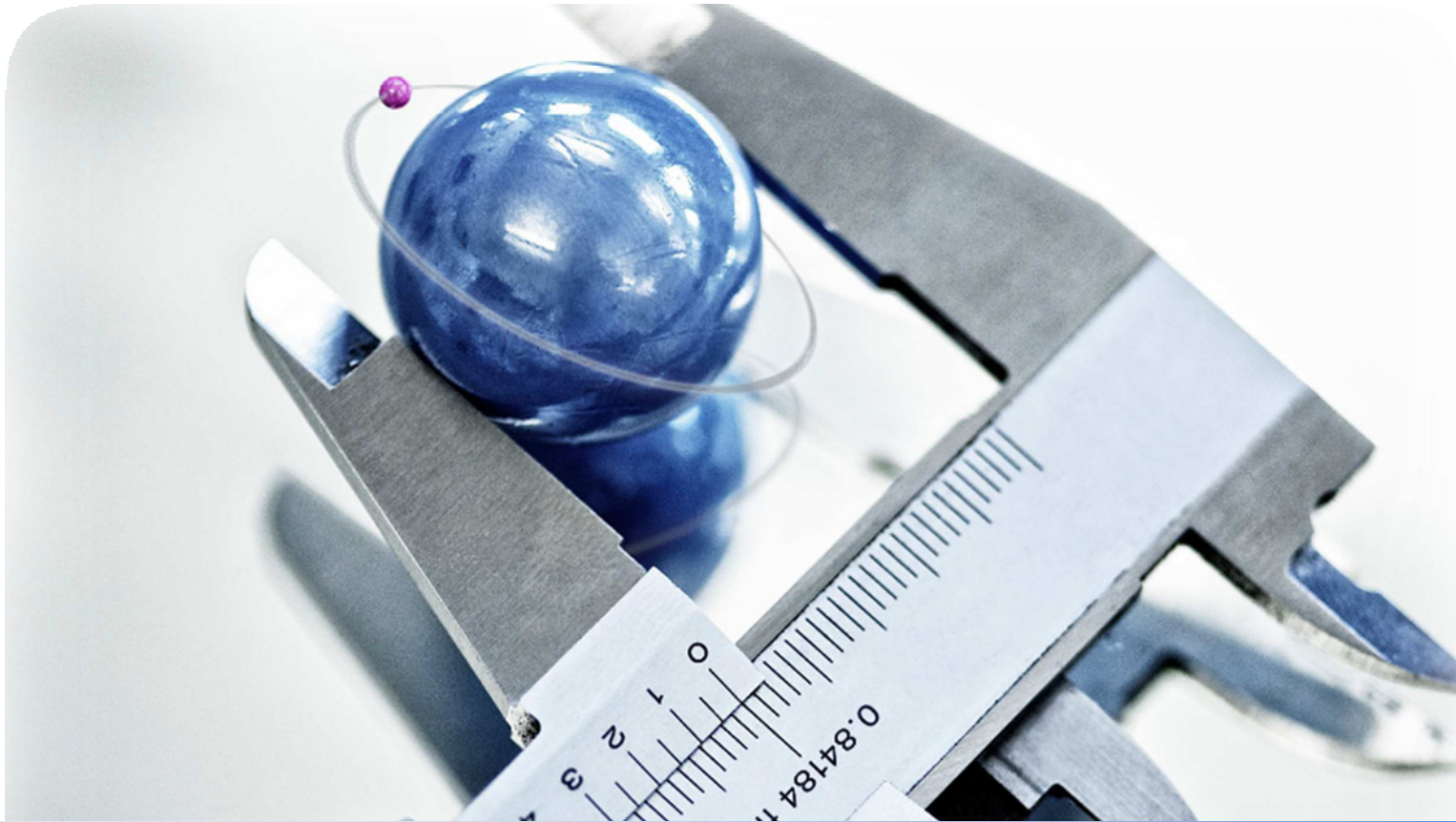
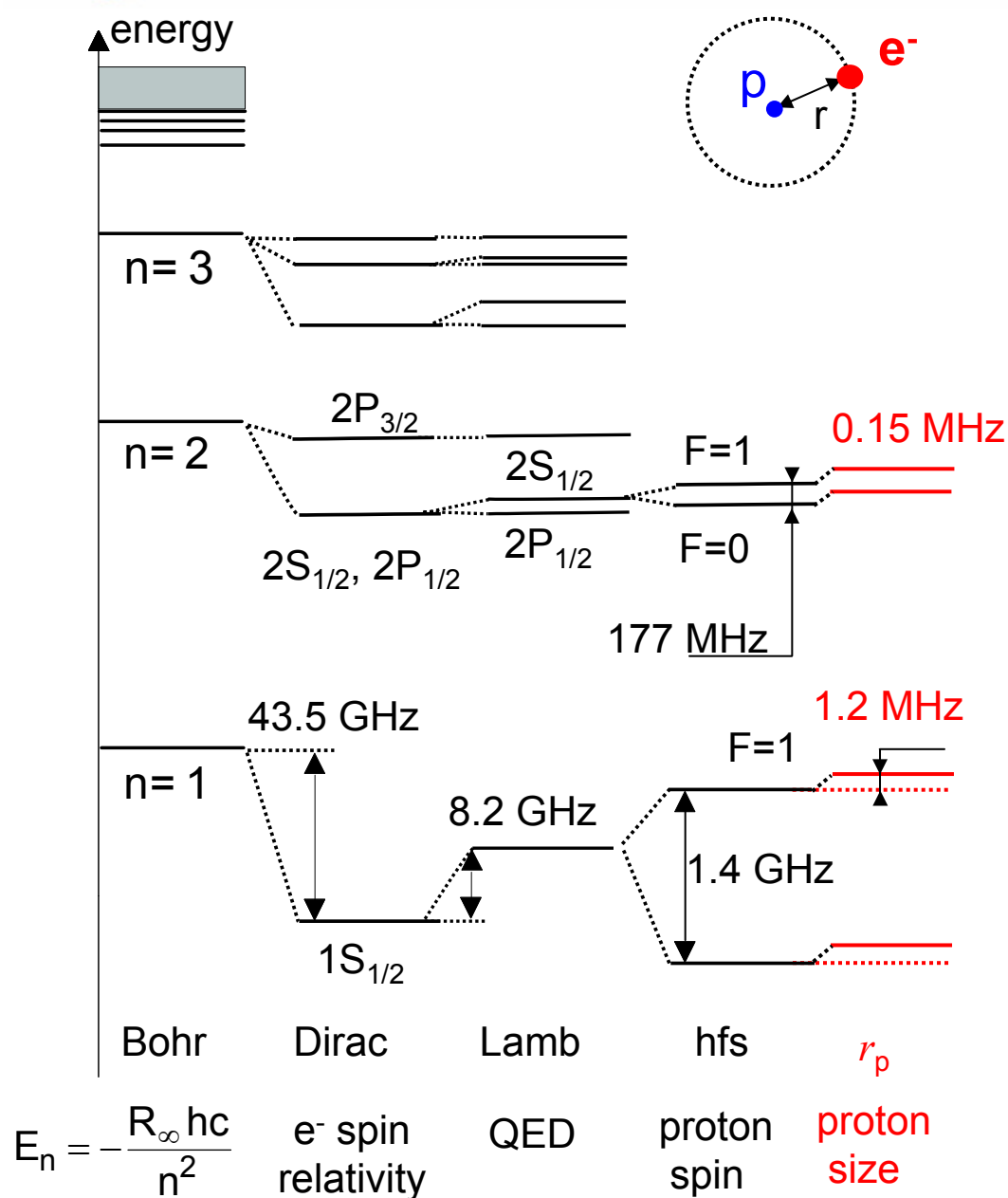


# Hydrogen 1S-3S spectroscopy to contribute to the proton charge radius puzzle



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# Hydrogen energy levels



$$E(n,l,j) = \underbrace{\text{Dirac} + \text{recoil}}_{\text{exact}} + \underbrace{L(n,j)}_{\text{not well known}}$$

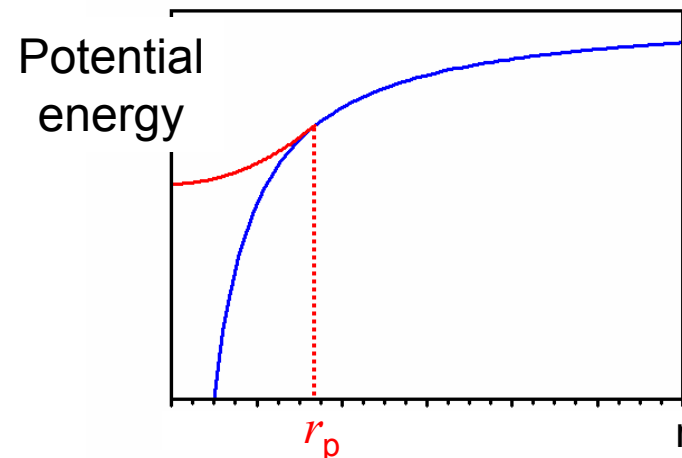
$$hcR_\infty f(\alpha, m_e/m_p, n,l,j)$$

exact

not well known

$$\Delta E^{FS} = \frac{2(Z\alpha)^4}{3n^3} m_r^3 r_p^2 \delta_{l0}$$

- QED corrections ( $1/n^3$ )
- relativistic recoil
- charge radius of the proton ( $1/n^3$ )



$$L^{\text{theo}}(1S_{1/2}) = 8172.903(4)(50) \text{ MHz}$$

QED scattering proton size



# H spectroscopy : $R_\infty$ and $L_{1S}$ determination

$$E(n,l,j) = hcR_\infty f(\alpha, m_e/m_p, n,l,j) + \text{recoil} + L(n,j,r_p) \approx \frac{R_\infty}{n^2} + L(n,r_p)$$

MPQ Garching	$\nu(1S - 2S) = (1 - \frac{1}{4})R_\infty + L(1S) - L(2S)$
LKB Paris	$\nu(2S - 8S) = (\frac{1}{4} - \frac{1}{64})R_\infty + L(2S) - L(8S)$
S. Karshenboim K. Pachucki	$L(1S) - 8L(2S) = \text{precisely calculated}$

Linear combinations  $\rightarrow R_\infty, L^{\text{exp}}(1S)$

$$L^{\text{exp}}(1S) = 8172.840(19) \text{ kHz} + \text{QED} \rightarrow r_p(1\%)$$

$$cR_\infty = 3\,289\,841\,960\,360.9(21.9) \text{ kHz} (6.6 \times 10^{-12})$$



# proton radius from H spectroscopy

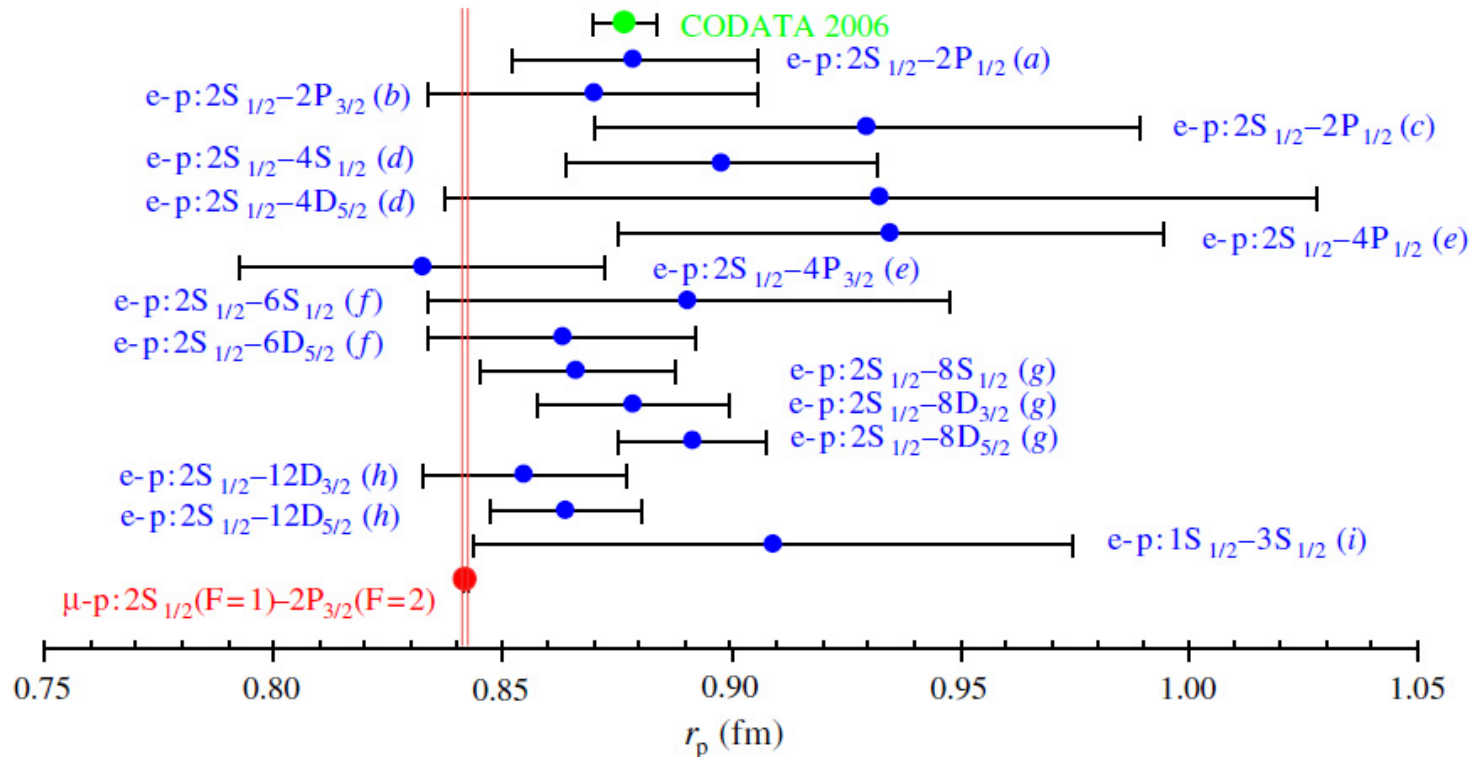
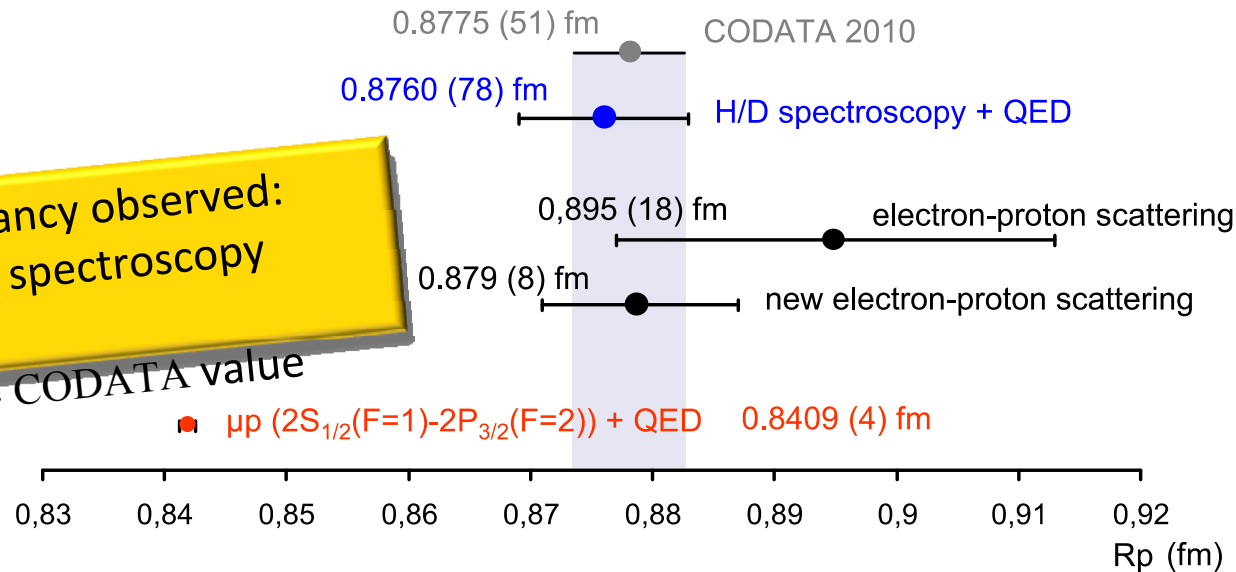


Figure 5. Comparison of various determinations of the proton radius from hydrogen spectroscopy. Each value is obtained from the  $1S-2S$  transition frequency, the  $1/n^3$  law and one of the other hydrogen experimental data from  $2S-n(S,P,D)$ . ((a) From Lundeen & Pipkin [55], (b) from Hagley & Pipkin [56], (c) from Newton *et al.* [57], (d) from Weitz *et al.* [58], (e) from Berkeland *et al.* [59], (f) from Bourzeix *et al.* [60] combined with Arnoult *et al.* [53], (g) from de Beauvoir *et al.* [24], (h) from Schwob *et al.* [61], and (i) from Arnoult *et al.* [53]). The double line corresponds to the uncertainty of the proton radius determination obtained from muonic hydrogen spectroscopy. (Online version in colour.)



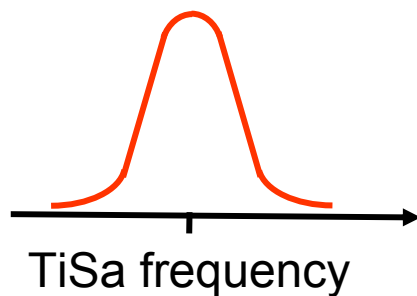
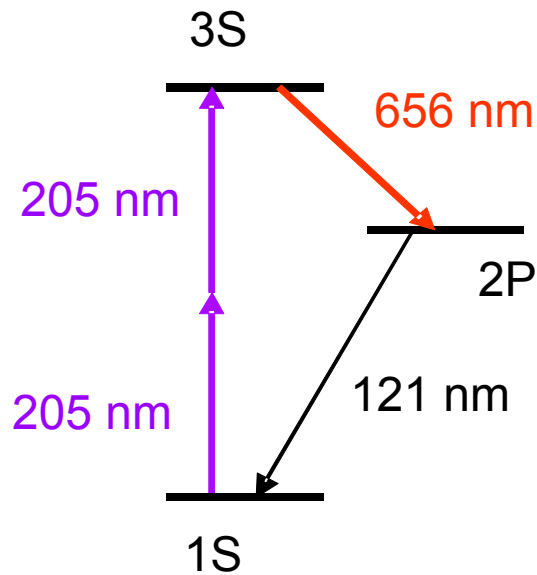
# « Proton radius puzzle »

Large discrepancy observed:  
▪  $4.5\sigma$  from H spectroscopy value  
▪  $7\sigma$  from the CODATA value



- Improve precision measurements on hydrogen:  
*1S-2S (MPQ Garching), 1S-3S (LKB and MPQ G.), 2S-6S (NPL), 2S-2P (York)*
- Improve electron-proton scattering experiment (*Newport News, Va and Mainz*)
- Improve the uncertainty of  $R_\infty$  ( *$^{20}\text{Ne}^{9+}$  Rydberg states at NIST-Gaithersburg*)
  
- Perform muon-proton scattering experiment (*MUSE project at PSI*)
- Perform  $\mu\text{-He}^+$  spectroscopy (*CREMA collaboration at PSI: 2S-2P*)
- Perform precise  $\text{He}^+$  spectroscopy (*1S-2S MPQ Garching and LaserLab Amsterdam*)

# 1S-3S/2S-8S spectroscopy of hydrogen

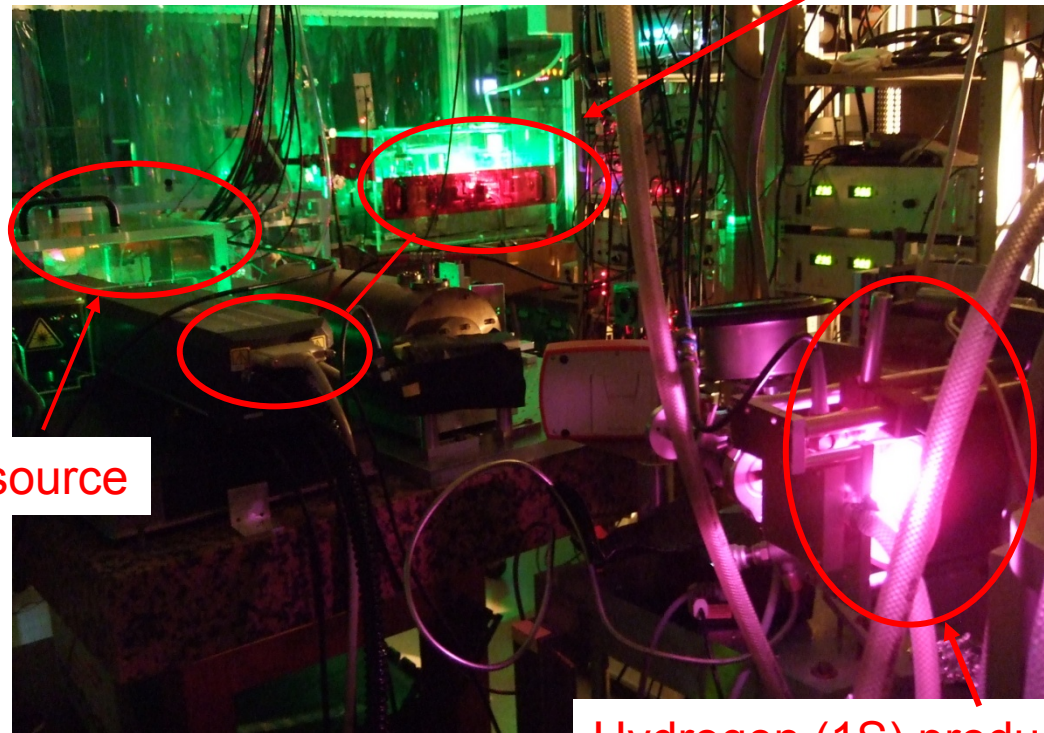
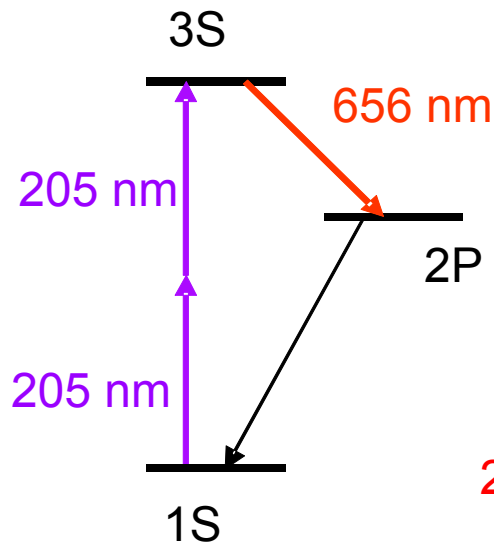
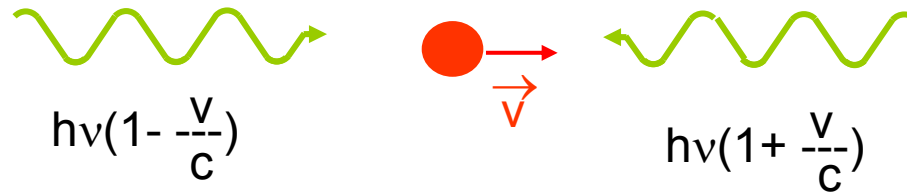


- 1S atomic beam ( $10^{14}$  at/cm<sup>3</sup>) ( $\sim 10^{11}$  at/s)  
2S atomic beam 17 at/cm<sup>3</sup> ! ( $2 \times 10^6$  at/s)
- transition probability  $\gamma_{fi} \propto \Gamma_f \gamma^2$ 
  - 1S-3S:  $\gamma = 2.14$  a.u.  
 $\Gamma_f = 1$  MHz
  - 2S-8S:  $\gamma = 14.921$  a.u.  
 $\Gamma_f = 144$  kHz
  - 1S-2S:  $\gamma = 7.85$  a.u.  
 $\Gamma_f = 1.2$  Hz
- 205 nm laser (<1mW) ( 820 nm  $\rightarrow$  410 nm  $\rightarrow$  205 nm )  
2S-8S 778 nm 1.6W !
- Velocity distribution measurement :  
No “easy” optical transition for Doppler spectroscopy (1S-2P : 121 nm ! )



# 1S-3S spectroscopy of hydrogen

Two photon spectroscopy :  
first order Doppler effect compensation

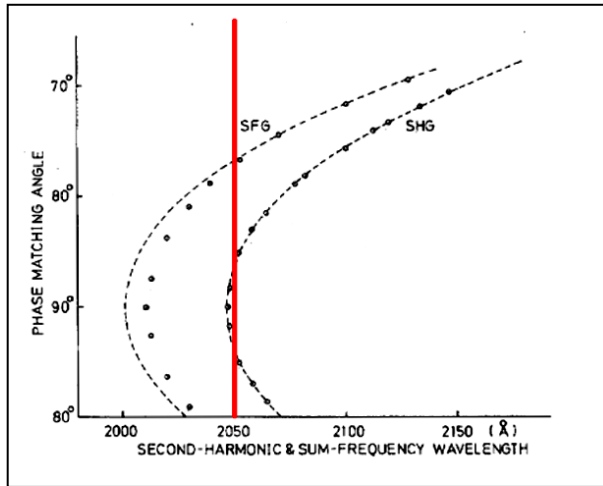


205 nm source

Laser sources

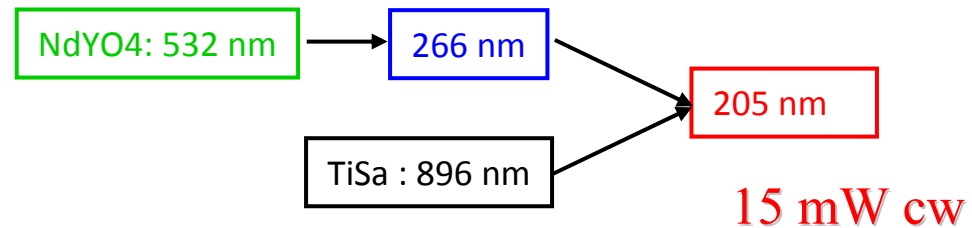
Hydrogen (1S) production

# The 205 nm cw light source

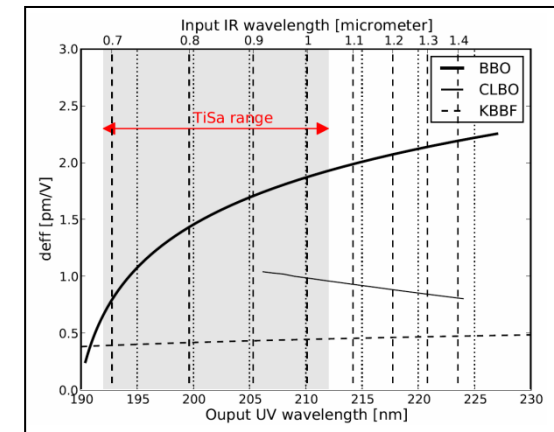


- **Two doubling stages :**  
TiSa: 820 nm → 410 nm in LBO → 205 nm in BBO  
< 1 mW quasi-continuous

- **Frequency mixing in BBO:**



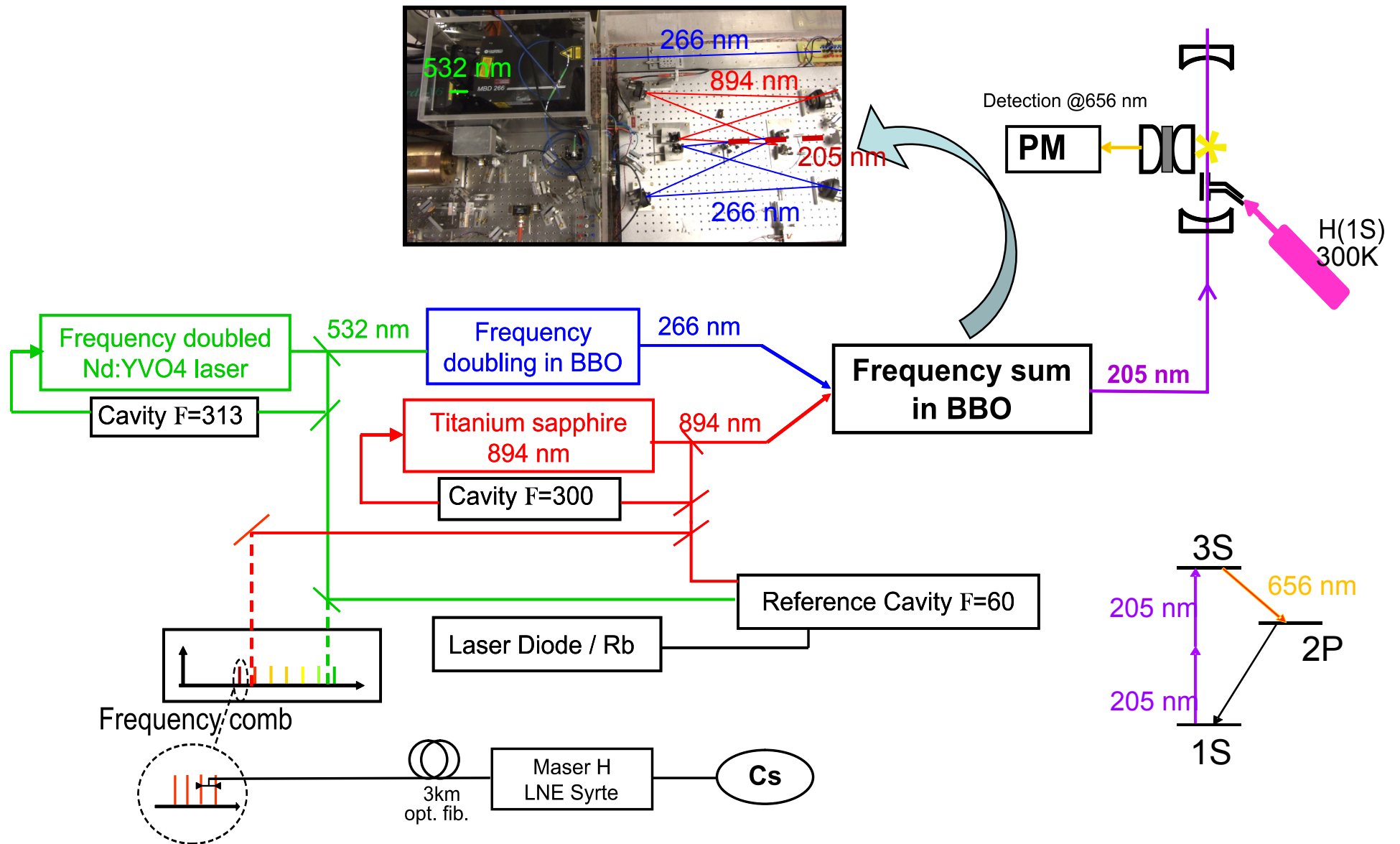
- Two-photon absorption probability proportional to  $P^2$ :  
→ Enhancement of the S/N ratio of the resonance signal
- **Continuous** laser beam:  
→ Easier spectroscopy (less systematic compared to pulsed spectroscopy)
- Possibility of generating 194 nm (1S-4S)



*S. Galtier, F. Nez, L. Julien and F. Biraben, Opt. Comm. 324 (2014) p.34-37 :  
"Ultraviolet continuous-wave laser source at 205 nm for hydrogen spectroscopy".*



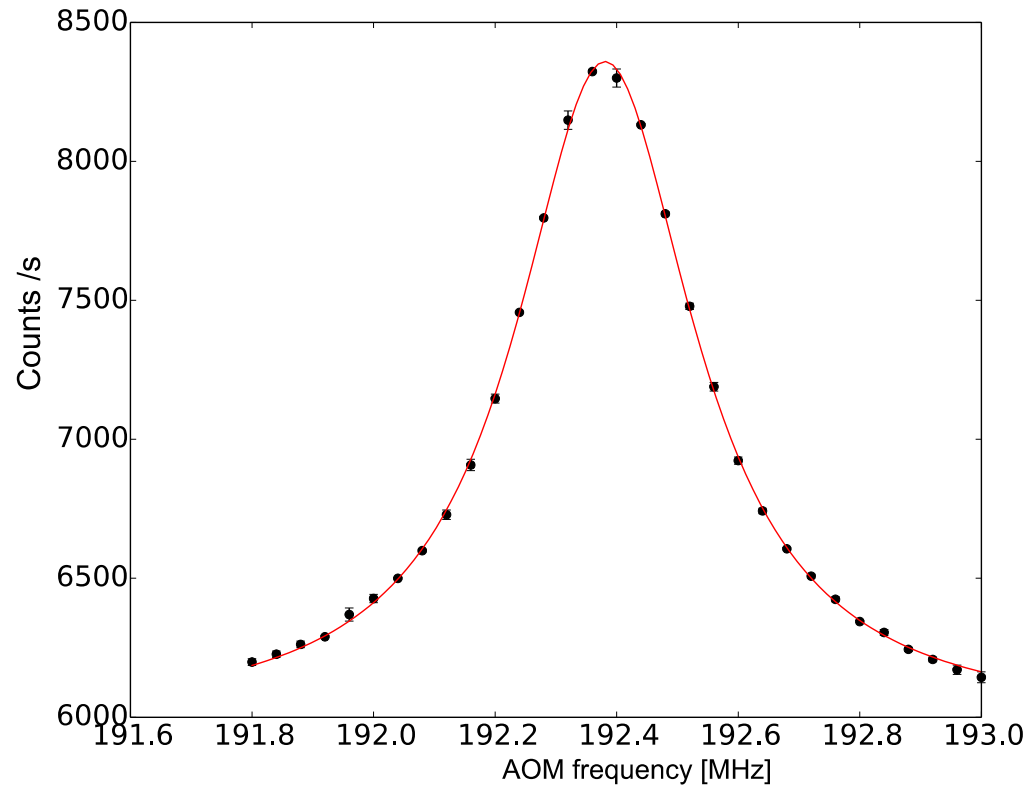
# The experimental setup



*Linewidth of TiSa laser and V6 laser : < 40 kHz*



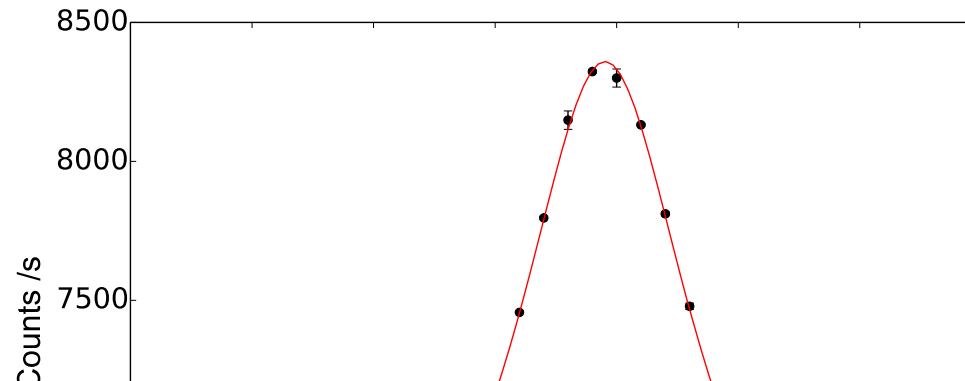
# The resonance signal



- The TiSa laser frequency is scanned over 2.4 MHz, with step of 80 kHz.  
→ curve obtained with an integration time of about 3.5 hours.
- fitted with a lorentzian function:  $\Gamma = 1.5$  MHz (1S-3S natural linewidth: 1MHz)  
(lorentzian fit on line      sophisticated fit after...)



# The resonance signal



## Frequency shifts sources:

- **Second order Doppler effect**

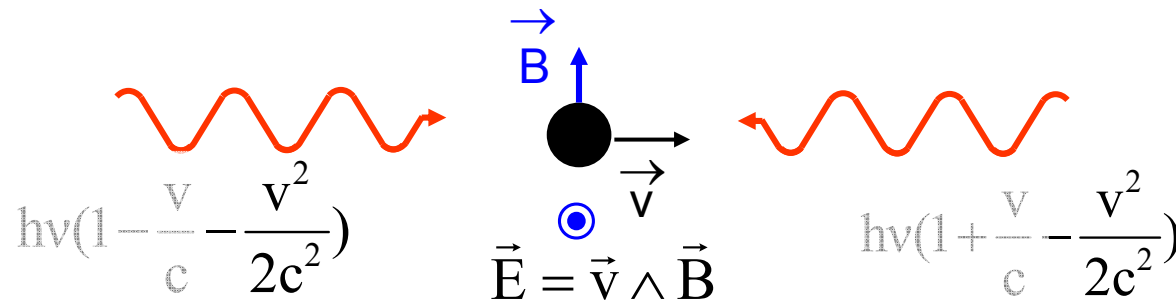
- **Light shift**

- **Collisional shift (thermal beam!)**

- We scan the TiSa laser frequency over 2.4 MHz, with step of 80 kHz.
  - curve obtained with an integration time of about 3.5 hours.
- fitted with a lorentzian function:  $\Gamma = 1.5$  MHz (1S-3S natural linewidth: 1MHz)  
(lorentzian fit on line      sophisticated fit after...)

# The 2<sup>nd</sup> order Doppler effect compensation

- **Relativistic effect**
- $v = 3\text{km/s} \rightarrow \delta_{\text{dop}} = 120 \text{ kHz} !$
- **Principle:** motional Stark effect for opposite parity levels (ex. S and P)



$$\delta_{\text{Stark}} = \frac{E^2}{\Delta v_{\text{SP}}} = \frac{v^2 B^2}{\Delta v_{\text{SP}}}$$

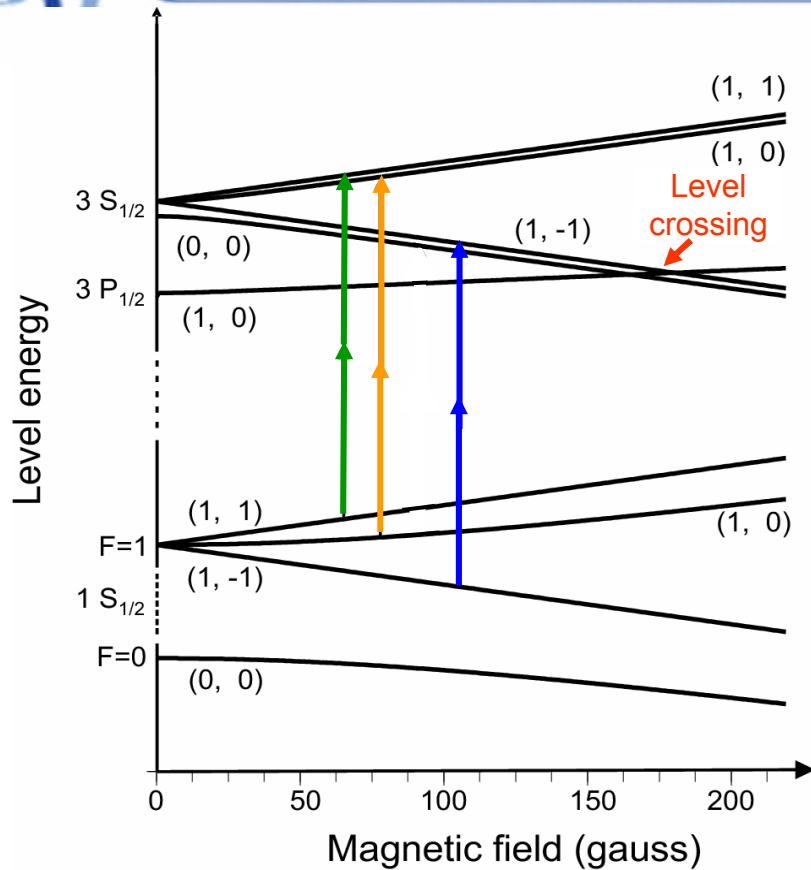
$$\delta_{\text{dop}} = -v_{\text{at}} \frac{v^2}{2c^2}$$

F. Biraben, L. Julien, J. Plon and F. Nez, Europhys. Lett., 15 (1991) p.831 :

"Compensation of the second Doppler effect in two photon spectroscopy of atomic hydrogen".



# The 2<sup>nd</sup> order Doppler effect compensation



- Two photon spectroscopy:  $\Delta F = 0$  and  $\Delta m_F = 0$

$$1S_{1/2} (F=1) \rightarrow 3S_{1/2} (F=1)$$

- Zeeman splitting:**

$$1S_{1/2} (F=1, m_F=1) \rightarrow 3S_{1/2} (F=1, m_F=1)$$

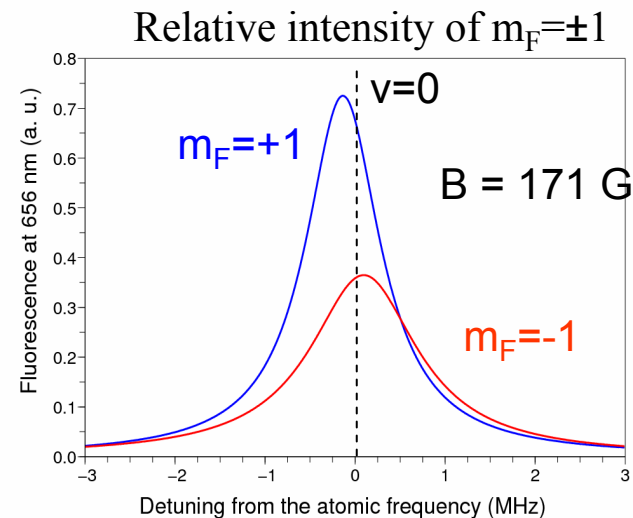
$$1S_{1/2} (F=1, m_F=-1) \rightarrow 3S_{1/2} (F=1, m_F=-1)$$

$$1S_{1/2} (F=1, m_F=0) \rightarrow 3S_{1/2} (F=1, m_F=0)$$

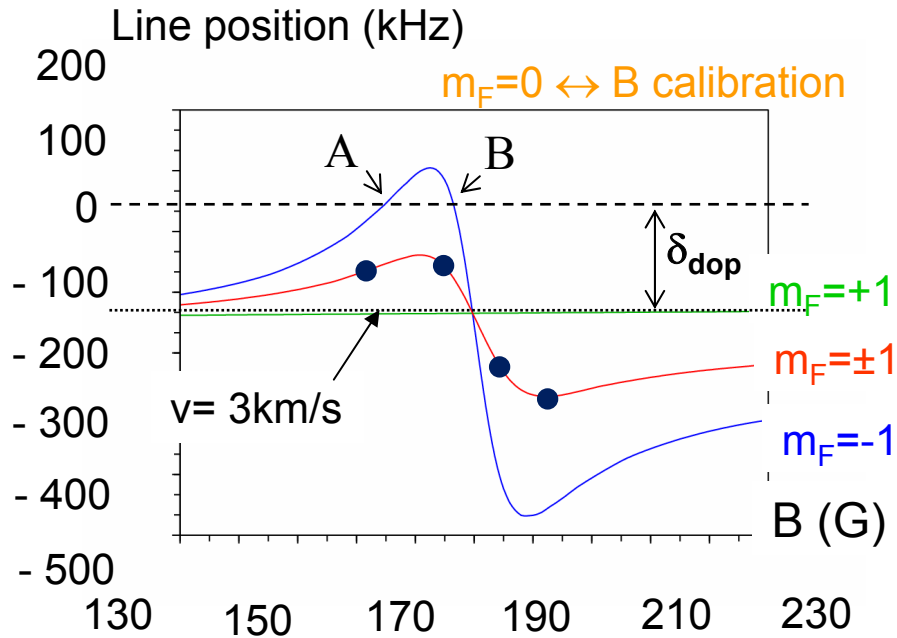
- Motional Stark effect - Level crossing 180G:**

$$3S_{1/2} (F=1, m_F = -1) \text{ coupled to } 3P_{1/2}$$

G. Hagel, R. Battesti, F. Nez, L. Julien and F. Biraben, Phys. Rev. Lett. 89 (2002) p.203001 : "Observation of a motional Stark effect to determine the second order Doppler effect".



# The 2<sup>nd</sup> order Doppler effect compensation



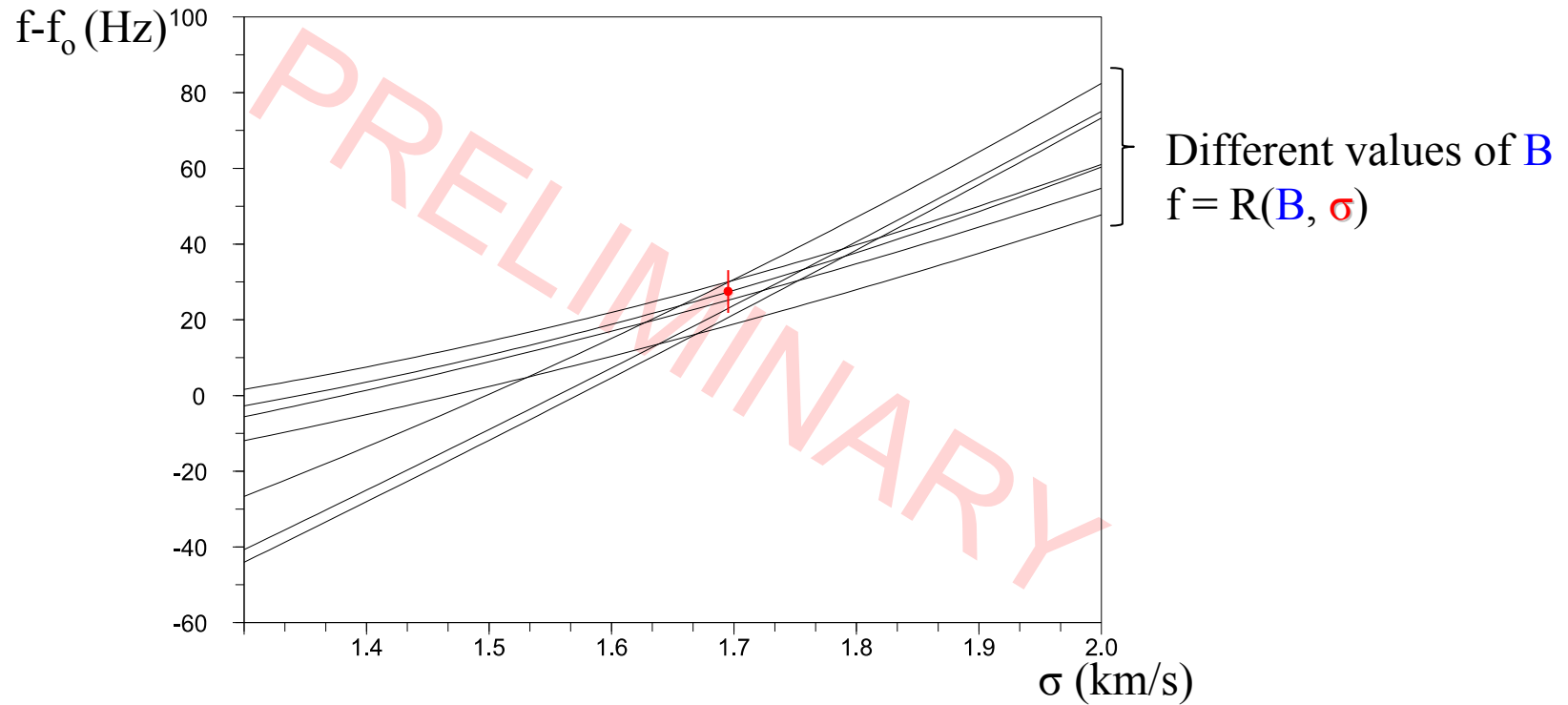
- **Two photon spectroscopy:  $\Delta F = 0$  and  $\Delta m_F = 0$**   
 $1S_{1/2} (F=1) \rightarrow 3S_{1/2} (F=1)$
- **Zeeman splitting:**  
 $1S_{1/2} (F=1, m_F=1) \rightarrow 3S_{1/2} (F=1, m_F=1)$   
 $1S_{1/2} (F=1, m_F=-1) \rightarrow 3S_{1/2} (F=1, m_F=-1)$   
 $1S_{1/2} (F=1, m_F=0) \rightarrow 3S_{1/2} (F=1, m_F=0)$
- **Motional Stark effect - Level crossing 180G:**  
 $3S_{1/2} (F=1, m_F = -1)$  coupled to  $3P_{1/2}$

Partial compensation at 171G  
 $\rightarrow$  2<sup>nd</sup> order Doppler effect determination  
 for a given velocity distribution





# 2014 first results

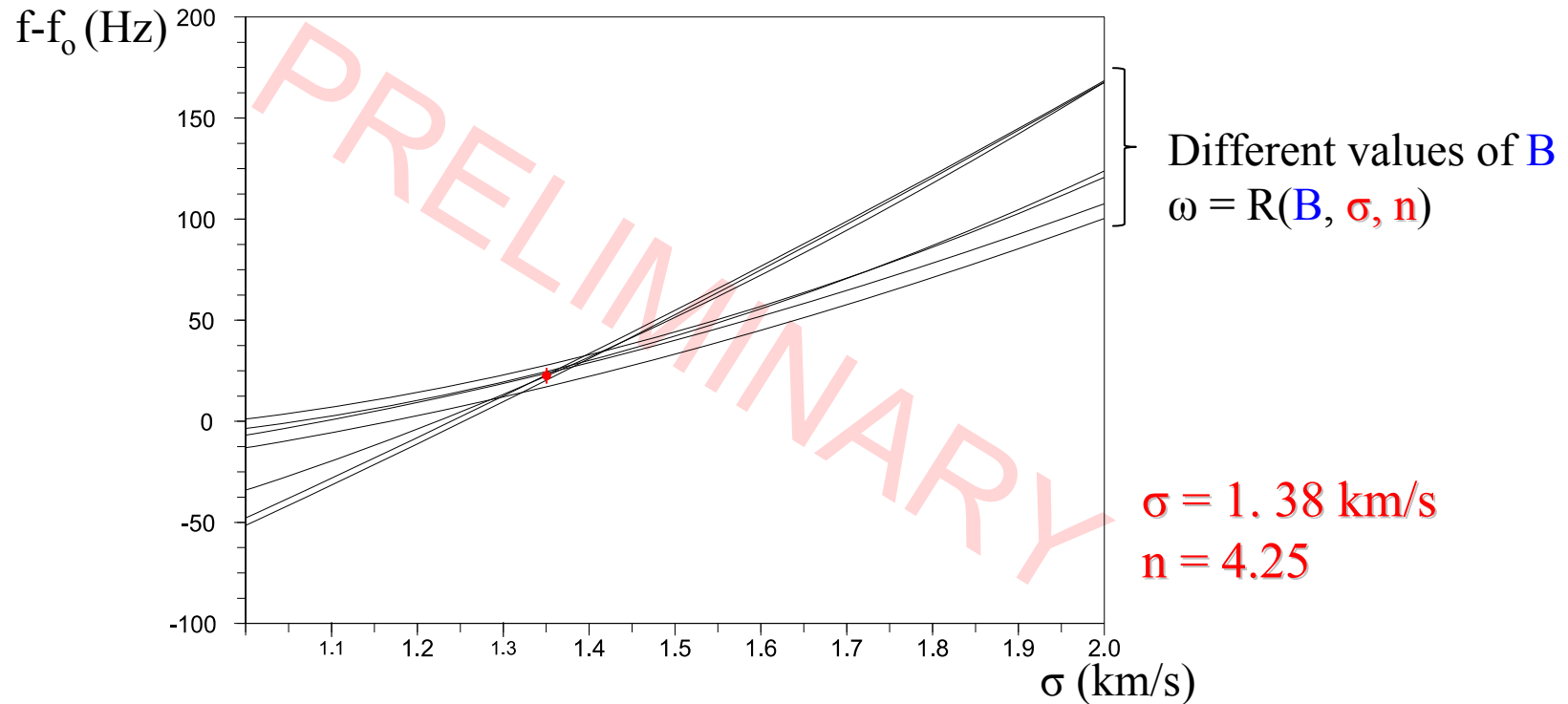


Velocity distribution?

Effusive jet:  $f(v, \sigma) = v^3 \exp(-v^2 / 2\sigma^2)$



# 2014 first results

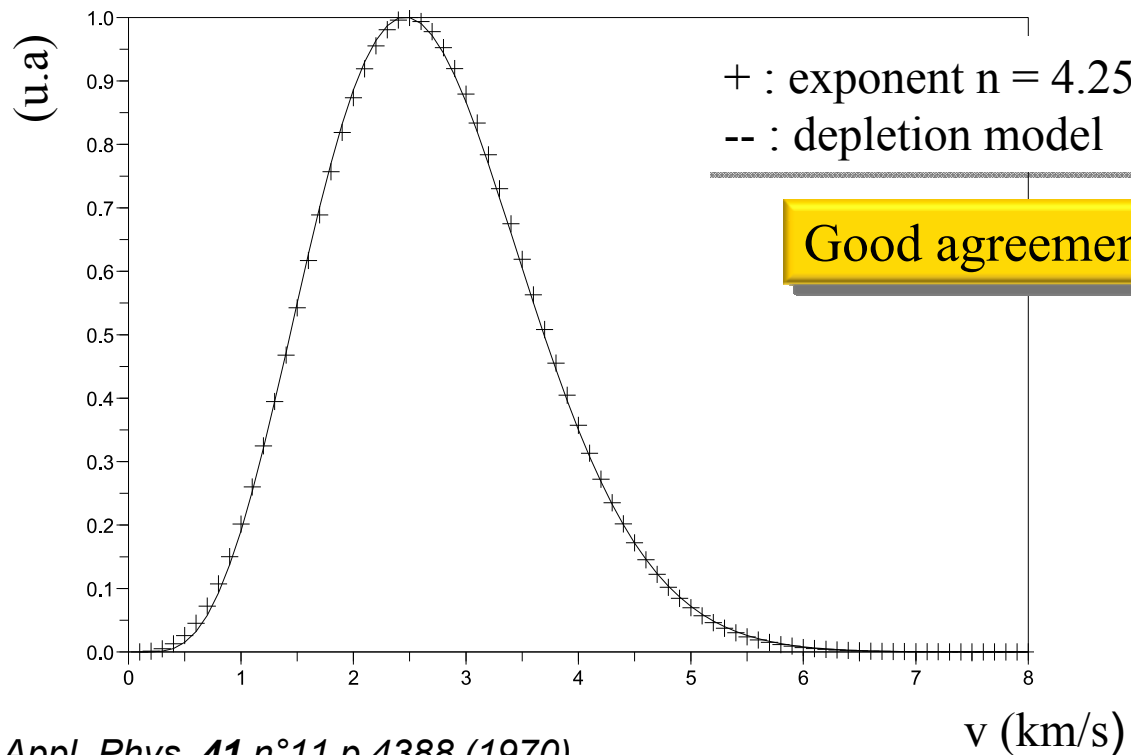
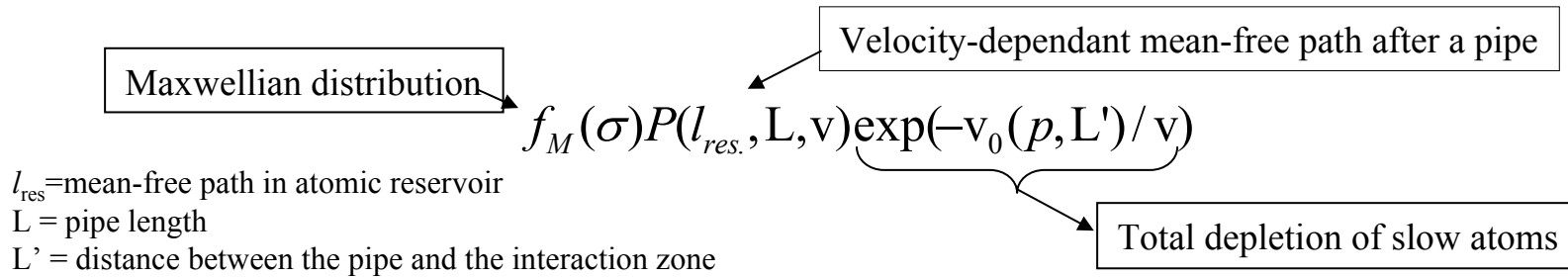


Velocity distribution?

~~Effusive jet:~~  $f(v, \sigma) = v^{\overset{n}{3}} \exp(-v^2 / 2\sigma^2)$

# The velocity distribution

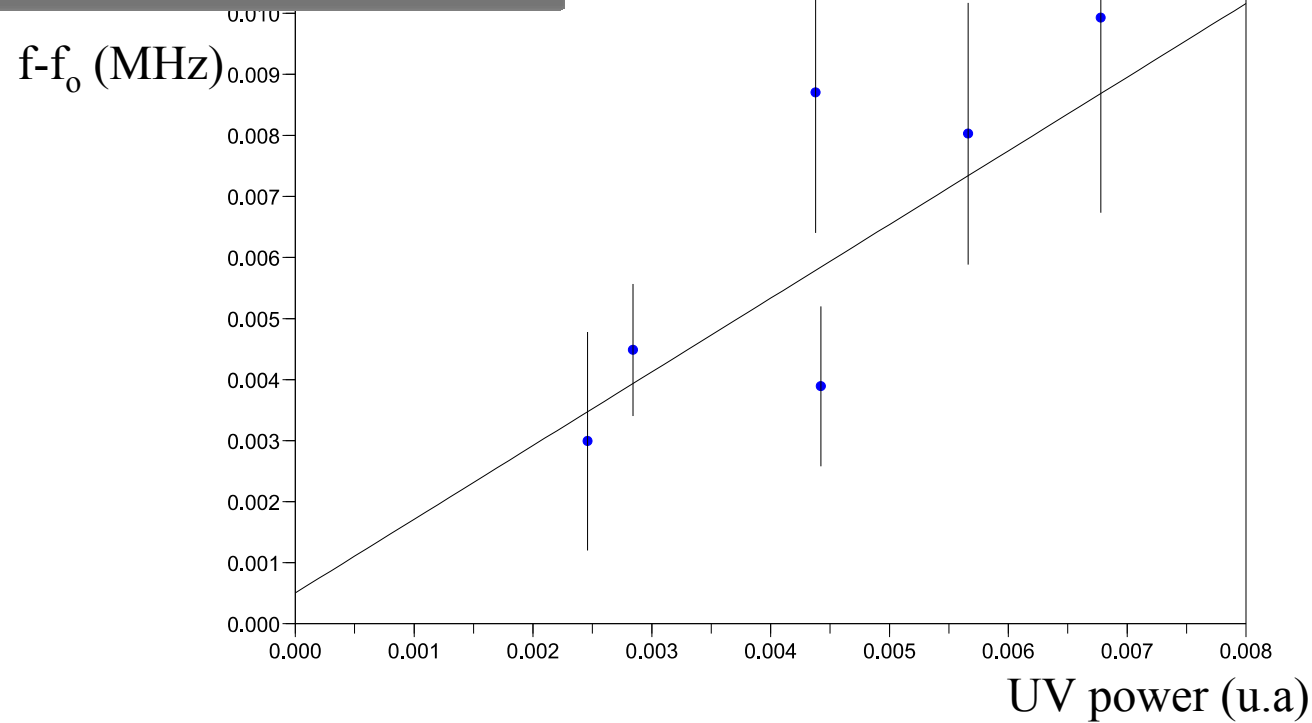
→ fast atoms pull along slower one:



D.R. Olander et al. *J. Appl. Phys.* **41** n°11 p.4388 (1970)  
 see also A. Huber et al *Phys Rev A* 59 1844 (1999)

# The light shift

## Preliminary analysis

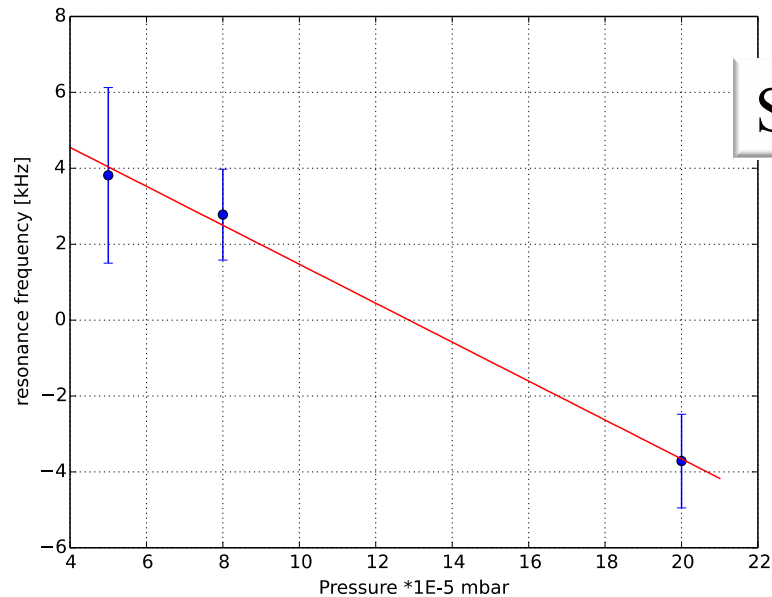


Light shift correction  $\square u_r \lesssim 10^{-12}$  (3 kHz)

$\Rightarrow$  Monitoring of the transmitted UV light from the FP cavity with a PMT instead of a UV-Si photodiode



# The collisional shift and conclusion



Shift at  $8 \cdot 10^{-5}$  mbar  $\approx 4(2)$  kHz

- Very preliminary results
- Collisional shift = the limiting effect!  
Improving the vacuum ( $1 \times 10^{-4}$  mbar currently).
- Cooling down the atomic jet ( $N_2$ ).
- Better UV power estimation

- Analysis on going
- next step: 1S-4S transitions at 194 nm ( $\Gamma=700$  kHz ).  
Laser to be checked

# Comments

## Which precision on 1S-3S transition to solve the proton puzzle ?

Taking into account the measured 1S-2S frequency:

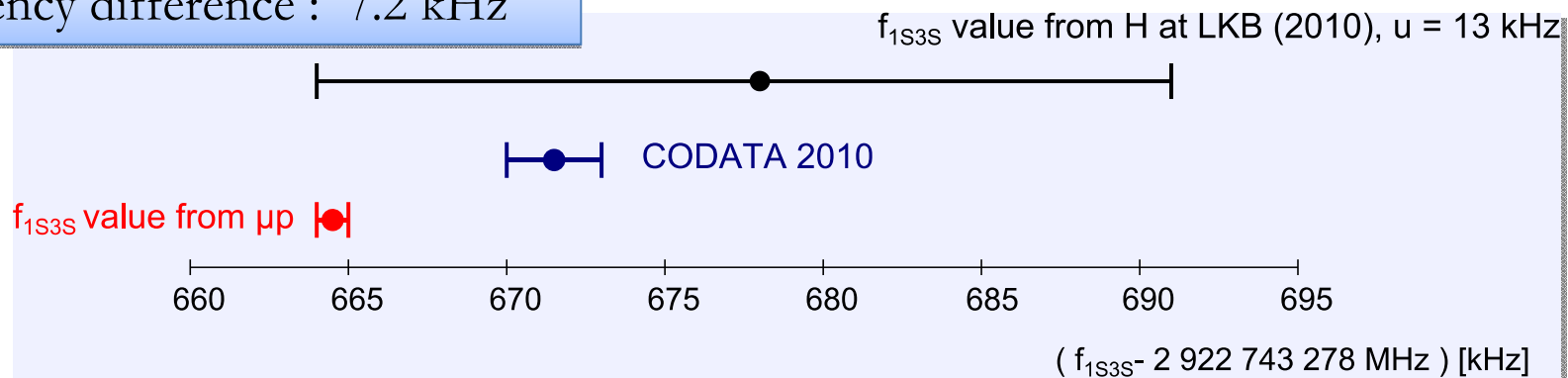
- with  $r_p$  deduced from hydrogen+scattering experiment (CODATA)

$$\nu[1S_{1/2}-3S_{1/2}] = 2\,922\,743\,278.6716 (14) \text{ MHz} \quad (4.8 \times 10^{-13})$$

- with  $r_p$  deduced from  $\mu p$  spectroscopy:

$$\nu[1S_{1/2}-3S_{1/2}] = 2\,922\,743\,278.6644 (5) \text{ MHz} \quad (1.7 \times 10^{-13})$$

Frequency difference : 7.2 kHz





I thank you  
for your attention!

Still missing  
pieces...

Bound state QED....

Proton structure

Experiments...

New theory...





## Codata Rydberg constant versus time

1998 :	109 737.315 685 9 (16) cm <sup>-1</sup>	without LKB
1998 :	109 737.315 685 3 (10) cm <sup>-1</sup>	without MPQG
1998 :	109 737.315 685 6 (96) cm <sup>-1</sup>	H only
1998 :	109 737.315 683 9 (13) cm <sup>-1</sup>	D only
1998 :	109 737.315 685 21 (81) cm <sup>-1</sup>	Codata
2002 :	109 737.315 685 59 (85) cm <sup>-1</sup>	H only
2002 :	109 737.315 683 9 (13) cm <sup>-1</sup>	D only
2002 :	109 737.315 685 25 (73) cm <sup>-1</sup>	Codata
2006 :	109 737.315 685 62 (85) cm <sup>-1</sup>	H only
2006 :	109 737.315 683 9 (13) cm <sup>-1</sup>	D only
2006 :	109 737.315 685 27 (73) cm <sup>-1</sup>	Codata
2010 :	109 737.315 685 61 (60) cm <sup>-1</sup>	H only
2010 :	109 737.315 683 7 (13) cm <sup>-1</sup>	D only
2010 :	109 737.315 685 39 (73) cm <sup>-1</sup>	Codata
2010 :	109 737.315 681 75 (12) cm <sup>-1</sup>	with $\mu\text{p}$

2006

TABLE XLV. Summary of the results of some of the least-squares adjustments used to analyze the input data related to  $R_\infty$ . The values of  $R_\infty$ ,  $R_p$ , and  $R_d$  are those obtained in the indicated adjustment,  $N$  is the number of input data,  $M$  is the number of adjusted constants,  $\nu = N - M$  is the degrees of freedom, and  $R_B = \sqrt{\chi^2/\nu}$  is the Birge ratio. See the text for an explanation and discussion of each adjustment, but, in brief, 4 is the final adjustment; 7 is 4 with the input data for  $R_p$  and  $R_d$  deleted; 8 is 4 with just the  $R_p$  datum deleted; 9 is 4 with just the  $R_d$  datum deleted; 10 is 4 but with only the hydrogen data included; and 11 is 4 but with only the deuterium data included.

Adj.	$N$	$M$	$\nu$	$\chi^2$	$R_B$	$R_\infty/\text{m}^{-1}$	$u_r(R_\infty)$	$R_p/\text{fm}$	$R_d/\text{fm}$
4	135	78	57	65.0	1.07	10 973 731.568 527(73)	$6.6 \times 10^{-12}$	0.8768(69)	2.1402(28)
7	133	78	55	63.0	1.07	10 973 731.568 518(82)	$7.5 \times 10^{-12}$	0.8760(78)	2.1398(32)
8	134	78	56	63.8	1.07	10 973 731.568 495(78)	$7.1 \times 10^{-12}$	0.8737(75)	2.1389(30)
9	134	78	56	63.9	1.07	10 973 731.568 549(76)	$6.9 \times 10^{-12}$	0.8790(71)	2.1411(29)
10	117	68	49	60.8	1.11	10 973 731.568 562(85)	$7.8 \times 10^{-12}$	0.8802(80)	
11	102	61	41	54.7	1.16	10 973 731.568 39(13)	$1.1 \times 10^{-11}$		2.1286(93)

2010

Phys. Med. Biol. 55: 973-979, 2010. doi:10.1088/0031-9155/55/9/000

TABLE XXXVIII. Summary of the results of some of the least-squares adjustments used to analyze the input data related to  $R_\infty$ . The values of  $R_\infty$ ,  $r_p$ , and  $r_d$  are those obtained in the indicated adjustment,  $N$  is the number of input data,  $M$  is the number of adjusted constants,  $\nu = N - M$  is the degrees of freedom, and  $R_B = \sqrt{\chi^2/\nu}$  is the Birge ratio. See the text for an explanation and discussion of each adjustment. In brief, adjustment 6 is 3 but the scattering data for the nuclear radii are omitted; 7 is 3, but with only the hydrogen data included (no isotope shift); 8 is 7 with the  $r_p$  data deleted; 9 and 10 are similar to 7 and 8, but for the deuterium data; 11 is 3 with the muonic Lamb-shift value of  $r_p$  included; and 12 is 11, but without the scattering values of  $r_p$  and  $r_d$ .

Adj.	$N$	$M$	$\nu$	$\chi^2$	$R_B$	$R_\infty (\text{m}^{-1})$	$u_r(R_\infty)$	$r_p (\text{fm})$	$r_d (\text{fm})$
3	149	82	67	58.1	0.93	10 973 731.568 539(55)	$5.0 \times 10^{-12}$	0.8775(51)	2.1424(21)
6	146	82	64	55.5	0.93	10 973 731.568 521(82)	$7.4 \times 10^{-12}$	0.8758(77)	2.1417(31)
7	131	72	59	53.4	0.95	10 973 731.568 561(60)	$5.5 \times 10^{-12}$	0.8796(56)	
8	129	72	57	52.5	0.96	10 973 731.568 528(94)	$8.6 \times 10^{-12}$	0.8764(89)	
9	114	65	49	46.9	0.98	10 973 731.568 37(13)	$1.1 \times 10^{-11}$		2.1288(93)
10	113	65	48	46.8	0.99	10 973 731.568 28(30)	$2.7 \times 10^{-11}$		2.121(25)
11	150	82	68	104.9	1.24	10 973 731.568 175(12)	$1.1 \times 10^{-12}$	0.842 25(65)	2.128 24(28)
12	147	82	65	74.3	1.07	10 973 731.568 171(12)	$1.1 \times 10^{-12}$	0.841 93(66)	2.128 11(28)

TABLE XVIII. Summary of the results of some of the least-squares adjustments used to analyze the input data related to  $R_\infty$  given in Tables XIV.A.1 and XIV.A.2. The values of  $R_\infty$ ,  $R_p$ , and  $R_d$  are those obtained in the indicated adjustment,  $N$  is the number of input data,  $M$  is the number of adjusted constants,  $\nu = N - M$  is the degrees of freedom,  $R_B = \sqrt{\chi^2/\nu}$  is the Birge ratio, and  $Q(\chi^2|\nu)$  is the probability that the observed value of  $\chi^2$  for  $\nu$  degrees of freedom would have exceeded that observed value.

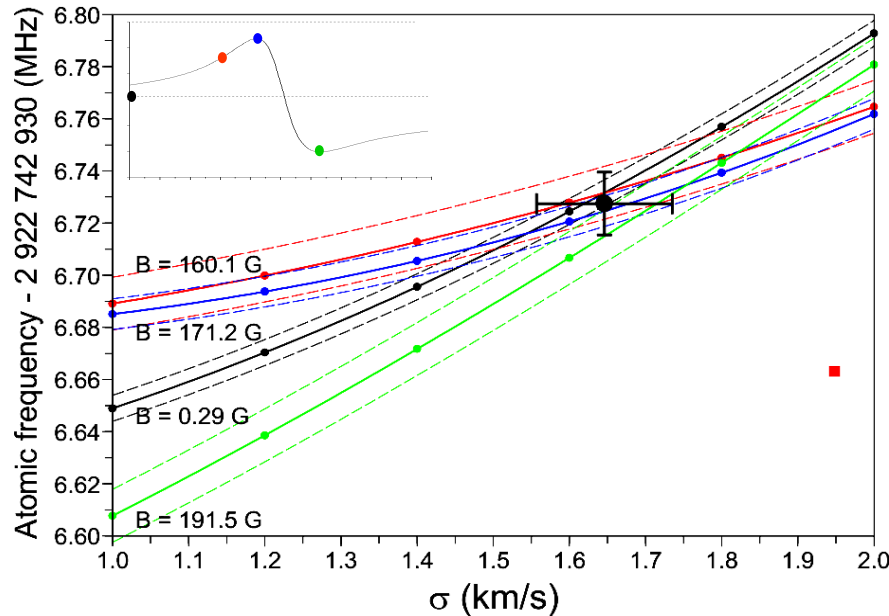
Adj.	$N$	$M$	$\nu$	$\chi^2$	$R_B$	$Q(\chi^2 \nu)$	$R_\infty/\text{m}^{-1}$	$u_r(R_\infty)$	$R_p/\text{fm}$	$R_d/\text{fm}$
1	50	28	22	12.7	0.76	0.94	10 973 731.568 521(81)	$7.3 \times 10^{-12}$	0.859(10)	2.1331(42)
2	48	28	20	10.4	0.72	0.96	10 973 731.568 549(83)	$7.5 \times 10^{-12}$	0.907(32)	2.153(14)
3	31	18	13	7.4	0.75	0.88	10 973 731.568 556(96)	$8.7 \times 10^{-12}$	0.908(33)	
4	16	11	5	2.1	0.65	0.84	10 973 731.568 32(30)	$2.7 \times 10^{-11}$		2.133(28)
5	36	28	8	4.8	0.78	0.78	10 973 731.568 59(16)	$1.5 \times 10^{-11}$	0.910(35)	2.154(15)
6	39	25	14	8.5	0.78	0.86	10 973 731.568 53(10)	$9.2 \times 10^{-12}$	0.903(35)	2.151(16)

TABLE XXIV. Summary of the results of some of the least-squares adjustments used to analyze the input data related to  $R_\infty$ . The values of  $R_\infty$ ,  $R_p$ , and  $R_d$  are those obtained in the indicated adjustment,  $N$  is the number of input data,  $M$  is the number of adjusted constants,  $\nu = N - M$  is the degrees of freedom, and  $R_B = \sqrt{\chi^2/\nu}$  is the Birge ratio.

Adj.	$N$	$M$	$\nu$	$\chi^2$	$R_B$	$R_\infty/\text{m}^{-1}$	$u_r(R_\infty)$	$R_p/\text{fm}$	$R_d/\text{fm}$
4	105	61	44	31.2	0.84	10 973 731.568 525(73)	$6.6 \times 10^{-12}$	0.8750(68)	2.1394(28)
7	103	61	42	29.0	0.83	10 973 731.568 511(82)	$7.5 \times 10^{-12}$	0.8736(77)	2.1389(32)
8	104	61	43	29.7	0.83	10 973 731.568 490(78)	$7.1 \times 10^{-12}$	0.8717(74)	2.1381(30)
9	104	61	43	30.2	0.84	10 973 731.568 546(76)	$6.9 \times 10^{-12}$	0.8769(71)	2.1402(29)
10	87	36	51	27.1	0.87	10 973 731.568 559(85)	$7.8 \times 10^{-12}$	0.8782(80)	
11	72	28	44	20.9	0.86	10 973 731.568 39(13)	$1.1 \times 10^{-11}$		2.1285(93)



## 2010 Results



- Velocity distribution:

$$f(v, \sigma) = v^3 \exp(-v^2 / 2\sigma^2)$$

- Line shape:

$$R(\omega_{\text{laser}}, \sigma, B)$$

$$\rightarrow \sigma = 1.646 (89) \text{ km/s}$$

LKB	$\nu[1S_{1/2}-3S_{1/2}] = 2\,922\,743\,278.6783$ (130) MHz	$(4.6 \times 10^{-12})$
NIST (data base)	$\nu[1S_{1/2}-3S_{1/2}] = 2\,922\,743\,278.6716$ (14) MHz	$(4.8 \times 10^{-13})$

We deduce:  $r_p = 0.911 (65) \text{ fm}$





# The velocity distribution

$$P[Kn, \psi(z)] = \frac{(\pi)^{1/2} \operatorname{erf}[\psi(z)/2Kn]^{1/2}}{2 [\psi(z)/2Kn]^{1/2}}$$

- $$\psi(z) = \frac{z \exp(-z^2) + [(\pi)^{1/2}/2](1+2z^2) \operatorname{erf}(z)}{(2\pi)^{1/2} z^2}$$

$\Psi(x)$  as for the mean number of collisions per second  $Z$  experienced by a molecule of speed  $c = x\alpha$  (where  $\alpha$  is the most probable speed) is given by:

$$Z = \sqrt{\pi} N \sigma^2 \alpha \frac{\Psi(x)}{x}$$

<sup>6</sup> E. H. Kennard, *Kinetic Theory of Gases* (McGraw-Hill, New York, 1938), pp. 97-113.

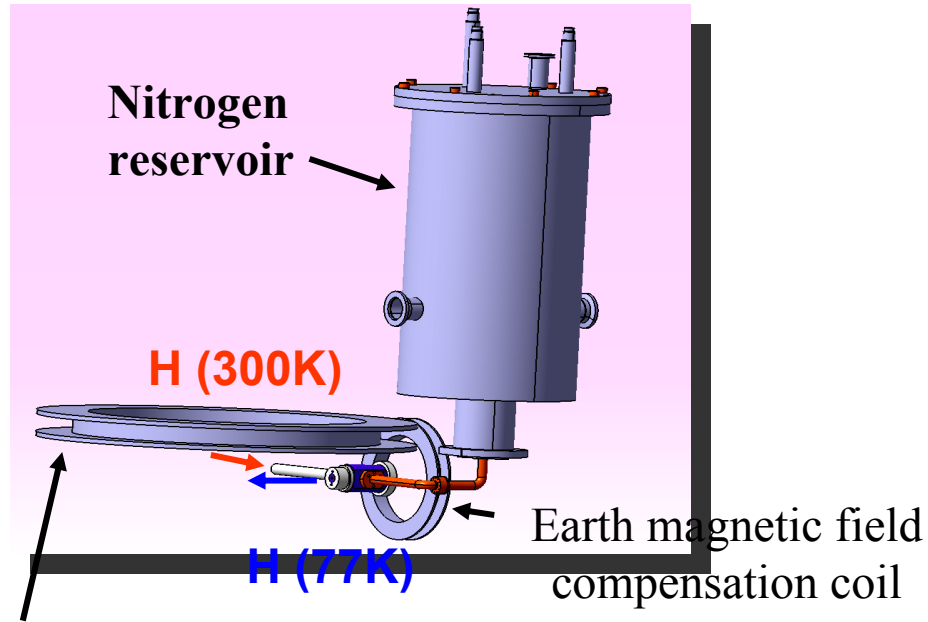
- $$Kn = \lambda_s / L.$$

The **Knudsen** number :  $L$ , the length of the circular tube from where escape the atoms.  
 $\lambda_s$ , the mean-free path in the source reservoir.

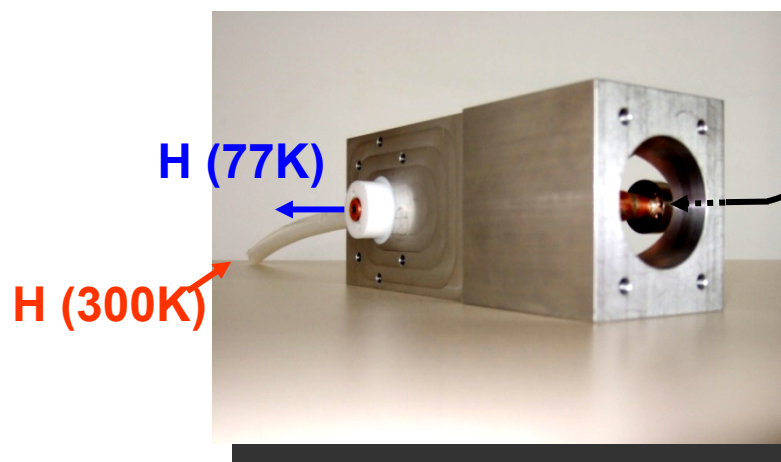
If  $Kn > 1 \rightarrow$  the velocity distribution deviates from a Maxwellian distribution  $\rightarrow$  more fast atoms



# Prospect :cooling down the hydrogen beam

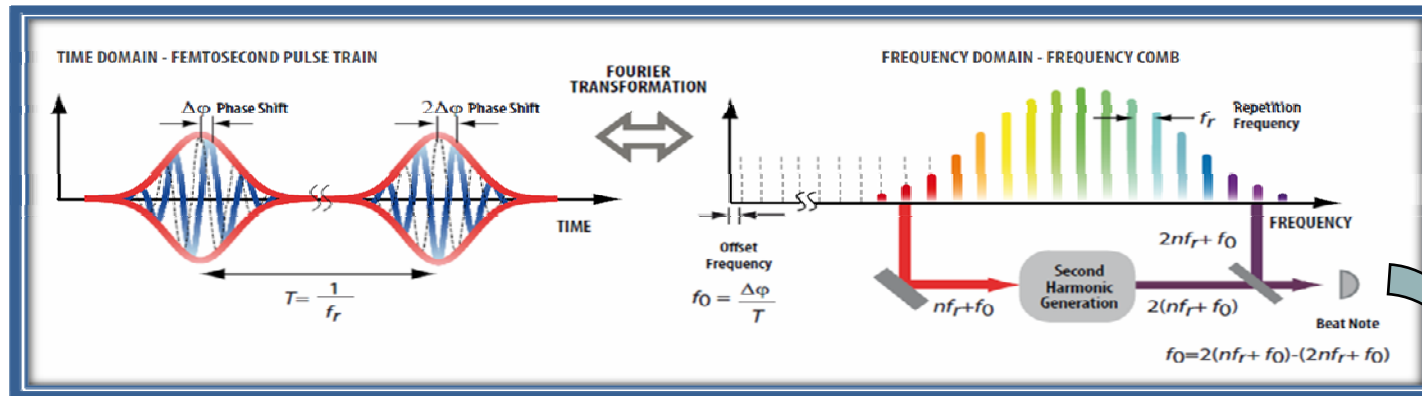


Coil to determine the atomic velocity



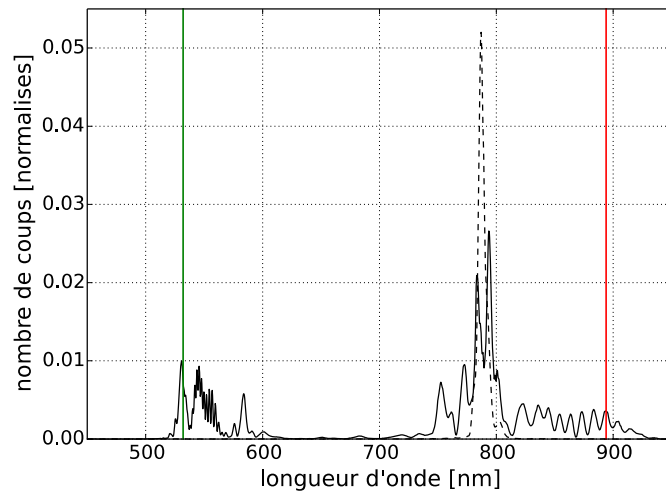
Liquid Nitrogen

# The frequency comb



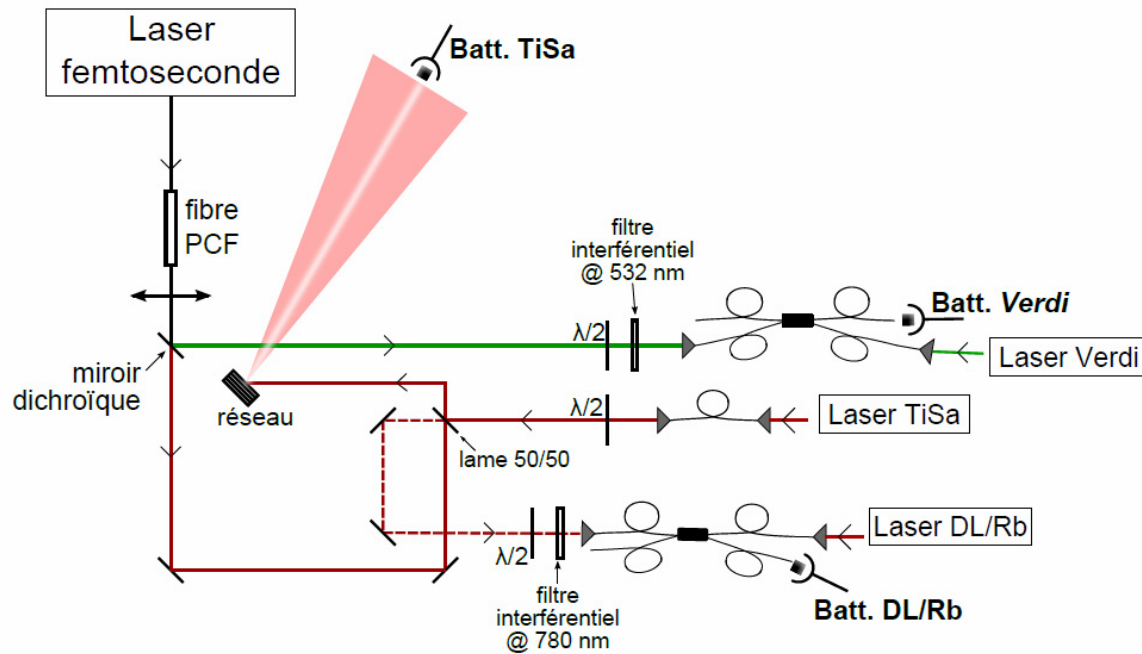
Sum frequency  $\Rightarrow$  simultaneous measurement of two laser frequencies

- Two photonic crystal fiber (PCF) spectrum:



Fiber laser frequency comb (MenloSystems)

# Absolute frequency measurements



Beat notes:

- TiSa laser:  
free space beat  
→ 30 dB with 1MHz RBW
- Verdi laser:  
fiber coupler  
→ 25 dB with 1MHz RBW

- Absolute frequency of the two lasers:

$$\begin{aligned} \text{TiSa laser: } & f_{\text{TiSa}} = 334\,797\,895\,352,900 \pm 0,994 \text{ kHz} \\ \text{Verdi laser: } & f_{\text{Verdi}} = 563\,286\,978\,440,6 \pm 2,6 \text{ kHz} \end{aligned}$$

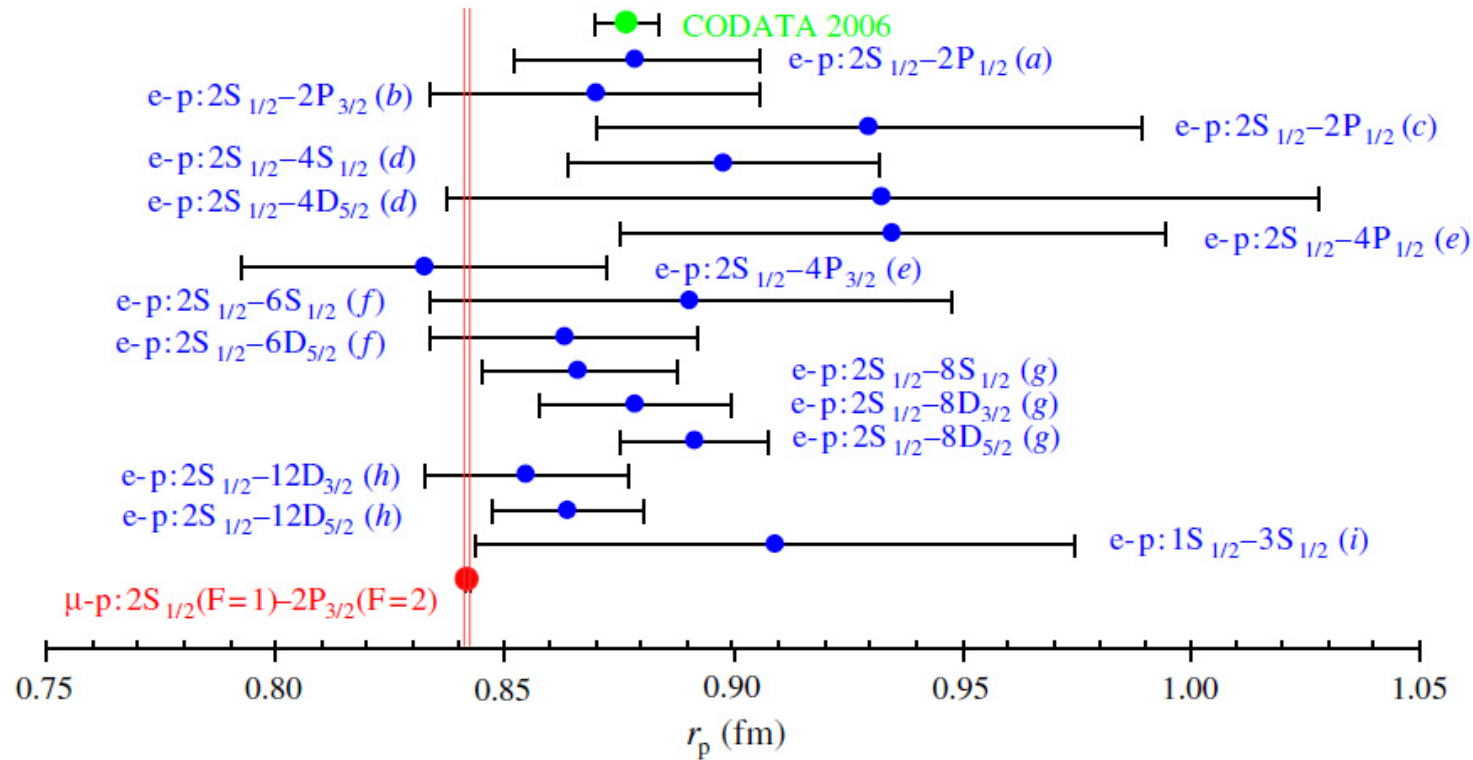


Figure 5. Comparison of various determinations of the proton radius from hydrogen spectroscopy. Each value is obtained from the  $1S-2S$  transition frequency, the  $1/n^3$  law and one of the other hydrogen experimental data from  $2S-n(S,P,D)$ . ((a) From Lundeen & Pipkin [55], (b) from Hagley & Pipkin [56], (c) from Newton *et al.* [57], (d) from Weitz *et al.* [58], (e) from Berkeland *et al.* [59], (f) from Bourzeix *et al.* [60] combined with Arnoult *et al.* [53], (g) from de Beauvoir *et al.* [24], (h) from Schwob *et al.* [61], and (i) from Arnoult *et al.* [53]). The double line corresponds to the uncertainty of the proton radius determination obtained from muonic hydrogen spectroscopy. (Online version in colour.)