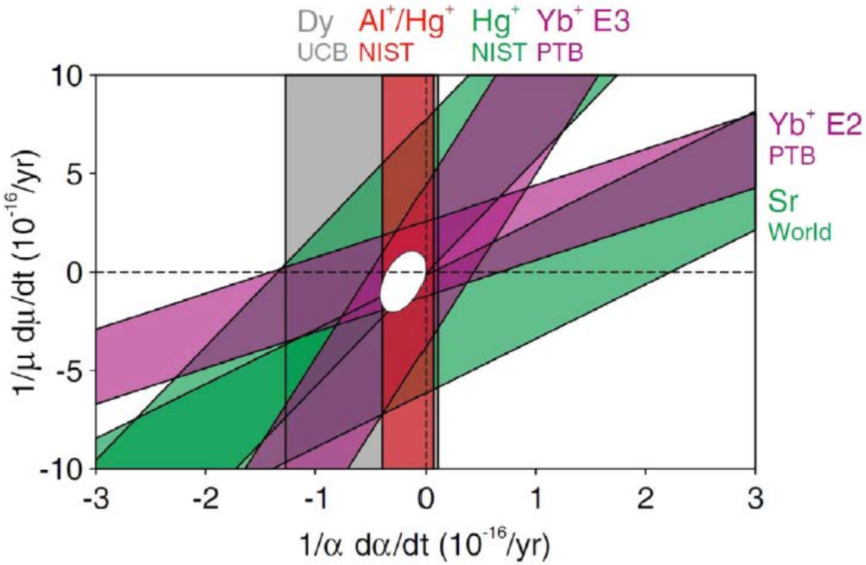
Optical clocks



N. Poli et al., Nuovo Cimento **36**, 555 (2013)

Fundamental tests with clocks

- Lorentz invariance
- Variation of fundamental constants



$$\boxed{\dot{\alpha} / \alpha = -2.0(2.0)\, 10^{-17}\, \mathrm{yr}^{-1}}$$

T. Rosenband et al., Science **319**, 1808 (2008)

$$\boxed{\dot{\mu} / \mu = 0.2(1.1)\, 10^{-16}\, \mathrm{yr}^{-1}}$$

N. Huntemann et al., PRL **113**, 210802 (2014)

Different types of clocks

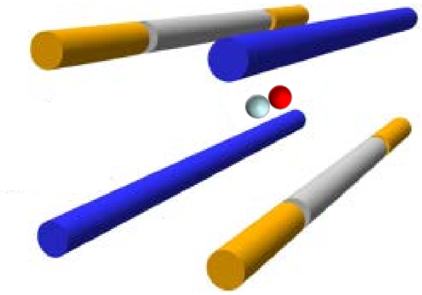
- Optical atomic clocks : $\boxed{f = cR_\infty C\, F(\alpha)}$
- Cs clock : $\boxed{f_{hfs} = \alpha^2\, cR_\infty C_{hfs}\, F_{hfs}(\alpha) \underbrace{G(\mu_N / \mu_B)}_{\propto 1/\mu}}$ μ_N : nuclear magnetic moment
- Vibrational transitions : $\boxed{f_{vibr} \approx cR_\infty M_r^{-1/2}\, \mu^{-1/2}}$

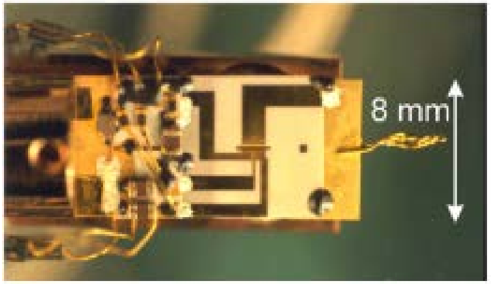
➢ ***Ratios of optical atomic clock frequencies :*** $\dot{\alpha}$ ***only***

➢ ***Ratios of optical to Cs frequencies :*** $\dot{\alpha}$ ***and*** $\dot{\mu}$ ***, limited by Cs performances***

➢ ***Ratios of atomic and molecular clock frequencies could set improved constraints on*** $\dot{\mu}$

Quantum-logic clocks





Al$^+$ clock, NIST

- Co-trapping of a laser-cooled “logic ion” (Be$^+$ or Mg$^+$)
- ***Sympathetic cooling*** to the motional ground state of the ion pair
- Internal state detection by mapping from the “clock ion” to the “logic ion” using quantum logic
- Allows investigation of *new candidate ion species*
  molecular ions
  highly charged ions

P.O. Schmidt et al., Science **309**, 749 (2005)
T. Rosenband et al., Phys. Rev. Lett. **98**, 220801 (2007)

Some attractive candidates

- ***Homonuclear diatomic molecules: H_2^+, N_2^+, O_2^+***

  - Extremely narrow (dipole forbidden) transitions
  - Production in selected ro-vibrational states by REMPI
  - Immune to BBR-induced transitions

H_2^+	S. Schiller et al., PRL **113**, 023004 (2014) ; J.-Ph. Karr, J. Mol. Spectrosc. **300**, 37 (2014)
N_2^+	M. Germann et al., Nat. Phys. **10**, 820 (2014) ; M. Kajita et al., PRA **89**, 032509 (2014) ; M. Kajita, PRA **92**, 043423 (2015)
O_2^+	D. Hanneke et al., PRA **94**, 050101(R) (2016) ; M. Kajita, PRA **95**, 023418 (2017)

- ***Weakly dipole-allowed vibrational overtones***

TeH^+,TeD^+	M.G. Kokish et al., arXiv:1710.08589v2 (23/04/2018)

E2 transitions in H_2^+: uncertainty budget

Unit: 10^{-17}

Transition	$(0,4) \to (2,2)$		$(0,0) \to (1,2)$		$(0,0) \to (2,2)$		$(0,0) \to (3,2)$	
Frequency (THz)	114.99		70.64		132.25		190.18	
Source	shift	uncert.	shift	uncert.	shift	uncert.	shift	uncert.
Quadrupole shift	0	0.049	0	1.7	0	1.0	0	0.82
Quadr. Zeeman shift	< 0.01	< 0.01	< 0.01	1.1	< 0.01	0.61	< 0.01	0.46
Blackbody radiation	-12	0.80	-9.3	0.62	-11	0.74	-13	0.88
AC Stark shift	-220	2.2	-5.0	0.050	-200	2.0	-4500	45
Total uncertainty		2.6		2.4		2.7		45

- With operating parameters of NIST Al^+ clock (Rabi interrogation, π pulse of duration $\tau = 100$ ms), the **AC Stark shift** is a big limitation.
- Can be cancelled to a high degree (**hyper-Ramsey** interrogation schemes).
- Then the overall uncertainty reduces to **1.6 10^{-17}** for the v=0⟶3 transition (including 1.0 10^{-17} from 2nd-order Doppler effect).
- Light ion ⟶ larger 2nd-order Doppler shift.

J.-Ph. Karr et al., J. Phys.: Conf. Ser. **723**, 012048 (2016)

Summary

- H_2^+ / HD^+ spectroscopy is a promising route towards
  - improved determination of m_p/m_e
  - a new independent determination of r_p , r_d and R_∞

- Candidates for molecular ion clocks are emerging, among which H_2^+, to probe m_p/m_e variations at the 10^{-17} … 10^{-18}/yr level.

Acknowledgements

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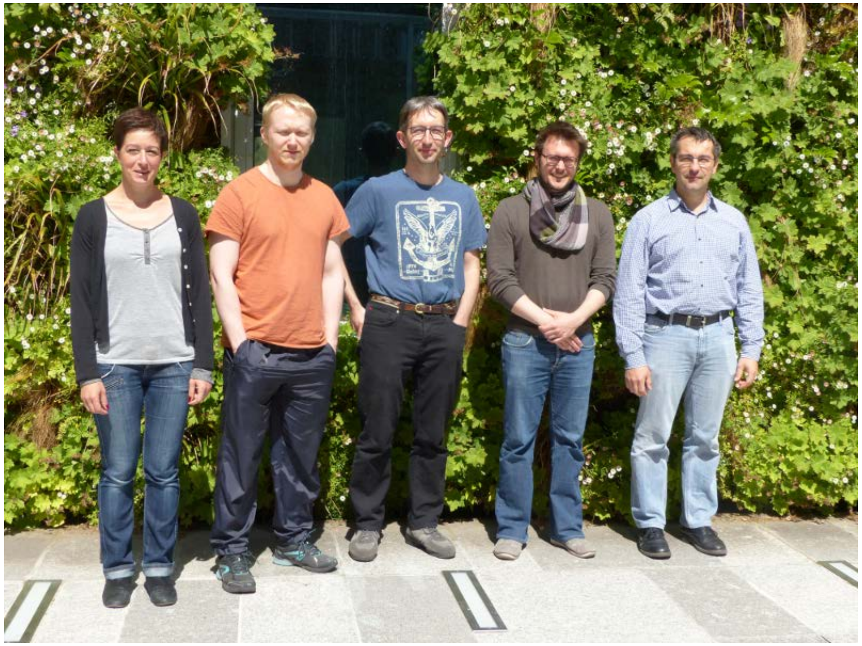

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