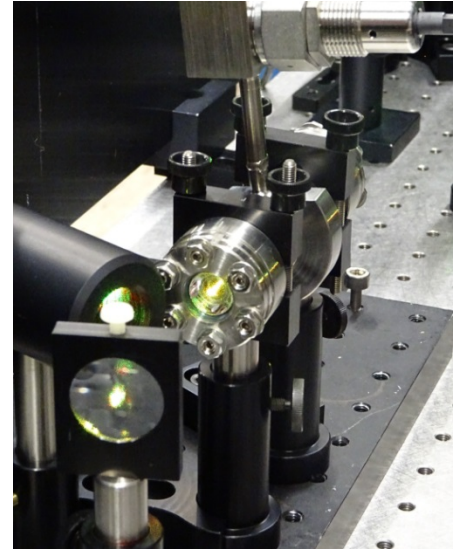
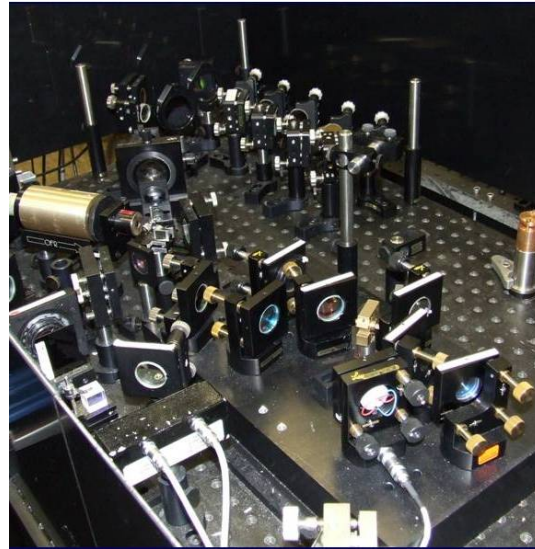
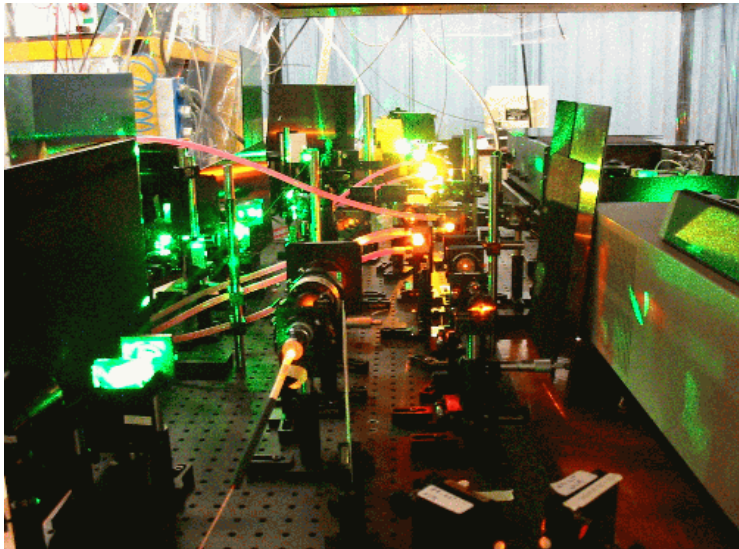


Physics beyond the Standard Model from hydrogen molecules



Wim Ubachs
Vrije Universiteit Amsterdam

Nikhef Colloquium
Amsterdam
22 Nov 2019

The Standard Model of Physics

What do we know ?

	Fermions			Bosons
Quarks	u up	c charm	t top	γ photon
	d down	s strange	b bottom	g gluon
Leptons	ν_e electron neutrino	ν_μ muon neutrino	ν_τ tau neutrino	Z Z boson
	e electron	μ muon	τ tau	W W boson
	I II III Three generations of matter			H Higgs boson

Force carriers

What do we not know ?

- Dark Matter
- Dark Energy
- How does Gravity fit to SM ?
- Why is Gravity so weak ?
- Are there only 3+1 dimensions ?
- Are there only 4 forces ?
- Amount of CP violation

Beyond SM ?

- Variation of fundamental constants
- Space-time dependence of physical law (breakdown of GR)

→ There is a precision molecular spectroscopy approach to these questions

Prelude

Empirical search for a drift of the Proton-Electron mass ratio μ

Lab

today

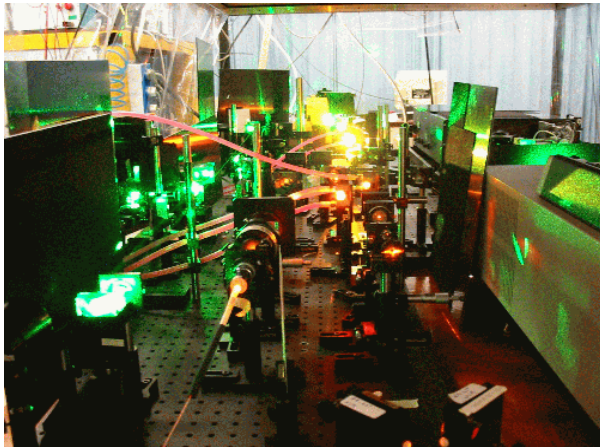
90-112 nm

Compare H₂ in different epochs

QSO

12 Gyr ago

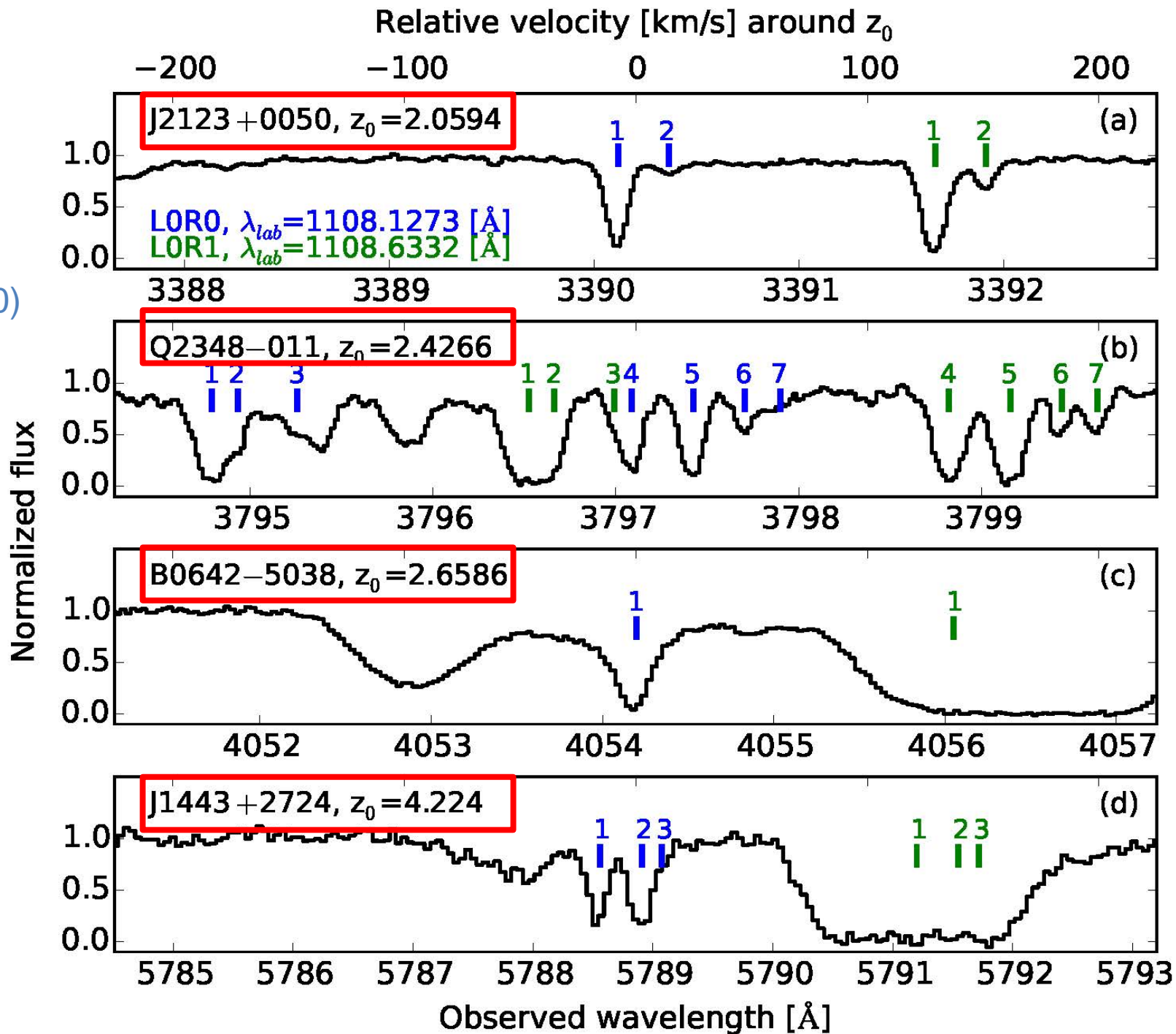
~275-350 nm



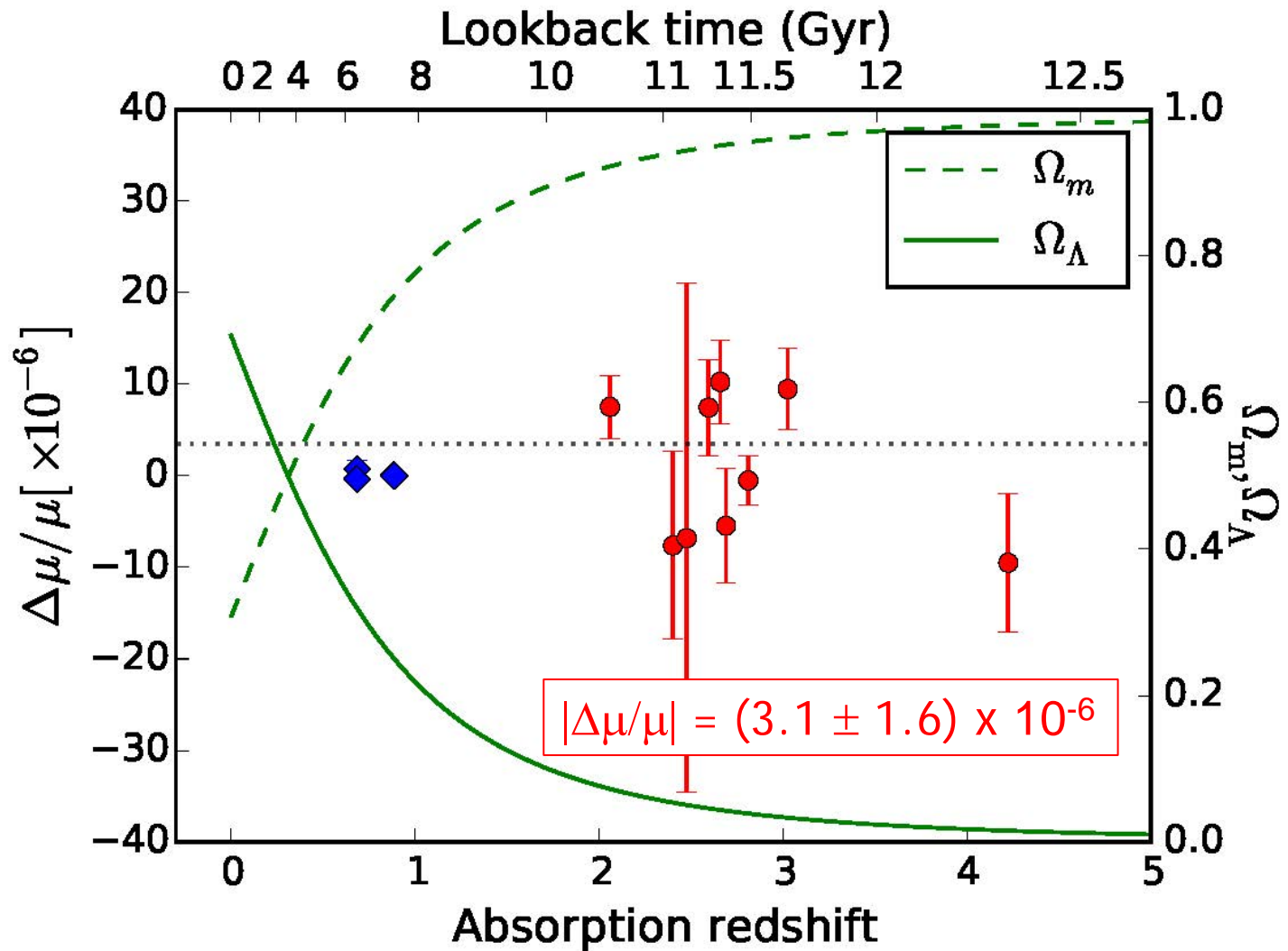
$$\frac{\lambda_i^z}{\lambda_i^0} \equiv 1 + z_i$$

Cosmological redshift





Status/Review: Varying Proton-electron mass ratio



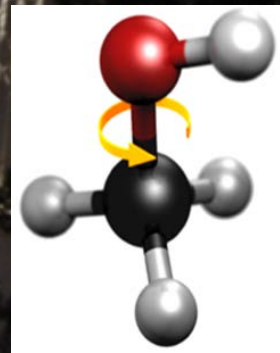
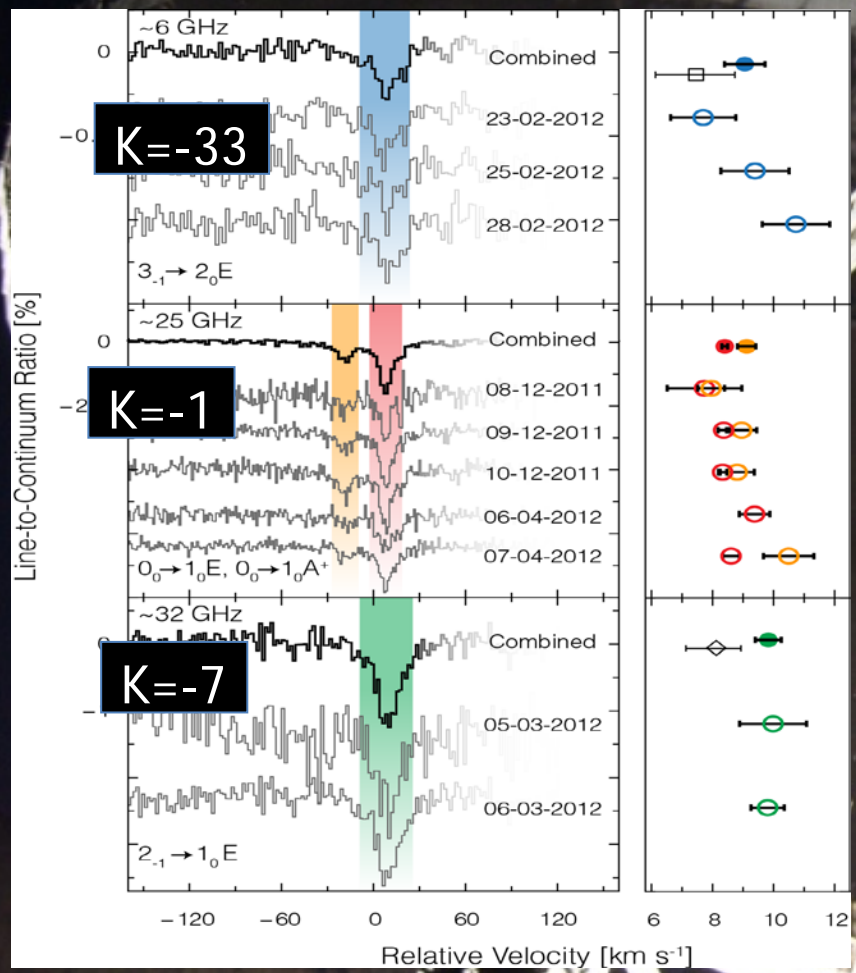
A Stringent Limit on a Drifting Proton-to-Electron Mass Ratio from Alcohol in the Early Universe

Bagdonaite, Jansen, Henkel, Bethlem, Menten, Ubachs, Science 339 (2013) 46

Effelsberg Radio Telescope

PKS-1830-211
"molecular factory"

at $z=0.88582$
(7.5 Gyrs look-back)



Methanol observations ongoing at ALMA

$$\left| \frac{\Delta\mu}{\mu} \right| < 10^{-7}$$

(Hydrogen) Molecules as a metrology test system

Search for BSM-physics from laboratory spectroscopy experiment

$$\Delta E = E_{\text{exp}} - E_{\text{theory}}$$
$$\delta E = \sqrt{\delta E_{\text{exp}}^2 + \delta E_{\text{theory}}^2}$$

$\Delta E < \delta E$ Validate theory (QED/SM)

$\Delta E > \delta E$ New Physics:

Discover new physics $\langle \Delta V_{\text{new}} \rangle > \delta E$

Constrain new physics $\langle \Delta V_{\text{new}} \rangle < \delta E$

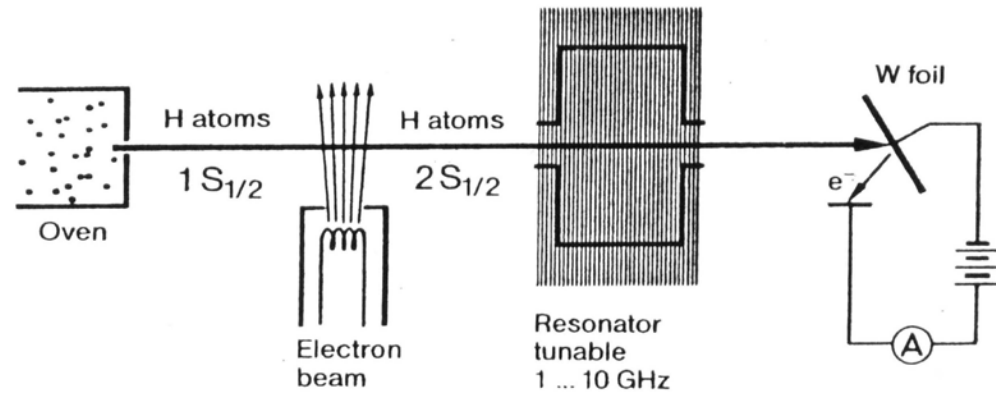
Theory is needed – this works for “calculable” systems

H₂ – Krzysztof Pachucki, Jacek Komasa, Mariusz Puchalski
H₂⁺/HD⁺ - Vladimir Korobov, Jean-Philippe Karr, Laurent Hilico,

Historical Inspiration



Willis E Lamb



Measurement of the tiny $2S_{1/2} - 2P_{1/2}$ splitting

Breakdown of the Dirac theory of the electron
The advent of Quantum Electro Dynamics

Molecules as a metrology test system

1) Test of QED in molecules
 H_2 , HD, D_2 , T_2

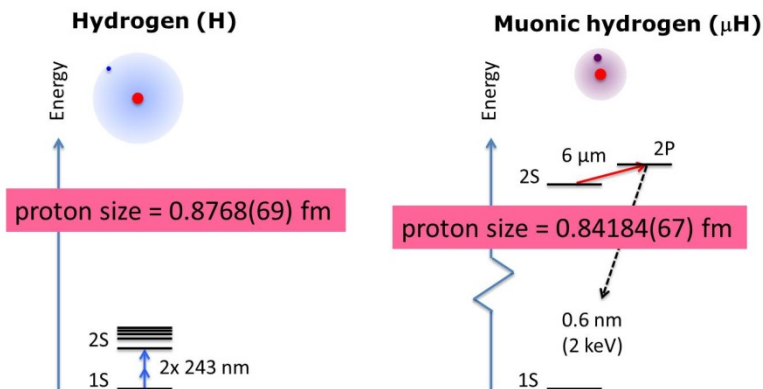
Born-Oppenheimer
 Adiabatic corrections
 Non-adiabatic corrections
 or **Full Nonadiabatic**
 Relativistic effects
 QED effects up to order: $m\alpha^n$

2) The values
 of fundamental constants

$$m_p / m_e \quad m_d / m_e$$

$$m_t / m_e \quad R_\infty \quad \alpha$$

3) The proton charge radius



4) New Physics


5th forces

Extra dimensions

Etc.

The proton size puzzle

$$E(n, l, j)/h = -\frac{Z^2 c R_\infty}{n^2} \frac{m_{\text{red}}}{m_e} + \frac{E_{\text{NS}}}{n^3} \delta_{l0} + \Delta(n, l, j)$$

$$E_{\text{NS}}^{(0)} = \frac{2}{3h} \left(\frac{m_{\text{red}}}{m_e} \right)^3 (Z\alpha)^4 m_e c^2 \left(\frac{r_N}{\lambda_C} \right)^2 \propto (Z\alpha)^6$$


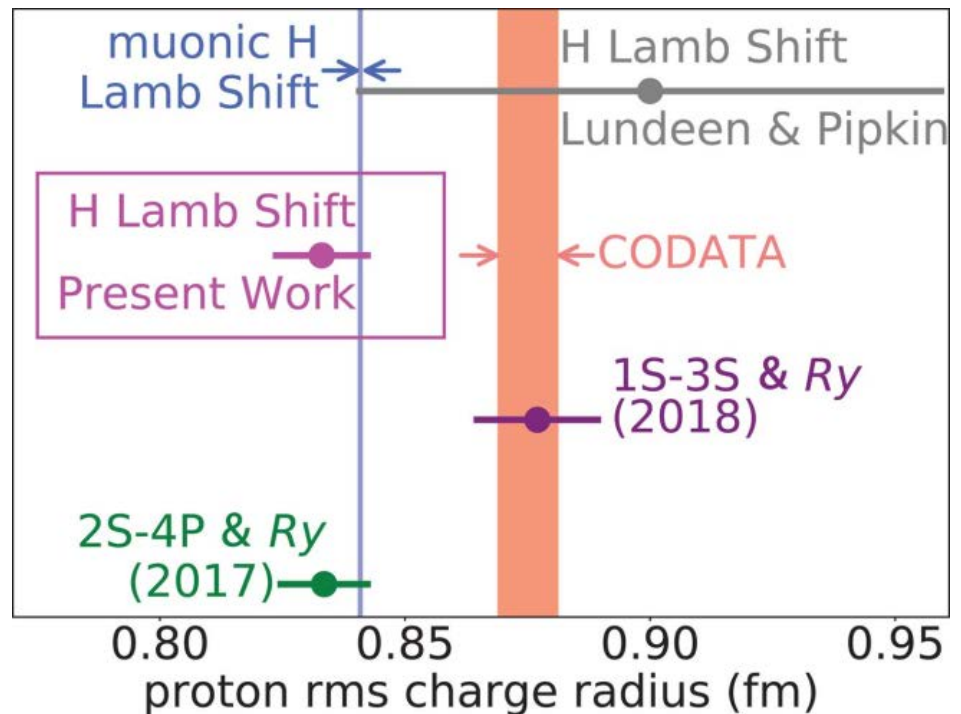
1s-2s in H
(15-digit accuracy)
= exact

+ 2s-2p Lamb shift

+ ns, np, nd states

+ molecular trans
(H₂, HD, HD⁺, H₂⁺)

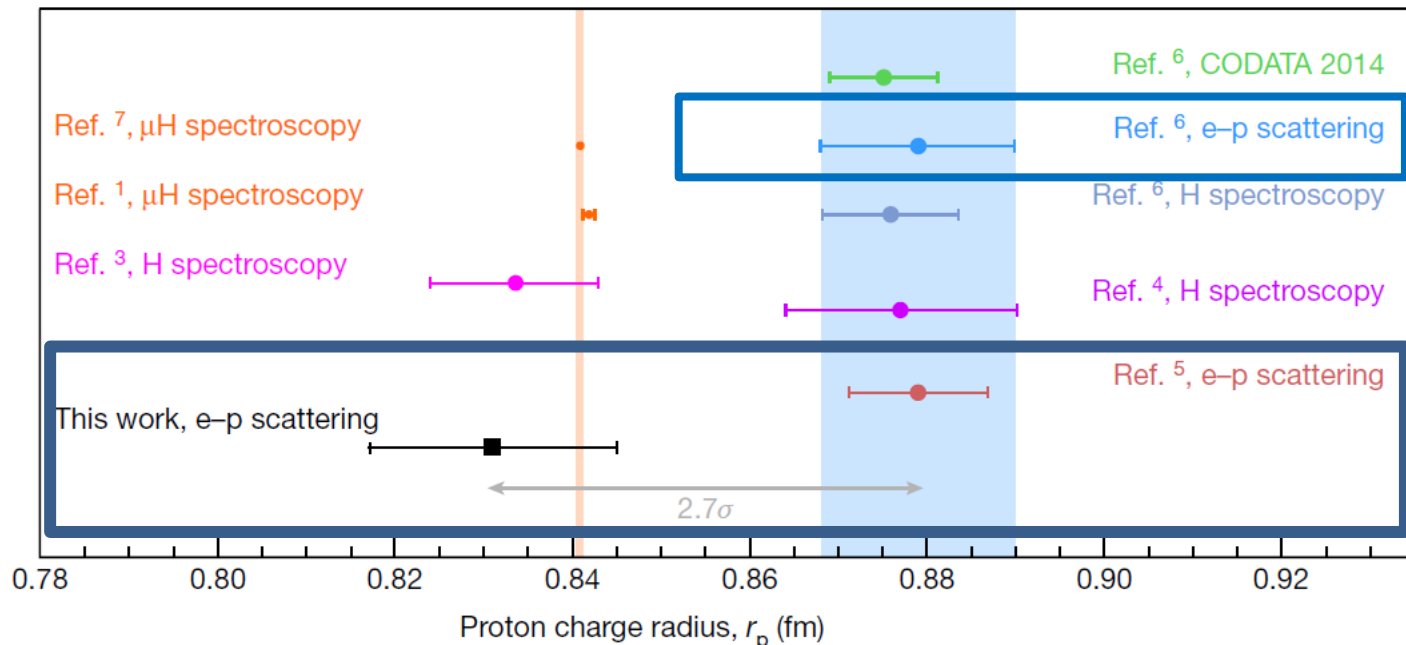
+ μH



The proton size puzzle

Remeasurement of e-p scattering (Jefferson Lab, Nature, Nov 2019)

+ 2s-4p
+ 1s-3s
Munich



- That seems to be settled, finally !
- So we trust the μ H value (most accurate)

Fifth-force searches

Assume: Extra *hadron-hadron* interaction

Parametrize as:

Yukawa potential
$$V_5(r) = N_1 N_2 \left\{ \alpha_5 \frac{\exp(-r/\lambda)}{r} \right\} \hbar c$$



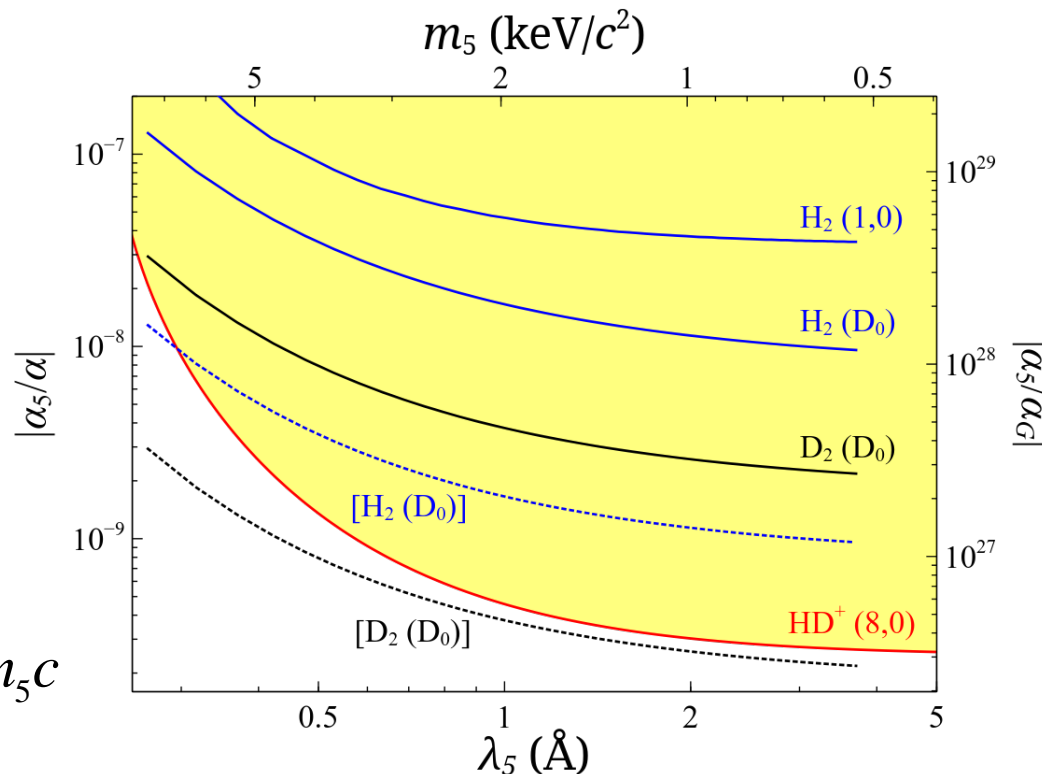
Hideki Yukawa

Constraint:

$$\langle \Delta V_5 \rangle < \delta E$$



$$\lambda = \hbar / m_5 c$$

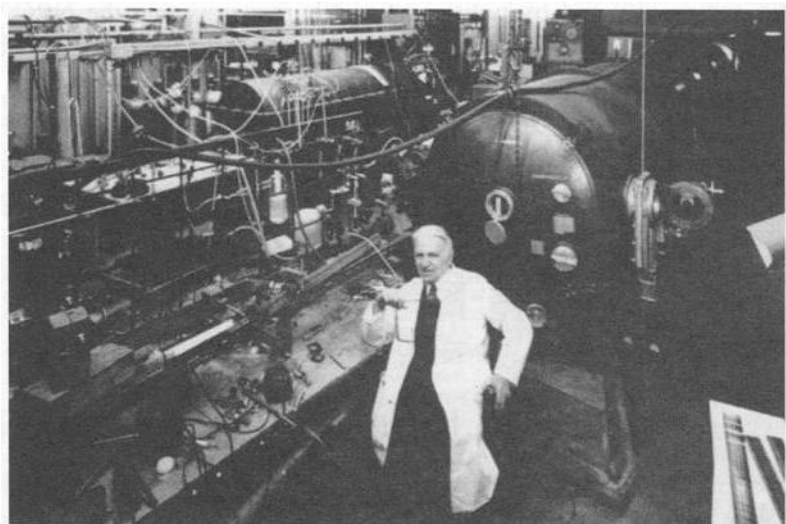


+ Search for higher Dimensions (ADD)

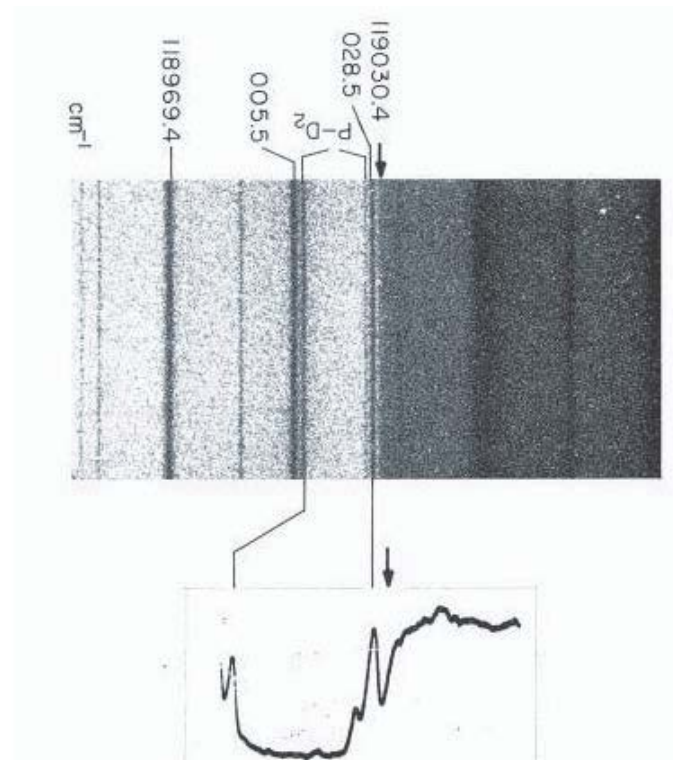
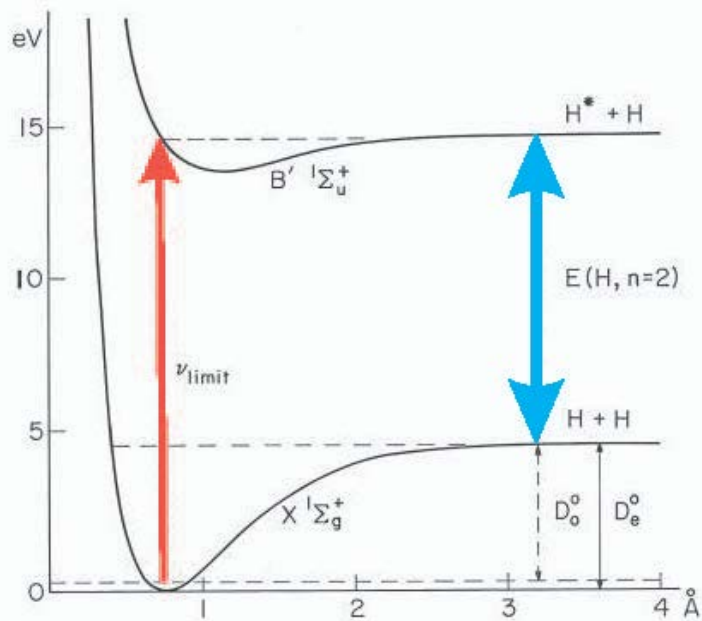
Salumbides
Gato-Rivera
Schellekens
Ubachs

New. J. Phys 17,
033015 (2015)

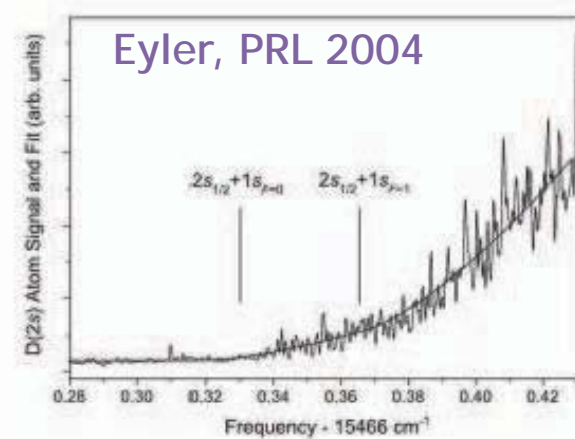
H₂ Dissociation energy; benchmark



Herzberg in his laboratory.

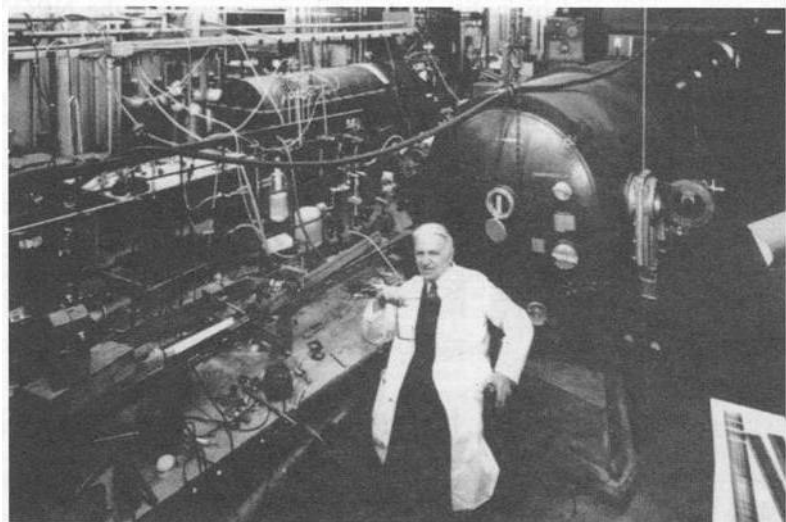


Herzberg
Nobel
Lecture



Eyler, PRL 2004

H₂ Dissociation energy; measurement of IP

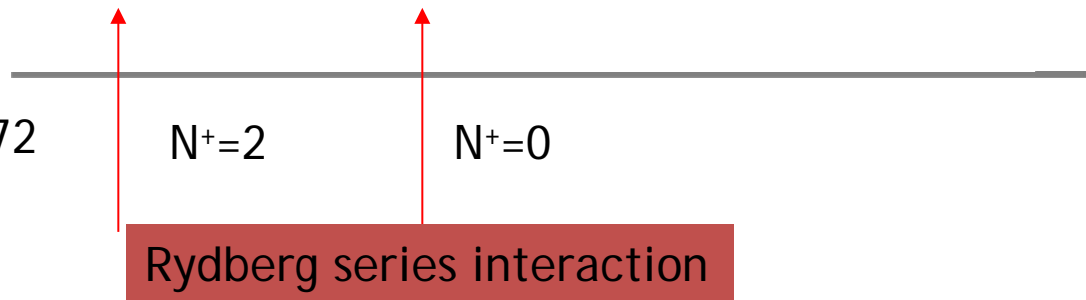
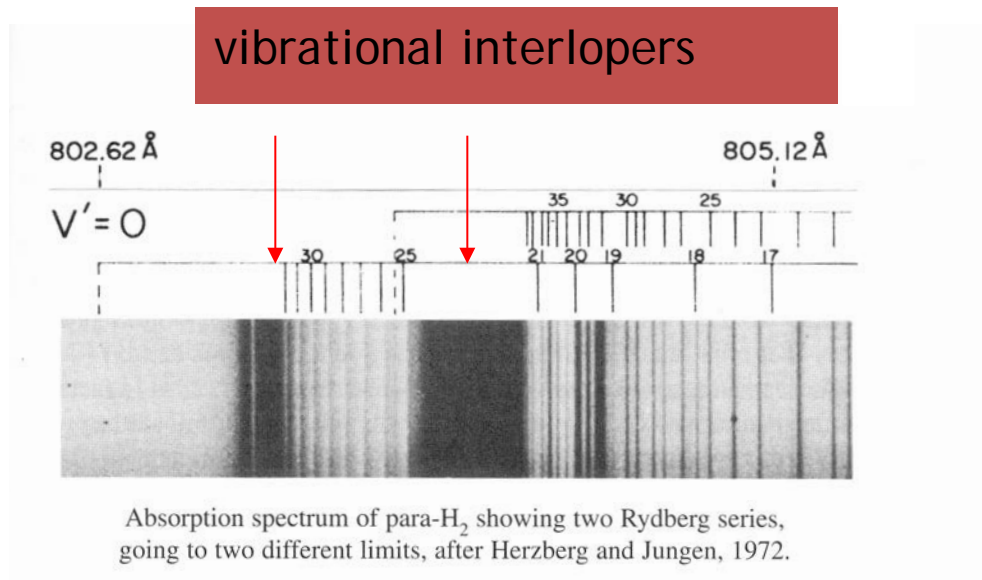


Herzberg in his laboratory.

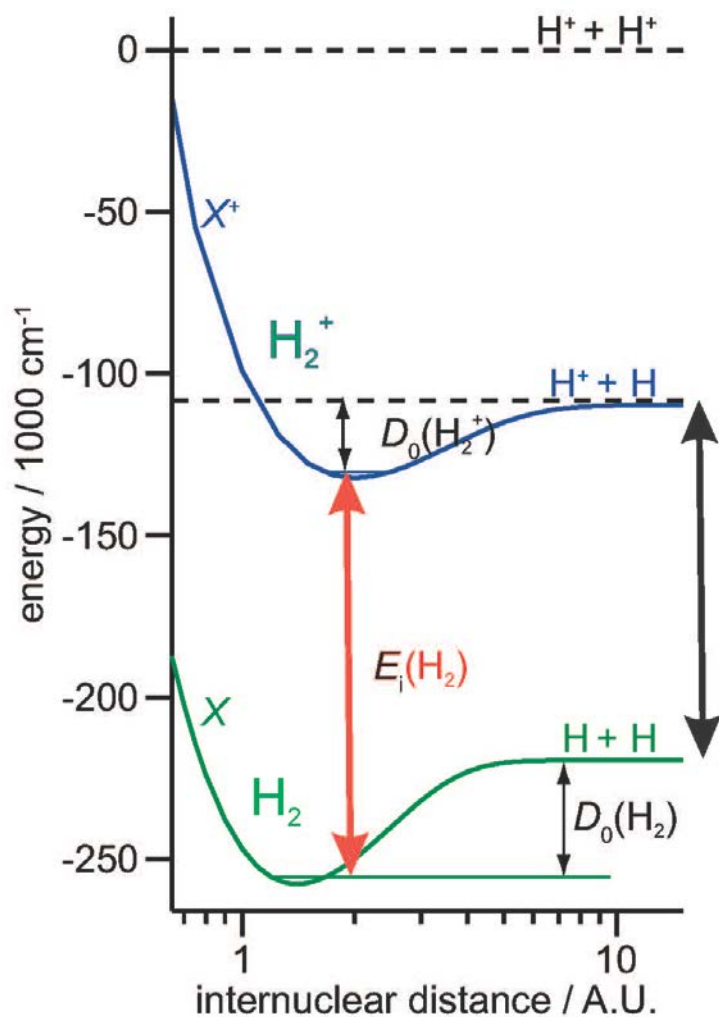


Christian Jungen

Herzberg and Jungen 1972
MQDT of molecules



Benchmark: Dissociation energy H_2



$$D_0(H_2) = E_{IP}(H_2) + D_0(H_2^+) - E_{IP}(H)$$

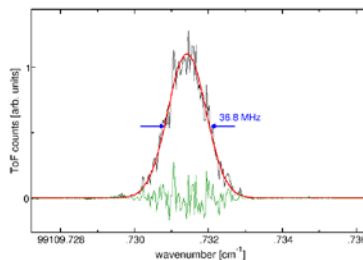
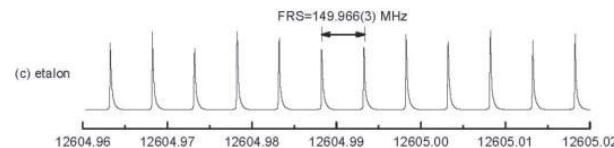
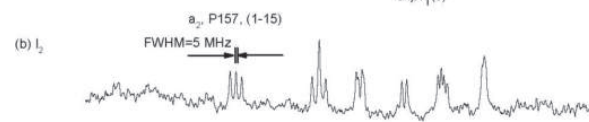
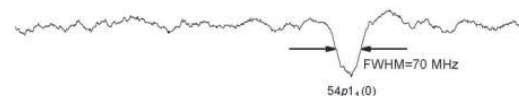
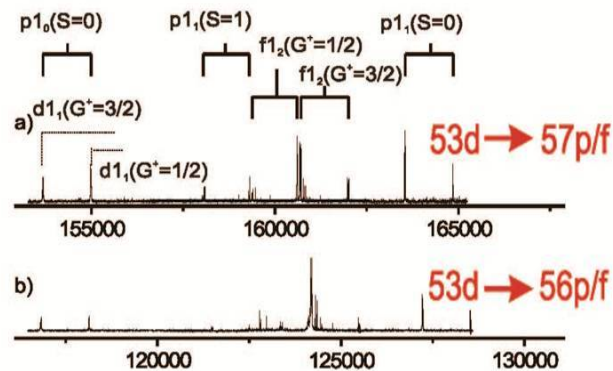
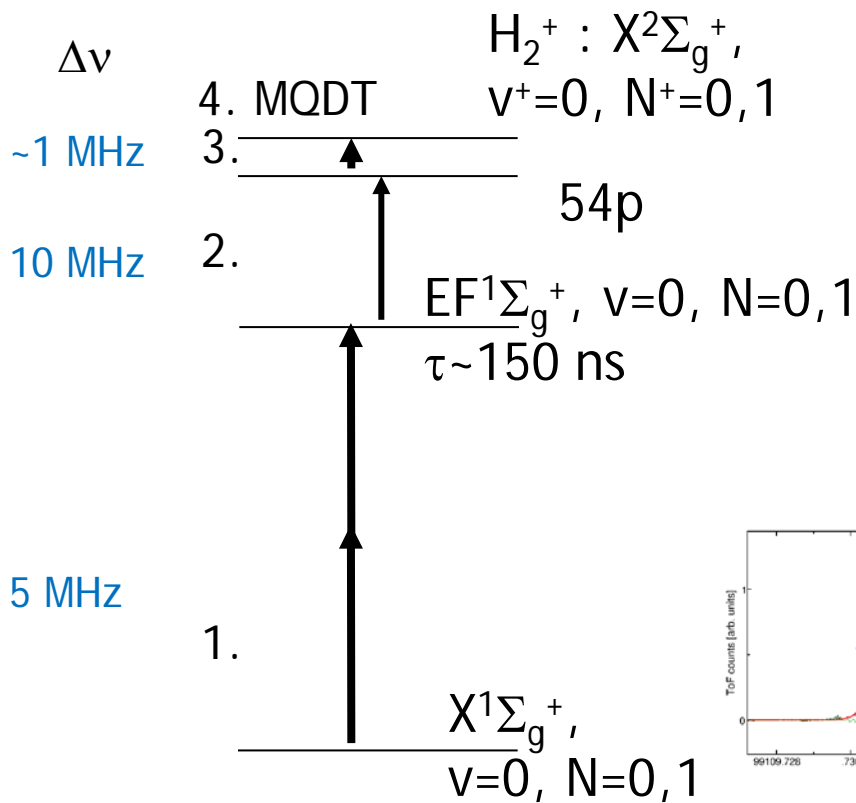
$$D_0(H_2^+) = 21379.350232(50) \text{ cm}^{-1}$$

$$E_{IP}(H) = 109678.77174307(10) \text{ cm}^{-1}$$

$$E_{IP}(H_2) \rightarrow D_0(H_2)$$

(2009 effort) Measurement of IP in H₂

3 step approach (Zürich-Amsterdam collaboration)



$$E_i \text{ (ortho)} = 124\,357.237\,97 \text{ (36) cm}^{-1}$$

$$E_i \text{ (para)} = 124\,417.491\,13 \text{ (37) cm}^{-1}$$

Comparison Theory/Experiment



(Theory: Pachucki, Komasa, et al.: 2010 values)

$D_0(\text{H}_2)$: Experiment [1] 36118.0696(4) cm⁻¹

Theory [2]:

Born–Oppenheimer 36112.5927(1) cm⁻¹

adiabatic + 5.7711(1) cm⁻¹

nonadiabatic + 0.4339(2) cm⁻¹

total α^0 36118.7978(2) cm⁻¹

α^2 all relativistic - 0.5319(5) cm⁻¹

α^3 all QED - 0.1948(3) cm⁻¹

α^4 one-loop term - 0.0016(8) cm⁻¹

Total theory 36118.0695(10) cm⁻¹

QED

$D_0(\text{D}_2)$: Total theory [2] 36748.3633(9) cm⁻¹

Experiment [4] 36748.3629(7) cm⁻¹

Towards measuring the ionisation and dissociation energies of molecular hydrogen with sub-MHz accuracy

Faraday Meeting 2011

Daniel Sprecher,^a Christian Jungen,^{†b} Wim Ubachs^c
and Frédéric Merkt^{*a}



Christian Jungen



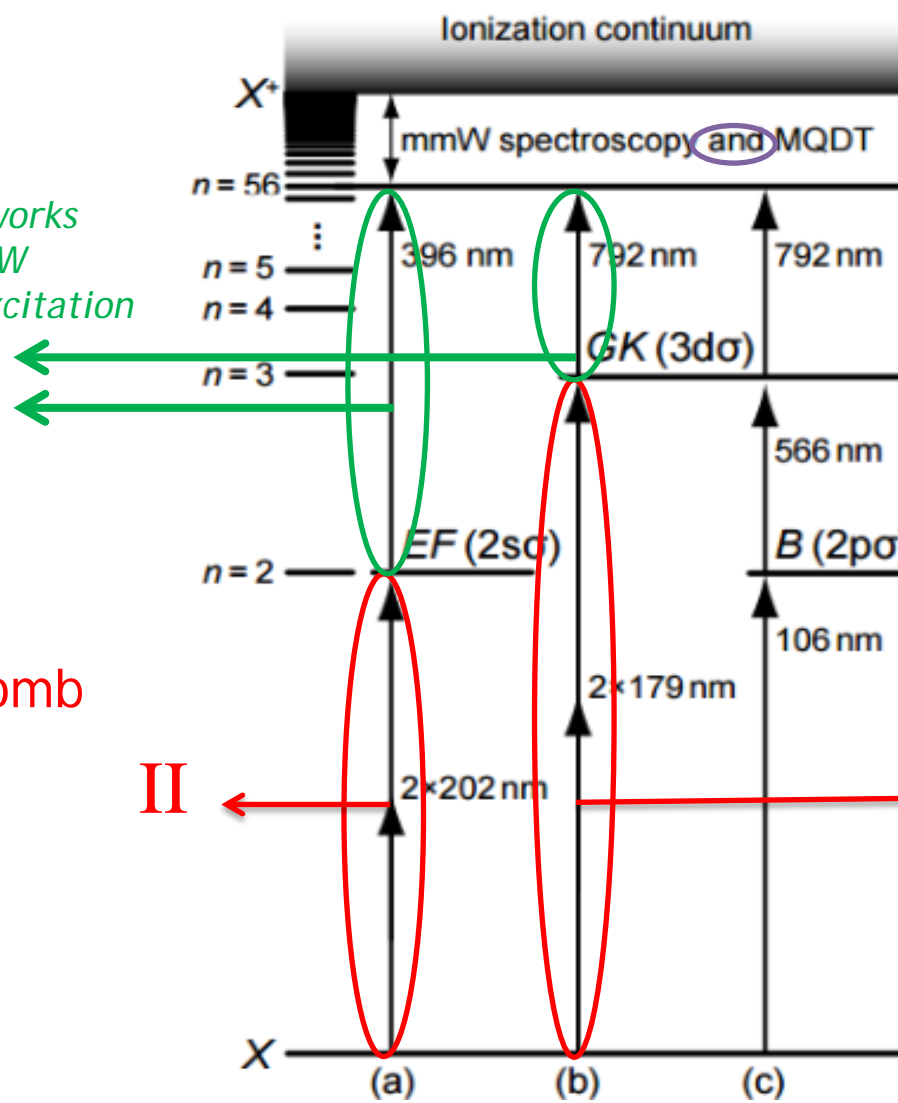
Frédéric Merkt

Zurich works
IR/UV CW
Laser excitation
+MMW

Direct frequency comb



Kjeld Eikema



Further Rydberg
MQDT-analyses

II ← 2x VUV → I

Amsterdam works

Progress in theory; NAPT and Full-NA

PHYSICAL REVIEW LETTERS **121**, 073001 (2018)

→ 4-particle variational

Nonadiabatic Relativistic Correction to the Dissociation Energy of H₂, D₂, and HD

Mariusz Puchalski,¹ Anna Spyszkievicz,¹ Jacek Komasa,¹ and Krzysztof Pachucki²

¹Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland

²Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland



(Received 5 June 2018; published 14 August 2018)

Contribution	$D_0(\text{H}_2)$	$D_0(\text{HD})$	$D_0(\text{D}_2)$
$\alpha^2 m$	36 118.797 746 10(3)	36 406.510 891 37(1)	36 749.090 990 99(2)
$\alpha^4 m$	-0.531 215 6(5)	-0.529 887 5(2)	-0.528 206 05(9)
$\alpha^5 m$	-0.194 8(2)	-0.196 4(2)	-0.198 2(2)
$\alpha^6 m$	-0.002 067(6)	-0.002 080(6)	-0.002 096(6)
$\mathcal{E}_{\text{rel}}^{(2)} \sim \alpha^6 m$	0.000 008 5	0.000 008 6	0.000 008 6
$\alpha^7 m$	0.000 12(6)	0.000 12(6)	0.000 12(6)
FS	-0.000 031	-0.000 117	-0.000 204
Total	36 118.069 76(21)	36 405.782 54(21)	36 748.362 41(21)
Exp.	36 118.069 62(37)	36 405.783 66(36)	36 748.362 86(68)

theory

Ams+Zur 2010

OK

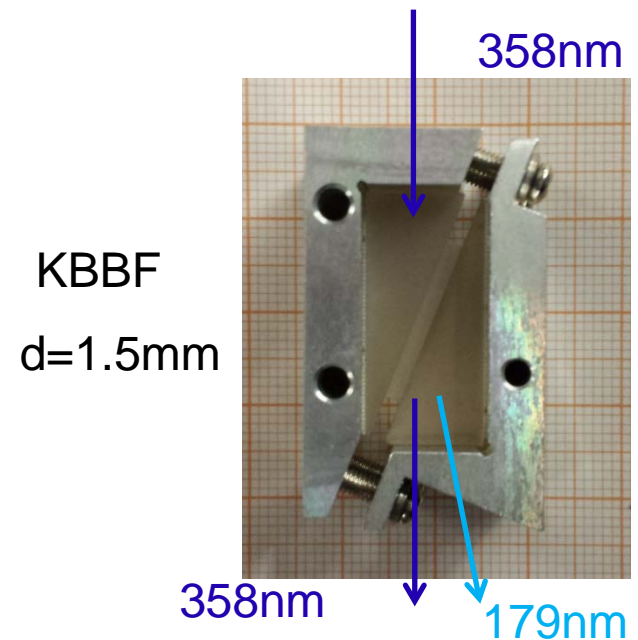
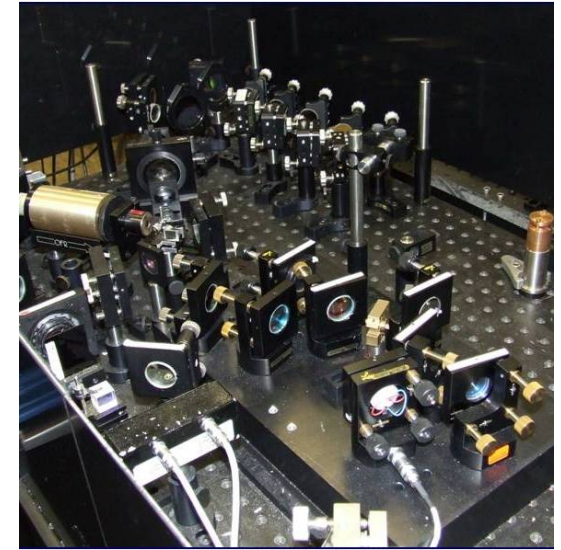
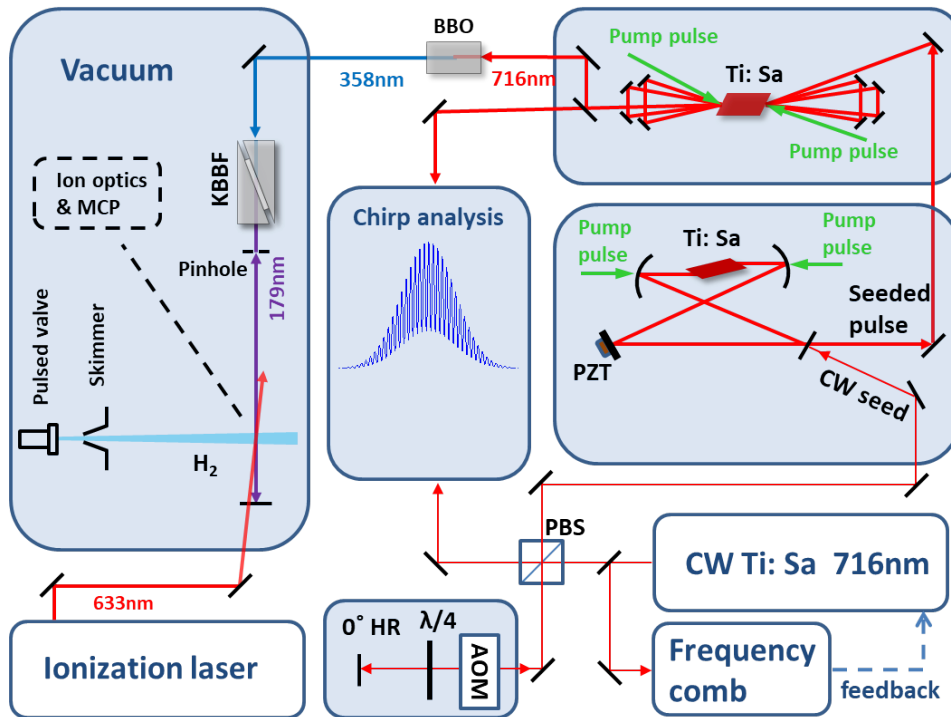
2.7 σ

OK

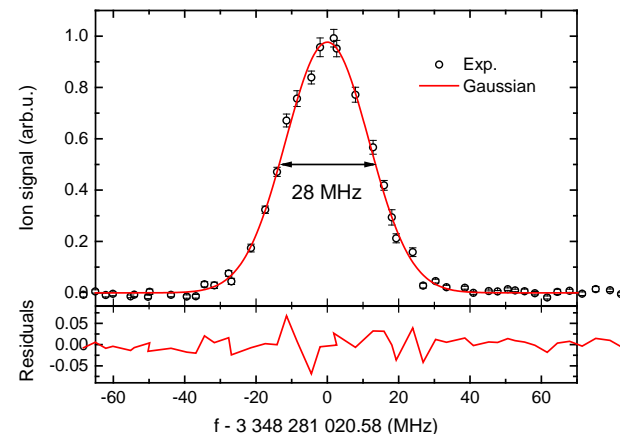
Path I

Two-photon Doppler-free at $\lambda = 179 \text{ nm}$

Experimental Scheme



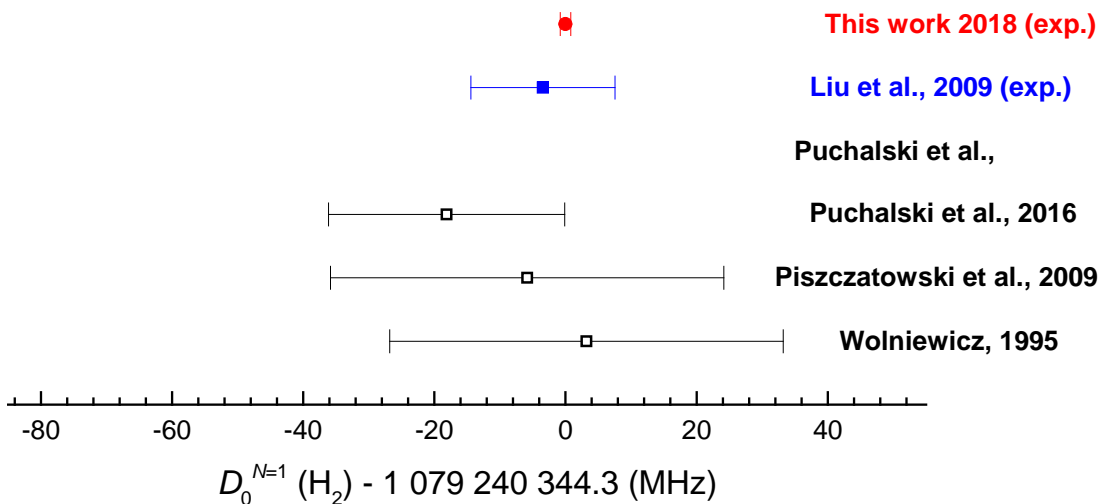
Ortho-H₂ Dissociation Energy of the Hydrogen Molecule at 10⁻⁹ Accuracy



GK-X (N=1): 3348281018.35(49)_{stat}(43)_{sys} MHz (2×10^{-10})

$E_i(\text{H}_2^{\text{ortho}}) = 124\,357.238\,062(25) \text{ cm}^{-1}$

$D_0(\text{H}_2^{\text{ortho}}) = 1\,079\,240\,344.3(8) \text{ MHz} \quad (8 \times 10^{-10})$



This work 2018 (exp.)

Liu et al., 2009 (exp.)

Puchalski et al.,

Puchalski et al., 2016

Piszczatowski et al., 2009

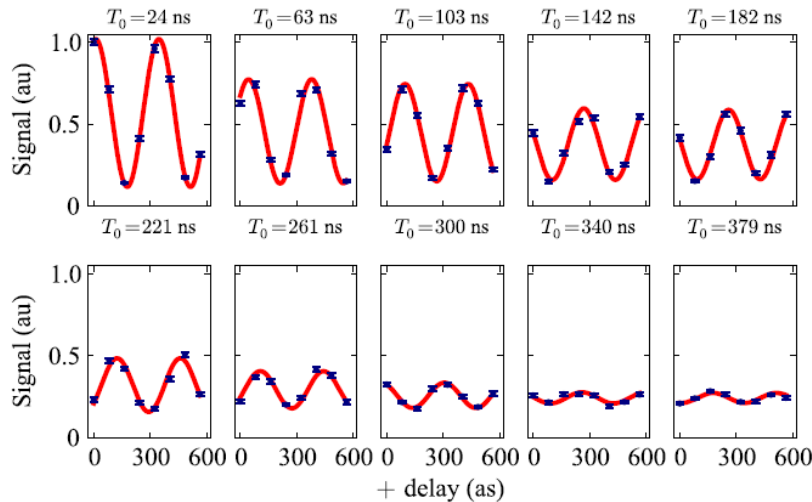
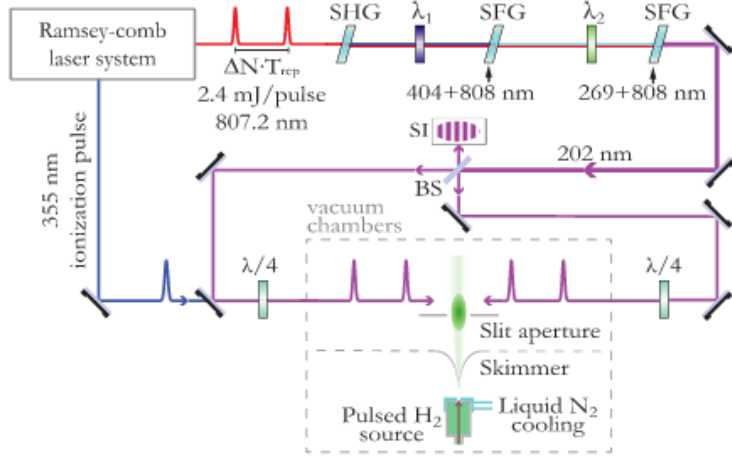
Wolniewicz, 1995

13 times better

Path II (direct frequency comb)

Amsterdam: EF-X(0,0) Q1

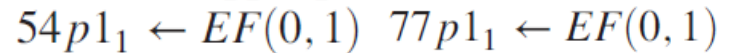
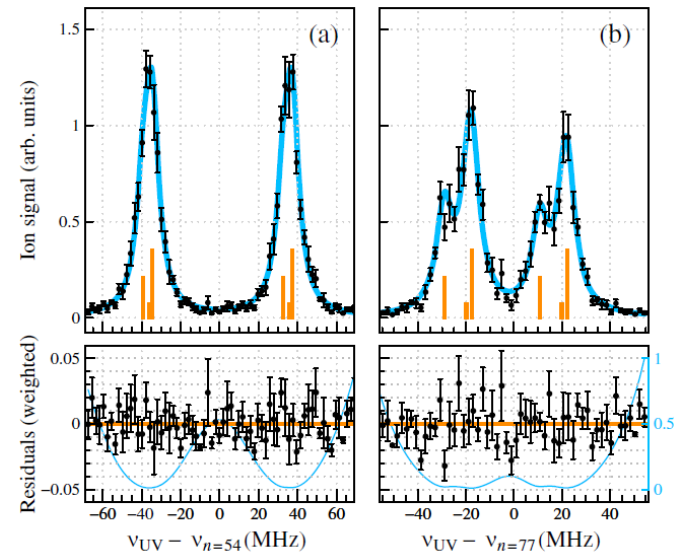
Direct Frequency-comb excitation;
Time-domain Ramsey ($\Delta\nu = 70$ kHz)



Altman et al, Phys. Rev. Lett. 120, 043204 (2018)

Zurich:

CW UV-laser excitation of Rydberg States; EF- np



Hölsch et al, Phys. Rev. Lett. 122, 123002 (2019)

Exp (2019):

$$D_0(\text{H}_2) = 35999.582834 (11) \text{ cm}^{-1}$$

340 kHz uncertainty

Theory (state-of-the-art):

$$D_0(\text{H}_2) = 35999.582820 (26) \text{ cm}^{-1}$$

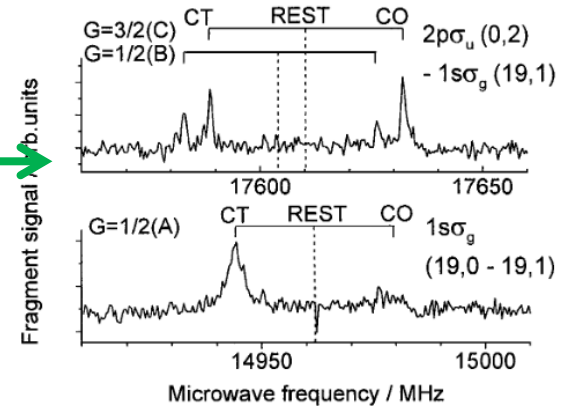
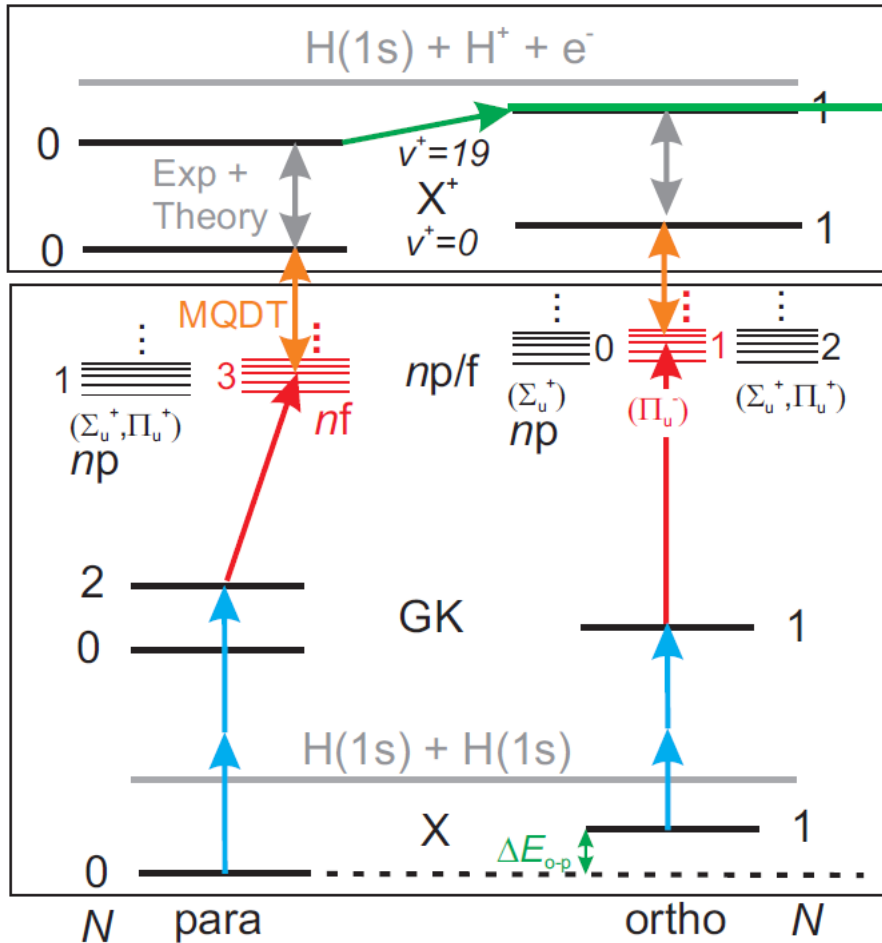
1 MHz uncertainty

Puchalski et al, Phys. Rev. Lett. 122, 1230023 (2019)

Editors' Suggestion

Determination of the Interval between the Ground States of Para- and Ortho-H₂

M. Beyer,^{1,*} N. Hölsch,¹ J. Hussels,² C.-F. Cheng,^{2,†} E. J. Salumbides,² K. S. E. Eikema,²
 W. Ubachs², Ch. Jungen,³ and F. Merkt¹



PRL
2001

Para-H₂

- $l=0$; no hyperfine
- problem with Q(0)
- problems with np series

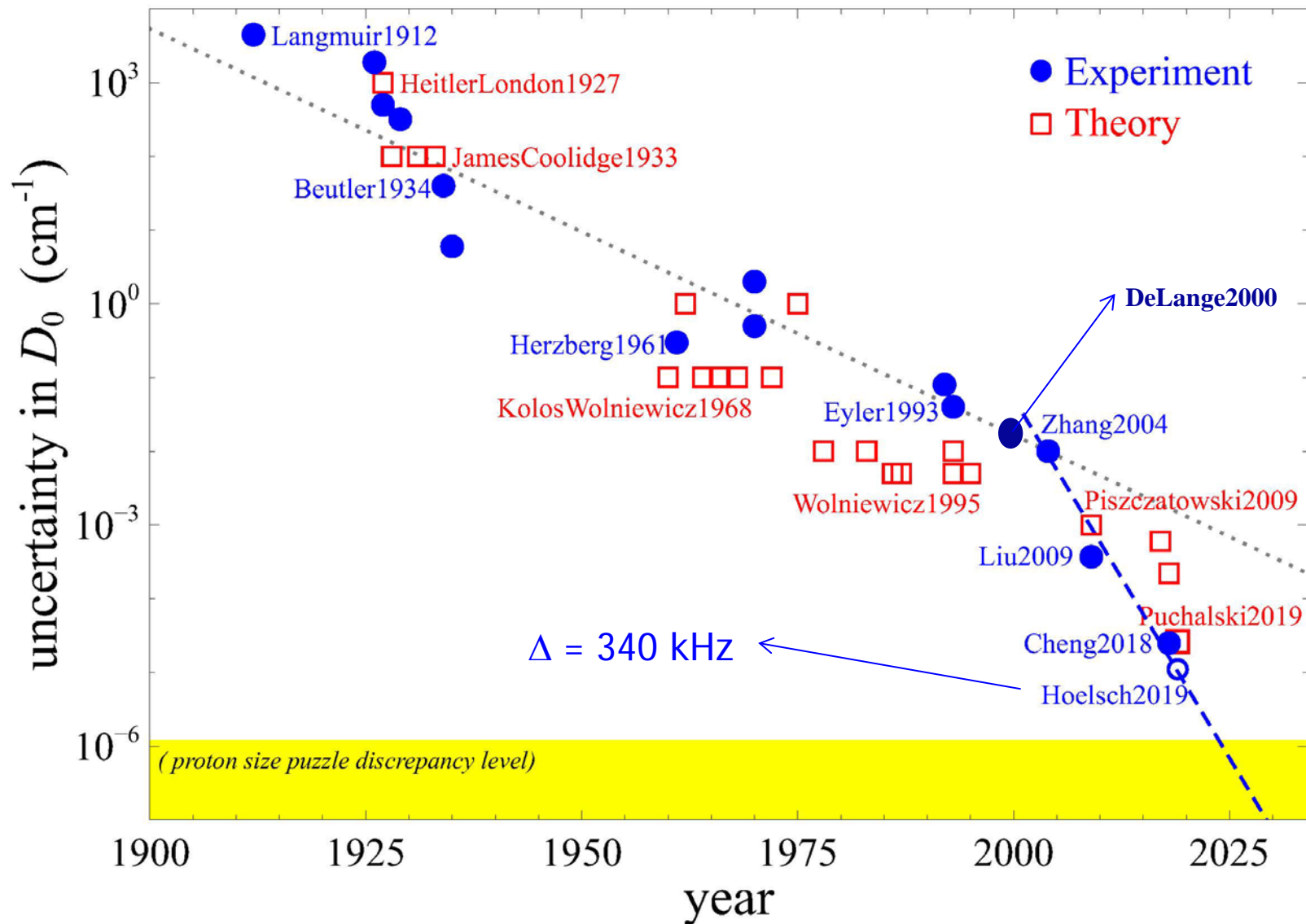
Ortho-para splitting in H₂

$$\Delta_{o-p} = 118.486773(33) \text{ cm}^{-1}$$

$$\Delta_{o-p}^{th} = 118.4868127(11) \text{ cm}^{-1}$$

Bound on spin-dependent forces ?

Pushing the precision frontier; history of $D_0(\text{H}_2)$



This is all pulsed lasers; can we do better ?

No. 4222 September 30, 1950

NATURE

563

Rotation-Vibration Spectrum of the HD Molecule

As is well known, the hydrogen molecule (H_2) has no ordinary dipole infra-red spectrum. Its quadrupole rotation-vibration spectrum is exceedingly weak, and has only recently been found¹. In the case of HD, on account of the asymmetry, there is no longer a distinction between symmetric and antisymmetric rotational levels, and a dipole rotation-vibration spectrum can occur at least in principle. However, the change of dipole moment associated with the vibration of HD is obviously very small. The main contribution to this change is due to the fact that the electrons lag slightly behind the nuclei during the vibrational motion. Wick² has calculated according to wave mechanics the intensity of the fundamental of HD. From his data it can be estimated that the minimum absorbing path required for an observation of the fundamental of HD is 30 m. atm. For the overtones, correspondingly longer path-lengths would be required.

With the technique of long optical paths recently developed³, it appeared promising to attempt an observation of the HD spectrum in the photographic infra-red. A glass absorption tube 5 metres long was filled with HD of 1 atm. pressure. Using approximately 140 and 200 traversals, that is, absorbing paths of 700 and 1,000 metres, seven and six lines respectively were found of the 3-0 and 4-0 bands of HD (second and third overtone) near 9650 and 7400 Å. The accompanying table lists the observed

$$\begin{aligned} B_v &= 45.638_6 - 1.9503(v + \frac{1}{2}) + 0.0140_0 (v + \frac{1}{2})^2. \\ D_v &= 0.02590 - 0.00084(v + \frac{1}{2}) + 0.00004_4 (v + \frac{1}{2})^2. \\ G(v) &= 3809.7_{45}(v + \frac{1}{2}) - 89.7668(v + \frac{1}{2})^2 + \\ &\quad 0.36567(v + \frac{1}{2})^3. \end{aligned}$$

A value $H_v = 0.0000219$ cm.⁻¹ was assumed. The differences between the observed wave-numbers and those calculated from the new constants are given in the last column of the table.

If a sufficient amount of hydrogen and deuterium were present in the atmospheres of the major planets, the 4-0 band of HD might be used for their detection⁷. However, this would require the highest possible resolution, because of the small width of these lines.

It is hoped to observe the fundamental and first overtone of HD in the near future with the aid of a new high-dispersion infra-red spectrometer which is being built in this laboratory.

G. HERZBERG

Division of Physics,
National Research Council,
Ottawa, Ontario, Canada.
May 30.

¹ Herzberg, G., *Nature*, **163**, 170 (1949); *Can. J. Res.*, A, **28**, 144 (1950).

² Wick, G. C., *Atti Reale Accad. Lincei*, **21**, 708 (1935).

³ White, J. U., *J. Opt. Soc. Amer.*, **32**, 285 (1942). Bernstein, H. J., and Herzberg, G., *J. Chem. Phys.*, **16**, 30 (1948).

⁴ Jeppesen, C. R., *Phys. Rev.*, **45**, 480 (1934).

⁵ Urey, H. C., and Teal, G. K., *Rev. Mod. Phys.*, **7**, 34 (1935).

⁶ Teal, G. K., and MacWood, G. E., *J. Chem. Phys.*, **3**, 760 (1950).

⁷ See Herzberg, G., *Astrophys. J.*, **87**, 428 (1938).

FORBIDDEN TRANSITIONS IN DIATOMIC MOLECULES

V. THE ROTATION-VIBRATION SPECTRUM OF THE HYDROGEN-DEUTERIDE (HD) MOLECULE¹

(1960)

HD (2-0) band

R. A. DURIE² AND G. HERZBERG

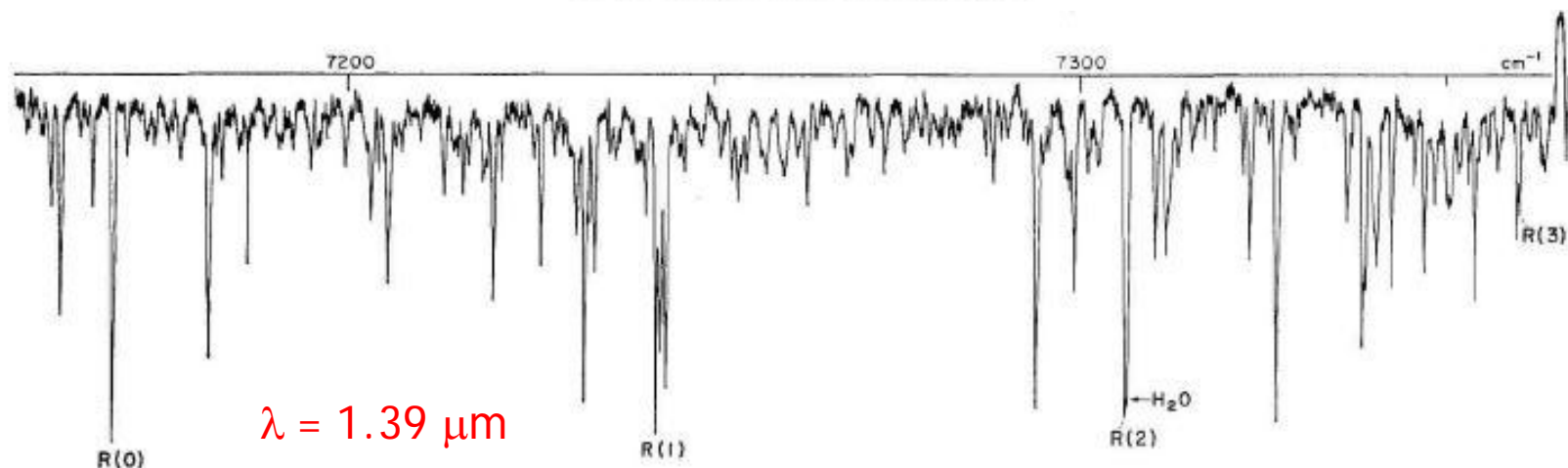
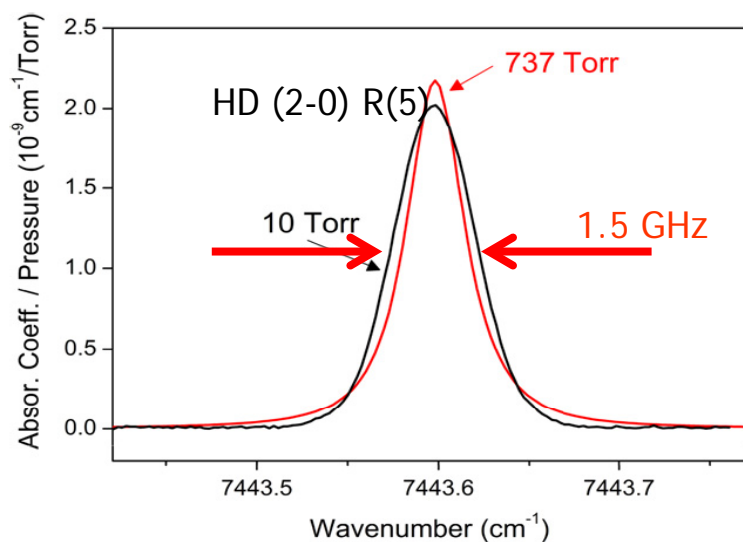


FIG. 2. R branch of 2-0 band of HD.

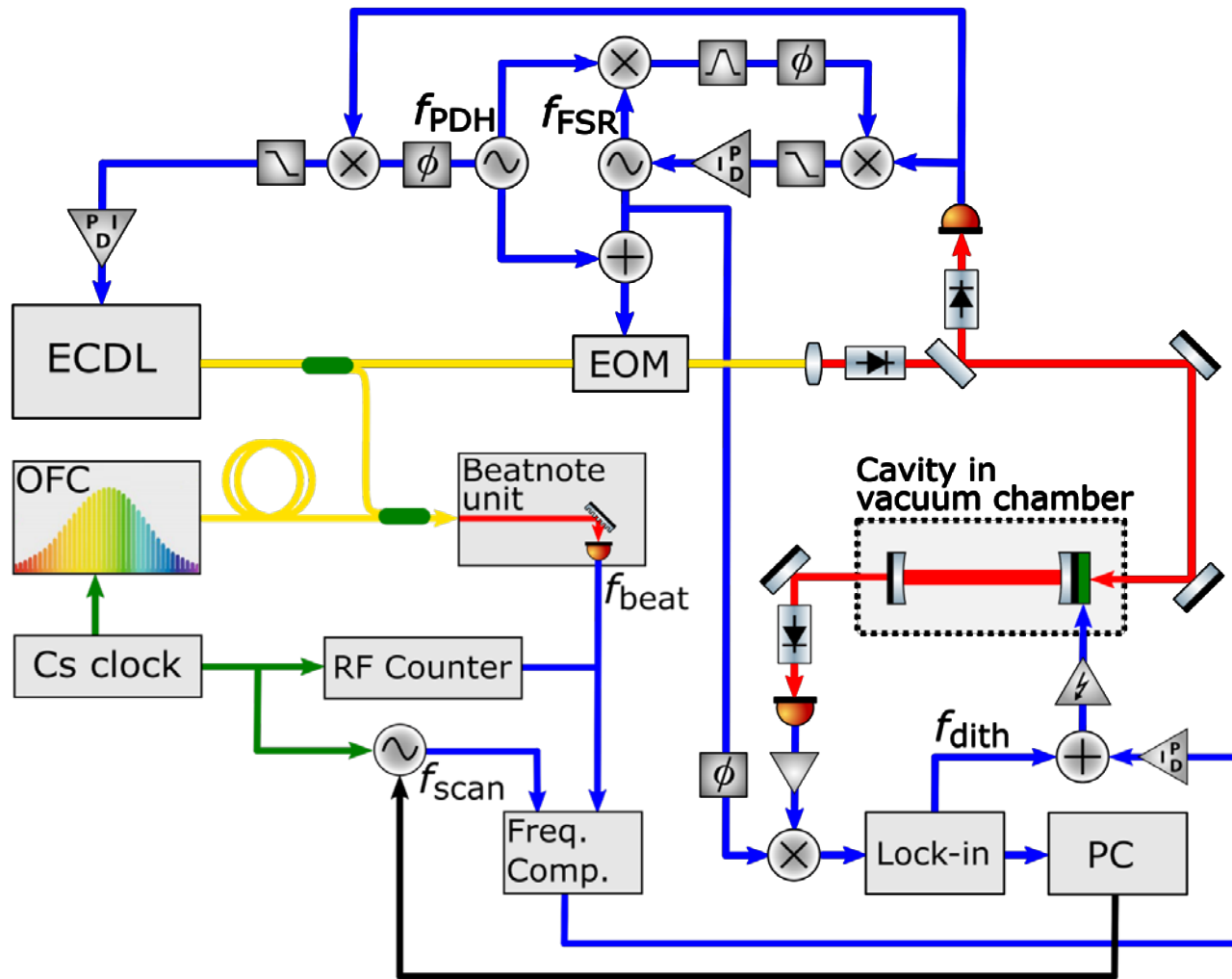
Absorbing path 200 m atm. Rapid scan in 2nd order of 7200 lines/inch grating. The unmarked absorption lines are due to H₂O.



Kassi & Campargue, Grenoble

Journal of Molecular Spectroscopy 267 (2011) 36-42

Frequency comb referenced NICE-OHMS



Cavity

- 130,000 finesse
- 40 km length
- ~ 100 W power

Diode laser

- Locked to cavity
- And to Comb

Detection

- Lock-in

OFC lock

- Stabilized to CS clock

Sub-Doppler Frequency Metrology in HD for Tests of Fundamental Physics

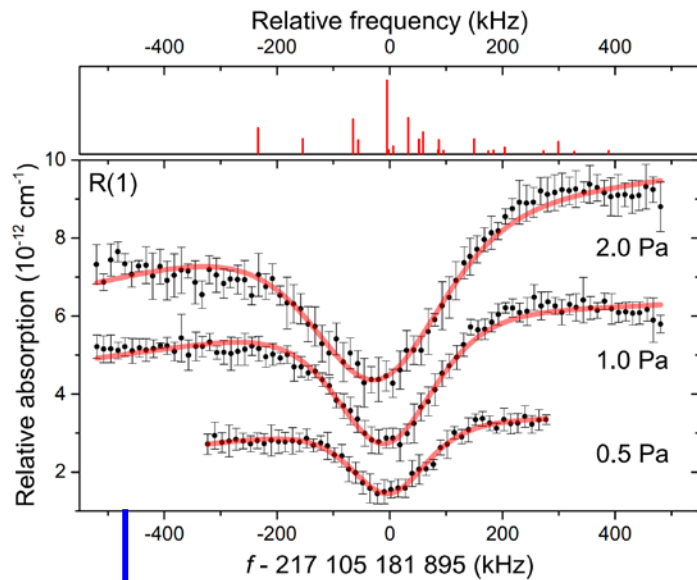
F. M. J. Cozijn,¹ P. Dupré,² E. J. Salumbides,¹ K. S. E. Eikema,¹ and W. Ubachs¹

¹*Department of Physics and Astronomy, LaserLaB, Vrije Universiteit Amsterdam, de Boelelaan 1081, 1081 HV Amsterdam, The Netherlands*

²*Laboratoire de Physico-Chimie de l'Atmosphère, Université du Littoral Côte d'Opale, 189A Avenue Maurice Schumann, 59140 Dunkerque, France*



(Received 22 December 2017; published 9 April 2018)



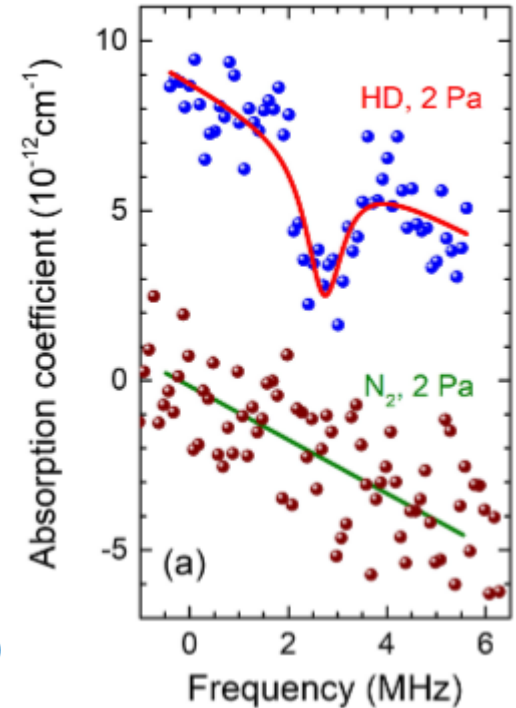
R(1) [2-0]

← AMS

Hefei →

$\Delta_{\text{AMS-Hefei}} = 900 \text{ kHz}$

(Discussed Opt. Lett. 2019)



Toward a Determination of the Proton-Electron Mass Ratio from the Lamb-Dip Measurement of HD

L.-G. Tao,¹ A.-W. Liu,^{1,2} K. Pachucki,³ J. Komasa,⁴ Y. R. Sun,^{1,2} J. Wang,¹ and S.-M. Hu^{1,2,*}

¹*Hefei National Laboratory for Physical Sciences at Microscale, iChem center, University of Science and Technology of China, Hefei, 230026 China*

²*CAS Center for Excellence in Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei, 230026 China*

³*Faculty of Physics, University of Warsaw, Pasteura 5, 02-093, Warsaw, Poland*

⁴*Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland*

Lines too narrow:

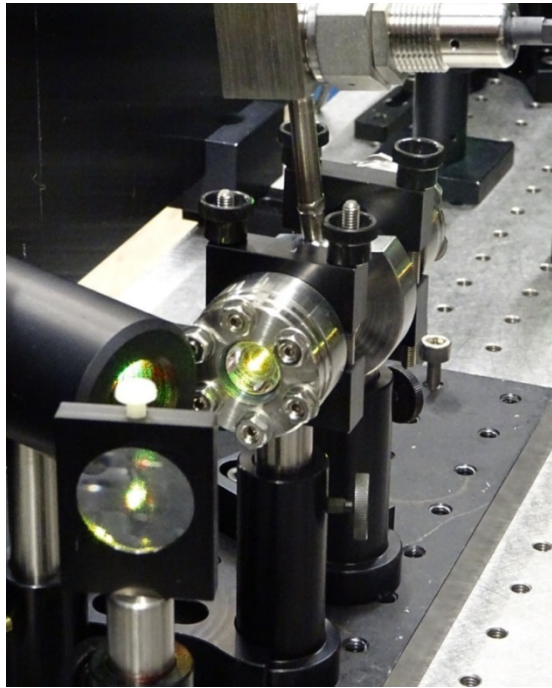
- transit time
- hyperfine
- recoil doublet

Section of cold molecules
(100 W intra / sat 10 kW)



(Received 22 December 2017; published 9 April 2018)

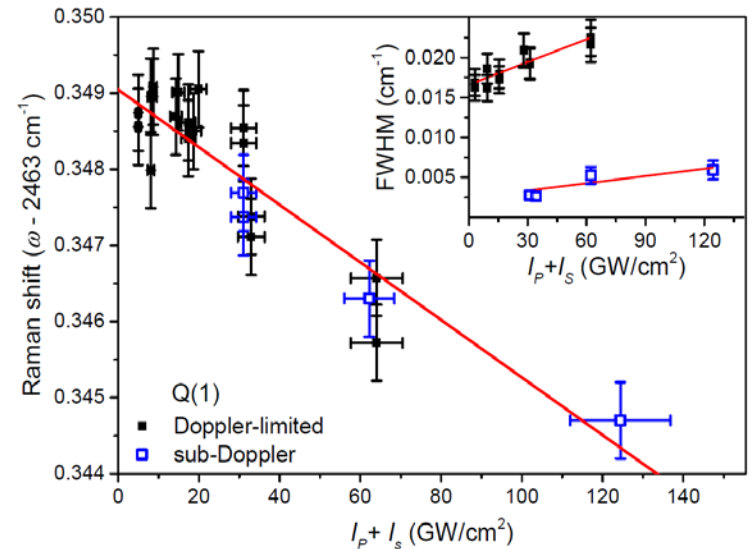
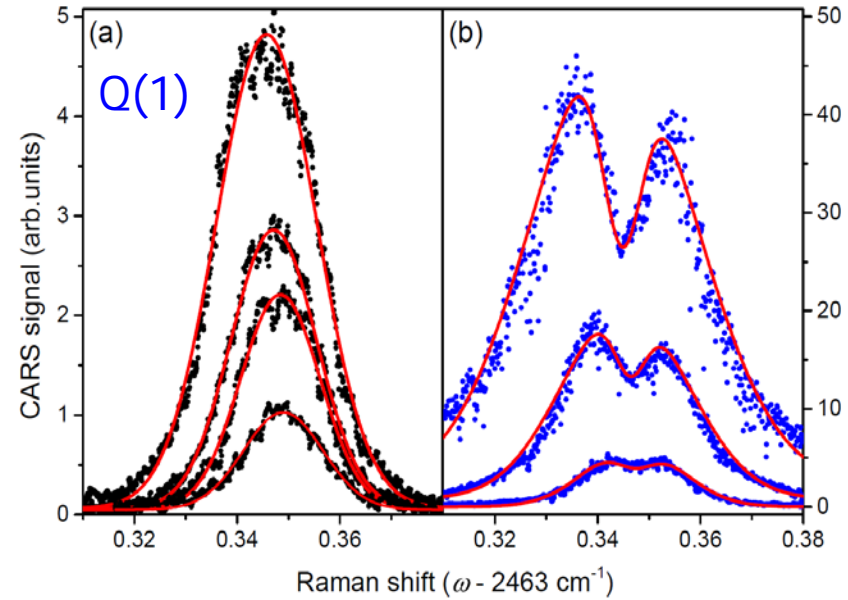
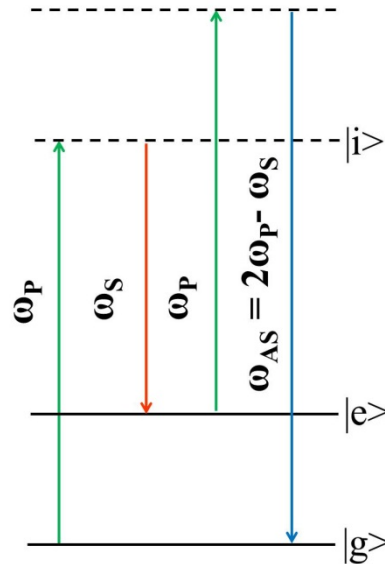
Pushing the experimental frontier : include Tritium



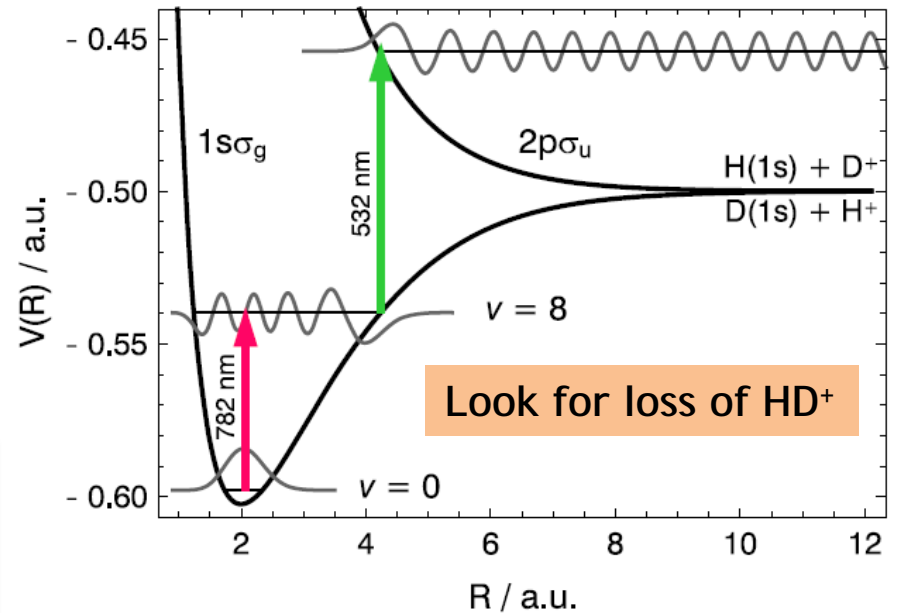
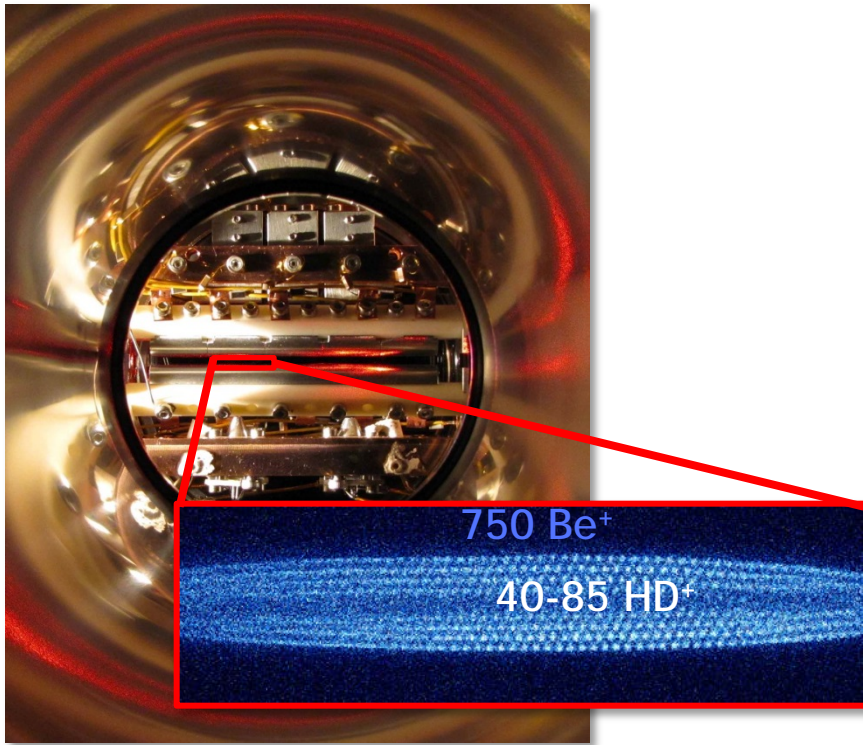
<1 GBq (15 mbar x cm³)
 All optical experiment
 CARS (saturated CARS)

T₂

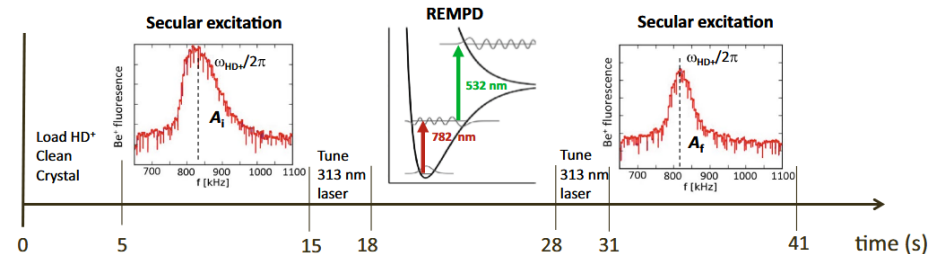
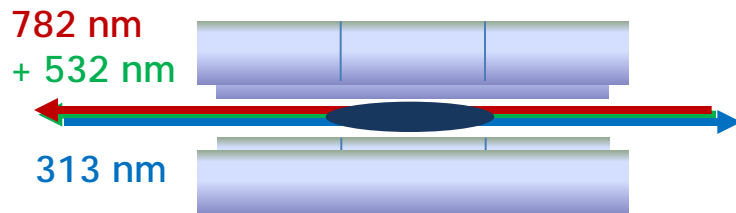
CARS scheme



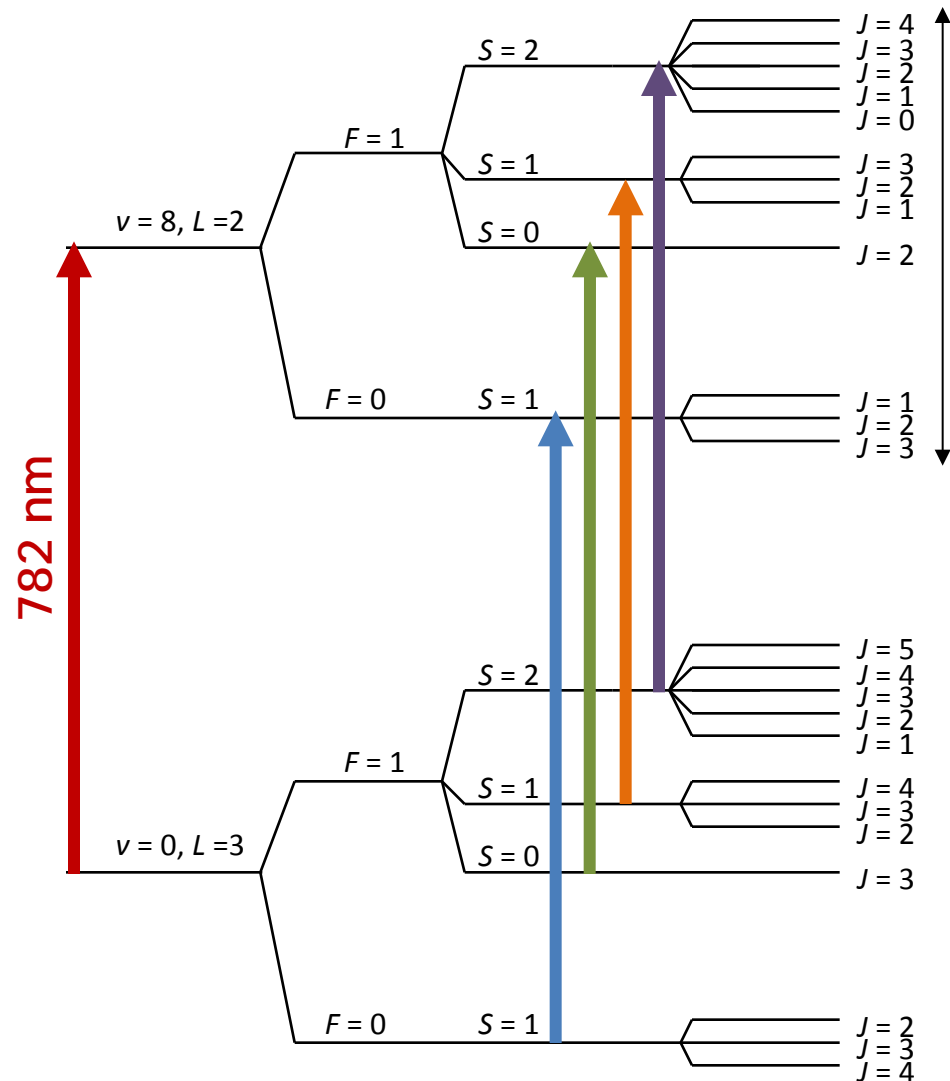
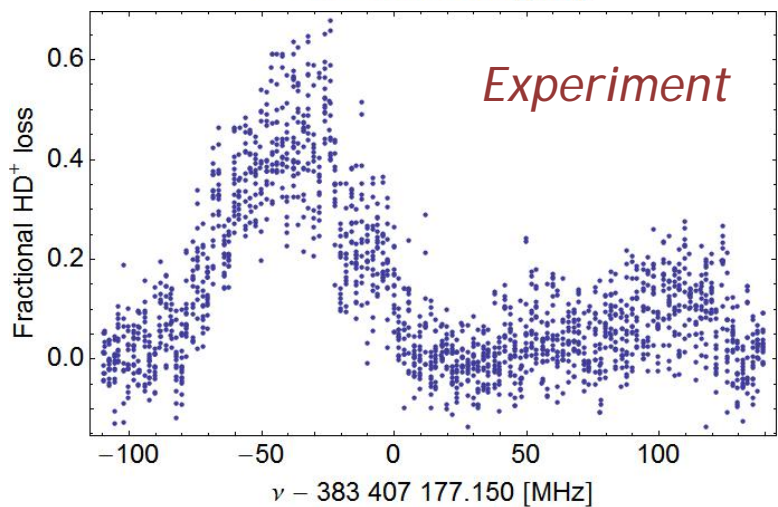
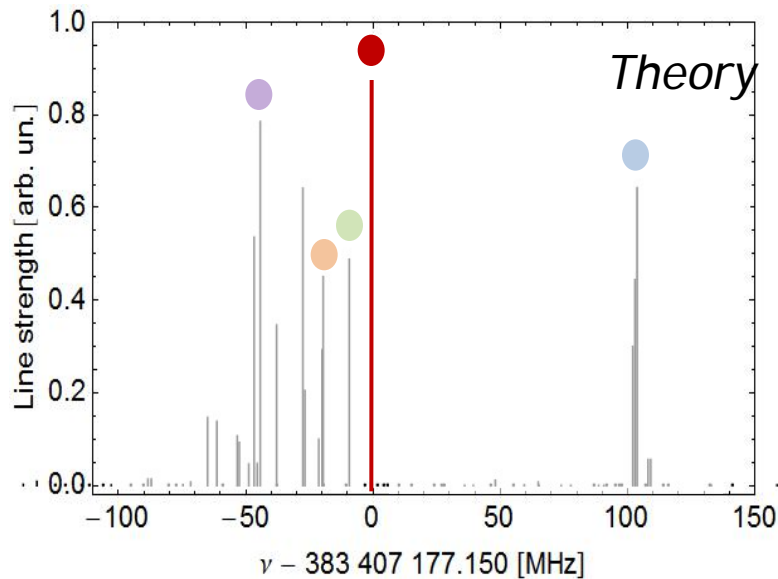
HD⁺ ions in a trap; measurement of (8,0)



Signal detection by REMPD



HD⁺ spectrum



Experiment: 383,407,177.38(41) MHz
 Theory*: 383,407,177.150(15) MHz

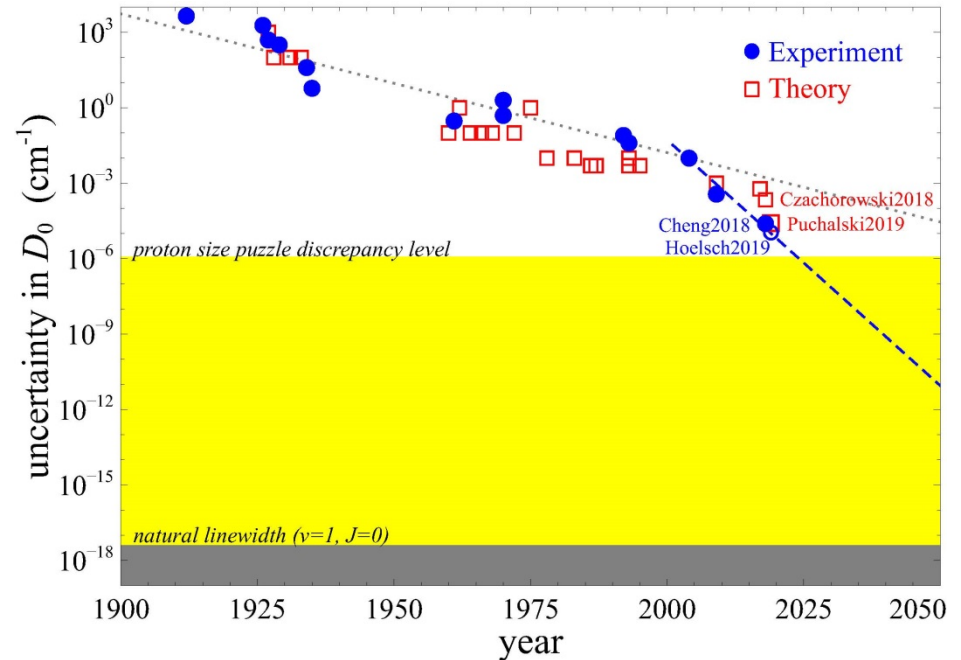
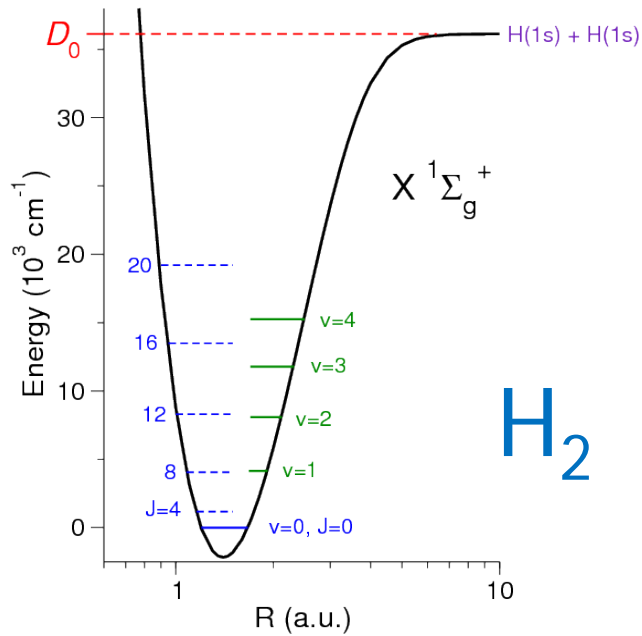
*Korobov, Hilico, Karr, Phys. Rev. A 89, 032511 (2014)

Conclusion:

Molecular Spectroscopy for Fundamental Physics

OUTLOOK:

A future molecular test system for physics



Lifetimes 10^6 seconds (!)

Quadrupole transitions $\sim 10^{14}$ Hz

Possible precision 20-digit



There is room at the bottom

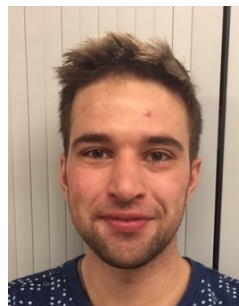
Thanks & Acknowledgement



Julija
Bagdonaite



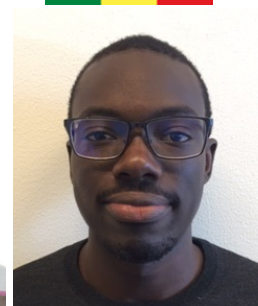
Edcel
Salumbides



Joel
Hussels



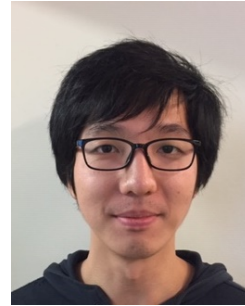
Madhu T
Trivikram



Meissa
Diouf



Frank
Cozijn



Kin Fung
Lai



Kjeld
Eikema



Jeroen
Koelemeij



Matthias
Germann



Frederic
Merkt



Krzysztof
Pachucki



Magnus
Schloesser