MPQ

Precision spectroscopy of atomic hydrogen and deuterium at MPQ

V. Wirthl, L. Maisenbacher, D. Taray, A. Grinin, A. Matveev, O. Amit, R. Pohl, T. W. Hänsch and Th. Udem
Max Planck Institute of Quantum Optics, Germany
PREN-2022 Paris, June 20, 2022

Motivation for hydrogen and deuterium spectroscopy

Hydrogen/deuterium energy levels including finite nuclear size and QED corrections:

$$E_{nlj} = hc\,R_\infty \frac{m_{\mathrm{red}}}{m_e}\left(-\frac{1}{n^2} + f_{nlj}\left(\alpha, \frac{m_e}{m_N}, \ldots\right) + \delta_{l0}\frac{C_{\mathrm{NS}}}{n^3} r_N^2\right)$$

Precisely calculated for hydrogen-like atoms (~12 digits)

Motivation: metrology, test QED and consistency of Standard Model, nuclear physics

Constants $\alpha, m_e/m_N, \cdots$ from e.g. Penning traps, atom interferometry

Considering spectroscopy only, two constants left for us:

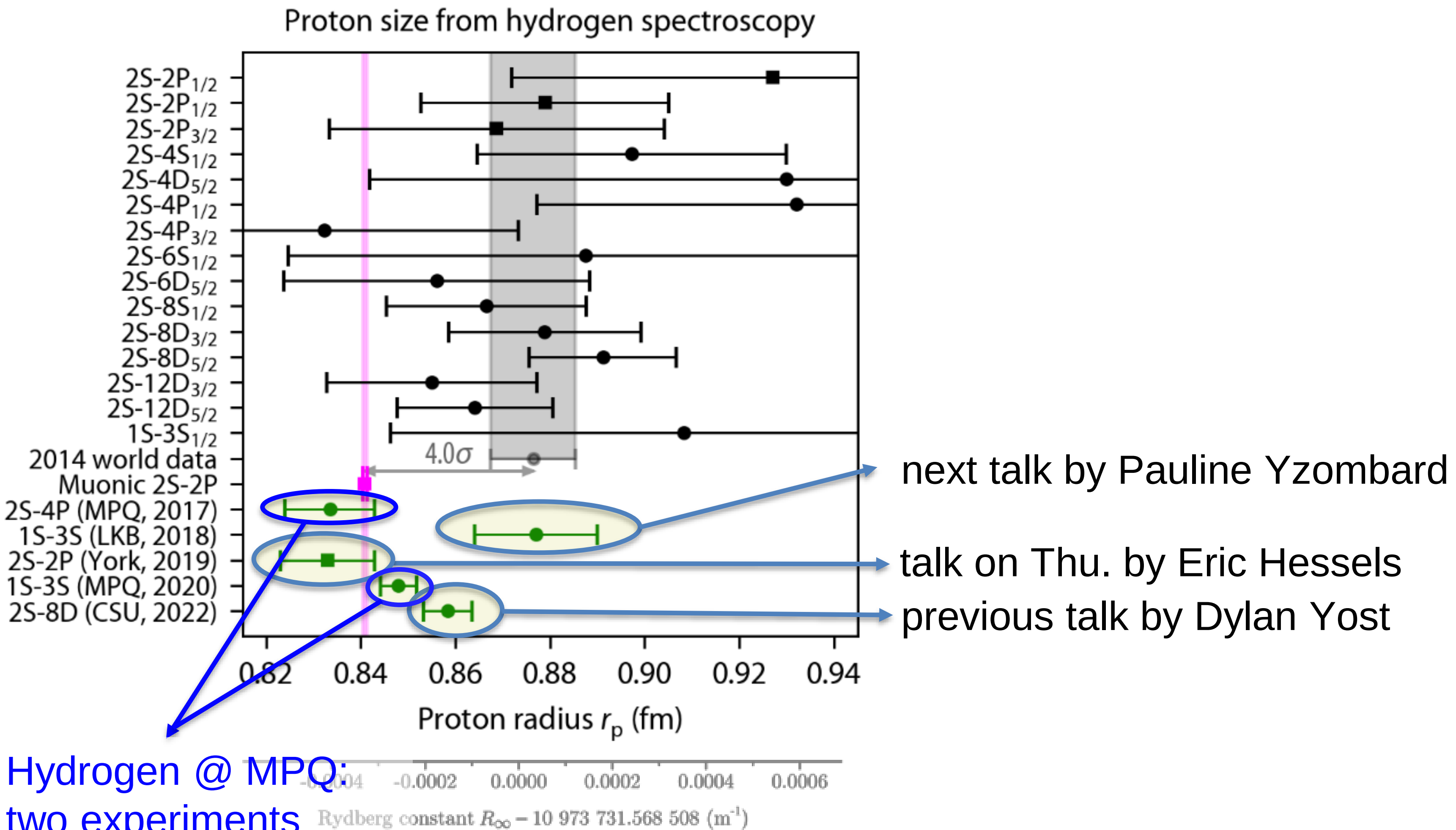
Rydberg constant R_∞ and RMS charge radius r_N^2

➔ need **at least 2 measurements**, more for tests

Measurement 1): e.g. narrow **1S-2S transition** using Doppler-free two-photon spectroscopy in hydrogen [1] and deuterium [2-3]

[1] C. G. Parthey *et al.*, PRL **107**, 203001 (2011); [2] C. G. Parthey *et al.*, PRL **104**, 233001 (2011); [3] R. Pohl *et al.*, Metrologia **54**, L1 (2017)

Considering hydrogen and deuterium separately: 1S-2S transition measurement in hydrogen or deuterium combined with other transition measurement:



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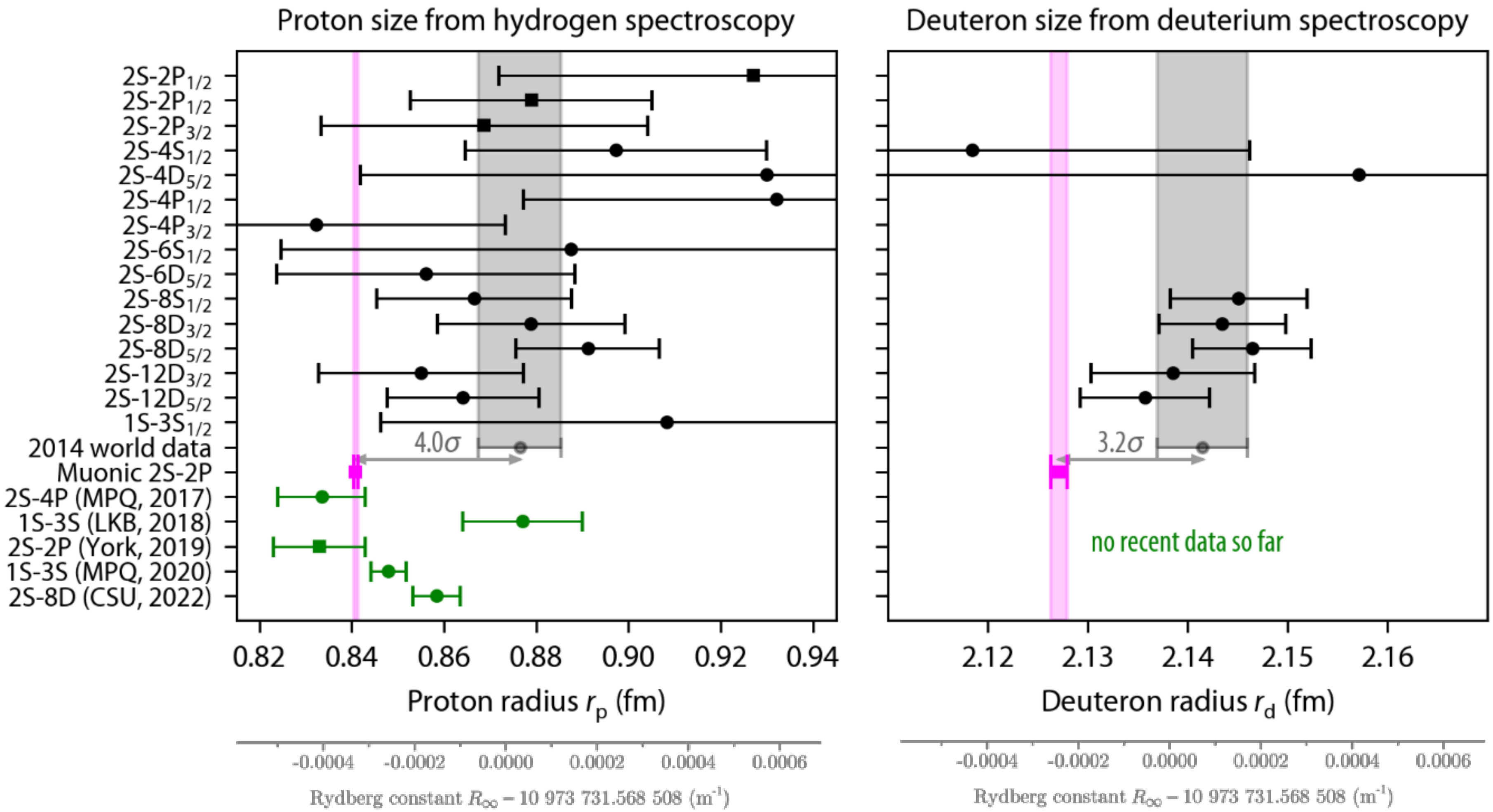
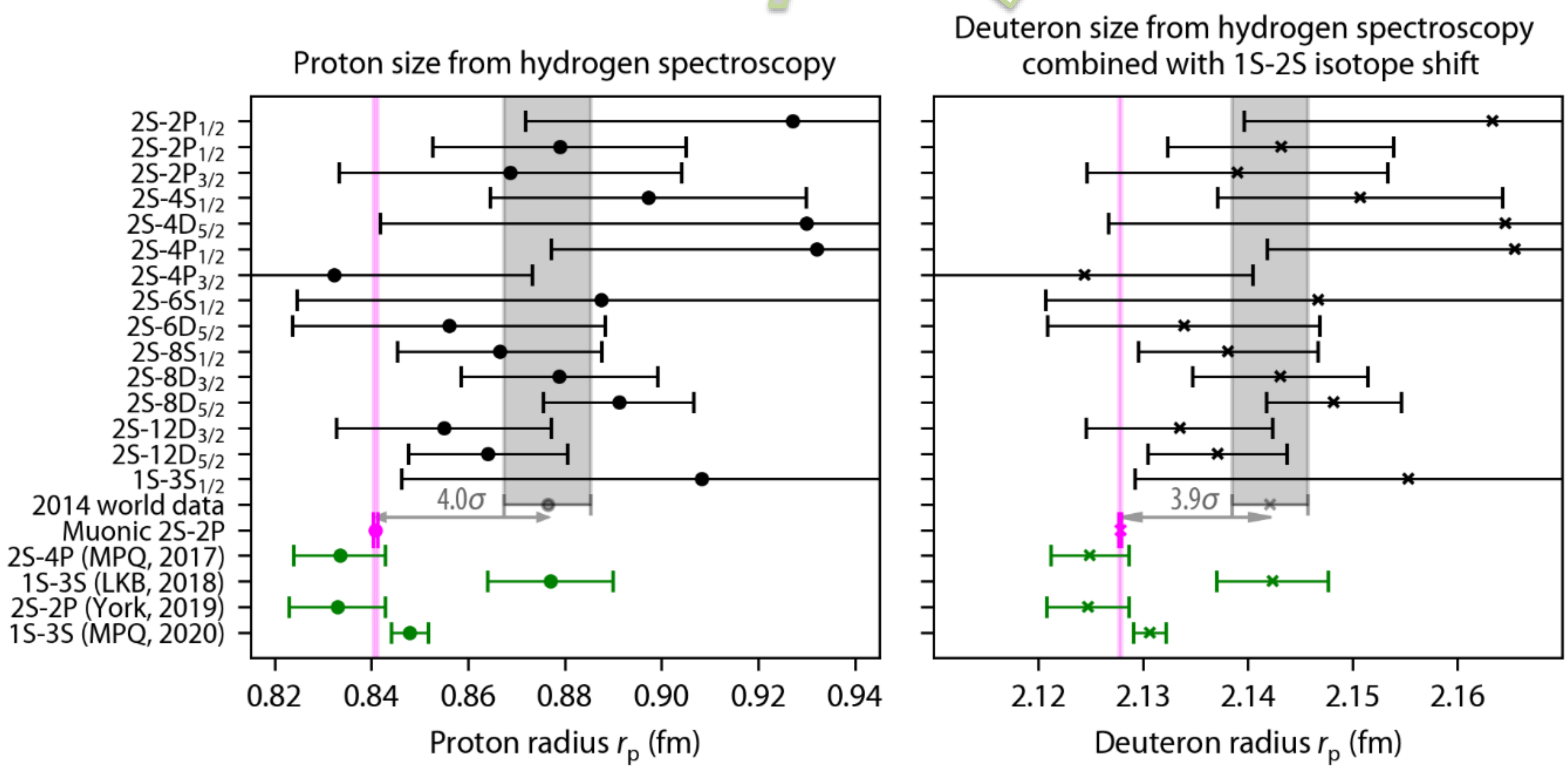


Similar discrepancy for the muonic and electronic deuterium, but so far no recent data from deuterium spectroscopy

Combination [1-3] of theory and experiment for the
1S-2S isotope shift links proton and deuteron radii:

$$r_{\mathrm{d}}^{2} = r_{\mathrm{struct.}}^{2} + r_{\mathrm{p}}^{2} + r_{\mathrm{n}}^{2} + \frac{3\hbar^2}{4m_p^2c^2}$$

$$r_d^2 - r_p^2 = 3.820\,70\,(31)\ \mathrm{fm}^2$$



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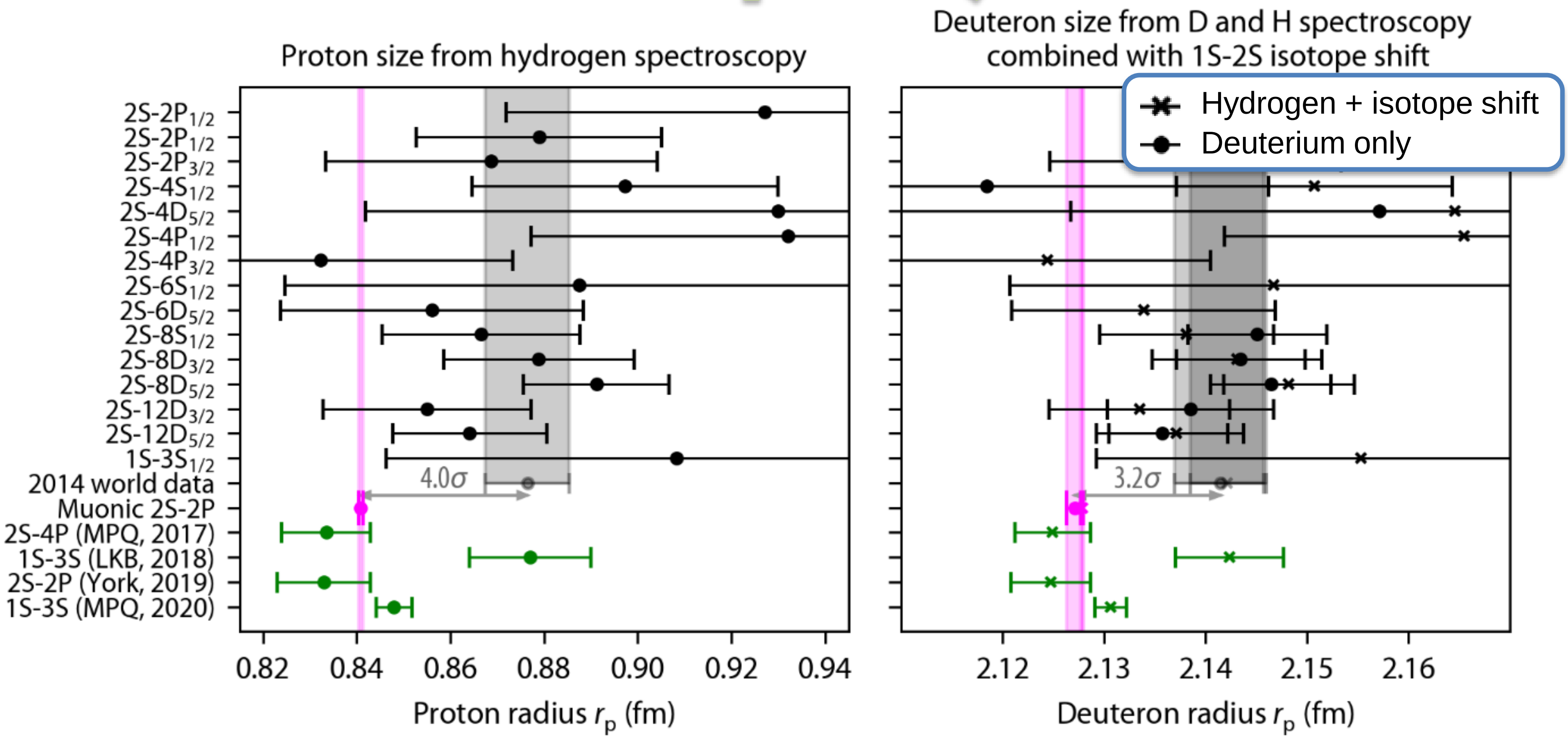

Direct deuteron radius determination also consistency check for hydrogen isotope shift theory

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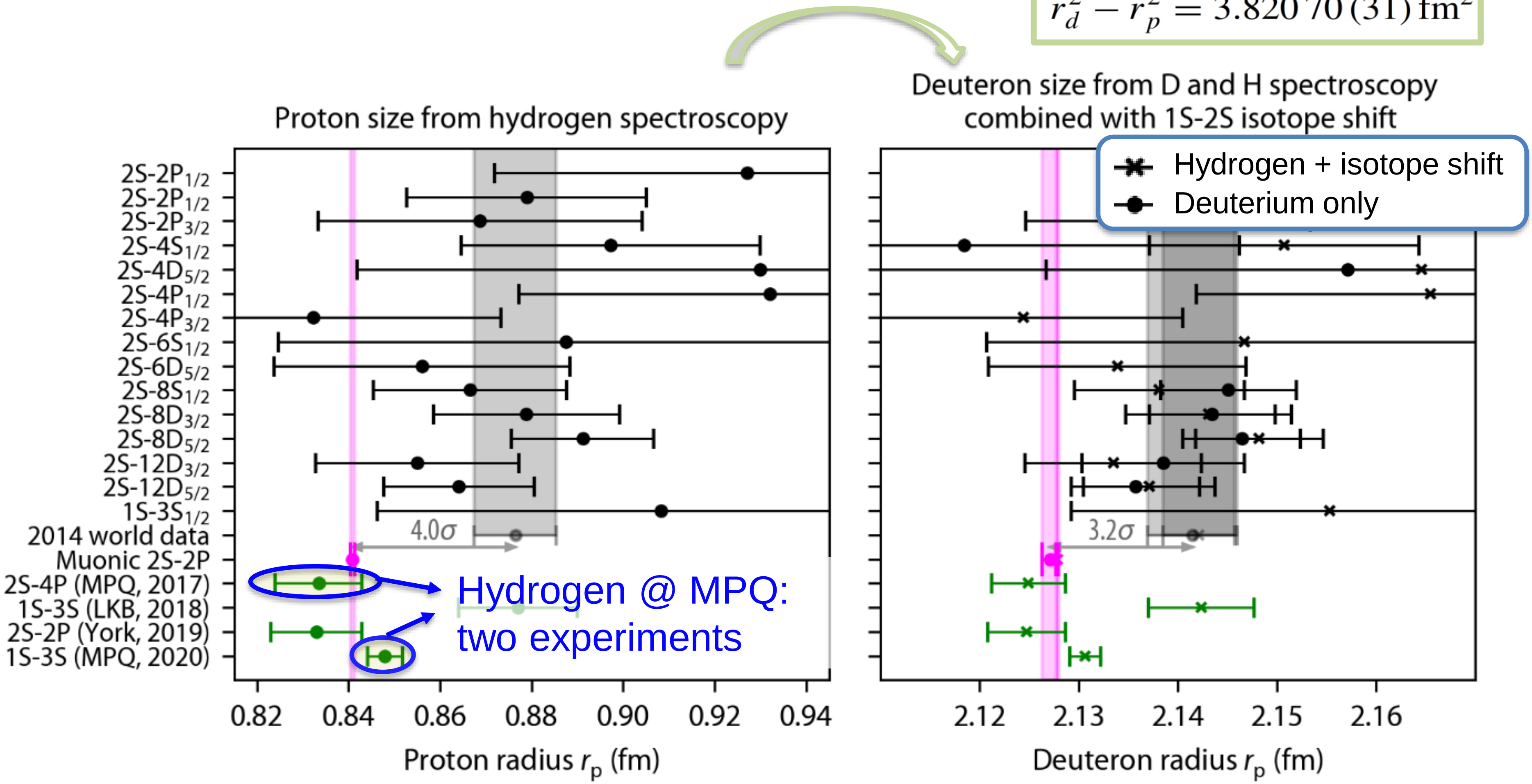

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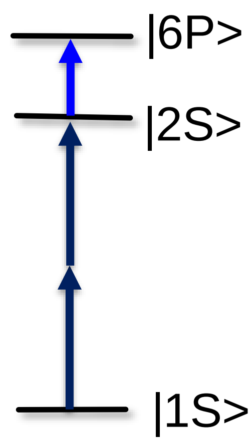
Two running hydrogen experiments at MPQ

1S-2S and 2S-*n*P experiment

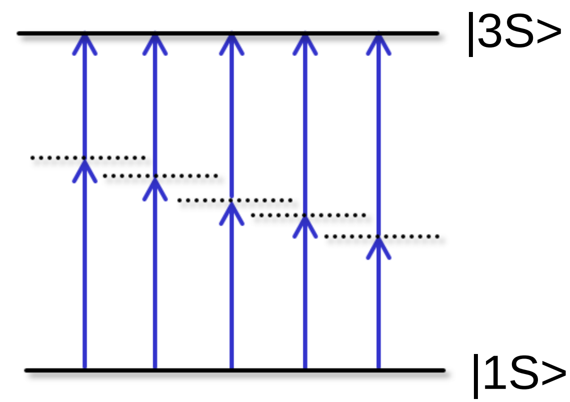
One-photon @ **410nm**

Two-photon @ **243nm**

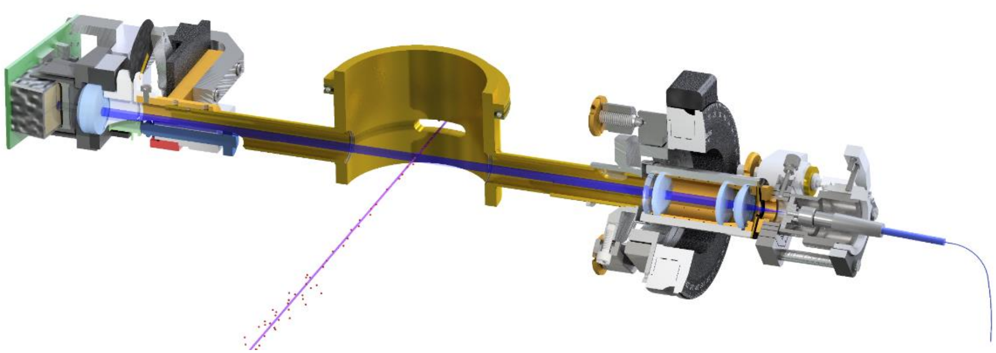


1S-3S experiment

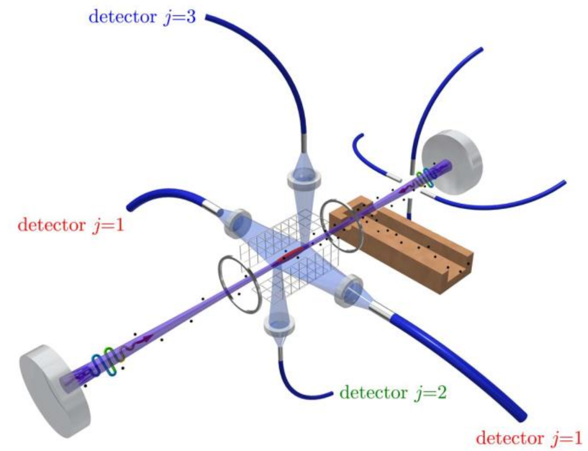
Two-photon
@ **205nm**



CW laser spectroscopy



Direct frequency comb spectroscopy



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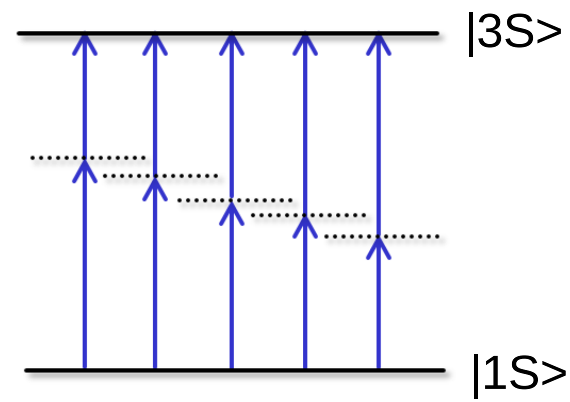

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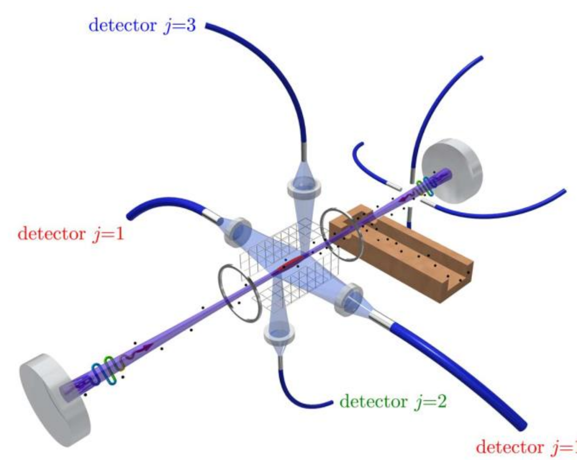


Published 1S-3S result in hydrogen:

FUNDAMENTAL PHYSICS

Two-photon frequency comb spectroscopy of atomic hydrogen

Alexey Grinin[1]*, Arthur Matveev[1], Dylan C. Yost[1]†, Lothar Maisenbacher[1], Vitaly Wirthl[1], Randolf Pohl[1]‡, Theodor W. Hänsch[1,2], Thomas Udem[1,2]

Grinin *et al.*, *Science* **370**, 1061–1066 (2020) 27 November 2020

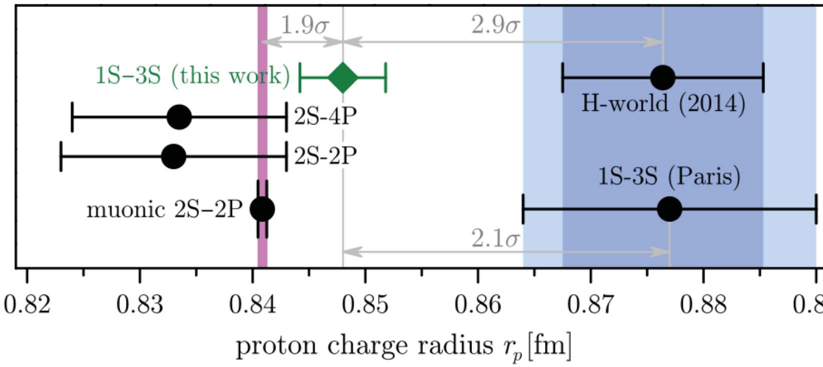


Table 1. Error budget of the 1S(*F*=1)–3S(*F*=1) measurement. All values are given in kilohertz. The average effect is the weighted mean of all evaluated data and quantifies the applied corrections. The multiparameter CIFODS (MP CIFODS) is an estimation of the uncertainty that may result from several independent sources of the laser chirp. This error budget is discussed in detail in the SM.

Contribution	Average effect	Correction	Uncertainty
Statistics	–	–	0.11
CIFODS	+0.79	–	0.08
SOD	–3.20	–	0.26
AC-Stark	+4.60	–	0.30
Pressure shift	+0.93	–	0.30
Residual Doppler	–	–	0.48
DC-Stark	+0.031	–0.031	0.015
Zeeman shift	–0.002	+0.002	0.002
Line pulling	–0.30	+0.30	0.050
MP CIFODS	–	–	0.10
Maser	–0.30	+0.30	0.030
Total		**+0.57**	**0.72**

AC Stark shift:
largest systematic effect

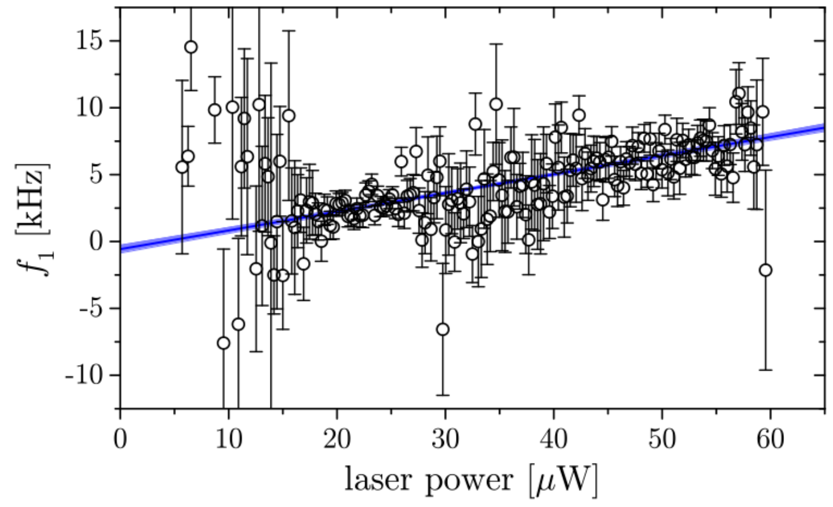
Status of the 1S-3S experiment at MPQ

- Hydrogen 1S-3S data: discrepancy between measured and modelled AC Stark shift *(model serves only for crosscheck, not used for the end result analysis)*

- Deuterium 1S-3S data: larger beam waist
  → smaller AC Stark shift expected

- Preliminary analysis of deuterium 1S-3S data: AC-Stark shift discrepancy even larger

AC Stark shift measurement in H 1S-3S



AC Stark shift coefficients	Measured	Modelled	
Hydrogen 1S-3S data	134(9) Hz/μW	33 Hz/μW	*4x discrepant*
Deuterium 1S-3S data	208(43) Hz/μW	~ 10 Hz/μW	*>10x discrepant*

Expected smaller AC Stark shift for Deuterium, but turned out to be even larger!

Though the **modelled value is not used in the data analysis**, it serves as a check of how well we understand our experiment: **need to resolve discrepancy**

...work in progress

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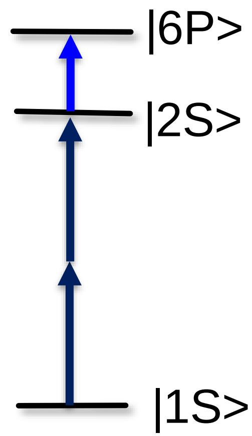

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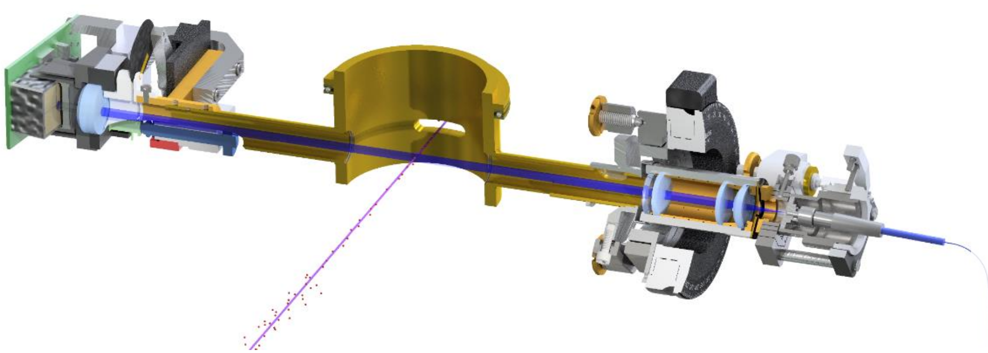

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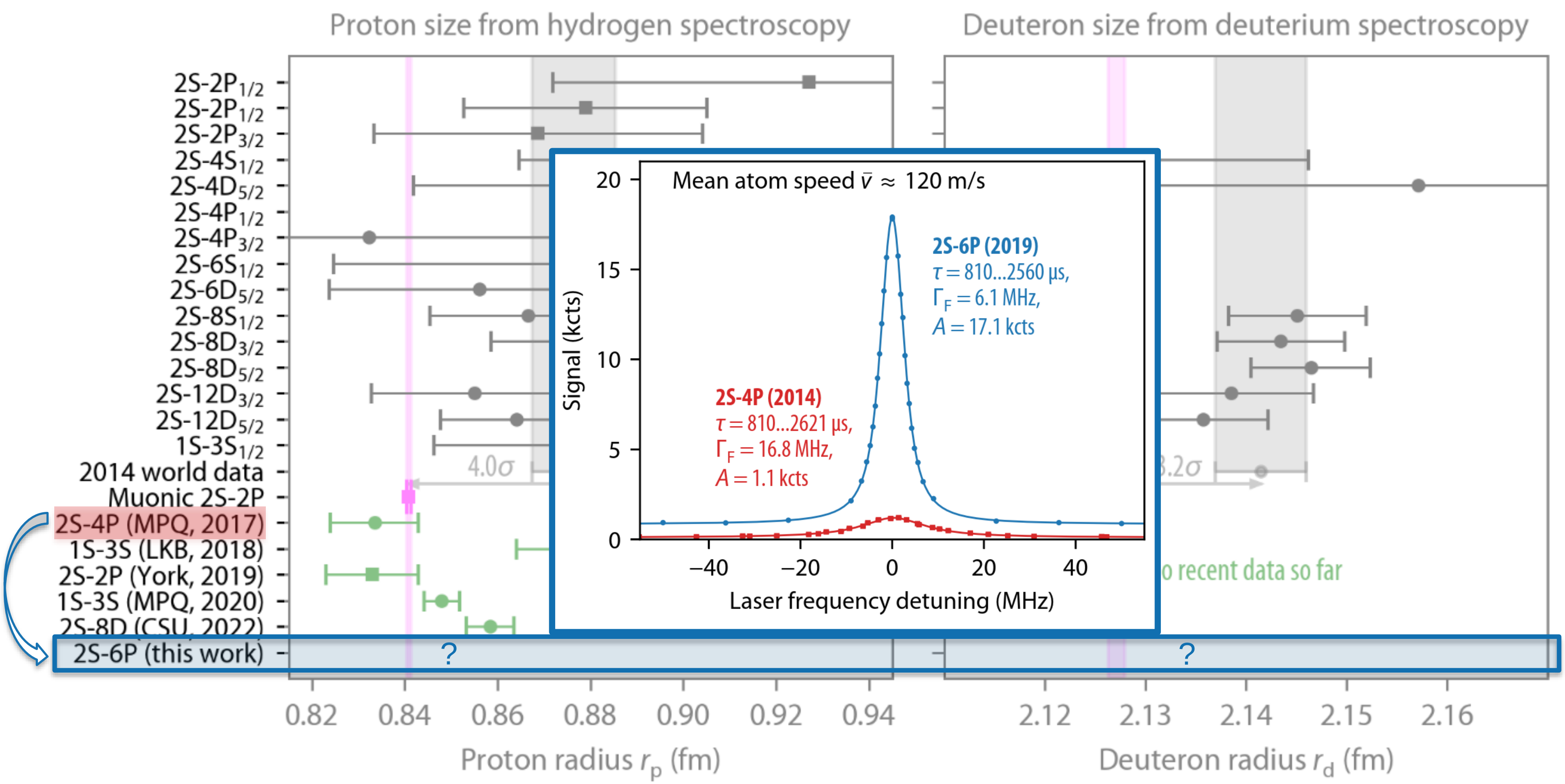
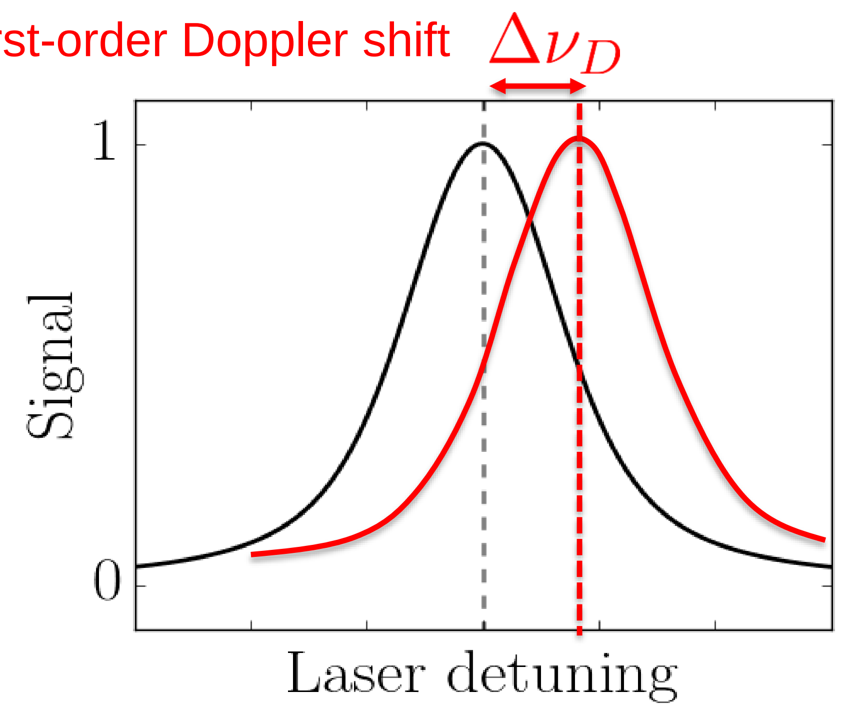


This work: 2S-6P transition measurement in hydrogen and deuterium (improves 2S-4P experiment: up to 16x higher signal and 3x lower linewidth)

Some properties of 2S-nP transitions in hydrogen and deuterium:

- dipole-allowed **one-photon** transitions between metastable *2S* level and short-lived *nP* level: **first-order Doppler shift**

  Cryostat & Active Fiber-based Retroreflector [1]



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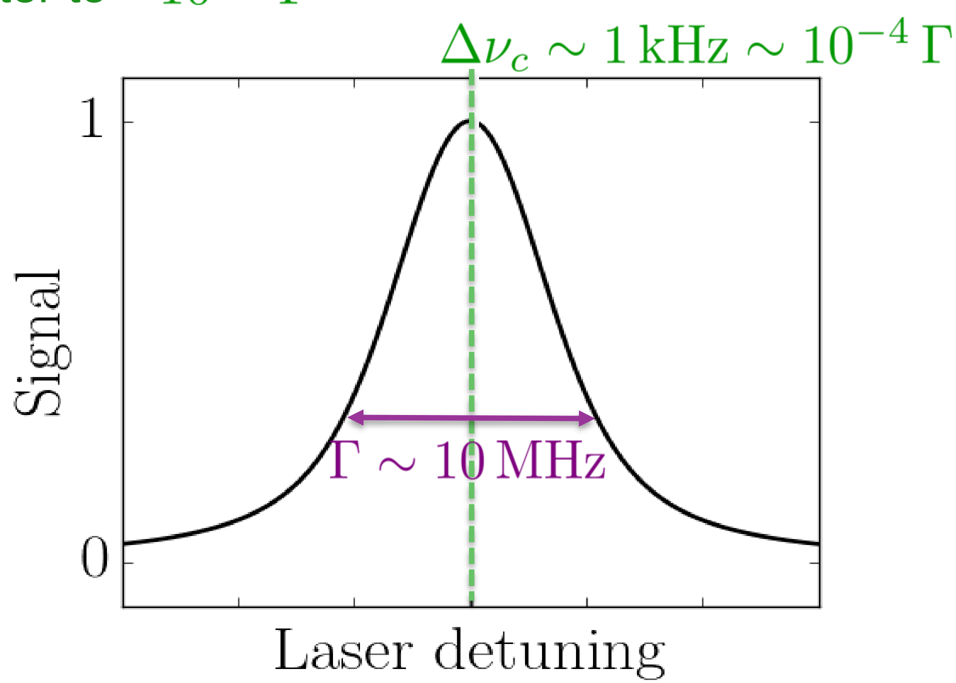
- **natural line width in the MHz range**: narrower for higher states (~n^3-scaling), ~ 4 MHz for 2S-6P

  Uncertainty goal: determine line center to ~ $10^{-4}\,\Gamma$



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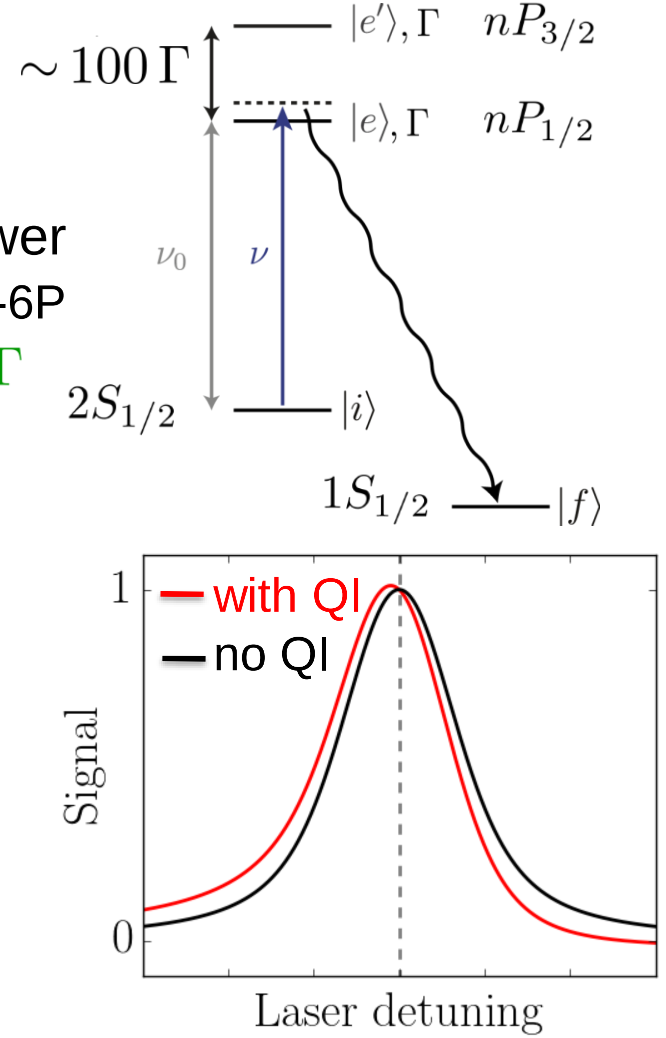
- **resolved quantum interference (QI)** from two fine structure components separated by ~100 Γ

  Studied in detail in Hydrogen 2S-4P [2]



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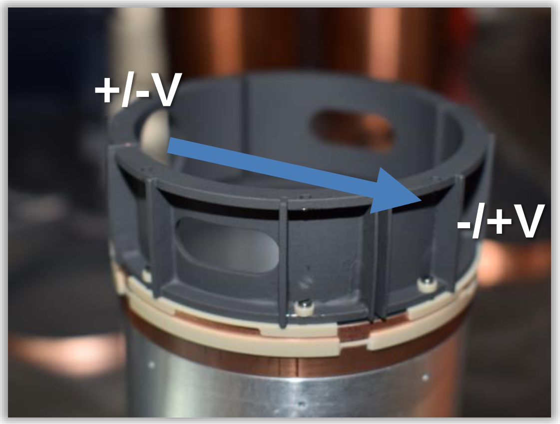
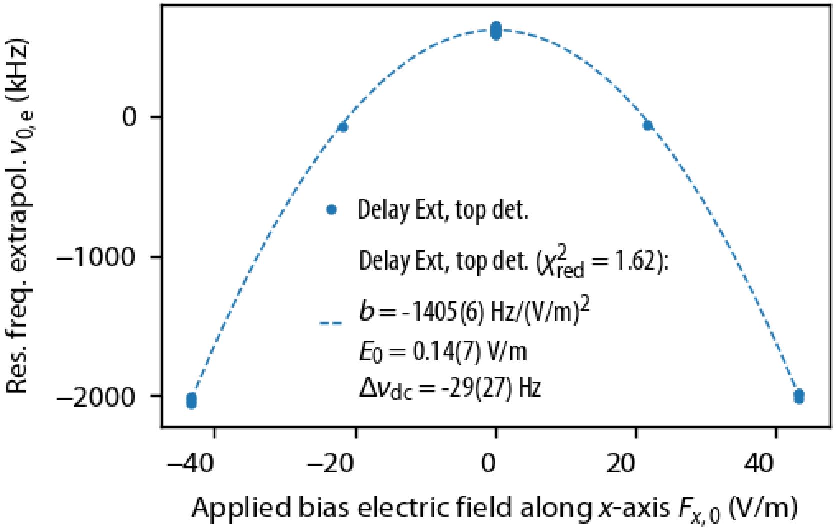
- **DC Stark shift**: n^7–scaling requires good control of stray electric fields

  Specially designed detector allows in-situ measurement of electric fields





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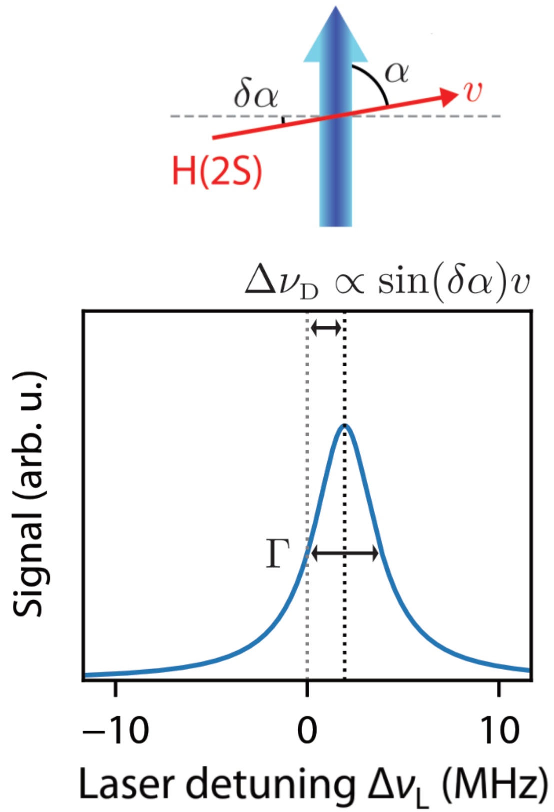
2S-6P transition is a dipole-allowed **one-photon** transition: the **first-order Doppler shift** is the main challenge



First-order Doppler shift suppression

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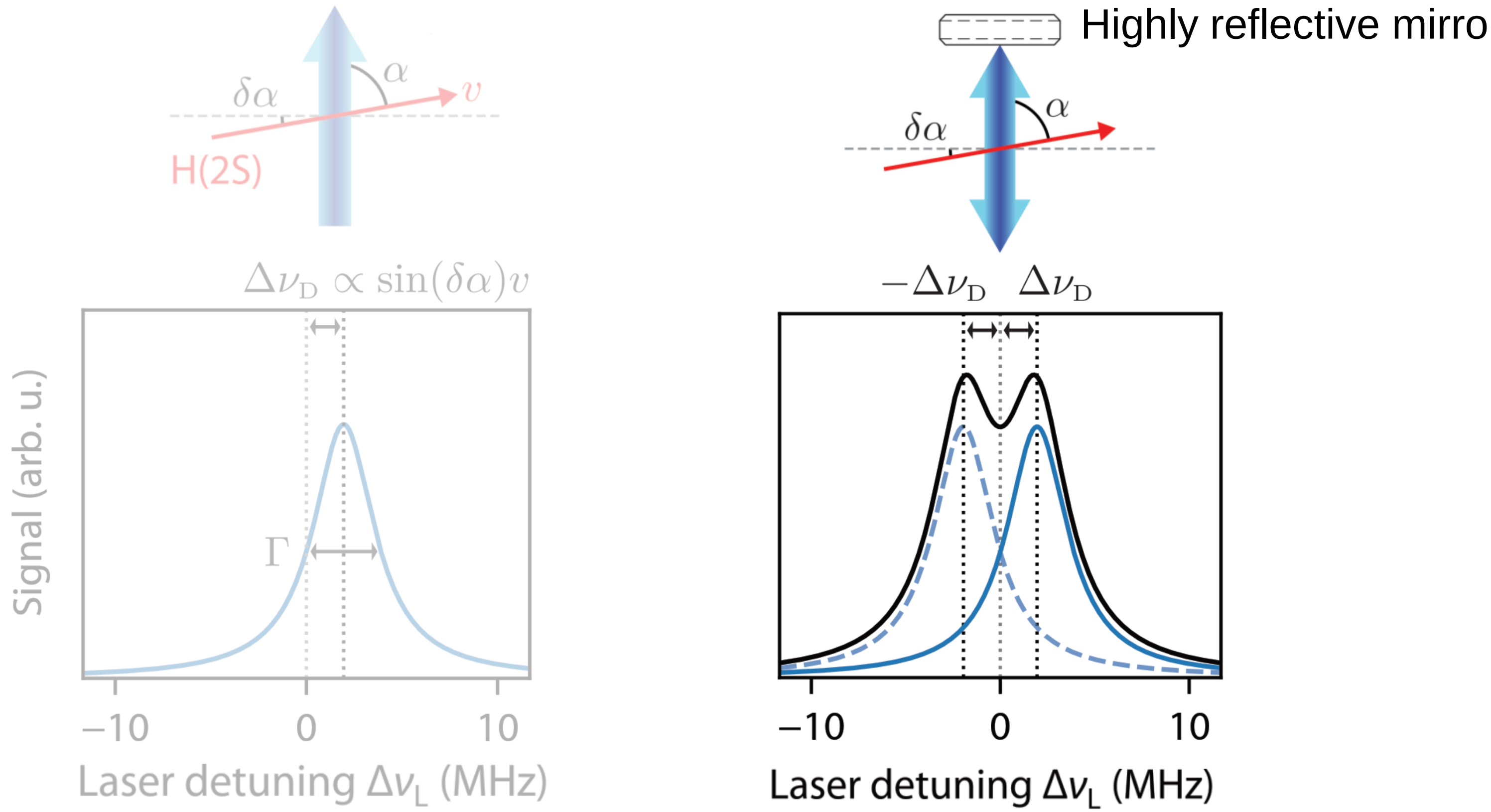
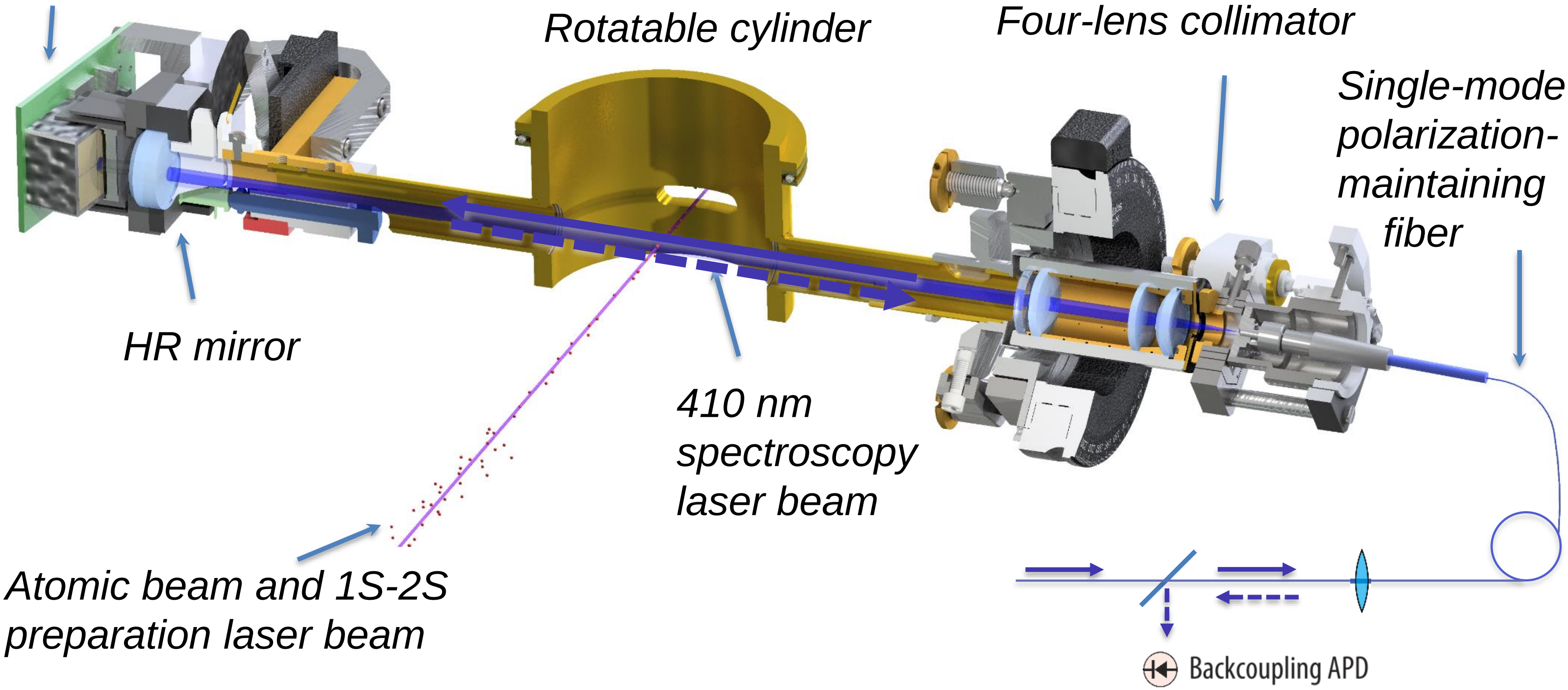


Basic principle of our techique to suppress Doppler shift:

Counter propagating beams with opposite wave-vectors:

Doppler shift adds to zero **for equal intensity and wavefront of both beams**

Improved active fiber-based retroreflector for near UV [1] provides high-quality wavefront-retracing anti-parallel laser beams:



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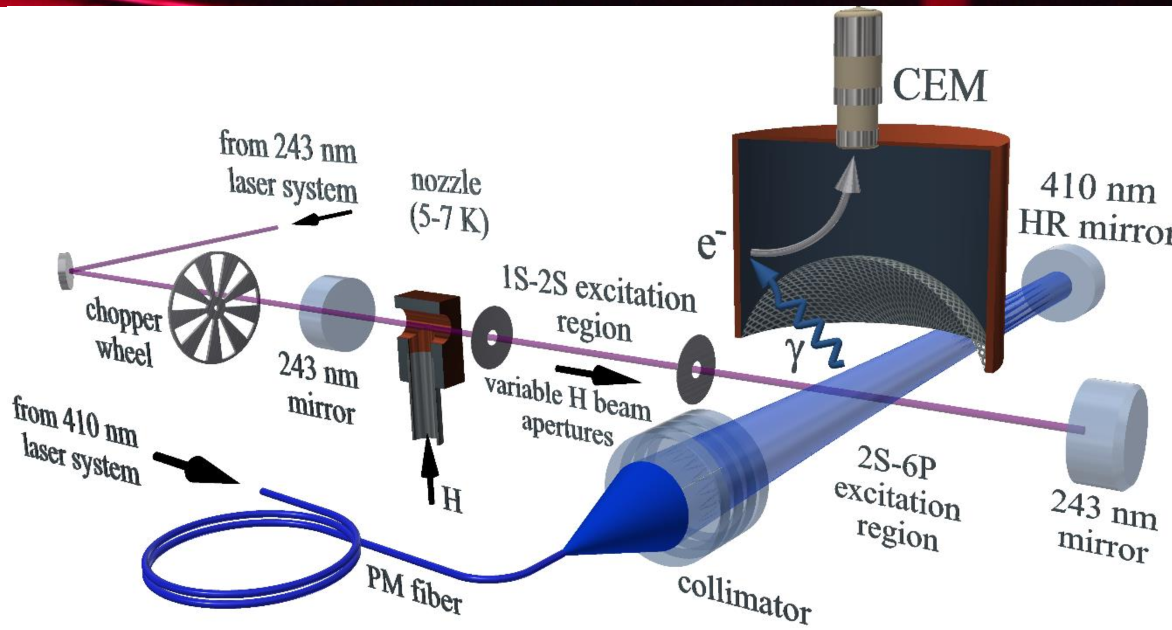
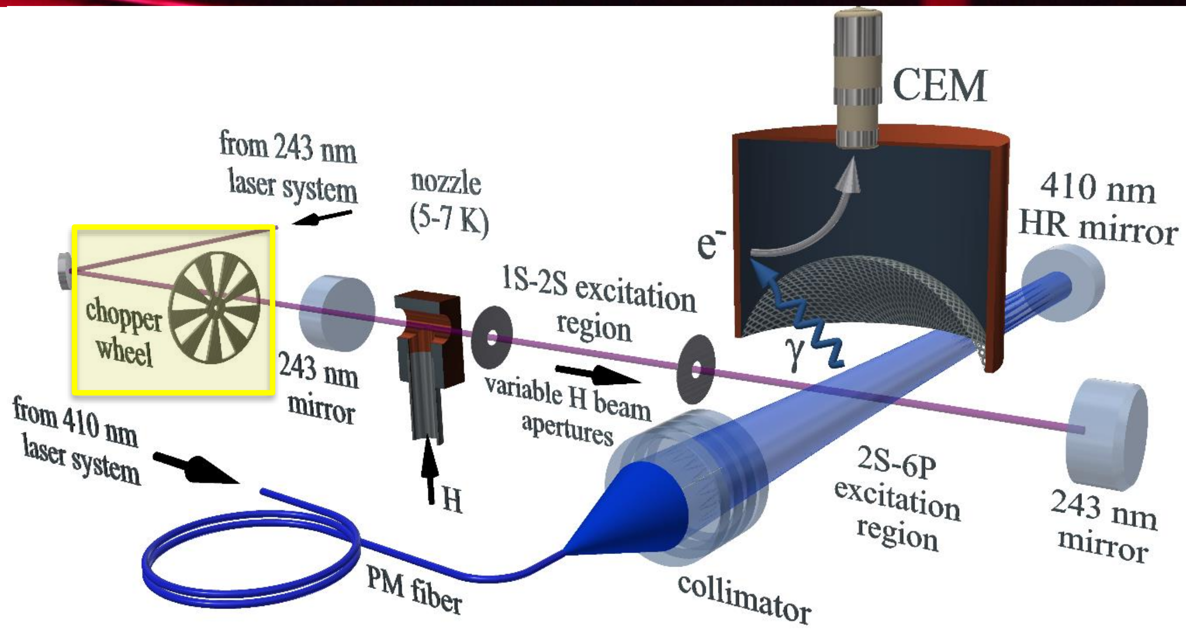

CEM
from 243 nm
laser system
nozzle
(5-7 K)
410 nm
HR mirror
e-
1S-2S excitation
region
chopper
wheel
243 nm
mirror
γ
variable H beam
apertures
from 410 nm
laser system
H
2S-6P
excitation
region
243 nm
mirror
PM fiber
collimator

Time-resolved detection: velocity-dependent signal

243nm chopper periodically stops the 1S-2S excitation ➔ signal is recorded as a function of **delay time after 243nm light is blocked**



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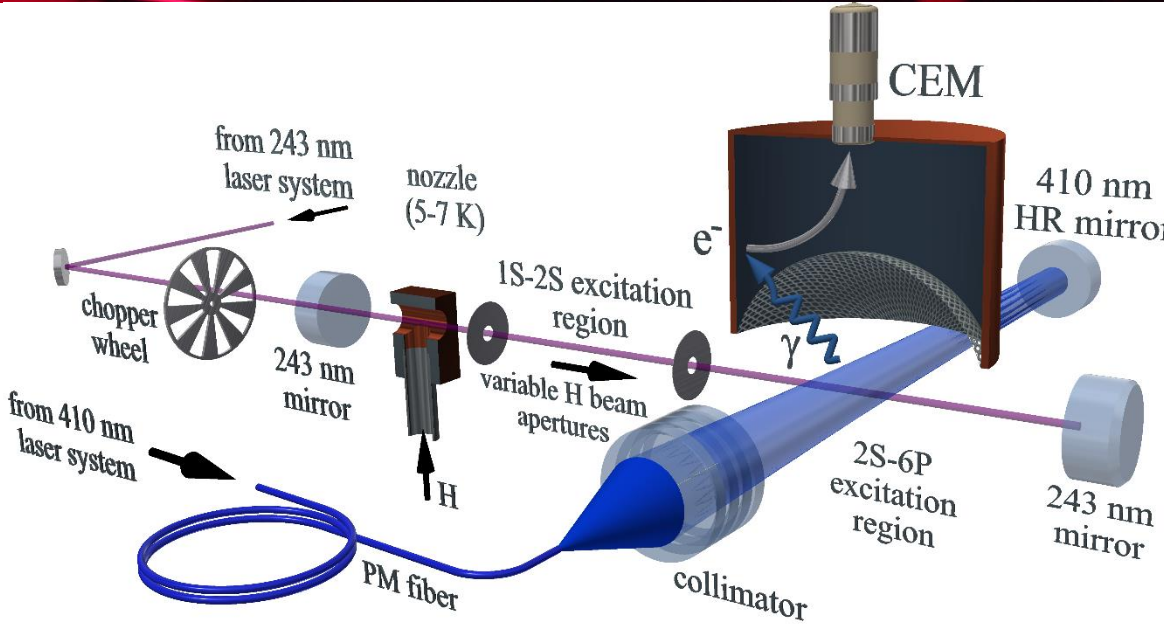

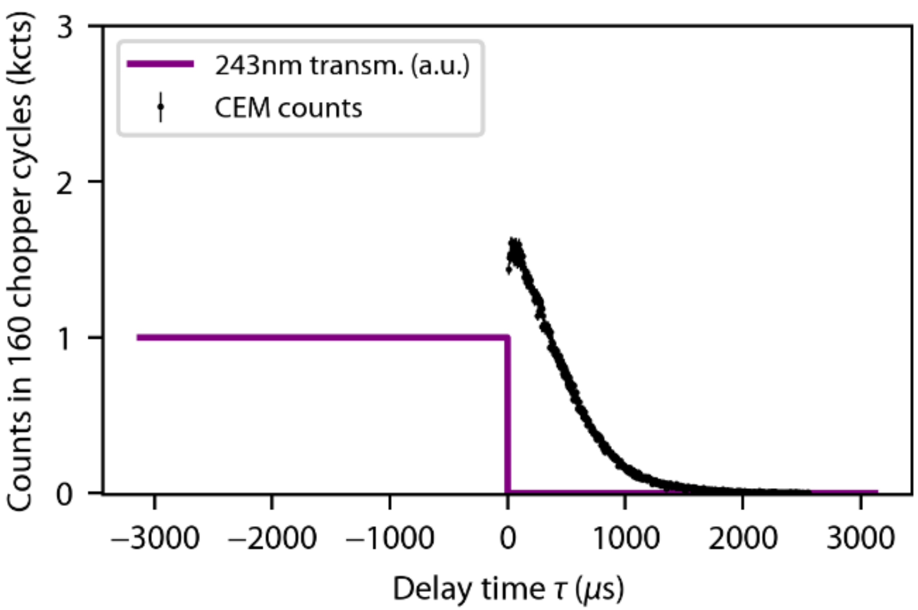

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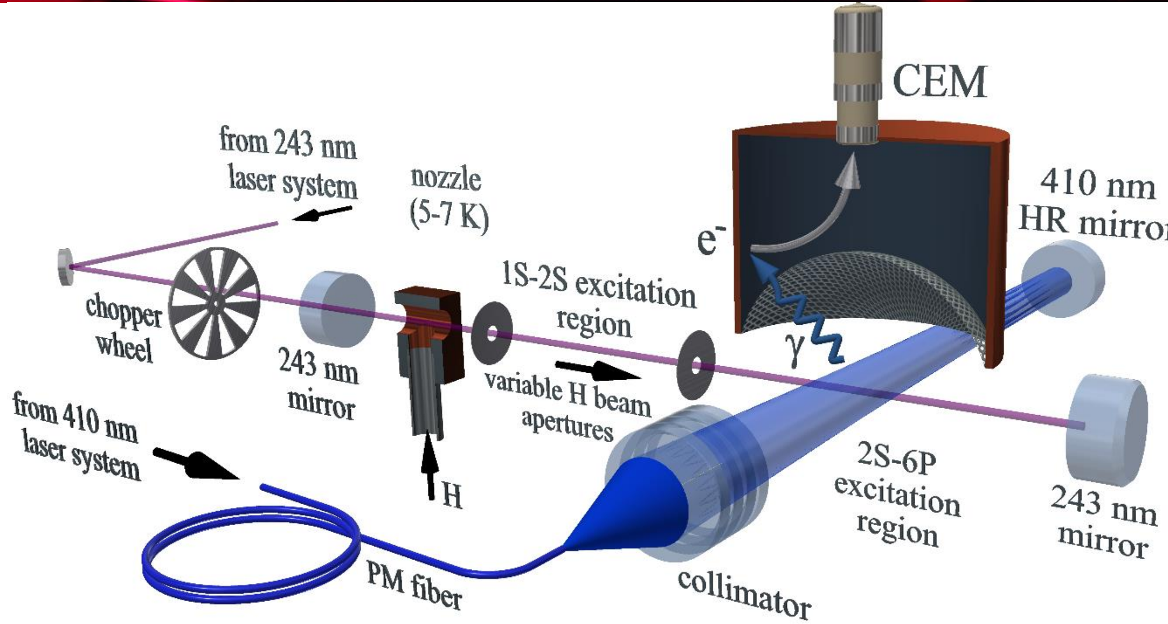

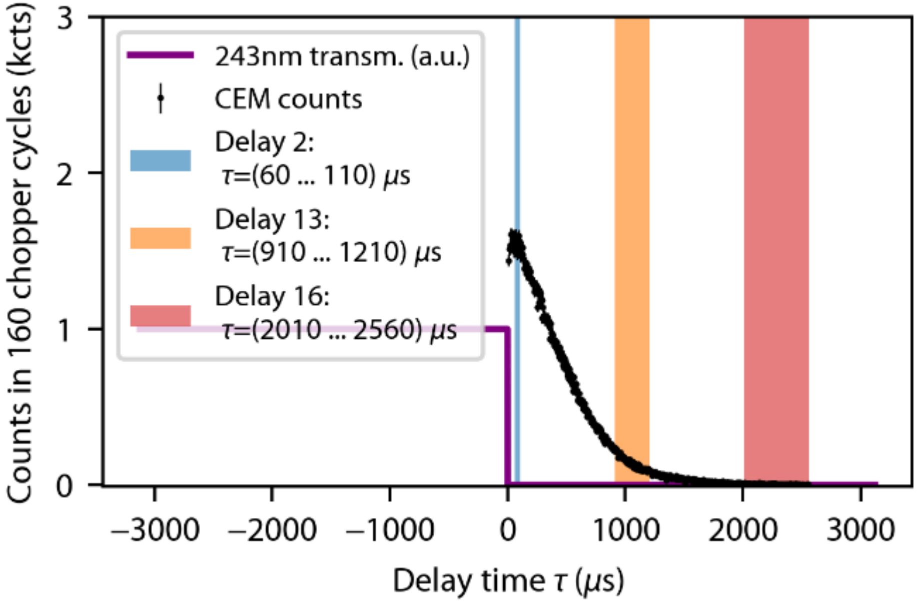

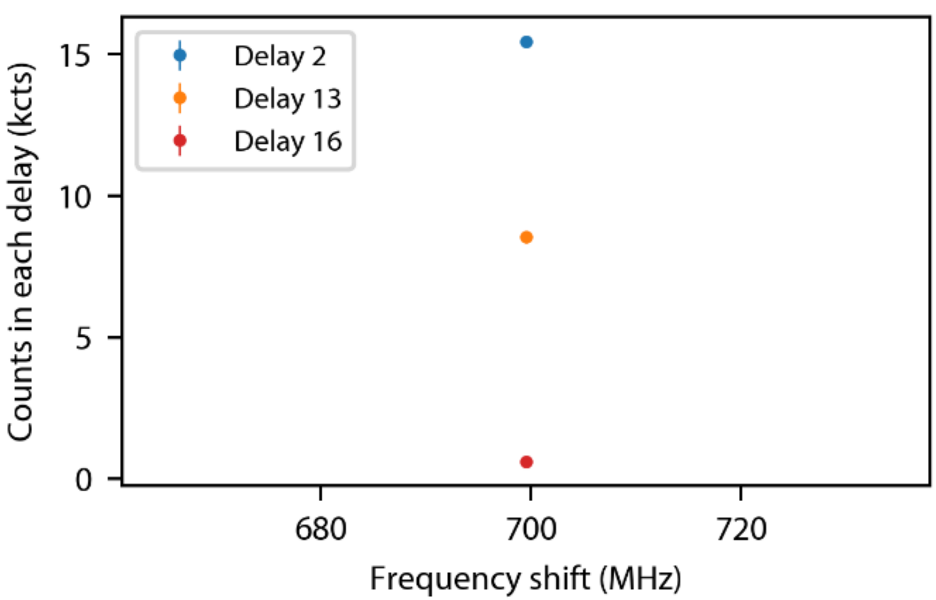

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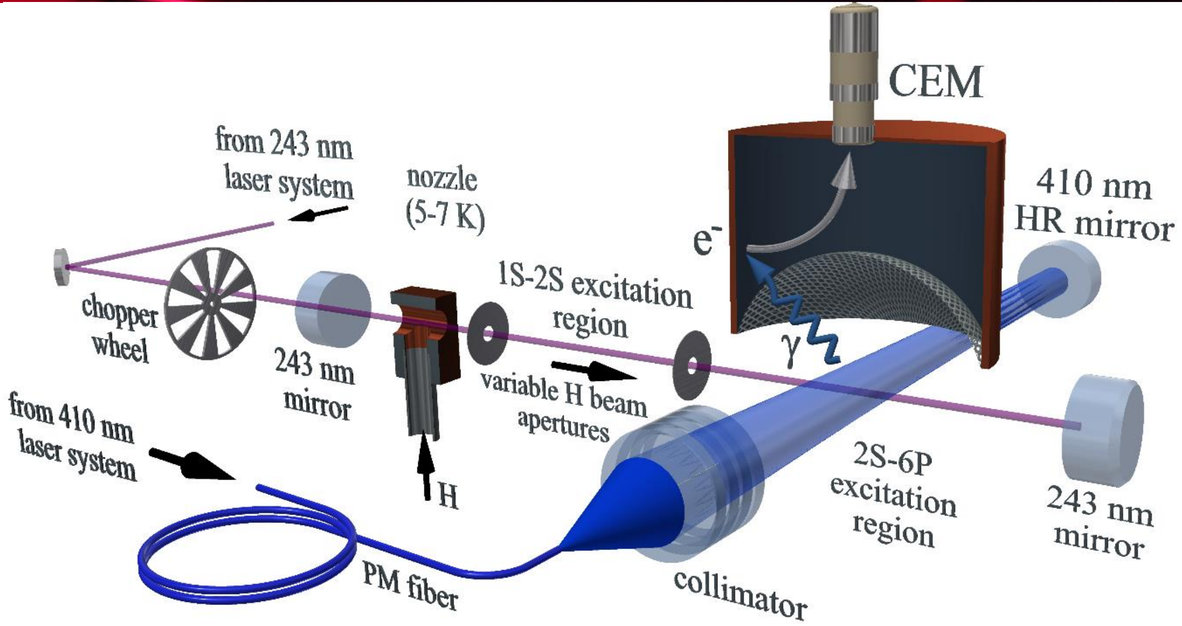

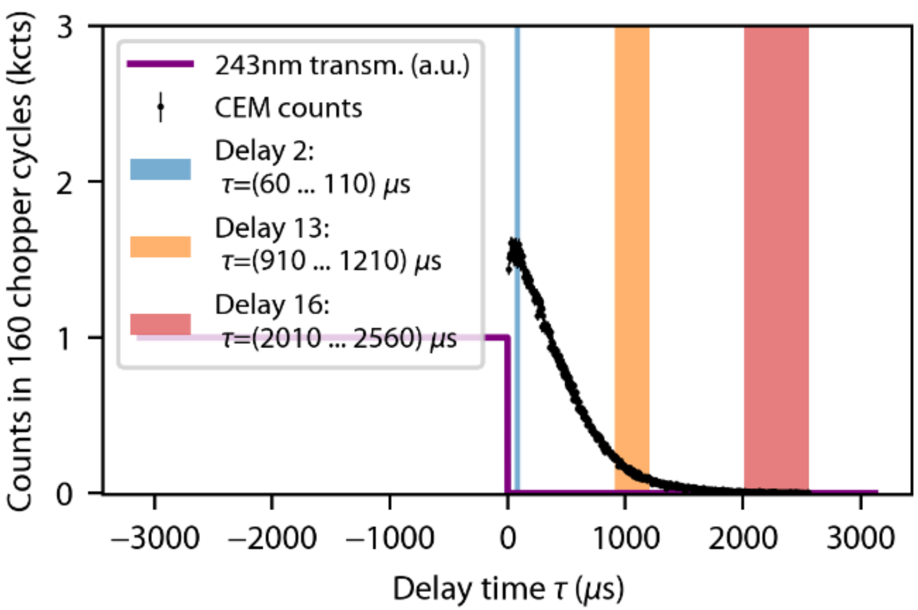

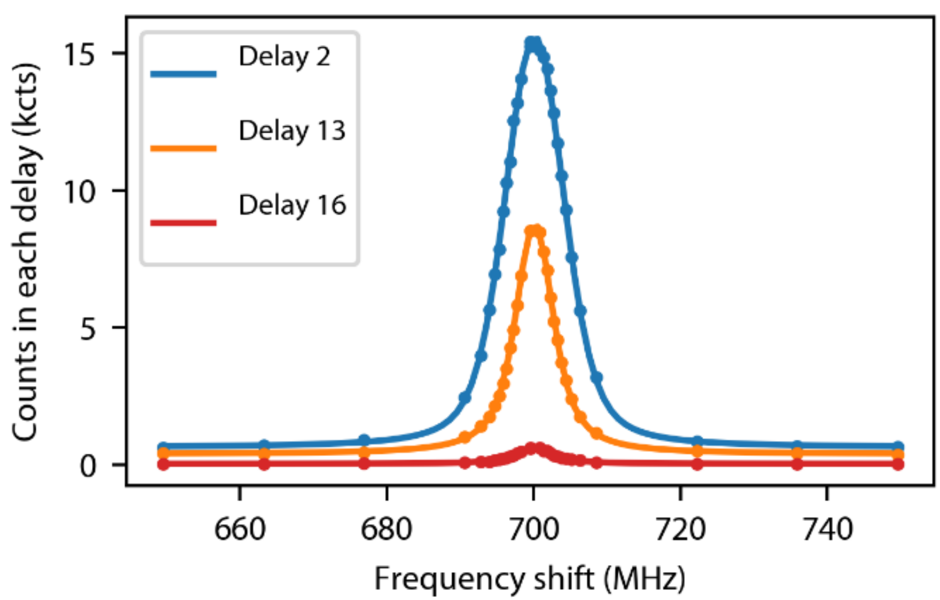

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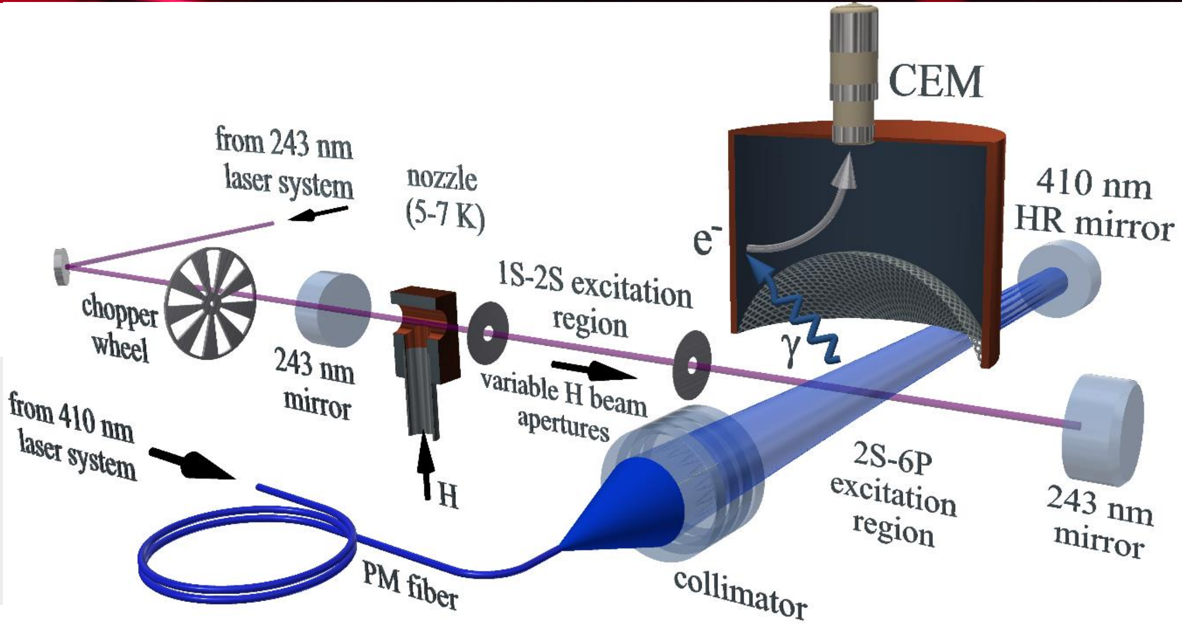
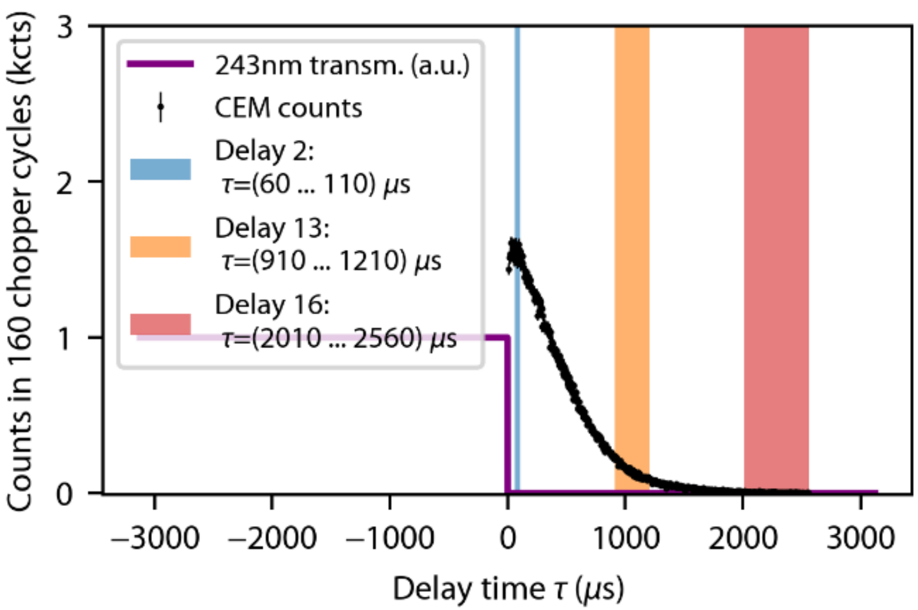


Time-resolved detection allows to access different velocity groups of atoms to **study velocity-dependent effects**



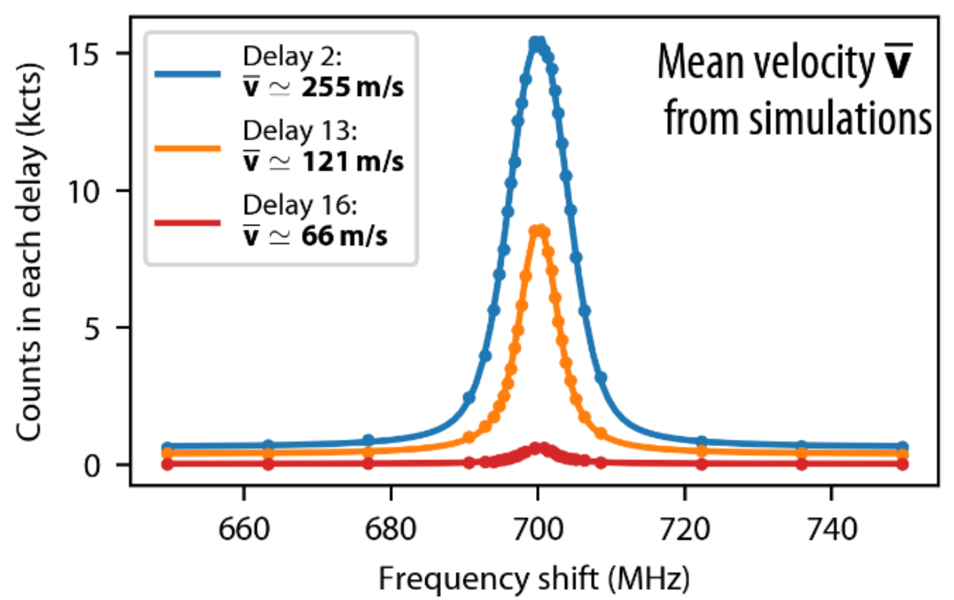
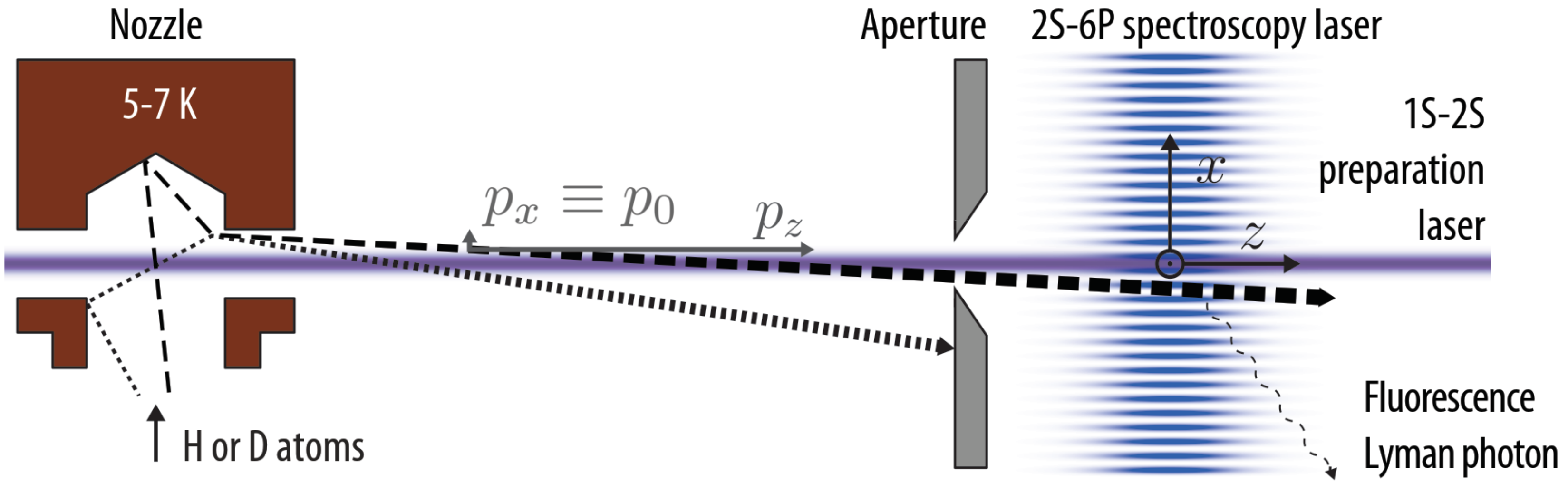


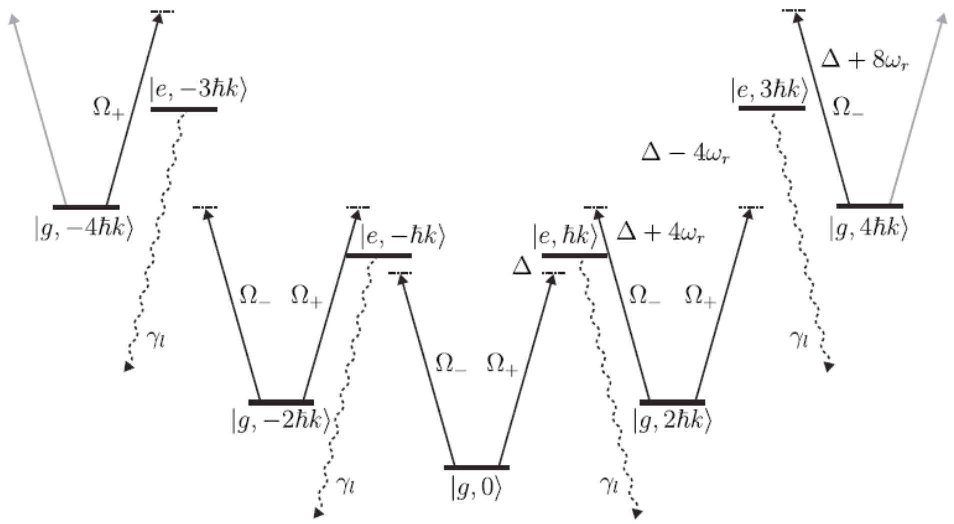
Table 2.3: List of corrections $\Delta\nu$ and uncertainties σ for the determination of the 2S-6P$_{1/2}$ ($\nu_{1/2}$) and the 2S-6P$_{3/2}$ ($\nu_{3/2}$) transition frequencies, as well as of the the 2S-6P fine-structure centroid $\nu_{\text{2S-6P}}$, formed by combining $\nu_{1/2}$ and $\nu_{3/2}$. All values are given in units of kHz. BBR: blackbody radiation, HFS: hyperfine structure, FS: fine structure.

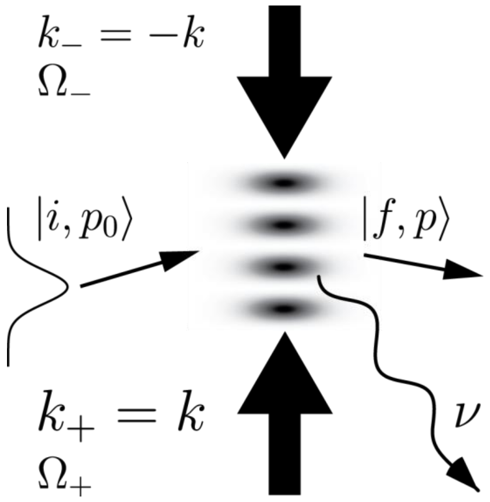
	$2S_{1/2}^{F=0}-6P_{1/2}^{F=1}$ ($\nu_{1/2}$)		$2S_{1/2}^{F=0}-6P_{3/2}^{F=1}$ ($\nu_{3/2}$)		2S-6P FS centroid ($\nu_{\text{2S-6P}}$)	
Contribution (kHz)	$\Delta\nu$	σ	$\Delta\nu$	σ	$\Delta\nu$	σ
Statistics (incl. Doppler shift)	—	0.49	—	0.60	—	0.43
Light force shift	0.70	0.21	1.31	0.39	1.11	0.33
				0.29	0.05	0.05
				0.02	-0.14	0.02
dc-Stark shift	0.20	0.20	0.05	0.05	0.10	0.10
BBR-induced shift	0.29	0.03	0.29	0.03	0.29	0.03
Zeeman shift	0.00	0.05	0.00	0.23	0.00	0.17
Pressure shift	0.00	0.01	0.00	0.01	0.00	0.01
Frequency standard	0.00	0.07	0.00	0.07	0.00	0.07
Total without recoil & HFS corr.	1.25	0.82	1.49	0.81	1.41	0.58
Recoil shift	-1176.03	0.00	-1176.03	0.00	-1176.03	0.00
Hyperfine structure corrections	—	—	—	—	-132985.252	0.007
Total	-1174.78	0.82	-1174.54	0.81	-134159.872	0.58

Largest systematic effect: light force shift

Light force shift





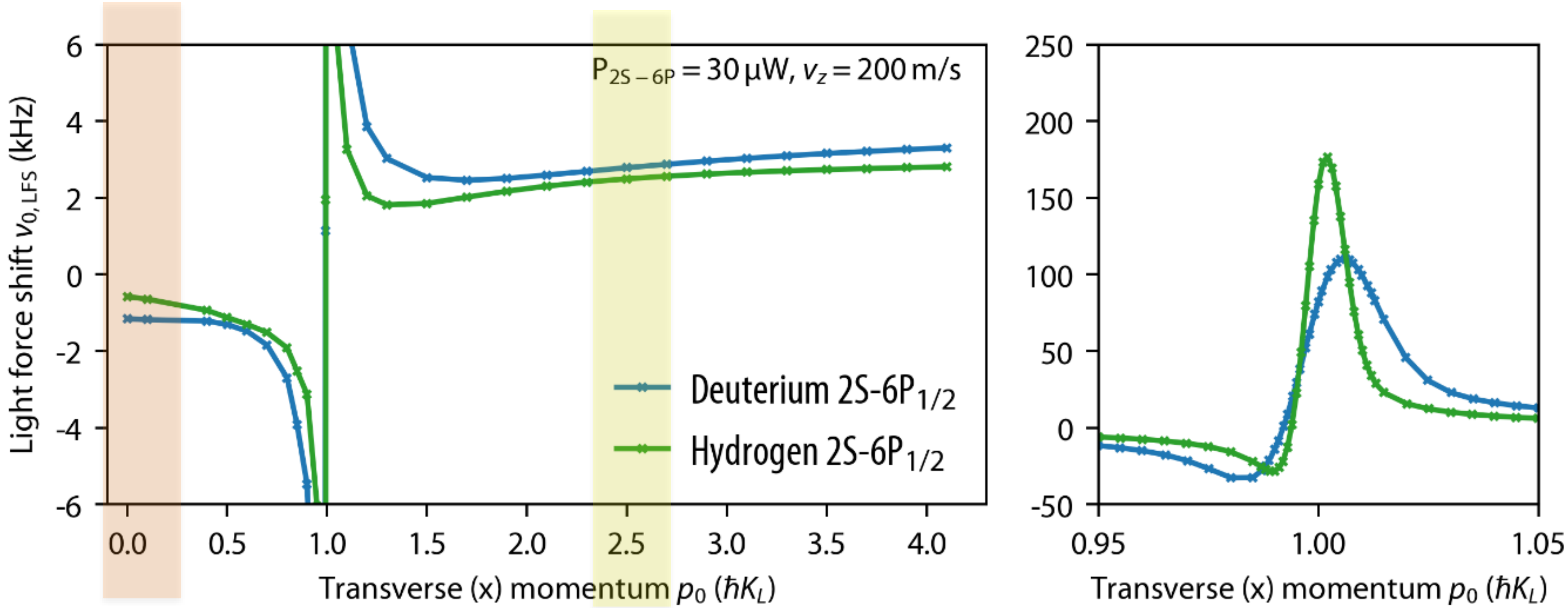


Atoms delocalized over standing wave (205 nm periodicity) and can be described as plane wave with defined transverse momentum p_0

Light force shift

Prediction: light force shift negative for p_0=0, positive shift for p_0 ~ 3 ħK_L

Exemplary simulation of the light force shift for a single atom at 200m/s:



−1.5 kHz
atomic beam crosses
laser beam at ~ 0 mrad
(regime for precision
measurement)

+2.8 kHz
atomic beam crosses
laser beam at ~ 12 mrad

(simulation results for an atomic hydrogen beam)

Simulated difference:
4.3 kHz

Measured light force shift in hydrogen
by taking data also at ~ 12 mrad:

Measured difference: 4.4(1.8) kHz

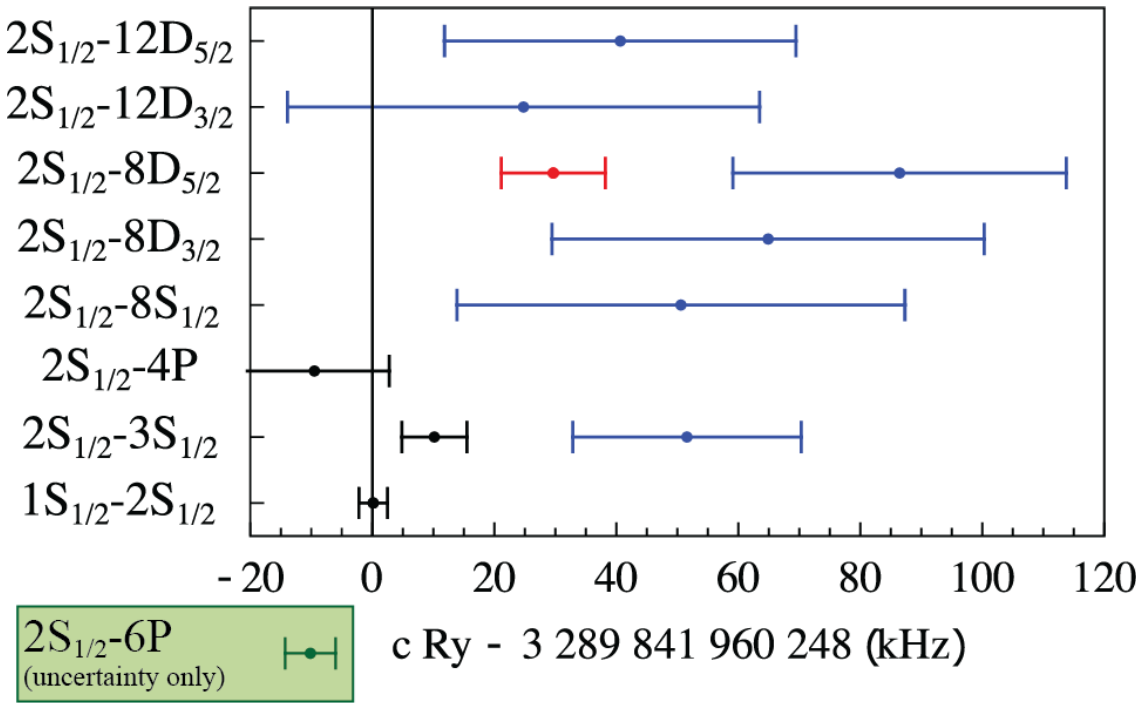
Table 2.3: List of corrections $\Delta\nu$ and uncertainties σ for the determination of the 2S-6P$_{1/2}$ ($\nu_{1/2}$) and the 2S-6P$_{3/2}$ ($\nu_{3/2}$) transition frequencies, as well as of the the 2S-6P fine-structure centroid $\nu_{\text{2S-6P}}$, formed by combining $\nu_{1/2}$ and $\nu_{3/2}$. All values are given in units of kHz. BBR: blackbody radiation, HFS: hyperfine structure, FS: fine structure.

	$2S_{1/2}^{F=0}-6P_{1/2}^{F=1}$ ($\nu_{1/2}$)		$2S_{1/2}^{F=0}-6P_{3/2}^{F=1}$ ($\nu_{3/2}$)		2S-6P FS centroid ($\nu_{\text{2S-6P}}$)	
Contribution (kHz)	$\Delta\nu$	σ	$\Delta\nu$	σ	$\Delta\nu$	σ
Statistics (incl. Doppler shift)	—	0.49	—	0.60	—	0.43
Light force shift	0.70	0.21	1.31	0.39	1.11	0.33
Quantum interference shifts	0.21	0.58	-0.02	0.29	0.05	0.05
Second-order Doppler shift	-0.15	0.02	-0.14	0.02	-0.14	0.02
dc-Stark shift	0.20	0.20	0.05	0.05	0.10	0.10
BBR-in	[illegible]	[illegible]	[illegible]	[illegible]	0.29	0.03
Zeeman	[illegible]	[illegible]	[illegible]	[illegible]	0.00	0.17
Pressur	[illegible]	[illegible]	[illegible]	[illegible]	0.00	0.01
Frequen	[illegible]	[illegible]	[illegible]	[illegible]	0.00	0.07
Total without recoil & HFS corr.	1.25	0.82	1.49	0.81	1.41	0.58
Recoil shift	-1176.03	0.00	-1176.03	0.00	-1176.03	0.00
Hyperfine structure corrections	—	—	—	—	-132985.252	0.007
Total	-1174.78	0.82	-1174.54	0.81	-134159.872	0.58

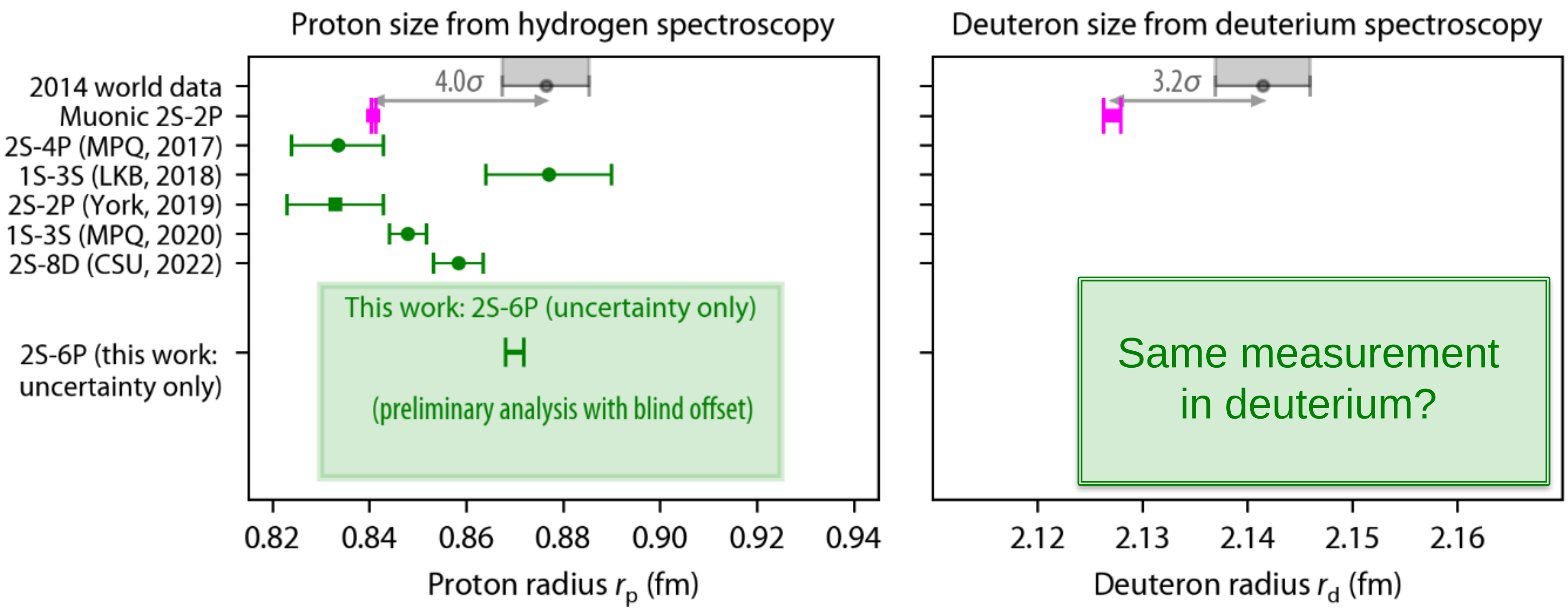
Preliminary Hydrogen 2S-6P uncertainty:
0.6 kHz with only 1.4 kHz corrections

Preliminary hydrogen 2S-6P measurement result

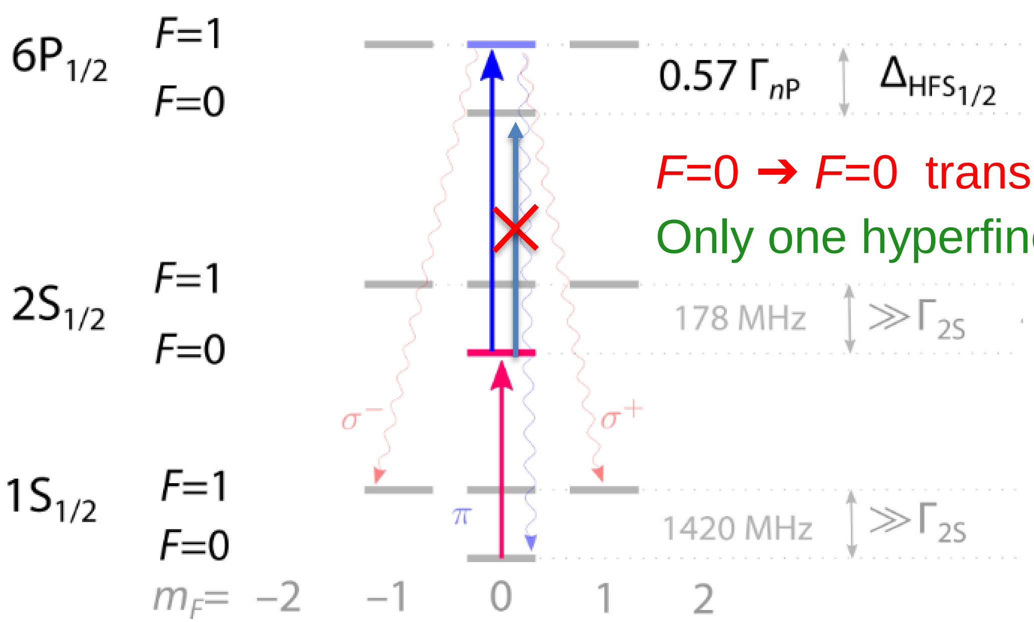
Data analysis of our hydrogen 2S-6P measurement campaign currently ongoing (with blind offset), preliminary uncertainty result:



Data analysis of our hydrogen 2S-6P measurement campaign currently ongoing (with blind offset), preliminary uncertainty result:



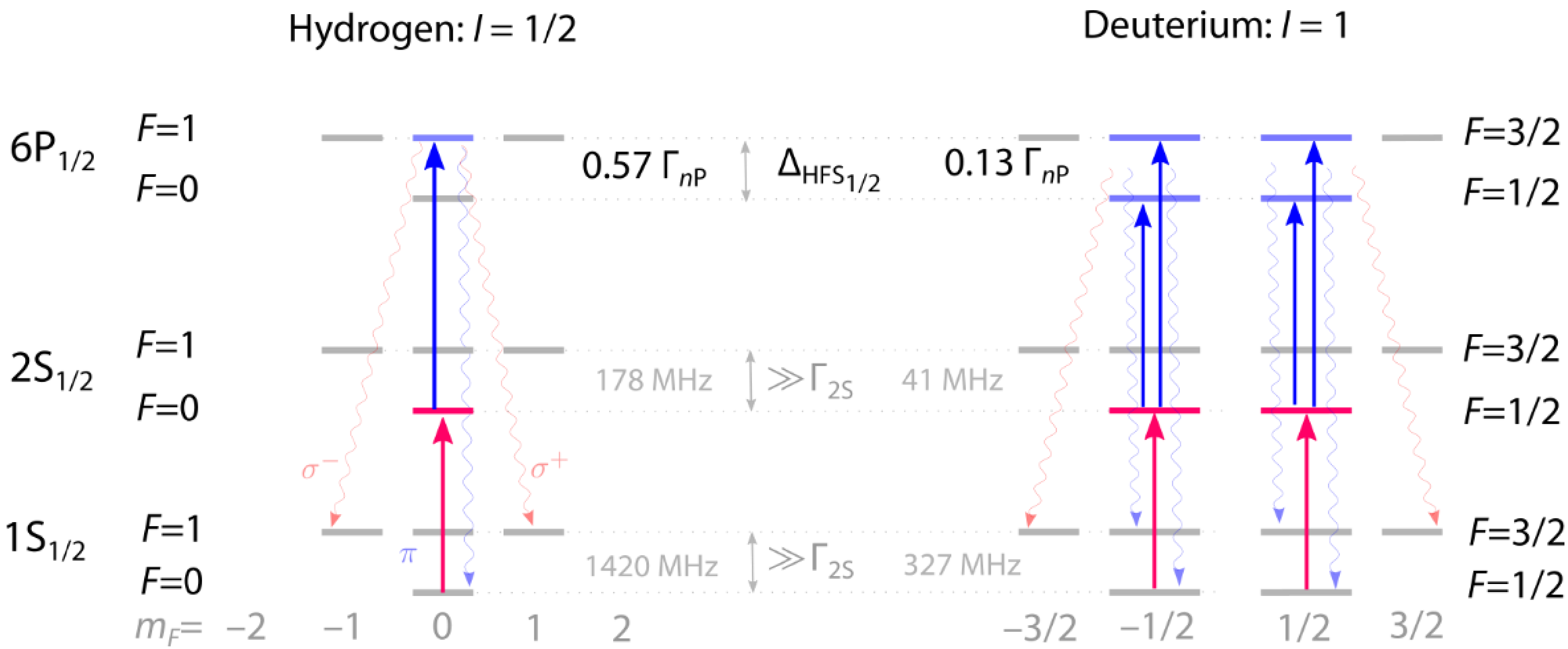
Hydrogen: $I = 1/2$



F=0 ➔ *F*=0 transitions forbidden

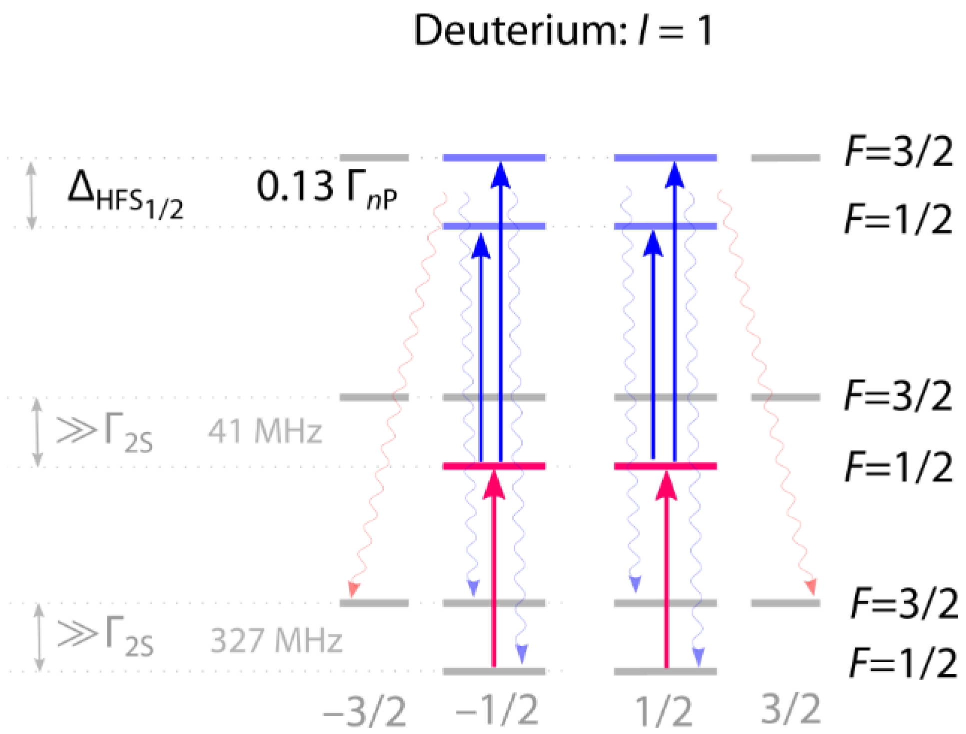
Only one hyperfine manifold excited

2S-6P in deuterium: complications



Additionally allowed transitions compared to hydrogen require to consider:

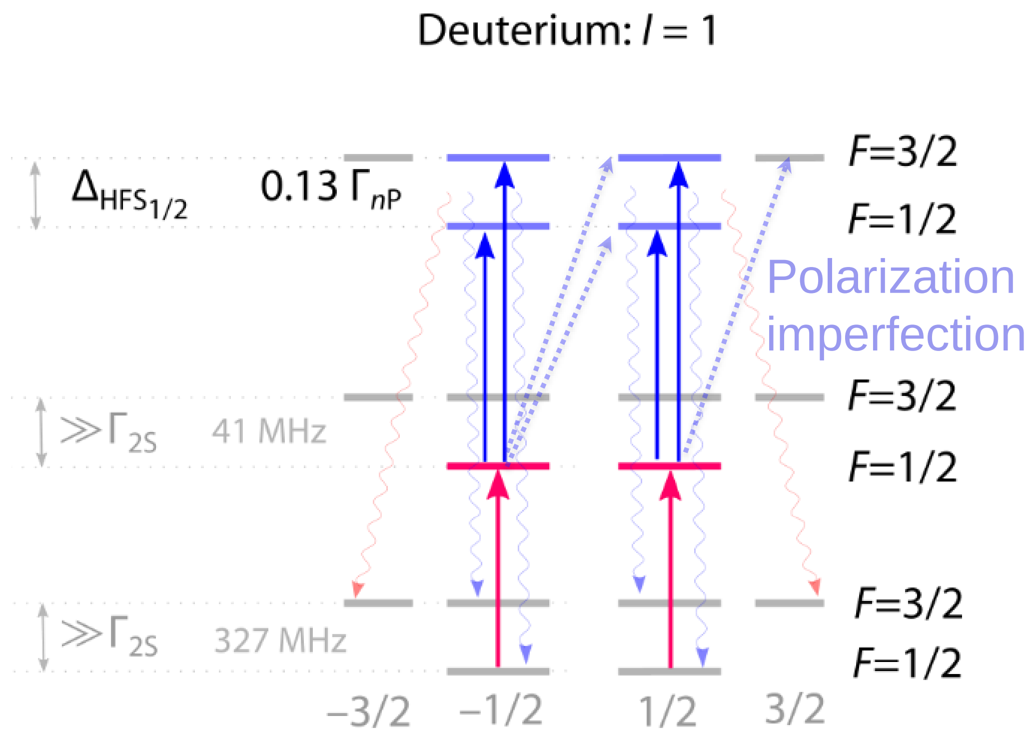
1) simultaneous excitation of different hyperfine levels



2S-6P in deuterium: complications

Additionally allowed transitions compared to hydrogen require to consider:

1) simultaneous excitation of different hyperfine levels

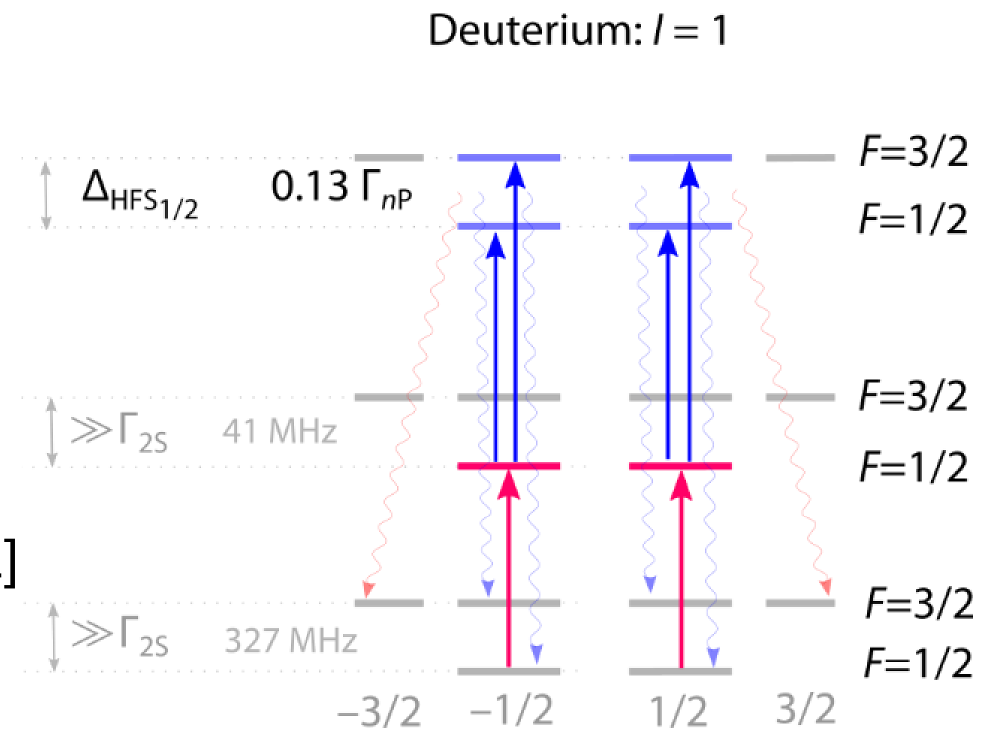


Residual circular polarization changes the dipole ratio of excited hyperfine state manifolds

Additionally allowed transitions compared to hydrogen require to consider:

1) simultaneous excitation of different hyperfine levels

2) quantum interference between unresolved hyperfine transitions [1]



Additionally allowed transitions compared to hydrogen require to consider:

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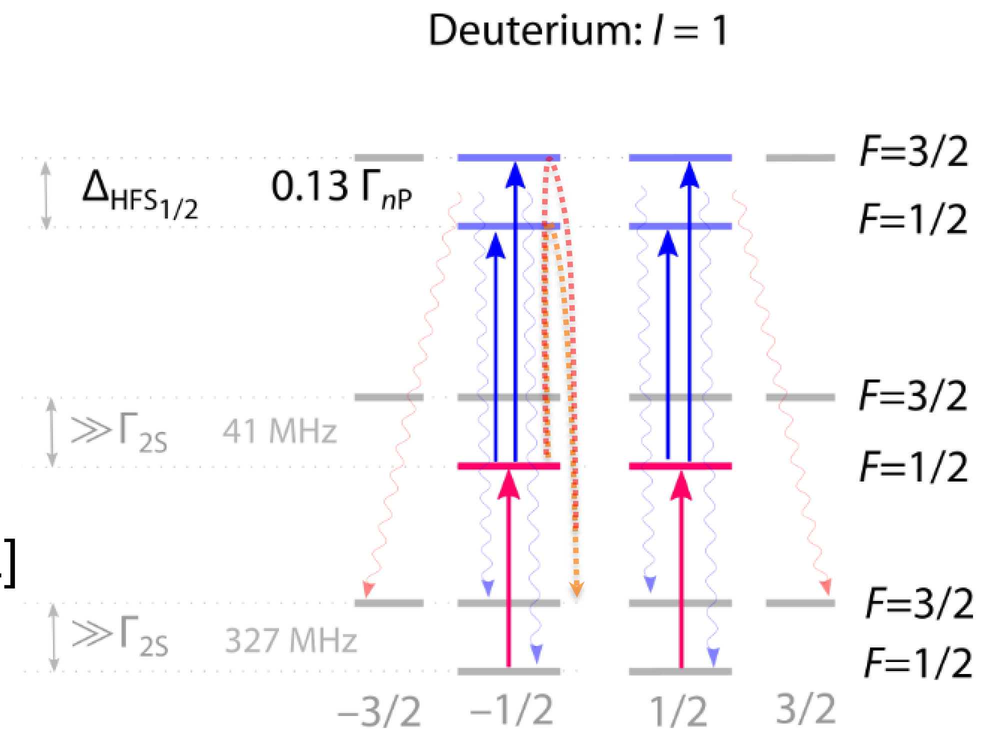


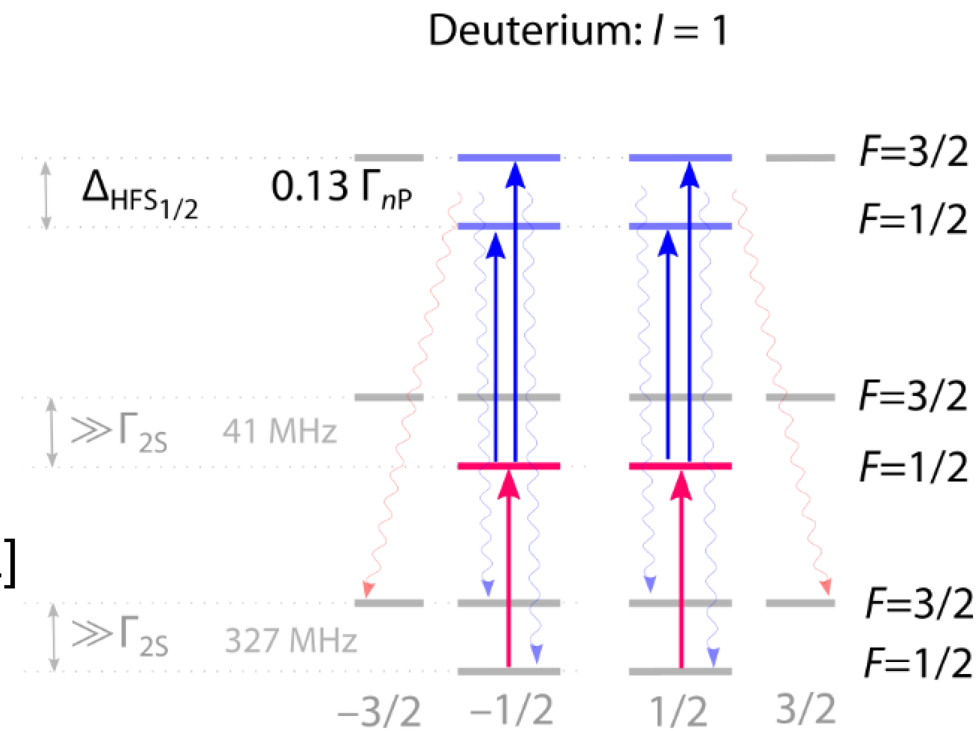
Possible quantum interference between the different signal paths from the two hyperfine manifolds

2S-6P in deuterium: complications

Additionally allowed transitions compared to hydrogen require to consider:

1) simultaneous excitation of different hyperfine levels

2) quantum interference between unresolved hyperfine transitions [1]

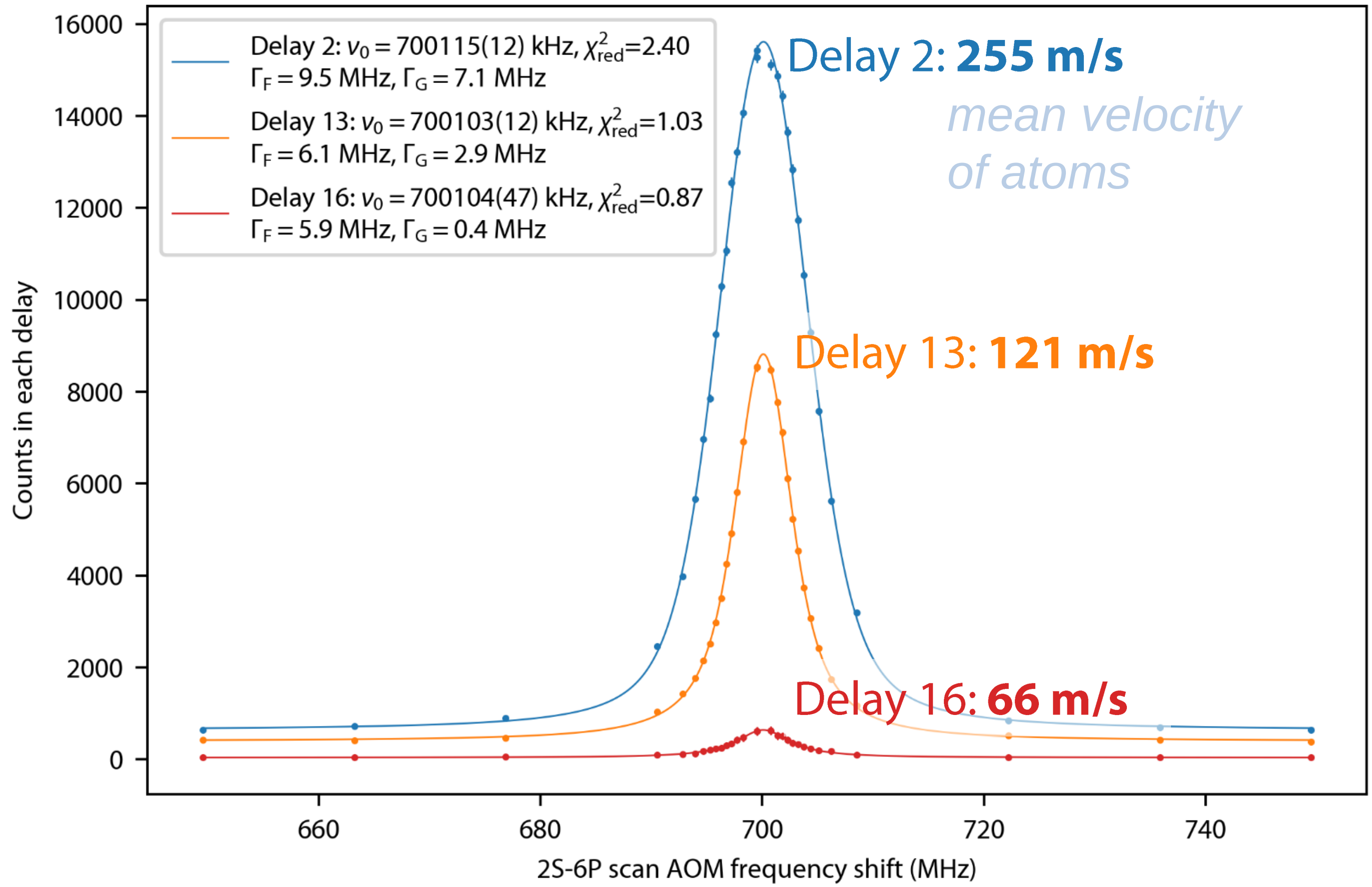


<table>
<tr><th></th><th>Detection different for LH/RH circular pol.</th><th>Initital state population asymmetry</th><th>Residual circular polarization</th></tr>
<tr><td>1) Shift from dipole ratio</td><td></td><td colspan="2">x</td></tr>
<tr><td>2) Unresolved Q.I.</td><td colspan="2">x</td><td></td></tr>
</table>

We find that both effects from additional transitions in deuterium **doubly suppressed**

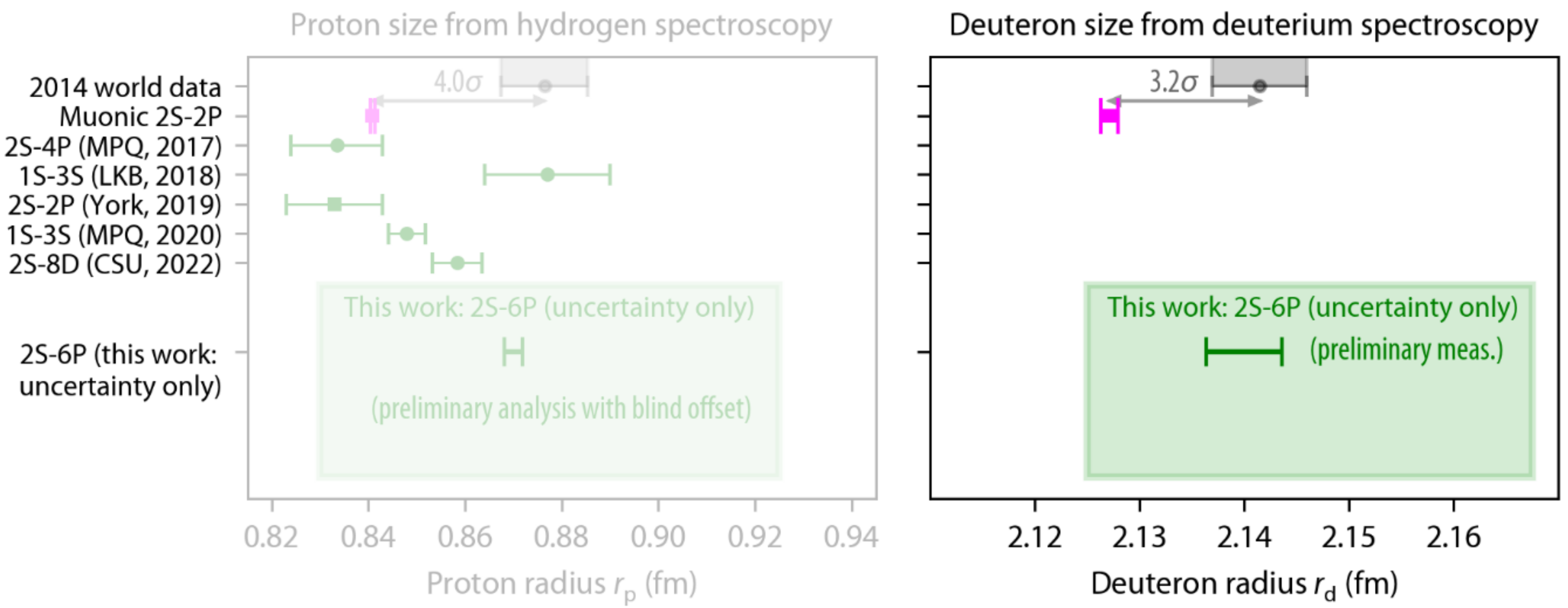
[1] Th. Udem *et al.*, *Ann. Phys. 531*(5), 1900044 (2019)

Observed deuterium 2S-6P transition signal with a high count rate, low background:

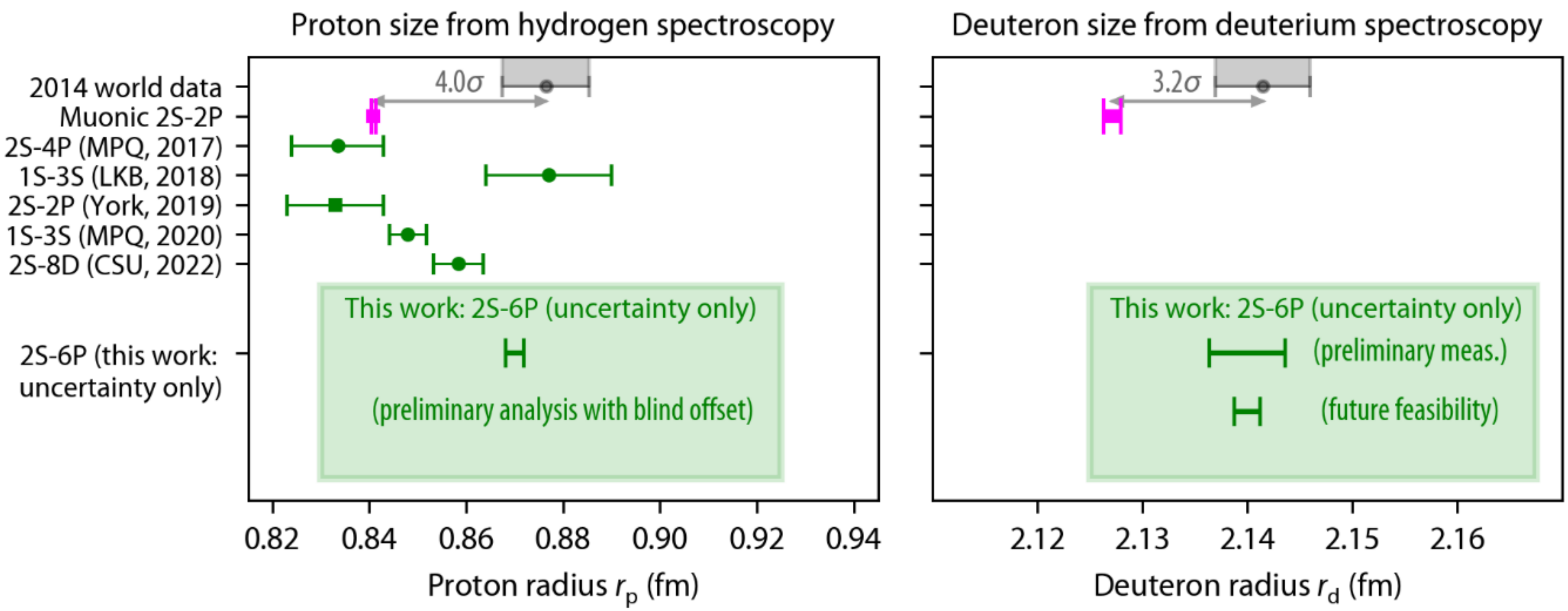


Preliminary measurement: ~ 500 deuterium 2S-6P precision line scans

Preliminary deuterium 2S-6P measurement campaign result:



Preliminary deuterium 2S-6P measurement campaign result:



Future deuterium 2S-6P measurement planned to reduce statistical uncertainty
➔ feasible with a similar precision as in hydrogen

Limiting systematic uncertainty: light force shift (currently under investigation in deuterium)

Thank you for your attention!

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