Spectroscopy of HD^+ and the Rydberg constant

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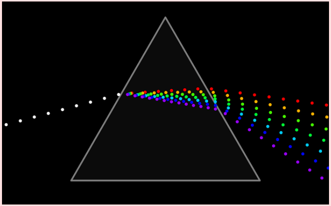


FFK19, Balaton, June 9-14, 2019

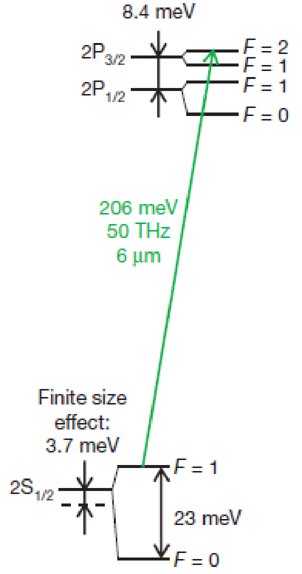
This work was done in collaboration with

- LKB: Laurent Hilico, Jean-Philippe Karr, Mohammad Haidar;
- Wuhan, WIPM CAS: Zhen-Xiang Zhong.

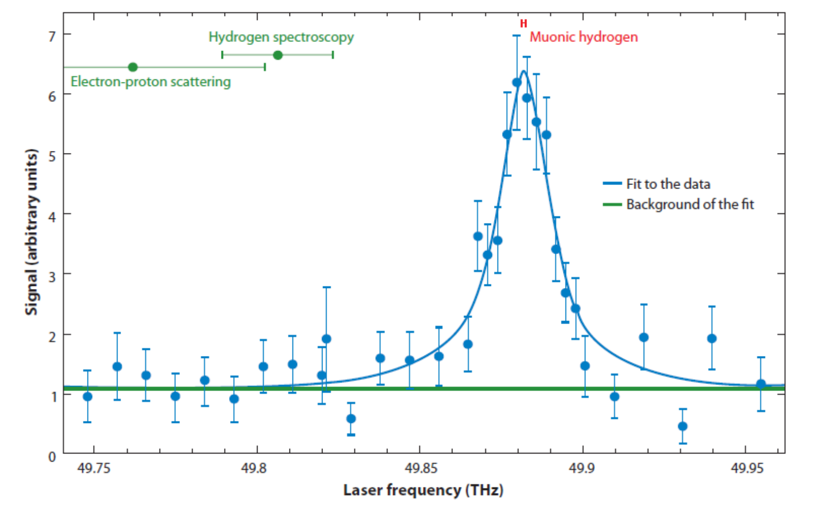
The proton charge radius and Rydberg constant

Proton charge radius puzzle. First experiment.

Randolf Pohl, *et al.* (CREMA Collab.)
The Size of the proton.
Nature **466**, 213 (2010).

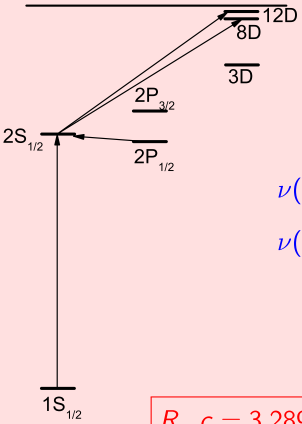


Proton charge radius puzzle



Rydberg constant

Rydberg constant from hydrogen atom



$$\nu(2S_{1/2}\text{–}12D_{5/2}) = 799\,191\,727.403\,7(47)\ \text{MHz}$$

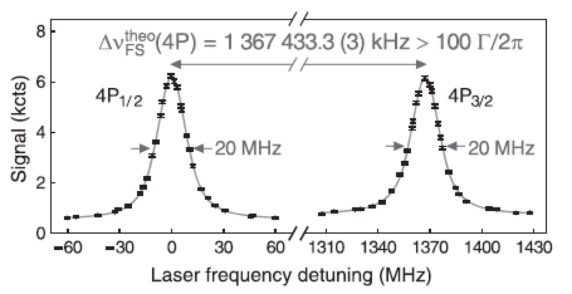
$$\nu(1S_{1/2}-2S_{1/2}) = 2\,466\,061\,413.187\,018(11)\ \text{MHz}$$

$$R_\infty c = 3.289\,841\,960\,355(19)\times 10^{15}\ \text{Hz}$$

Rydberg constant. MPQ experiment, Garching.

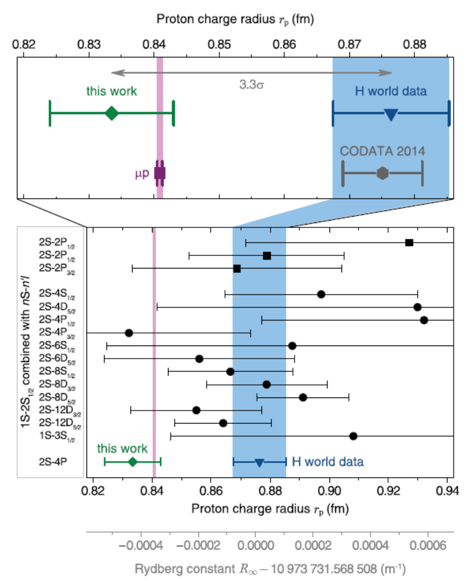
$1S$-$4P$ transition.

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



conservation. (**B**) Typical experimental fluorescence signal from a single line scan over the 2S-$4P_{1/2}$ (left) and 2S-$4P_{3/2}$ (right) resonance (black diamonds). The observed line width (full width at half maximum) of ~2π × 20 MHz is larger than the natural line width Γ = 2π × 12.9 MHz because of Doppler and power broadening. The accuracy of our measurement corresponds to almost 1 part in 10,000 of the observed line width. The constant background counts are caused by the decay of 2S atoms inside the detector (*17*). kcts, kilocounts.

Rydberg constant. MPQ experiment.



Rydberg constant. LKB experiment, Paris.

$1S$-$3S$ transition.
H. Fleurbaey, *et al.* Phys. Rev. Lett. **120**, 183001 (2018).

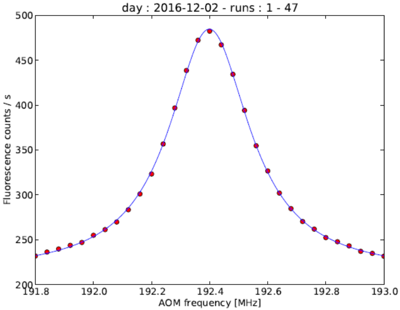


Figure 2.17: Hydrogen $1S - 3S$ signal. This is the mean over one day of recording using the 1.2-MHz (31 points) scan width, corresponding to an integration time of 4 hours. No magnetic field was applied to the atoms apart from the residual offset of -0.3 G, and the pressure was 2.7×10^{-5} mbar. Red points are experimental data, the blue line is a Lorenztian fit.

Rydberg constant. LKB experiment, Paris.

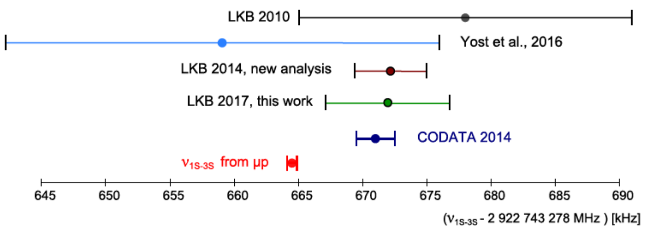


Figure 4.7: Comparison of our measurement (LKB 2017) with other determinations of the $1S - 3S$ transition frequency.

Precision spectroscopy of HD^+. Experiment

Pure rotational transition

Lamb-Dicke regime

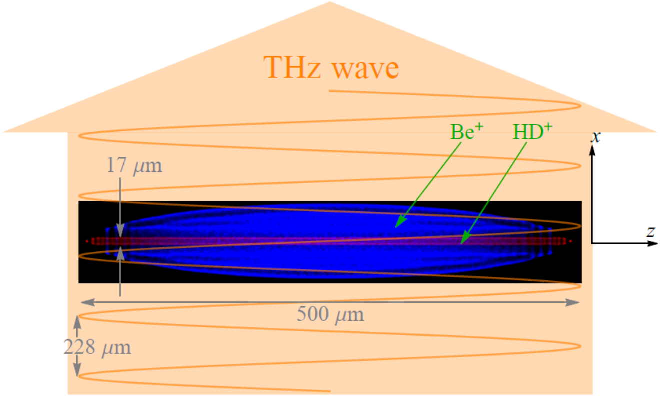


FIG. 1. **Principle of the Lamb-Dicke rotational spectroscopy of sympathetically cooled molecular ions.** The ion cluster is prolate, and the sympathetically cooled ions exhibit a relatively small motional range in the directions x, y perpendicular to the trap axis z. The spectroscopy radiation propagates perpendicular to z. The ion cluster image is a time average of ion trajectories

Pure rotational transition $(N=0, v=0) \to (1,0)$ in HD^+

Pure rotational transition in HD^+ (in kHz).
CODATA14 recommended values of constants.

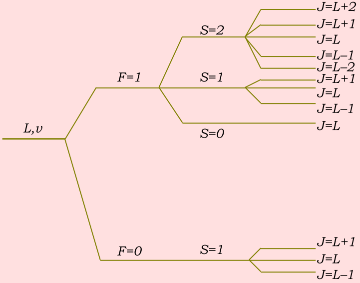
	HD^+
ΔE_{nr}	1 314 886 776.354
ΔE_{α^2}	48 416.164
ΔE_{α^3}	−9 378.125
ΔE_{α^4}	−65.625(2)
ΔE_{α^5}	3.923(3)
ΔE_{α^6}	−0.070(18)
ΔE_{tot}	1 314 925 752.627(18)

$$u_r(\nu) = u_r(\mu) = 1.35 \times 10^{-11}$$

HFS of HD$^+$ molecular ion

Coupling scheme:

$$\begin{cases} \mathbf{F} = \mathbf{I}_p + \mathbf{s}_e, \\ \mathbf{S} = \mathbf{F} + \mathbf{I}_d, \\ \mathbf{J} = \mathbf{S} + \mathbf{N}. \end{cases}$$



Effective Hamiltonian:

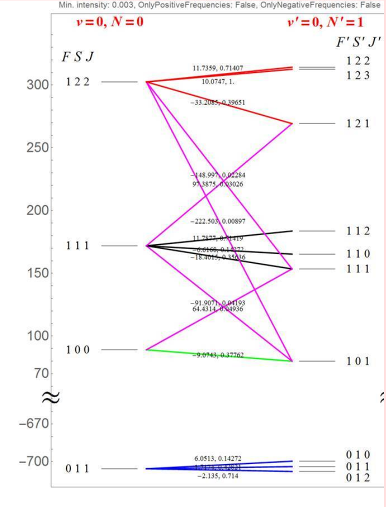
$$H_{\mathrm{HFS}} = E_1(\mathbf{N}\cdot\mathbf{s}_e) + E_2(\mathbf{N}\cdot\mathbf{I}_p) + E_3(\mathbf{N}\cdot\mathbf{I}_d) + E_4(\mathbf{I}_p\cdot\mathbf{s}_e) + E_5(\mathbf{I}_d\cdot\mathbf{s}_e)$$
$$+E_6\left\{2\mathbf{N}^2(\mathbf{I}_p\cdot\mathbf{s}_e) - 3[(\mathbf{N}\cdot\mathbf{I}_p)(\mathbf{N}\cdot\mathbf{s}_e) + (\mathbf{N}\cdot\mathbf{s}_e)(\mathbf{N}\cdot\mathbf{I}_p)]\right\} + E_7\left\{2\mathbf{N}^2(\mathbf{I}_d\cdot\mathbf{s}_e) - 3[(\mathbf{N}\cdot\mathbf{I}_d)(\mathbf{N}\cdot\mathbf{s}_e) + (\mathbf{N}\cdot\mathbf{s}_e)(\mathbf{N}\cdot\mathbf{I}_d)]\right\}$$
$$+E_8\left\{2\mathbf{N}^2(\mathbf{I}_p\cdot\mathbf{I}_d) - 3[(\mathbf{N}\cdot\mathbf{I}_p)(\mathbf{N}\cdot\mathbf{I}_d) + (\mathbf{N}\cdot\mathbf{I}_d)(\mathbf{N}\cdot\mathbf{I}_p)]\right\} + E_9\left[\mathbf{N}^2\mathbf{I}_d^2 - \frac{3}{2}(\mathbf{N}\cdot\mathbf{I}_d) - 3(\mathbf{N}\cdot\mathbf{I}_d)^2\right].$$

Coefficients E_i of the effective spin Hamiltonian (in MHz)

E_1	E_2	E_4	E_5	E_6	E_7
31.9846	−3.134[−02]	924.629	142.146	8.6111	1.3218

Pure rotational transition. HF structure

Transition: $(N = 0, v = 0) \to (1, 0)$



Theory for stretch states

Spin-averaged transition frequency:

$$f_{\mathrm{spin-avg}} = 1\,314\,925.752\,627(18)\ \mathrm{MHz}$$

Hyperfine shift:

$$\begin{aligned}\Delta E(vL, F=1, S=2, J=N+2)/h &= E_4/4 + E_5/2 \\ &+ (E_1 + E_2 + 2E_3 + E_6 + 2E_7 + 2E_8 + E_9)\,N/2 \\ &- (2E_6 + 4E_7 + 4E_8 + 2E_9)\,N^2/2 \\ &= 10.0747(10)\ \mathrm{MHz},\end{aligned}$$

So far, the coefficients have been calculated within the Breit-Pauli approximation. Taking into account the higher order correction to E_1, E_6, and E_7, we have

$$\Delta E(vL, F=1, S=2, J=N+2)/h = 10.07540(2)\ \mathrm{MHz},$$

Experiment at Düsseldorf. Preliminary results.

Pure rotational transition $(N=0, v=0) \to (1,0)$ in HD^+.

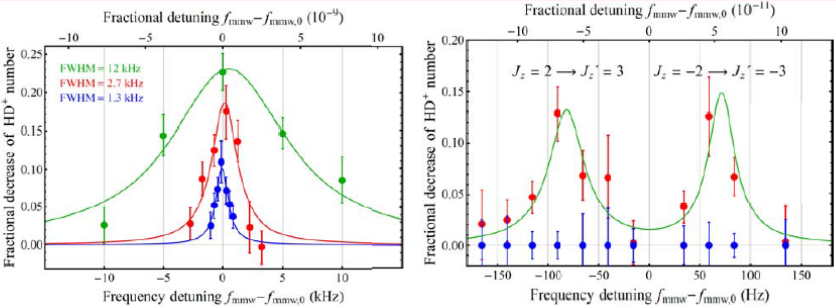


Figure: Lamb-Dicke rotational spectroscopy of an ensemble of sympathetically cooled HD^+ at 1.3 THz. *Left*: power-broadened transition, with linewidth of 1×10^{-9}. *Right*: at strongly reduced intensity, linewidth of 3×10^{-11}. [Ali17].

Pure vibrational transition

Pure vibrational transition $(N=3, v=0) \to (3,9)$ in HD^+

Pure vibrational transition in HD^+ (in kHz).
CODATA14 recommended values of constants.

	HD^+
ΔE_{nr}	415 260 910 661.2
ΔE_{α^2}	5666 203.4
ΔE_{α^3}	−1640 449.4
ΔE_{α^4}	−11 666.7(4)
ΔE_{α^5}	732.2(5)
ΔE_{α^6}	−14.8(3.1)
ΔE_{tot}	415 264 925 467.0(3.1)

$$u_r(\nu) = 7.5 \times 10^{-12}$$

Experiment at Amsterdam. Schematic diagram of the experiment.

Pure vibrational two-photon transition $(N=3, v=0) \to (3,9)$ in HD^+.

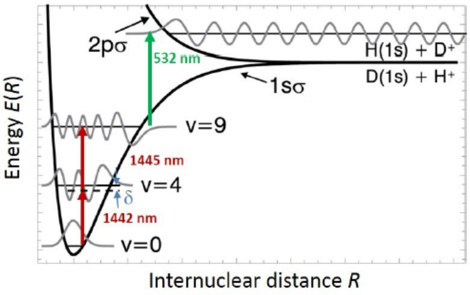


Figure 1. (colour online) Schematic of quasi-degenerate two-photon spectroscopy of the (v, L) : $(0,3) \to (9,3)$ transition in HD^+ (the hyperfine structure of the rovibrational levels is not shown). The transition is detected by resonance-enhanced multiphoton dissociation (REMPD) using an additional laser at 532 nm; the REMPD signal is the fractional loss of HD^+ ions.

Experiment at Amsterdam. Profile line.

Pure vibrational two-photon transition $(N=3, v=0) \to (3, 9)$ in HD^+.

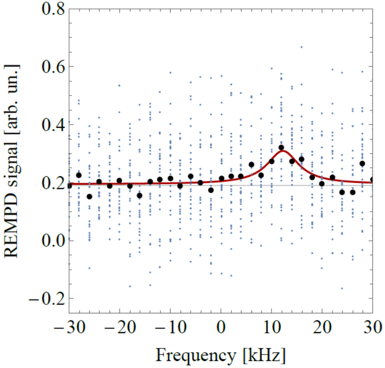


Figure 7.8: Measured spectrum of the hyperfine component $(F, S, J) : (1, 2, 5) \to (1, 2, 5)$ of the two-photon $(v, L) : (0, 3) \to (9, 3)$ ro-vibrational transition in HD^+ is fitted with a Lorentzian lineshape model as mentioned in the text. The width of the fitted lineshape is found to be 7.63 ± 2.53 kHz and the uncertainty (standard deviation) of the fitted centre frequency is 0.75 kHz. The origin of the frequency axis is at an arbitrary position as more measurements are required to assess all the systematic effects.

Rydberg constant

Energies of diatomic molecule

Adiabatic approximation:

$$\left\{\frac{1}{2\mu_n}\left[-\frac{1}{R^2}\frac{d}{dR}R^2\frac{d}{dR}+\frac{N(N+1)}{R^2}\right]+\mathcal{E}_{\rm el}(R)+\mathcal{E}_{\rm ad}(R)\right\}\chi_{\rm ad}=E_{\rm ad}\,\chi_{\rm ad},$$

The energy structure of diatomic molecular states:

$$E_i = U_e + B_e\,N(N+1) + \hbar\omega_e(v+\frac{1}{2}),$$

where $B_e \sim 1/M$ and $\omega_e \sim 1/\sqrt{M}$.

Inferring the Rydberg constant

Master equation

$$\begin{cases} \dfrac{\nu_N^{\mathrm{exp}} - \nu_N^{\mathrm{th}}}{\nu_N^{\mathrm{th}}} = \dfrac{\Delta R_\infty}{R_\infty} - \dfrac{\Delta\mu}{\mu} \\ \dfrac{\nu_v^{\mathrm{exp}} - \nu_v^{\mathrm{th}}}{\nu_v^{\mathrm{th}}} = \dfrac{\Delta R_\infty}{R_\infty} - \dfrac{1}{2}\dfrac{\Delta\mu}{\mu} \end{cases}$$

Inverting it one gets

$$\begin{cases} \dfrac{\Delta R_\infty}{R_\infty} = -\dfrac{\nu_N^{\mathrm{exp}} - \nu_N^{\mathrm{th}}}{\nu_N^{\mathrm{th}}} + 2\dfrac{\nu_v^{\mathrm{exp}} - \nu_v^{\mathrm{th}}}{\nu_v^{\mathrm{th}}} \\ \dfrac{\Delta\mu}{\mu} = -2\dfrac{\nu_N^{\mathrm{exp}} - \nu_N^{\mathrm{th}}}{\nu_N^{\mathrm{th}}} + 2\dfrac{\nu_v^{\mathrm{exp}} - \nu_v^{\mathrm{th}}}{\nu_v^{\mathrm{th}}} \end{cases}$$

Uncertainty for the Rydberg constant from the present theoretical uncertainty is $\boxed{u_r = 1.9 \times 10^{-11}}$.

Experiment at Düsseldorf. Preliminary results.

The first very preliminary result:

$$R_\infty(m_e/m_p + m_e/m_d) = 8966.2051500(9) \qquad (u_r = 1.0 \cdot 10^{-10})$$

CODATA14 value (mainly limited by the uncertainty in m_p):

$$R_\infty(m_e/m_p + m_e/m_d) = 8966.20514923(56) \qquad (u_r = 6.2 \cdot 10^{-11})$$

And now the Blaum experiment on m_p. They've got a value, which is 3σ away off the CODATA14 m_p!

$$R_\infty(m_e/m_p + m_e/m_d) = 8966.20515001(28) \qquad (u_r = 3.1 \cdot 10^{-11})$$

Thank you for your attention!