PLASMA & MATERIALS PROCESSING

The role of negative ions on the density of excited hydrogen atoms in a hydrogen plasma jet

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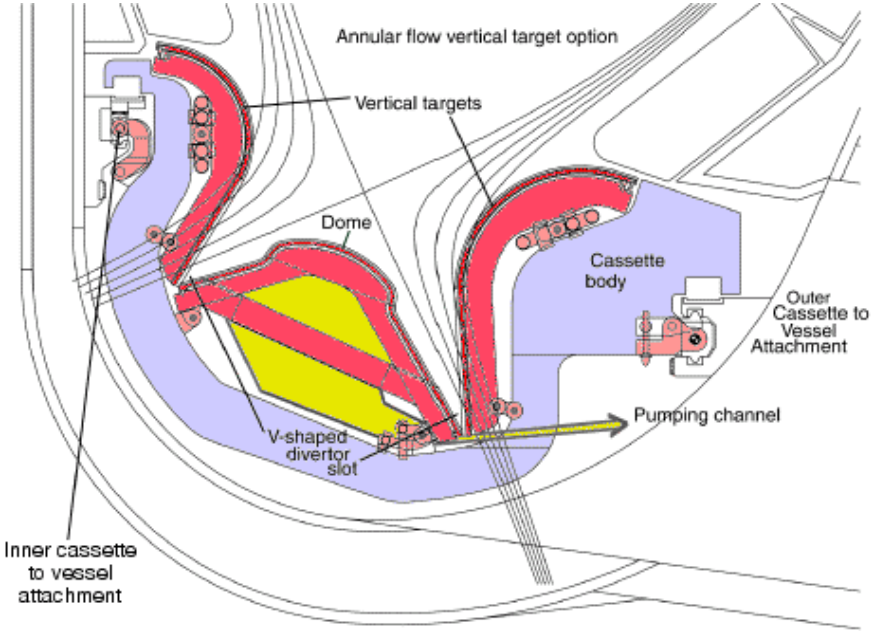
Technische Universiteit Eindhoven University of Technology

Where innovation starts

GDR Arches, May 4 – 7, 2009, Agelonde

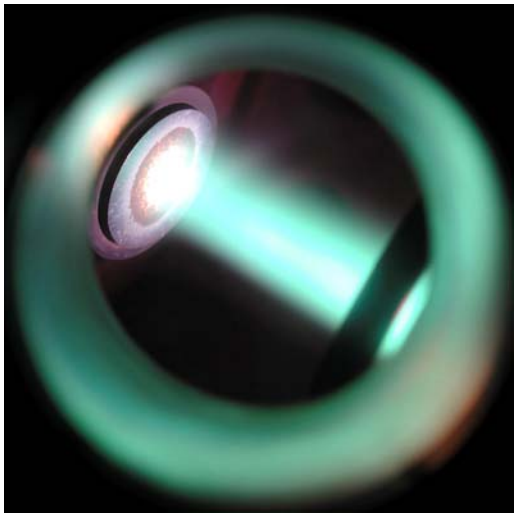
Hydrogen containing plasma



http://www.iter.org/divertor.htm

Divertor region of a fusion reactor

- Cool enough for H_2 to survive long enough; loss of dissociation and ionization processes.
- H_2^{rv} can reduce the ion flux due to molecular assisted recombinations (MAR)
- Hydrogen surface association on carbon surfaces?



H / H_2 in thin film deposition of polycrystalline / amorphous silicon

Why study expanding hydrogen plasma ?
(produced from a cascaded arc)

1. Use of H_2 gas in processing plasma application

- etching and cleaning
- passivation during deposition

2. Astrophysical interest

- 'hot' H_2, formed at grains through surface association, and acts as precursor in astro-chemistry

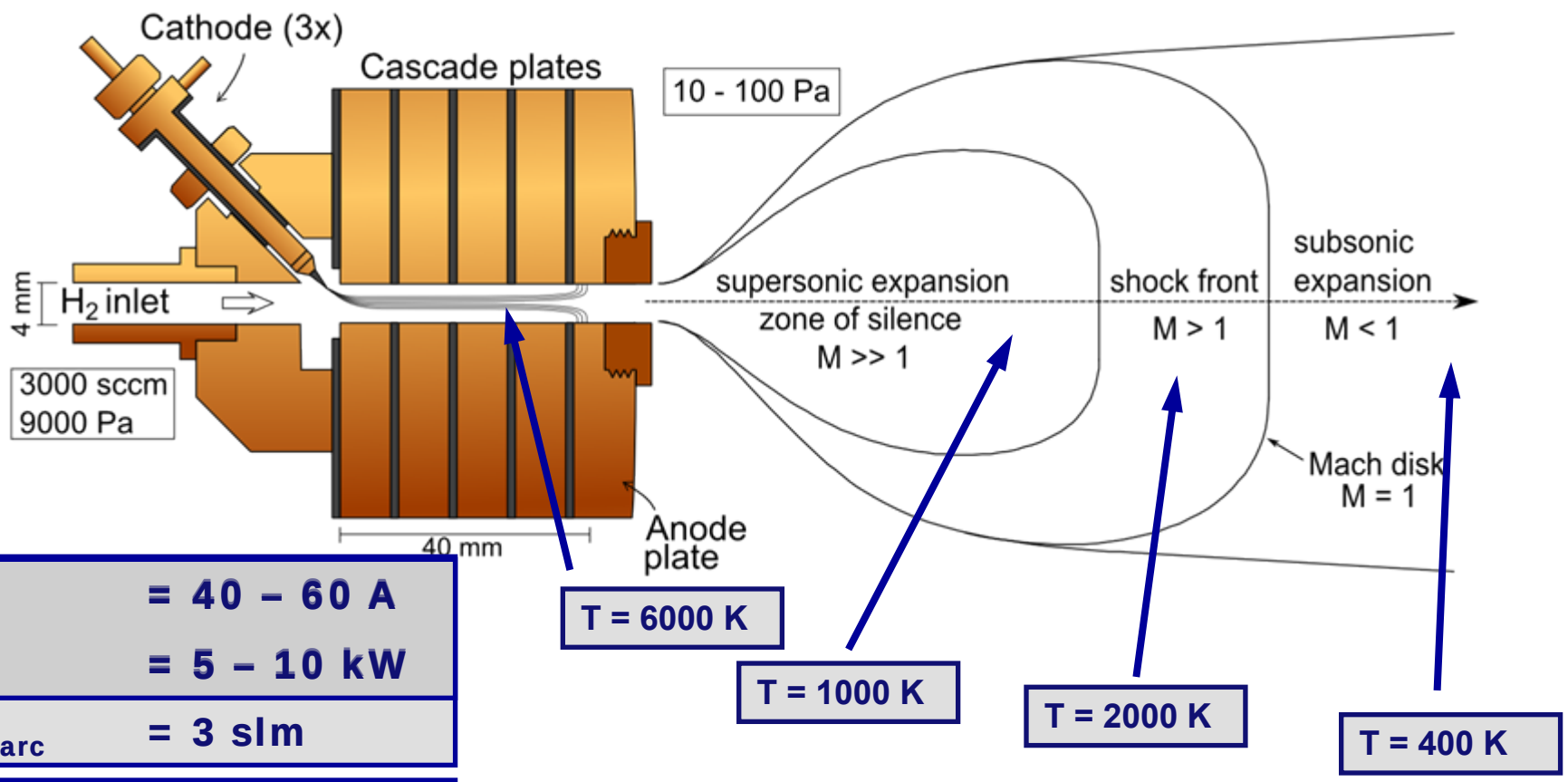
3. Fundamental study of H_2 / HD / D_2 Lyman transitions

- extension of database

4. The cascaded arc might be used as H^- ion source, because of high fluxes of $H_2^{r,v}$

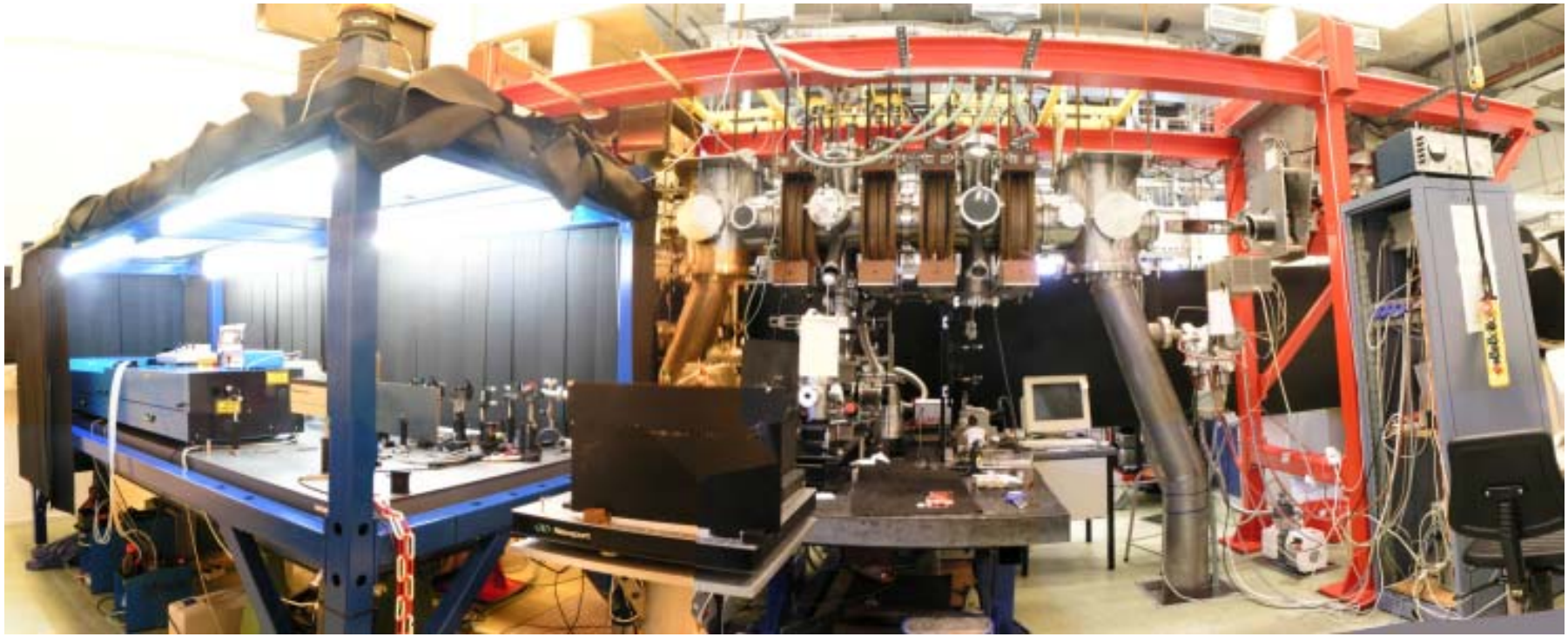
Plasma source and expansion

Plasma expansion



I	= 40 – 60 A
P	= 5 – 10 kW
Φ_{arc}	= 3 slm
p_{arc}	= 0.2×10^5 Pa
p_{bg}	= 100 Pa

PLEXIS setup



Laser table
Nd:YAG
(450 mJ/shot @ 355 nm)
dye laser
(50 mJ/shot @ 460 nm)

Vacuum chamber
cylindrical (2m x 0.3m)
7 Pa / 2000 sccm H_2

- Movable plasma source and substrate
- Axial magnetic field B_{max} = 0.2 T

Measurements performed on ETP (H_2/D_2)

1. Two-photon absorption LIF (TALIF)

- H-atom densities, velocities and temperatures (z,r)

2. VUV-LIF

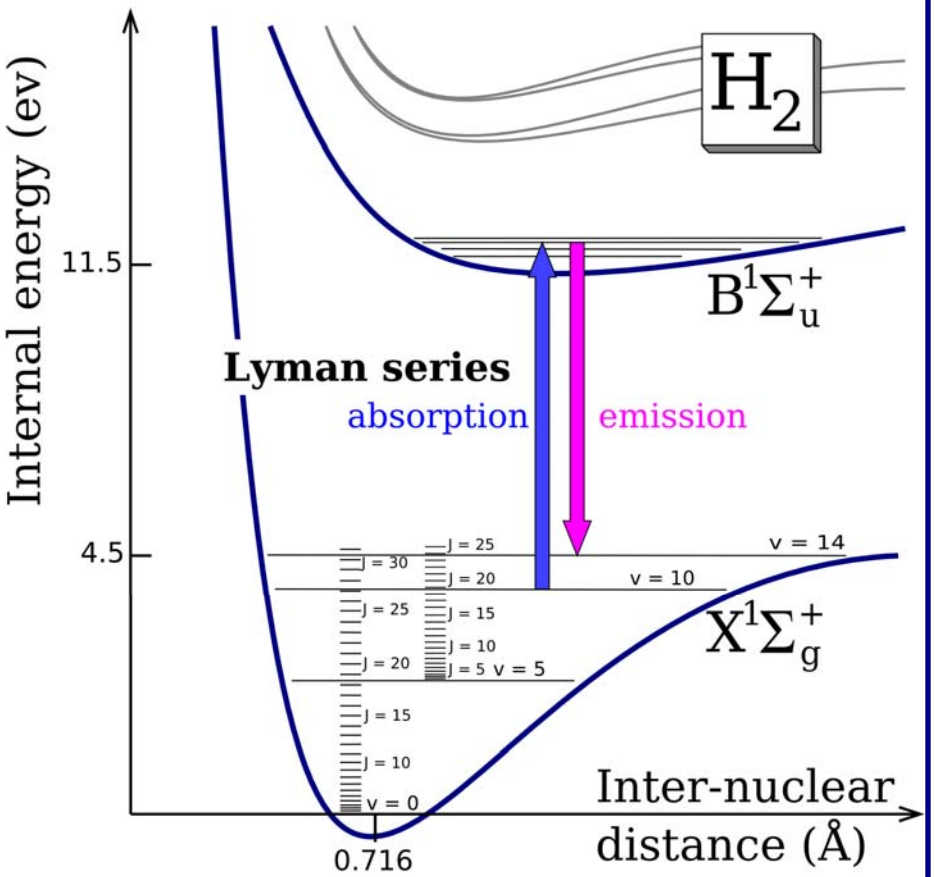
- $H_2^{r,v}$, $D_2^{r,v}$, $HD^{r,v}$ Lyman spectra (z)
- (non-) Boltzmann density distributions

3. Optical emission spectroscopy

- H(n), D(n) absolute density (z)

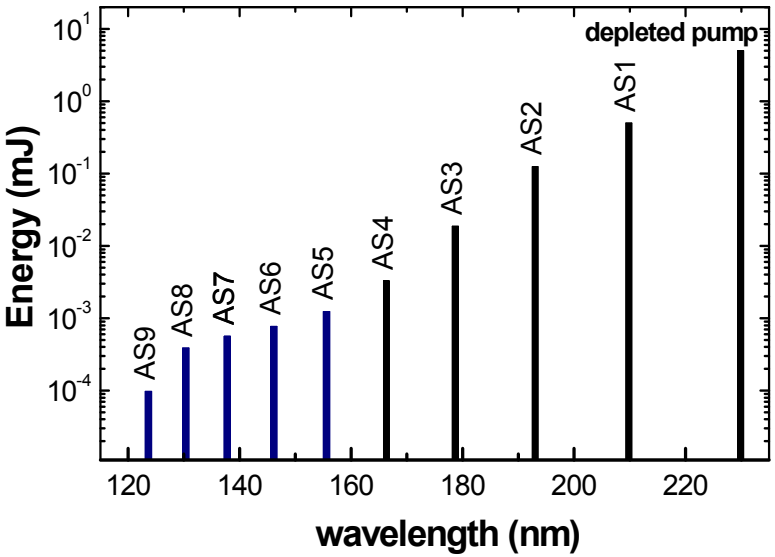
VUV-LIF detection of $H_2^{r,v}$

H_2 energy scheme

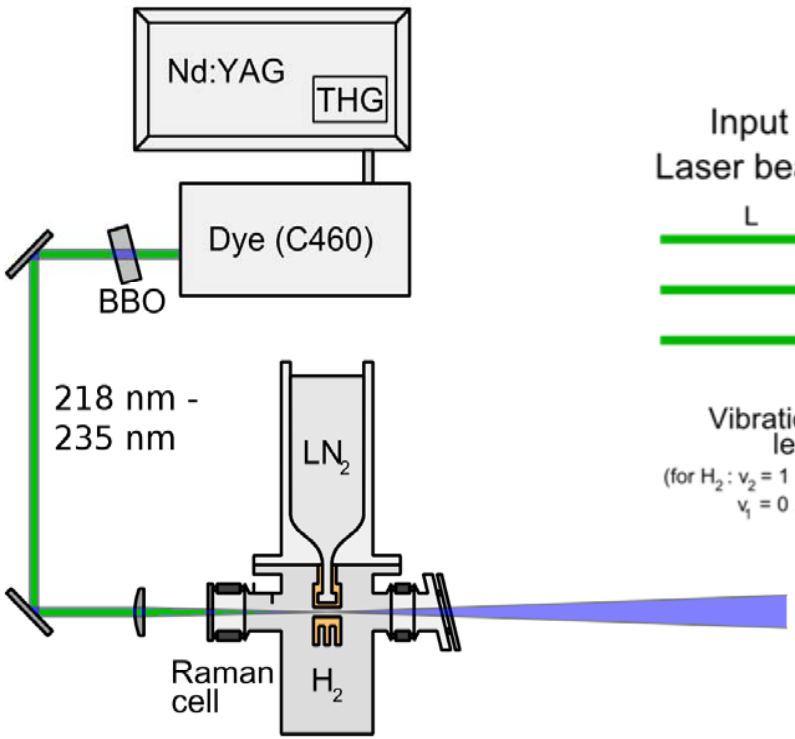


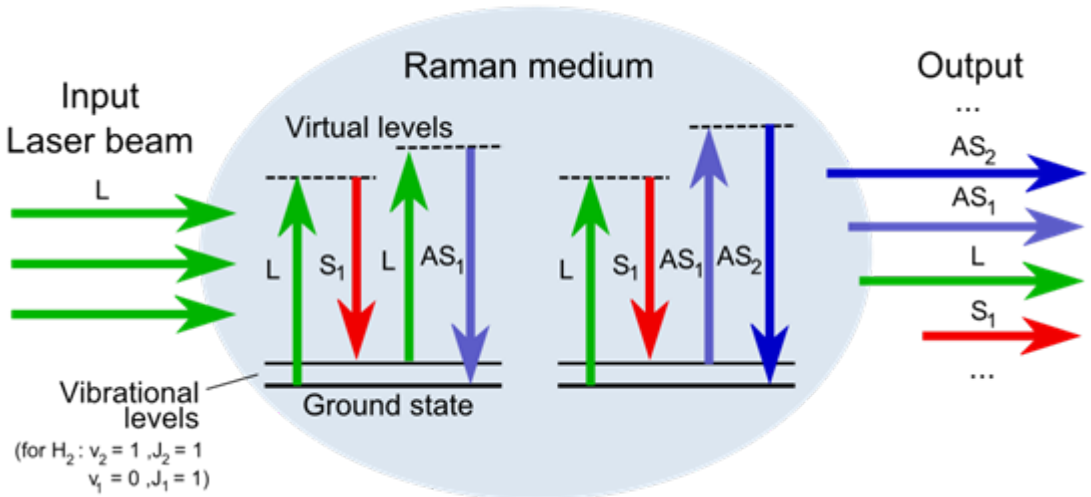
Excitation and detection

- X → B transition in H_2 (~11 eV)
- Detection of fluorescence in the VUV range
- Excitation with 120 – 165 nm photons, produced via SARS



SARS technique

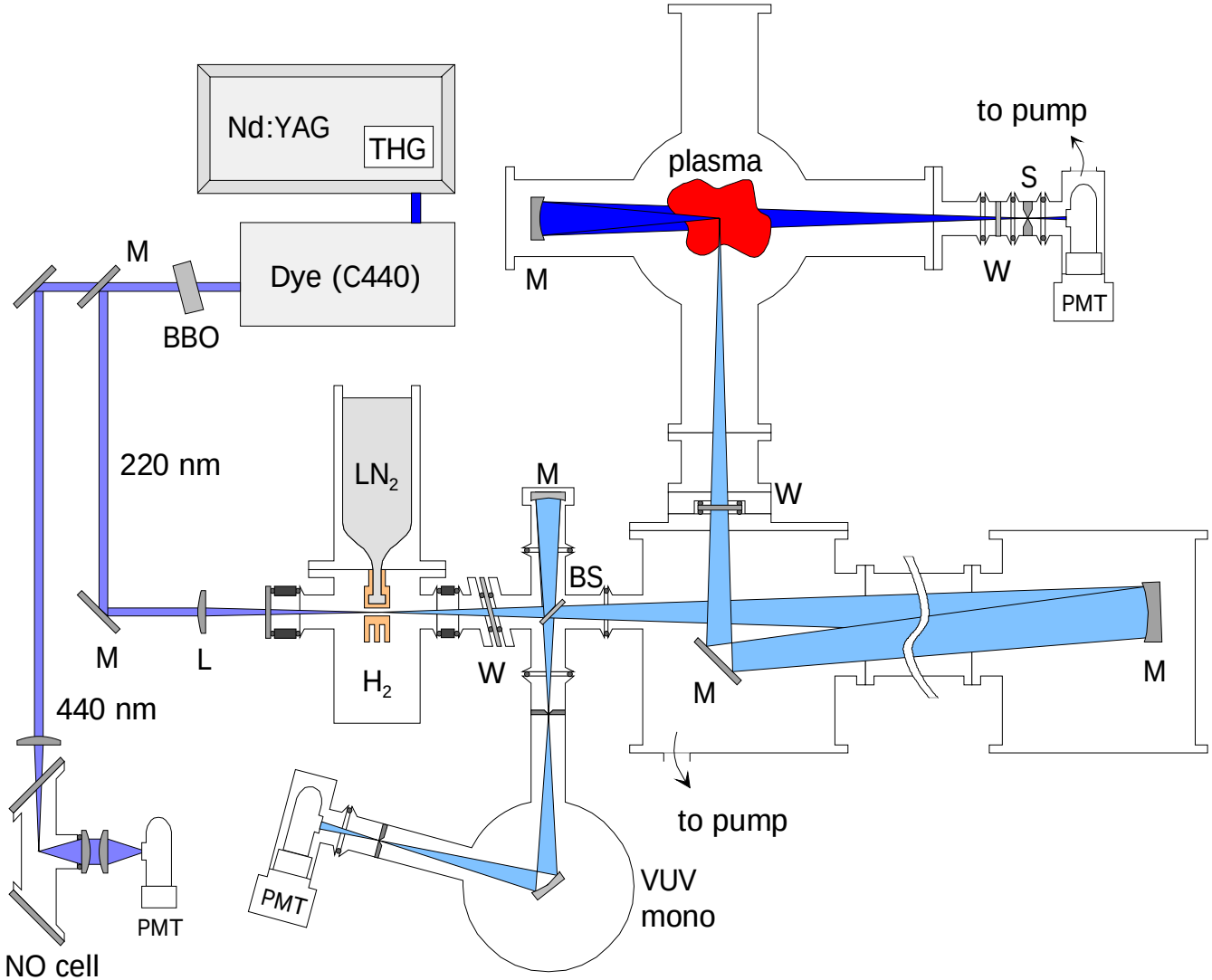




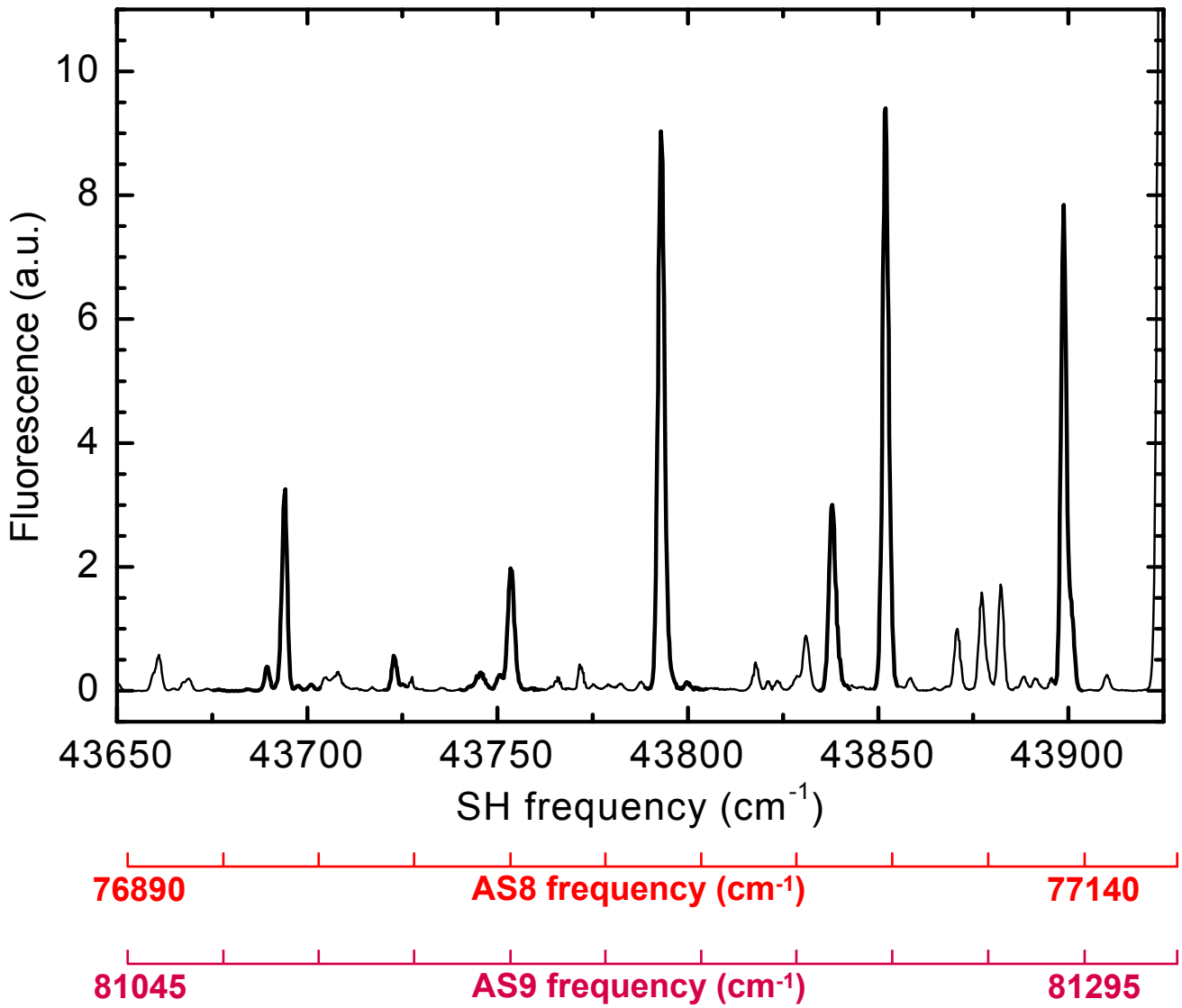
Four-wave mixing process

M. Spaan, A. Goehlich, V. Schultz-von der Gathen, H. F. Döbele, *Applied Optics* **33** (1994) 3865
T. Mosbach, H. M. Katsch, H. F. Döbele, *Rev. Sci. Instrum*. **85** (2000) 3420
P. Vankan, S.B.S. Heil, S. Mazouffre, R. Engeln and D.C. Schram, H. F. Döbele, *Rev. Sci. Instrum*. **75** (2004) 996

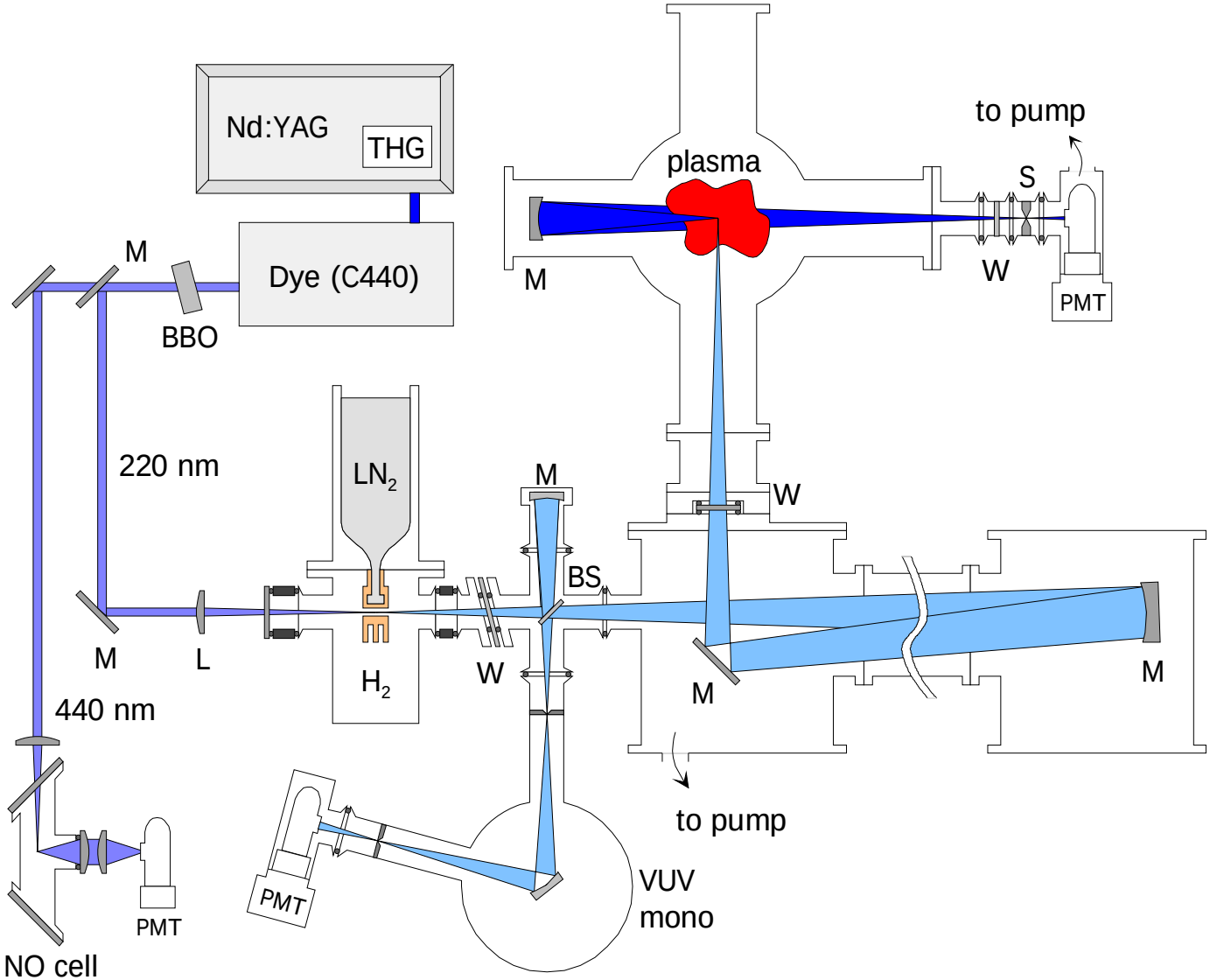
VUV-LIF setup



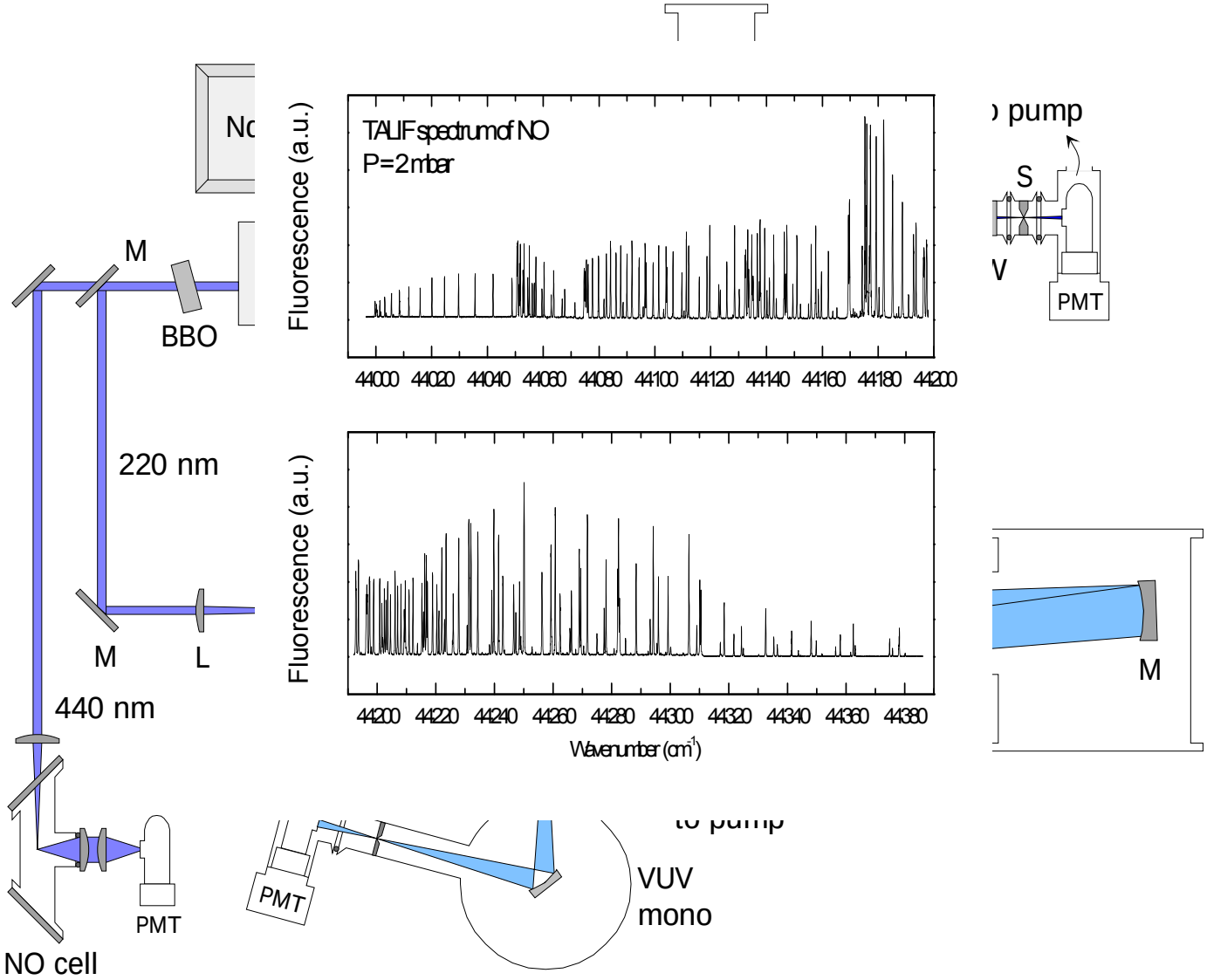
Measured H_2 Lyman spectrum



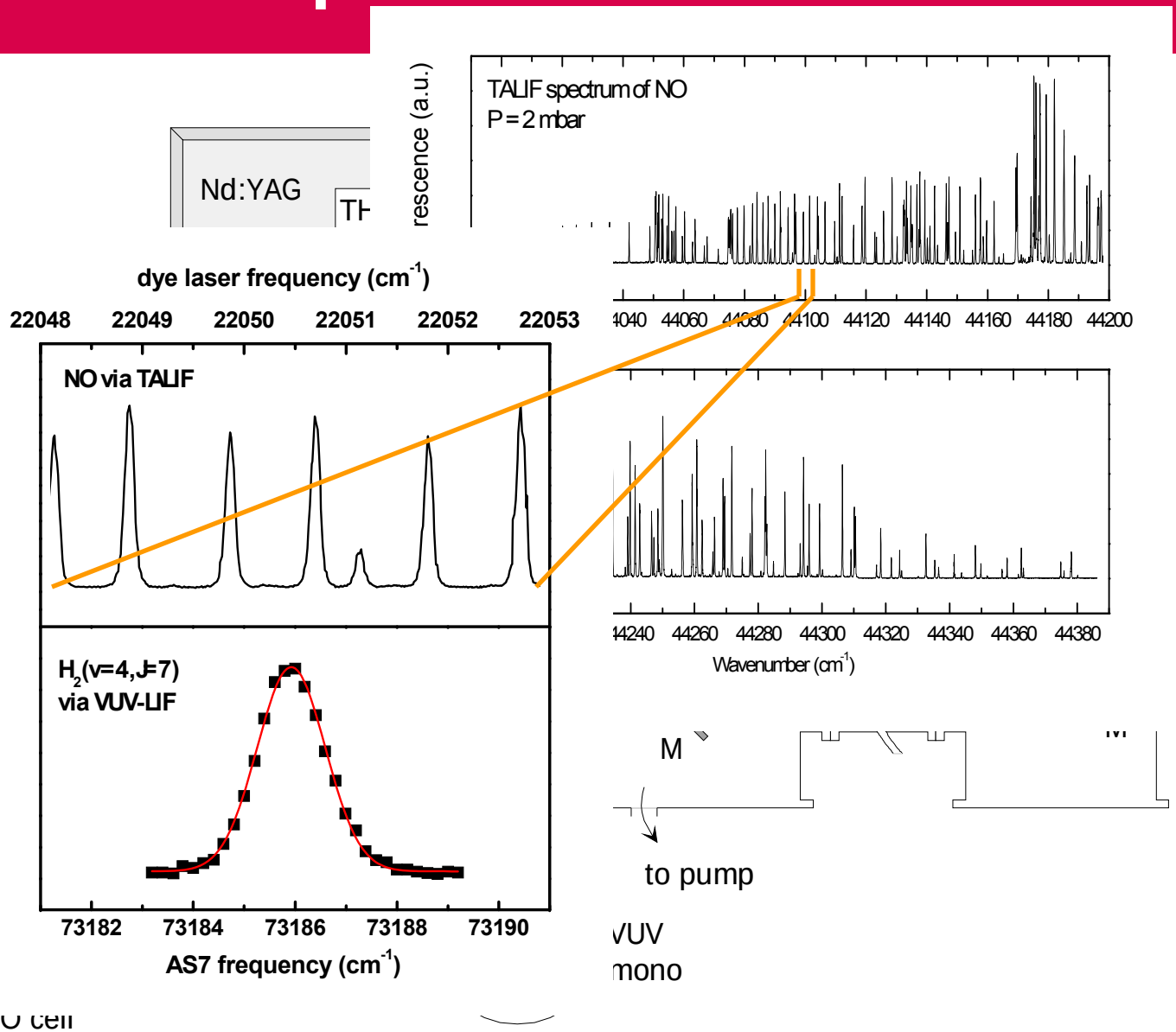
VUV LIF setup



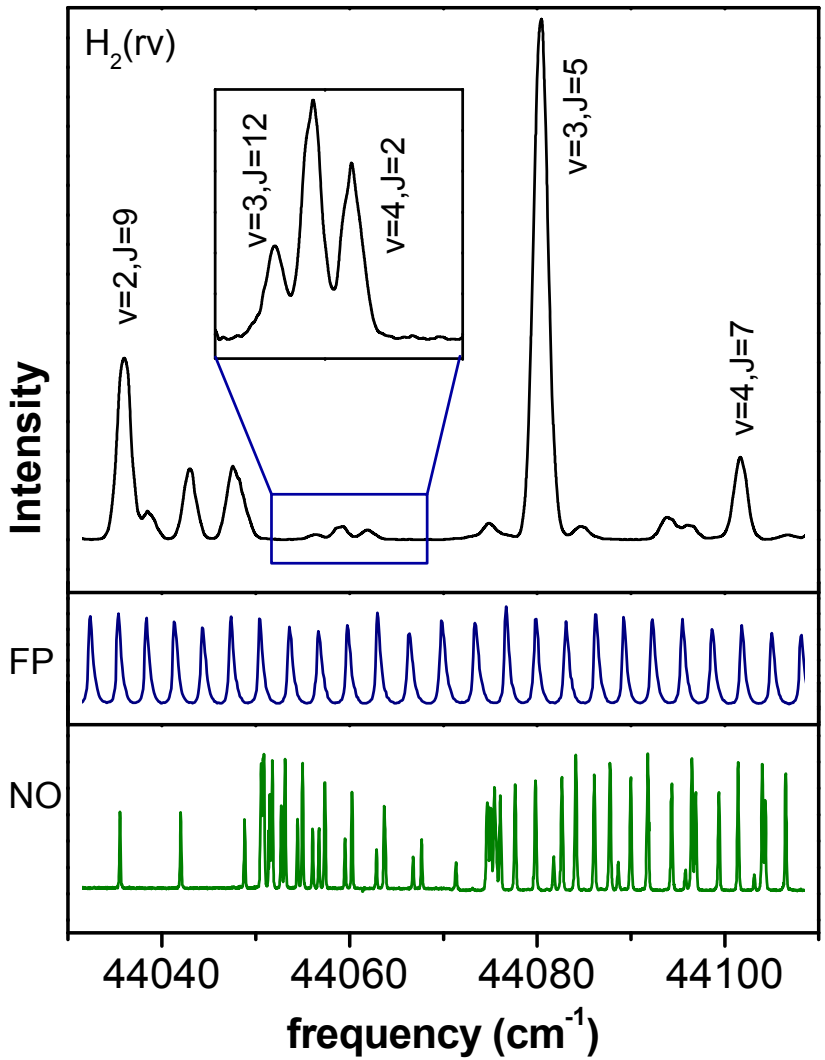
VUV LIF setup



VUV LIF setup



Measured H_2 Lyman spectrum



state-selective

spatially resolved

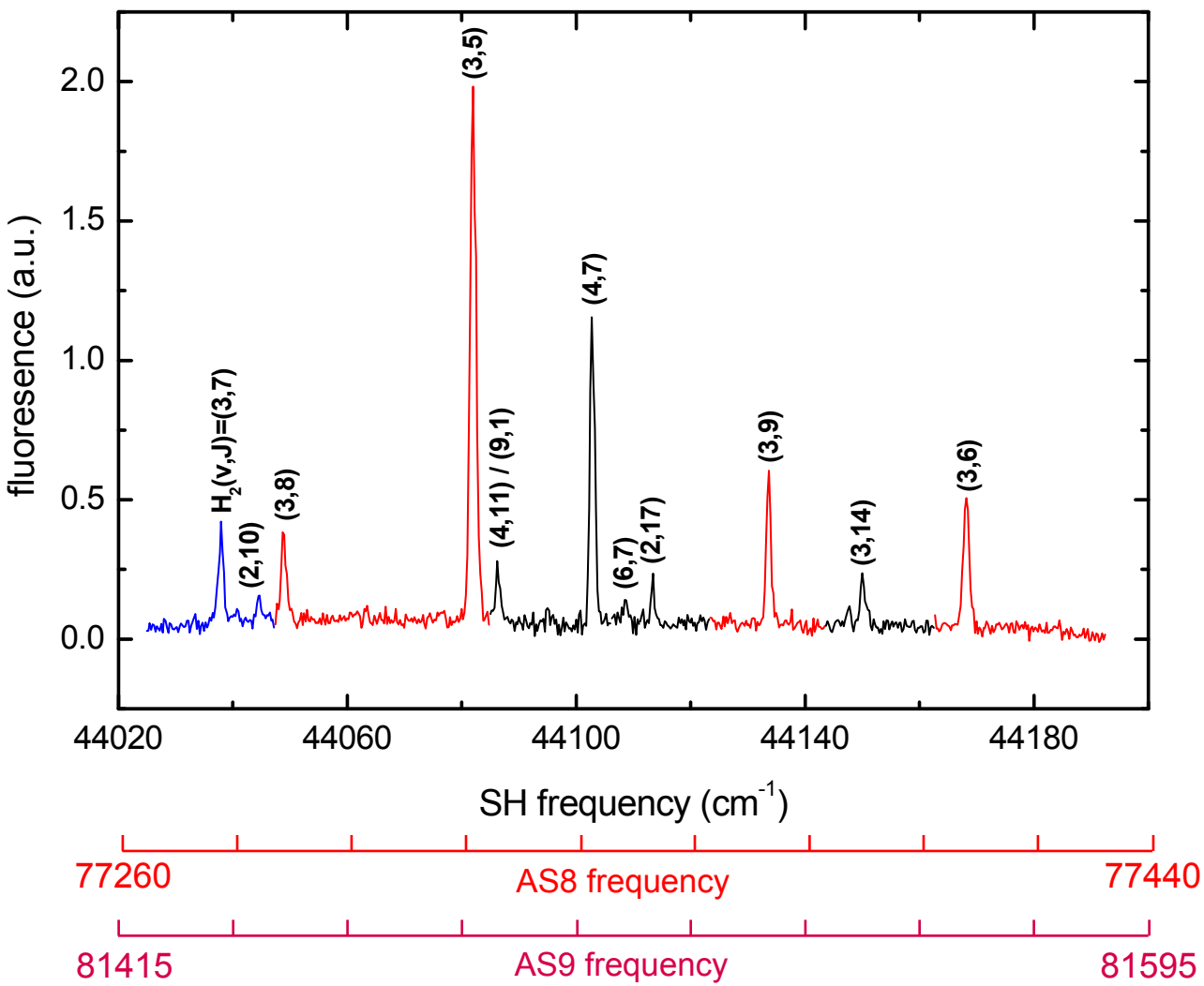
non-intrusive

dynamic range > 4 orders

detection limit ~ 10^{13} m^{-3}

P. Vankan et al., *Rev. Sci. Instrum.* **75** (2004) 996

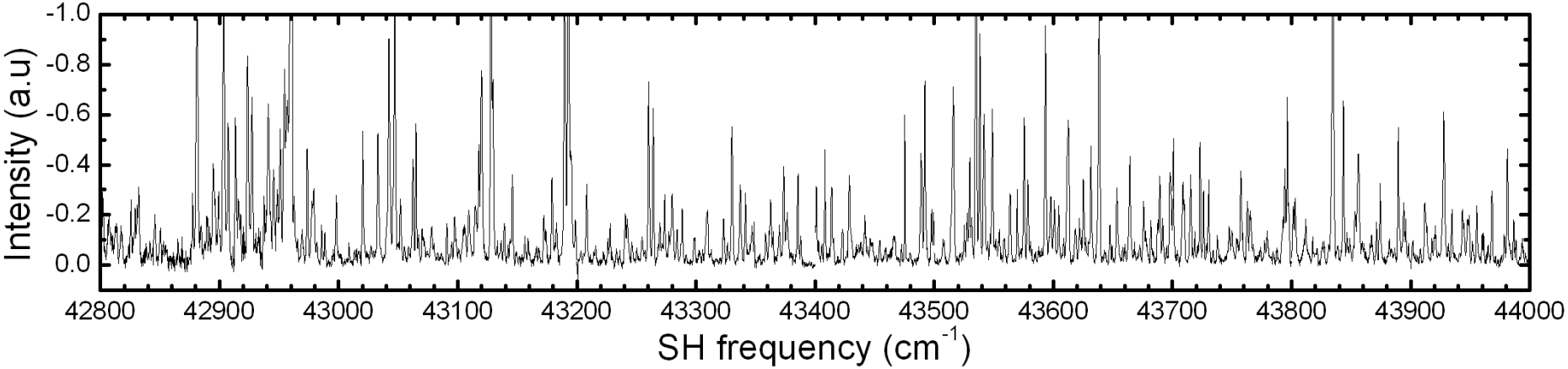
Measured H_2 Lyman spectrum

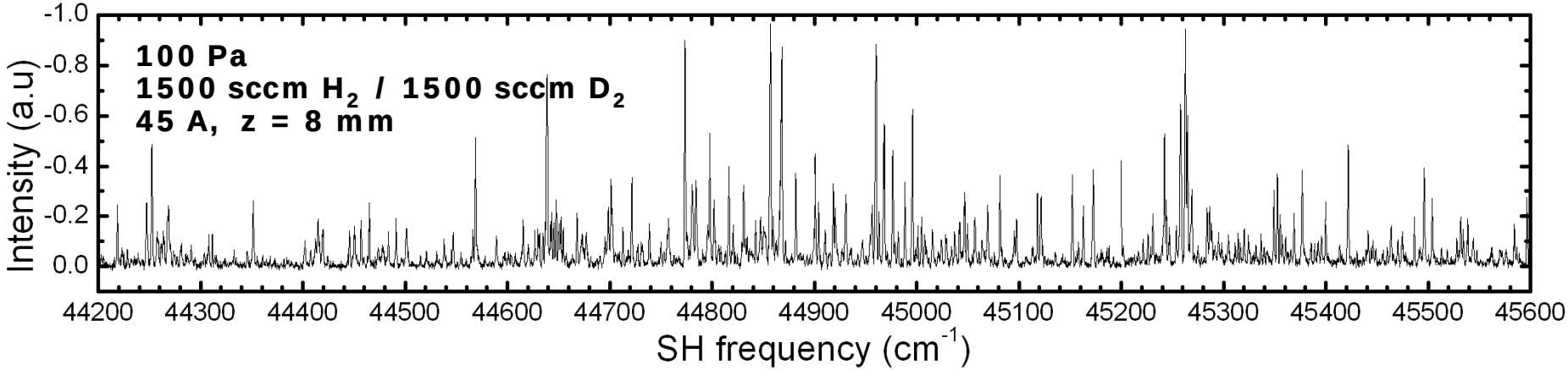


Advantages multiplexing:

- ✓ **spectrum more dense**
- ✓ **efficient measurement**

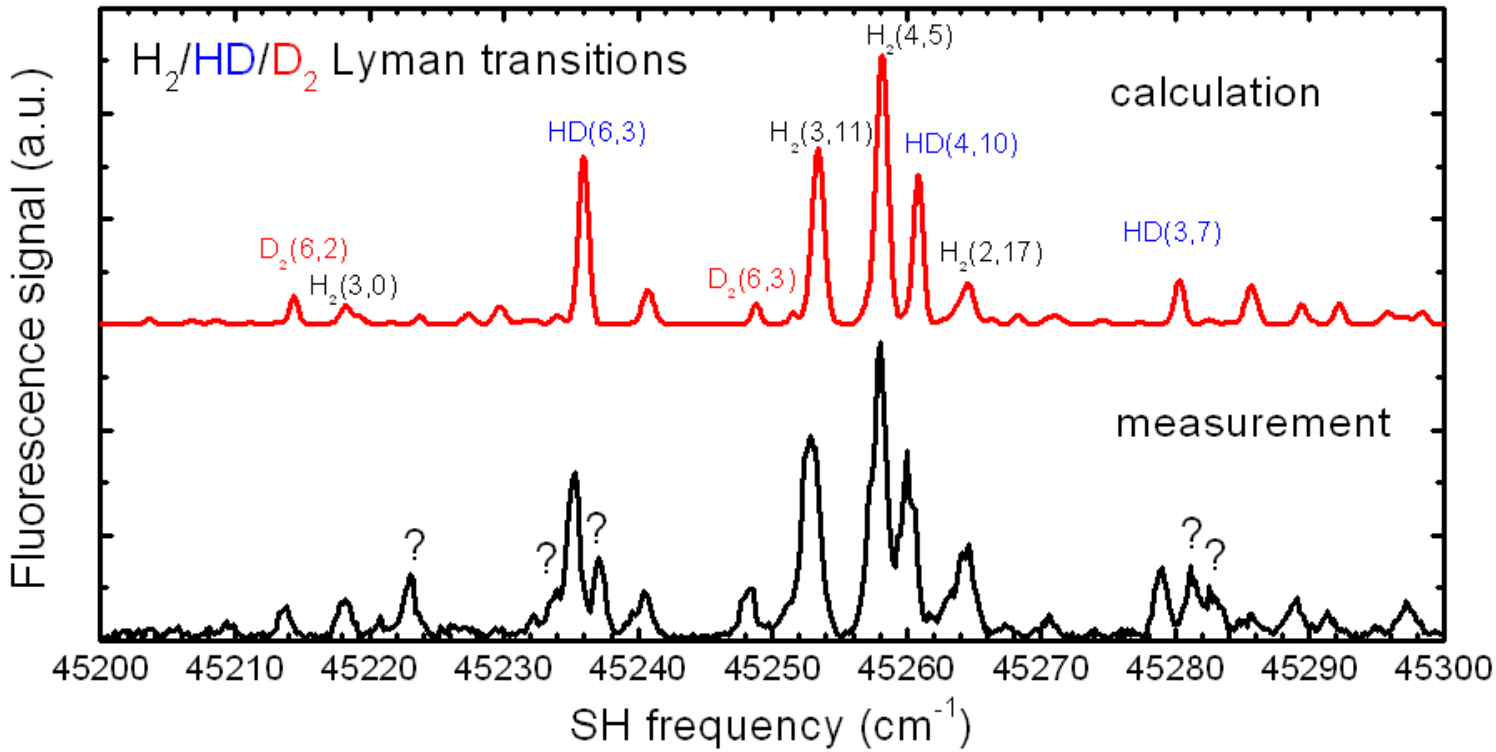
Measured H_2/HD/D_2 Lyman spectra





O. Gabriel et al. *Chemical Physics Letters* 451 (2008) 204

Measured and calculated Lyman spectra



Spectroscopic data for H_2

H. Abgrall et al. *Astron. Astrophys. Suppl. Ser.* 101 (1993) 273

All H_2 Lyman transitions

Spectroscopic data for HD

H. Abgrall, E. Roueff, *Astron. Astrophys.* 445 (2006) 361

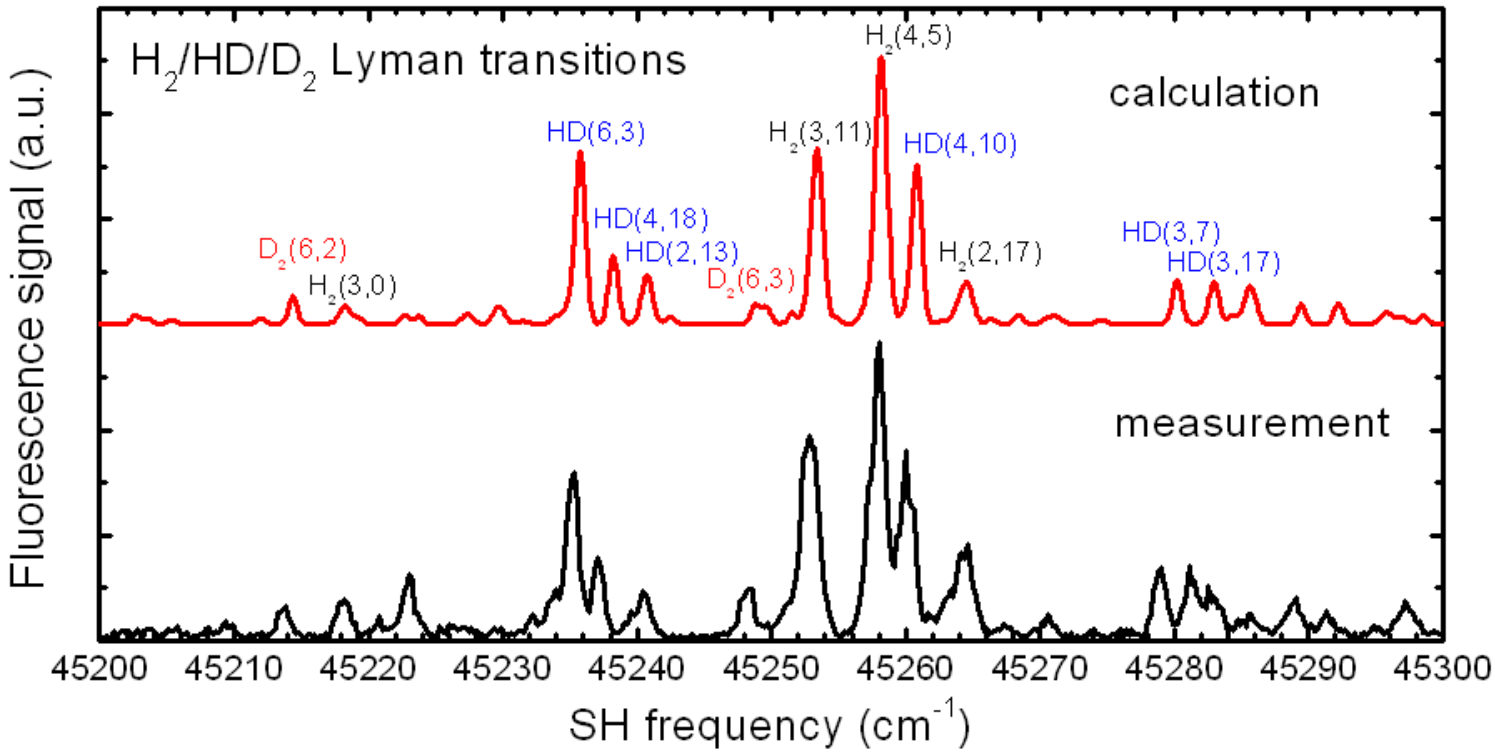
HD Lyman transitions J < 11

Spectroscopic data for D_2

H. Abgrall et al. *J. Phys. B: At., Mol. Opt. Phys.* 32 (1999) 3813

D_2 Lyman transitions J < 12

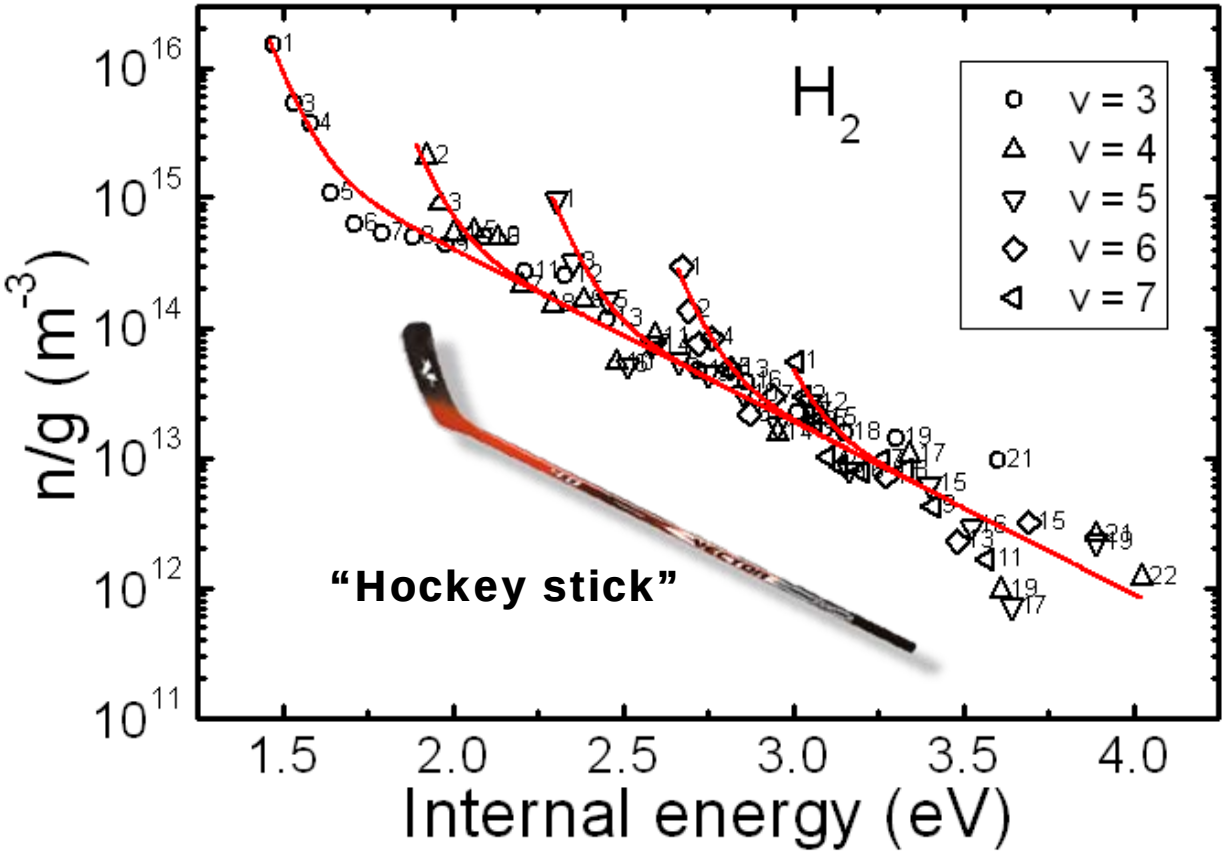
Measured and calculated Lyman spectra



New calculated Lyman transitions including higher rotational states (J > 10), Abgrall/Roueff, private communication

O. Gabriel et al. *J. Mol. Spectrosc* ***253*** *(2009) 64*

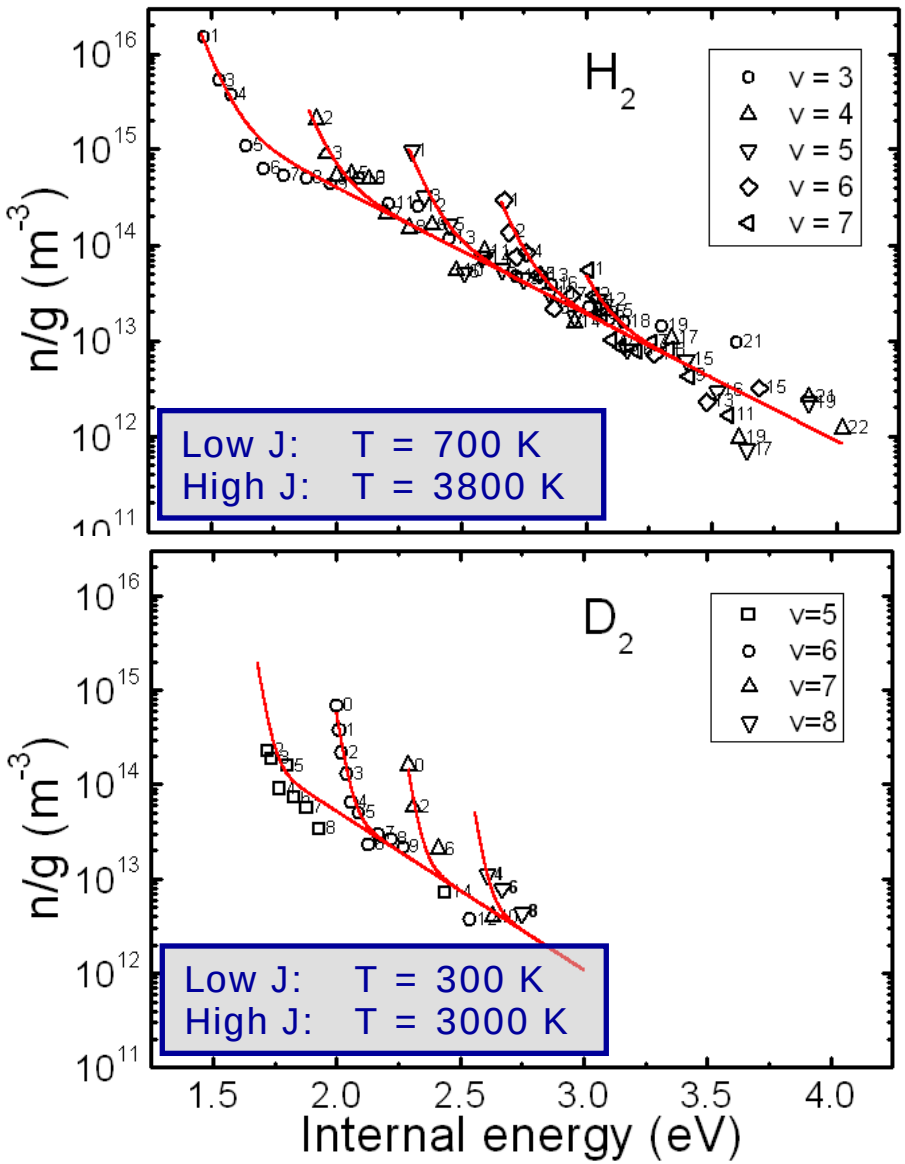
Non-Boltzmann distribution for H_2

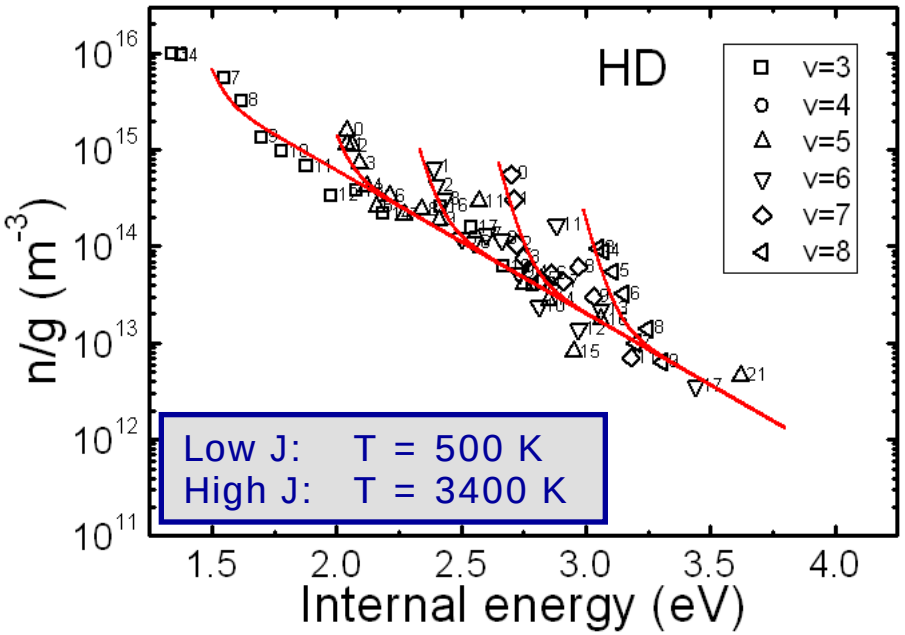


700 K for low J

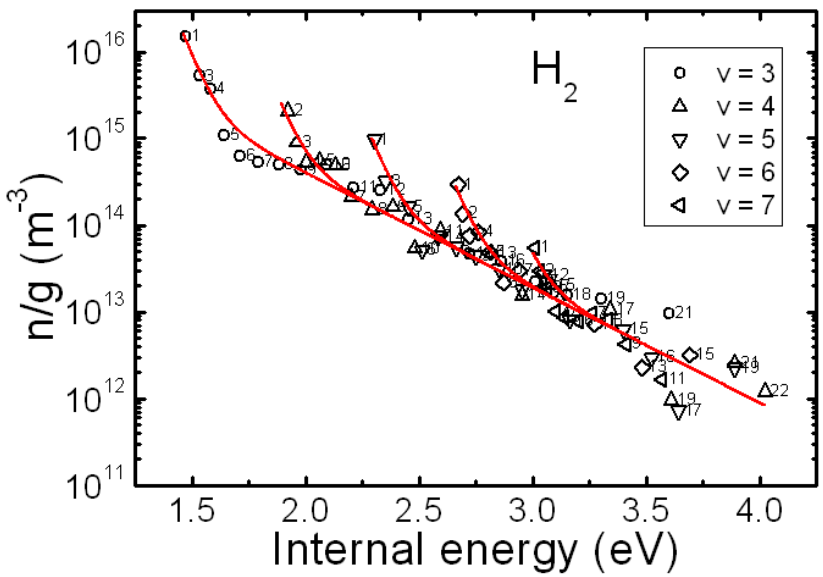
3800 K for high J

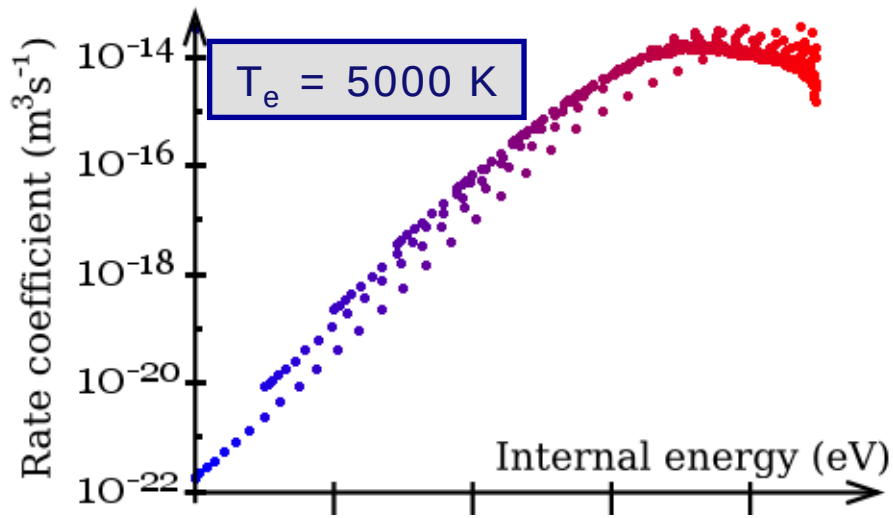
Non-Boltzmann distributions in H_2/D_2 jet

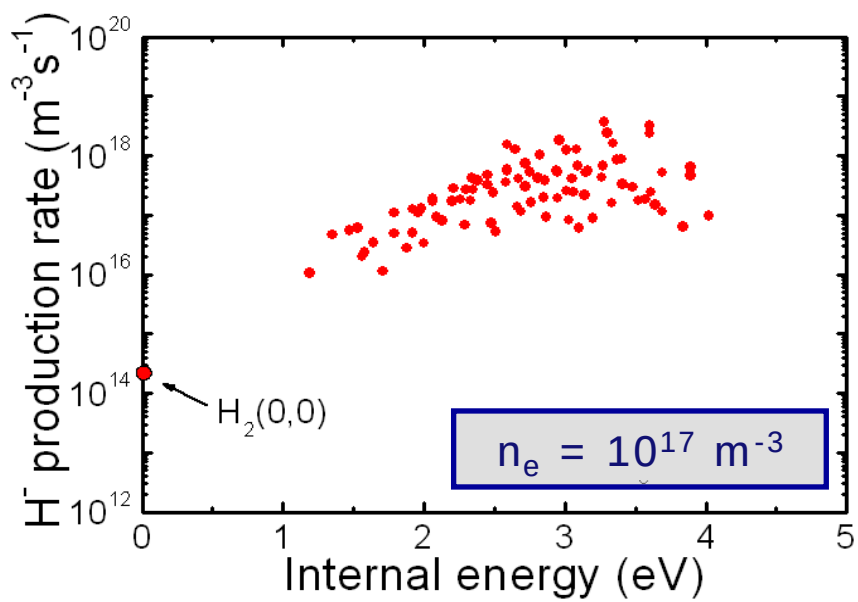




Results on H^- production



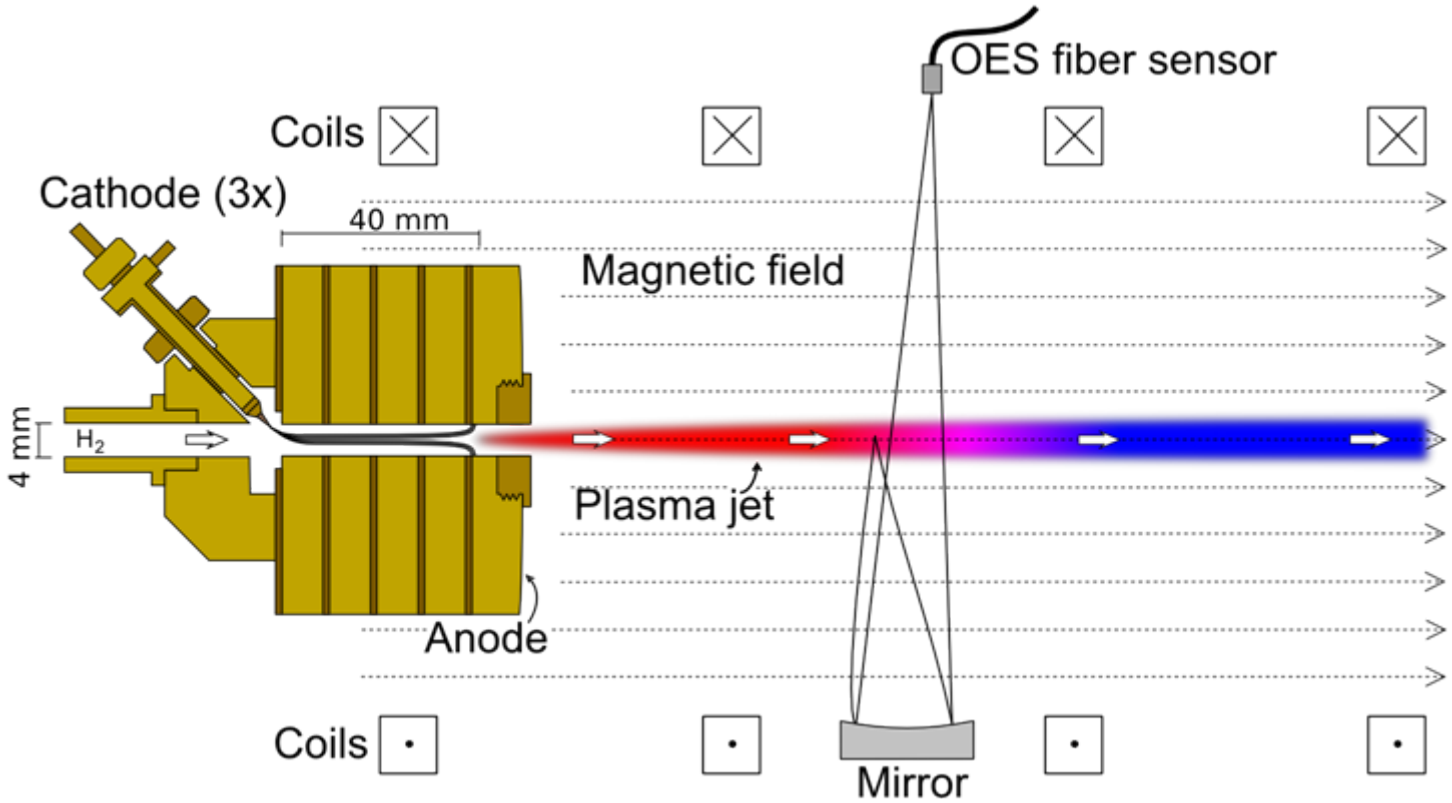




Production rate of H^- ions by dissociative attachment

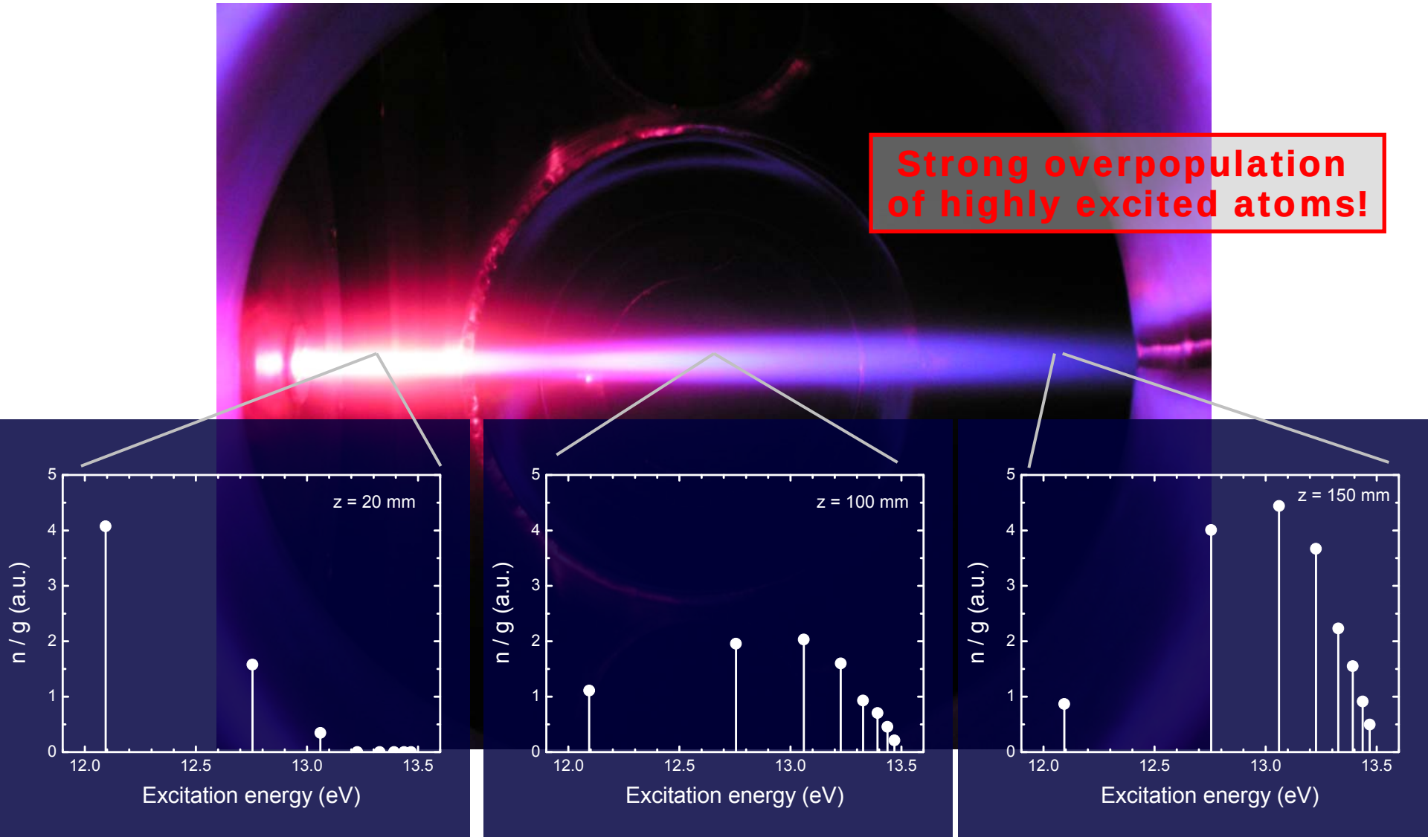
J. Horáček et al, *Phys. Rev. A*, 70 (2004) 052712

Effect of magnetic field on ETP



Background pressure: 7 … 300 Pa
Gas flow through arc: 1000 … 5000 sccm
Arc power: 5 … 10 kW
Magnetic field: 0 … 160 mT

Effect of magnetic field on ETP



Excited atom formation

Three body recombination:

$H^+ + e + e \rightarrow H(n) + e$ (n high)

Ion-ion recombination:

$H^+ + H^- \rightarrow H + H(n)$ (n = 2,3)

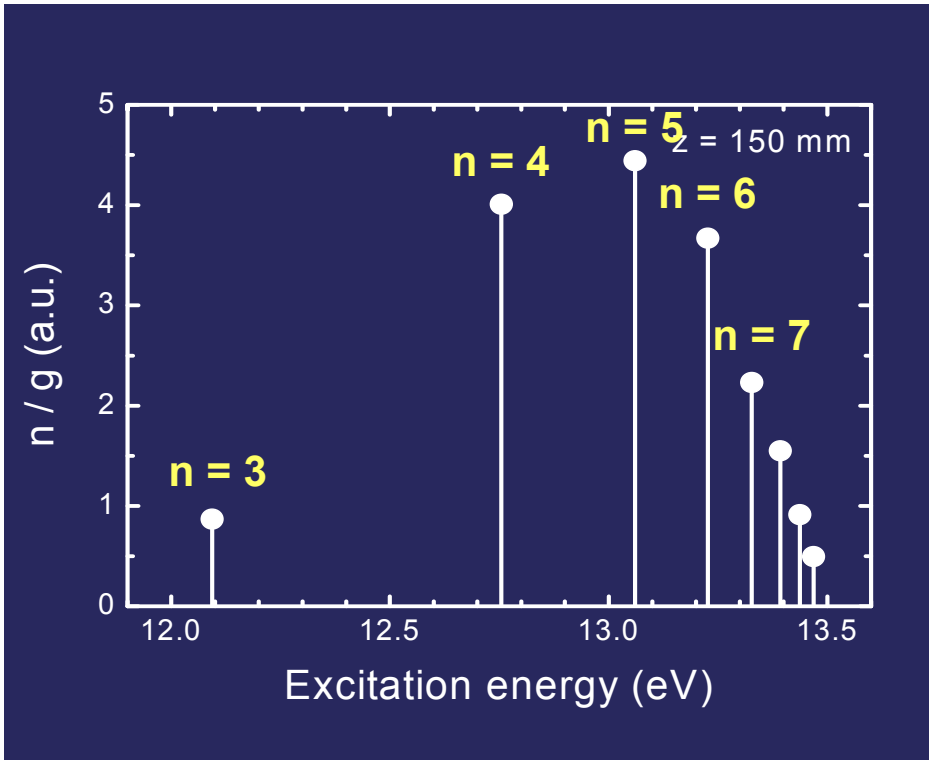
Molecular ion recombination:

$H_2^+ + e \rightarrow H + H(n)$ ($n \leq 2,3$)

Mutual recombination:

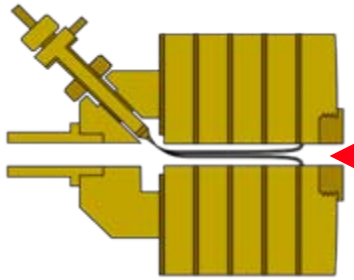
$H_2^+ + H^- \rightarrow H + H(n)$ ($n \leq 5,6$?)

$H_3^+ + H^- \rightarrow H + H + H$ (excitation(?))



How to proof importance of H^- ?

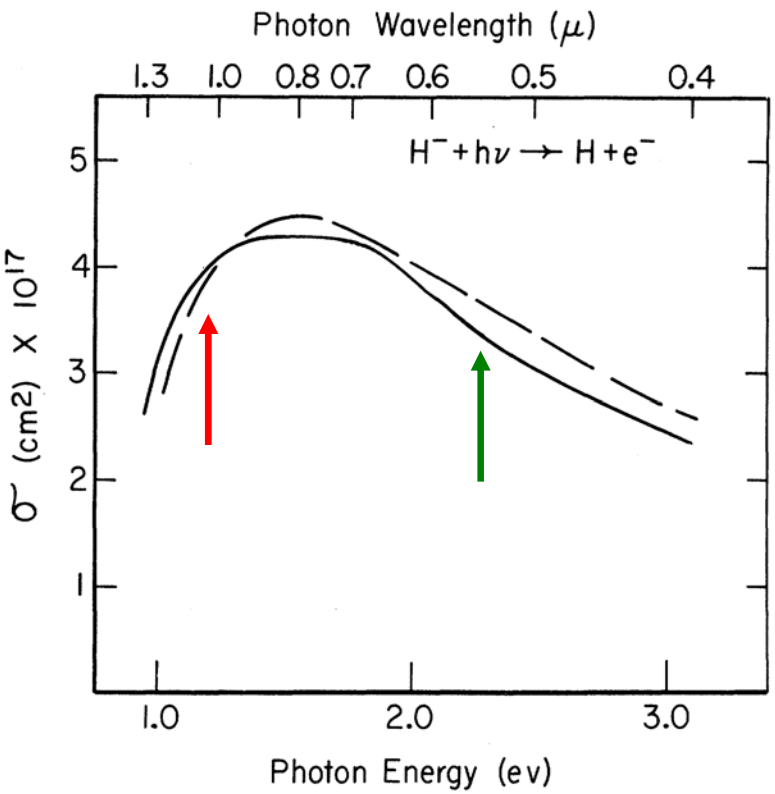
Photo-detachment



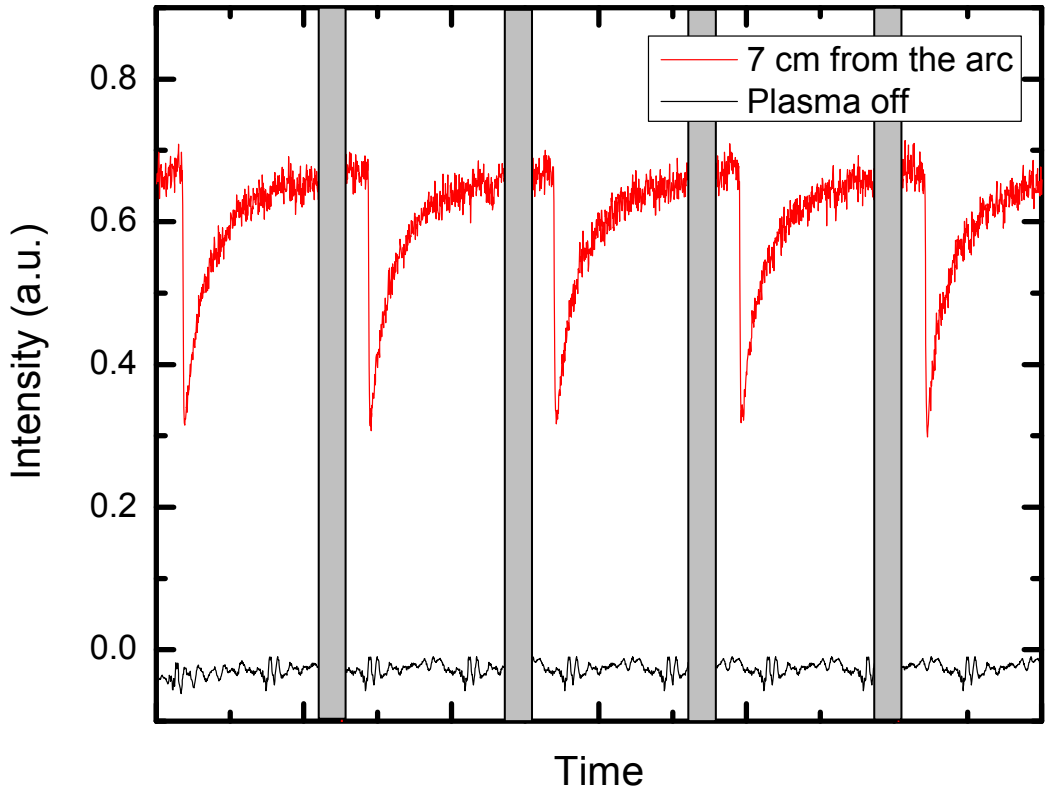
$H^- + h\nu \rightarrow H + e$

emission spectrograph

Photo detachment at 1064 nm and 532 nm



H_α emission



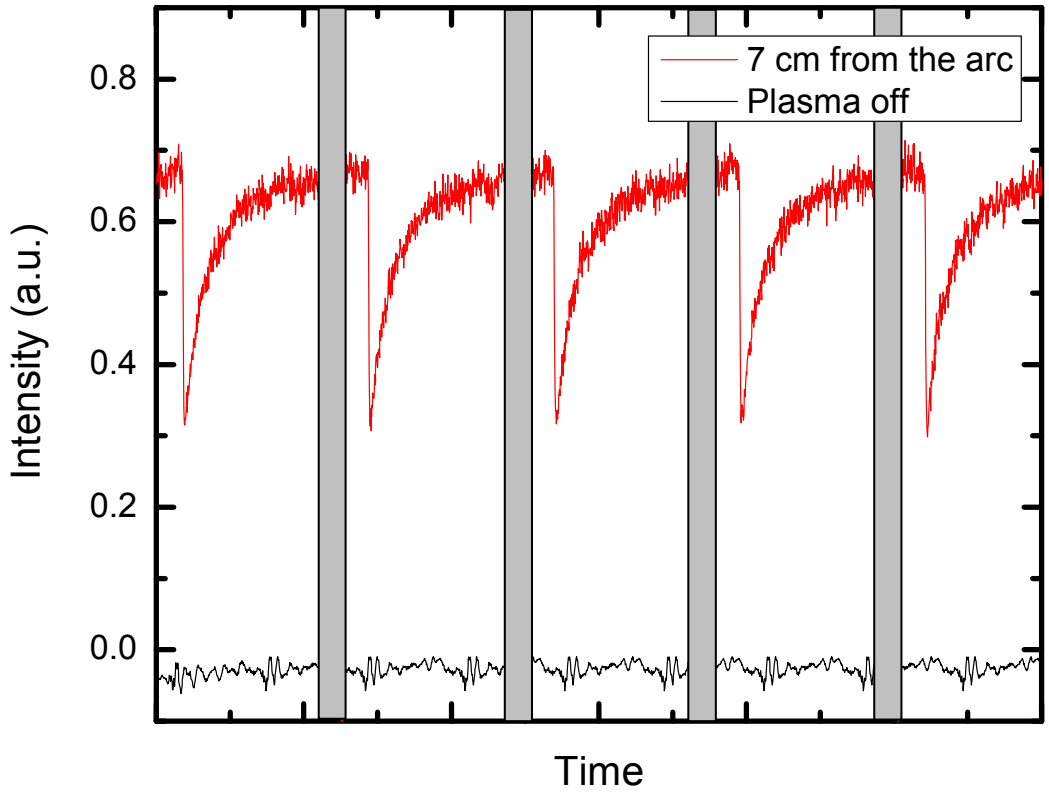
Loss of H^-

(due to detachment laser)

Less production H(n=3)

(via H^+ + H^-)

H_α emission

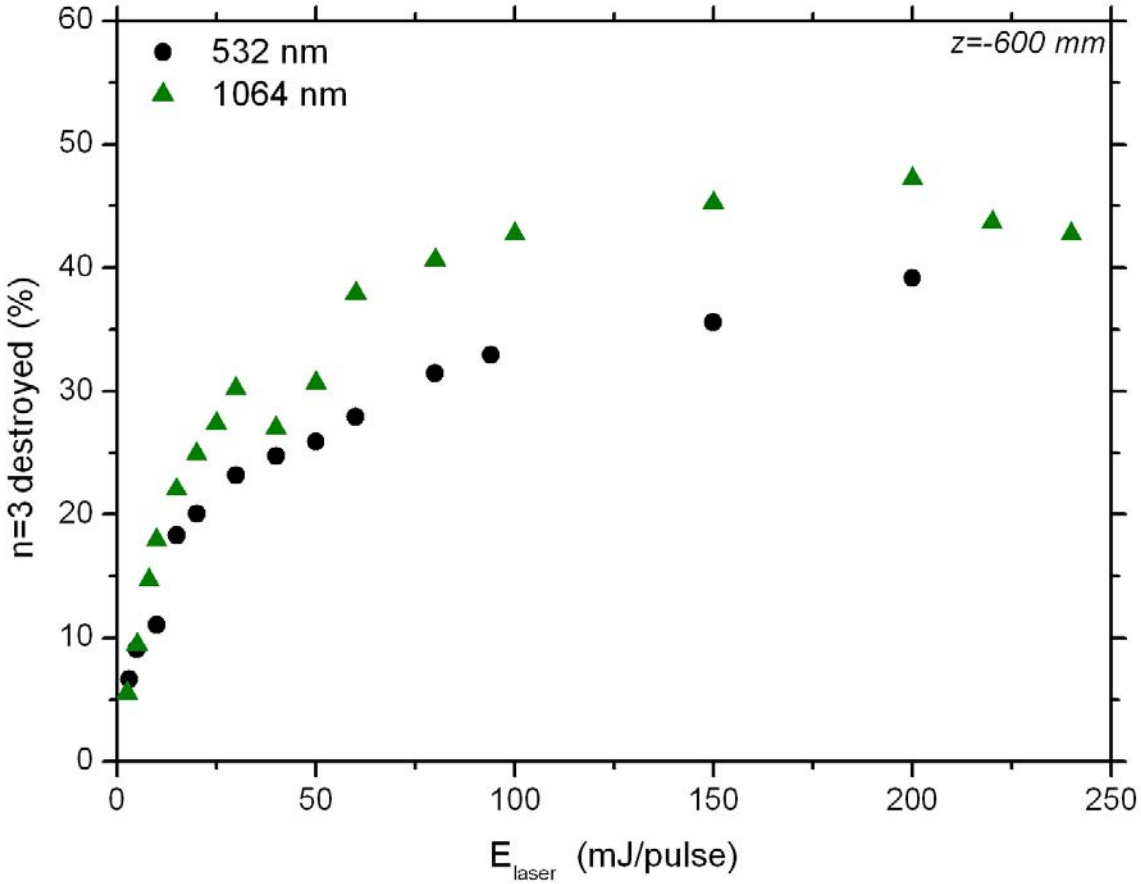


Due to detachment laser

More destruction H(n=3)

(due to photo-ionization)

Photo-detachment of H^- (at 1064 nm and 532 nm)



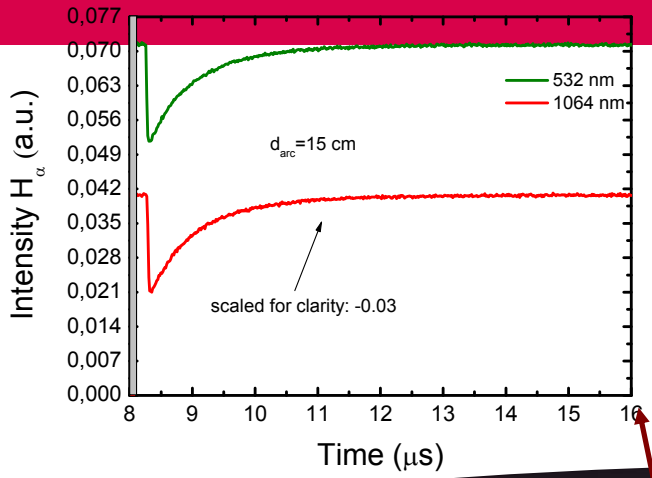
Cross-section of photo-detachment:

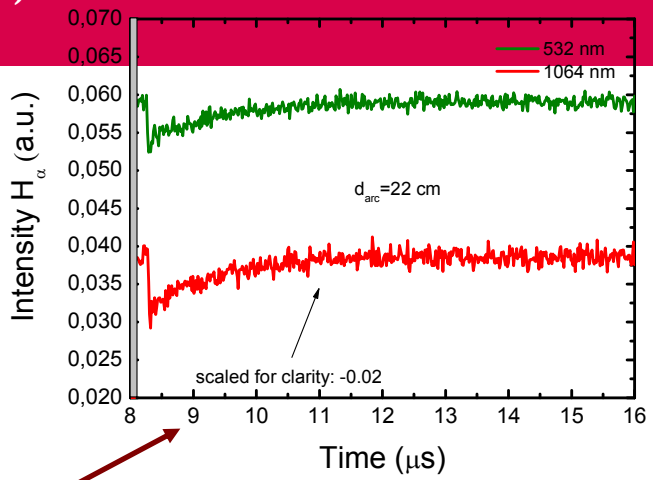
$3.8\ 10^{-21}$ m^2 at 1064 nm

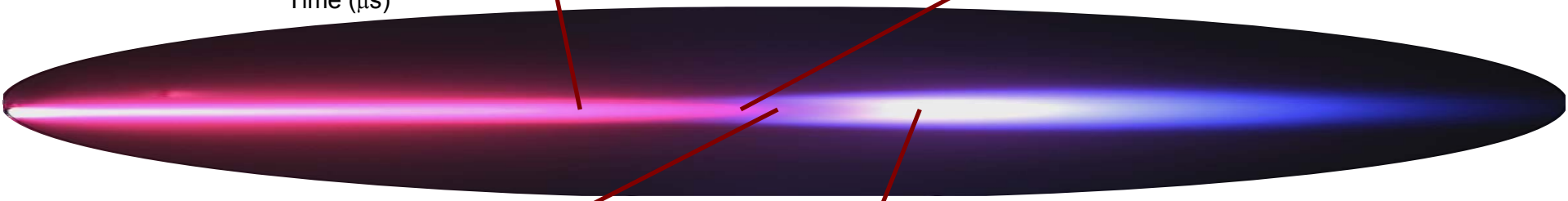
$3.3\ 10^{-21}$ m^2 at 532 nm

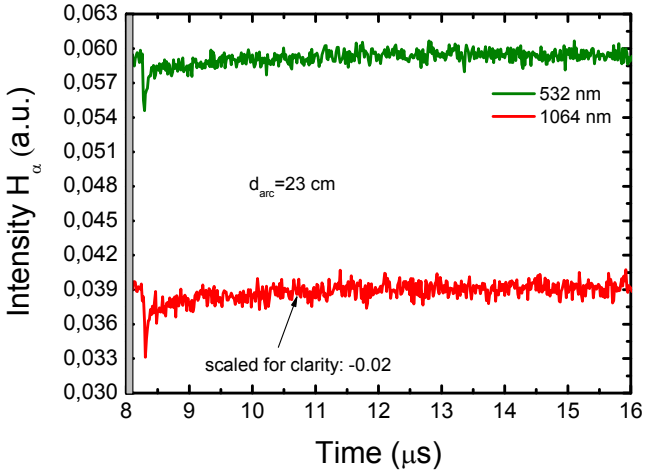
Photo-ionization not (very) important

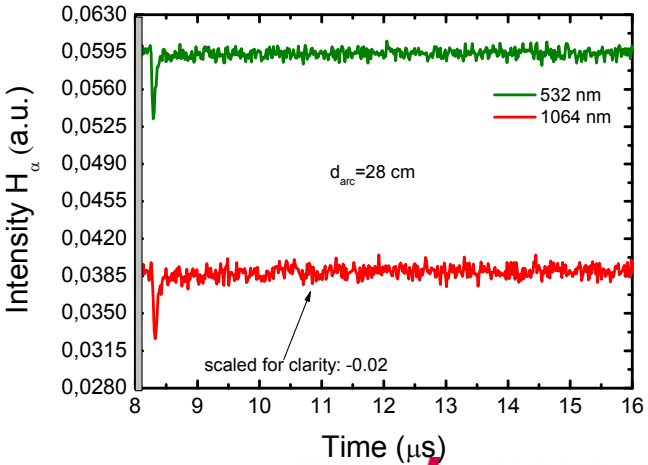
Experiments (Axial photo-detachment p=3)





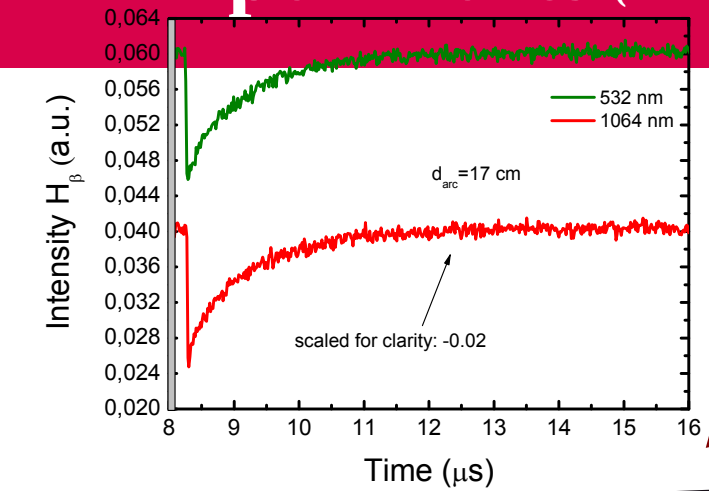


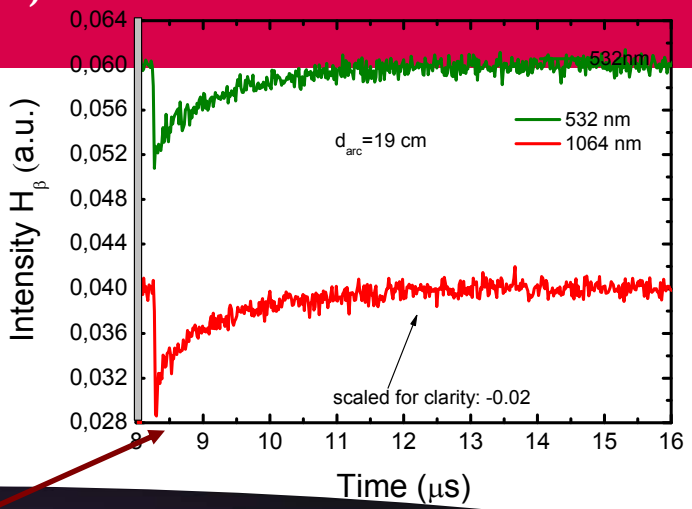


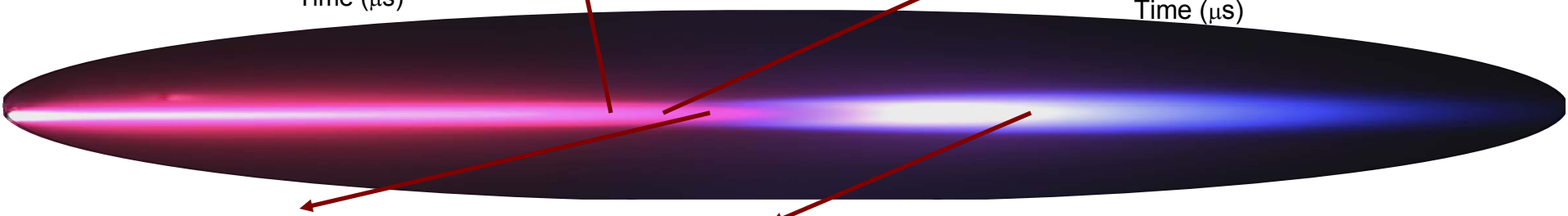


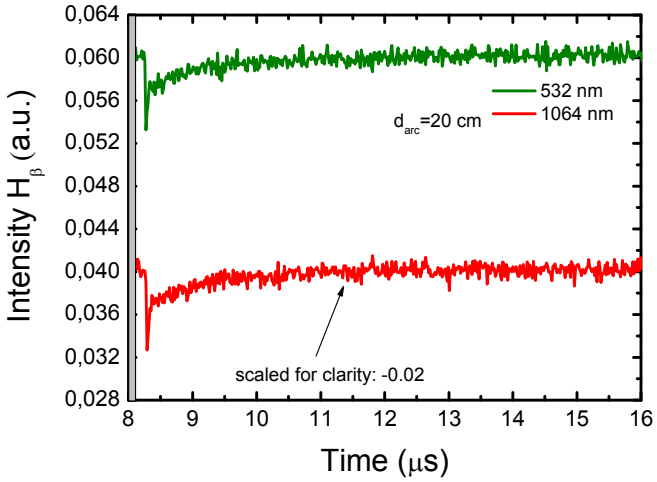
Laser: 1064 and 532 nm
Detection on Hα
P_{laser} = 100 mJ

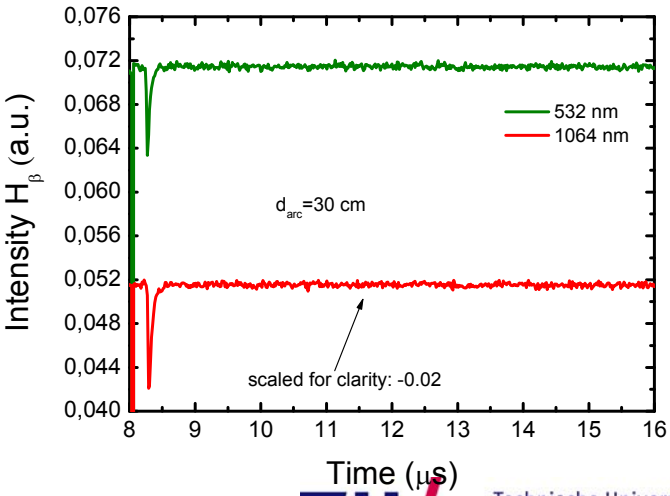
Experiments (Axial photo-detachment p=4)





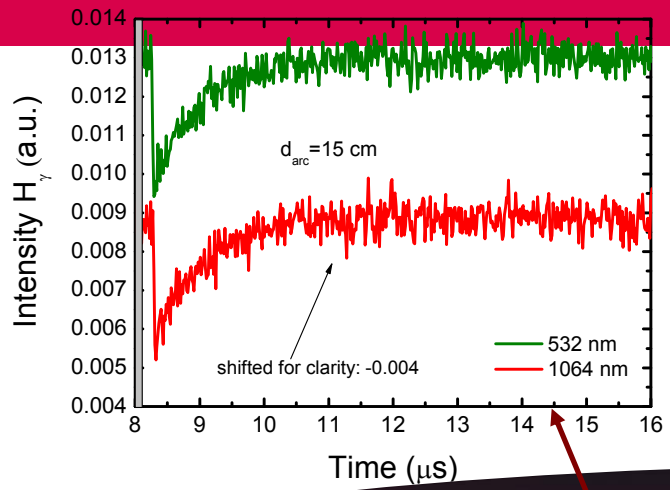


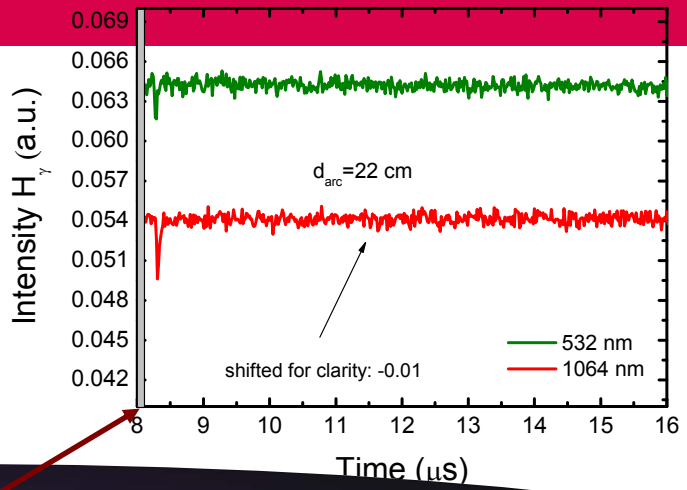


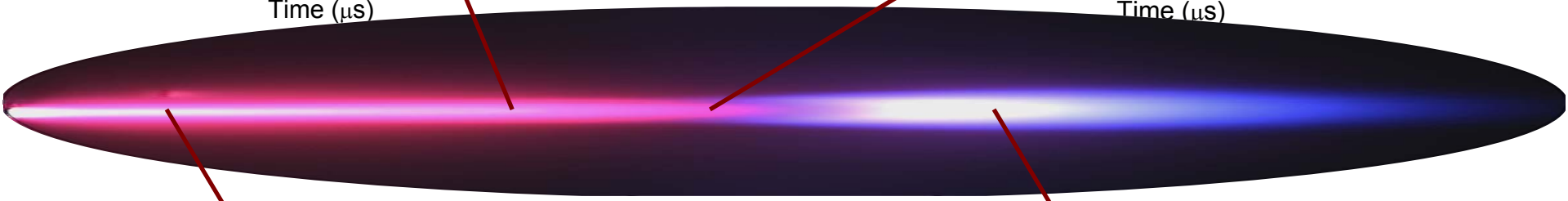


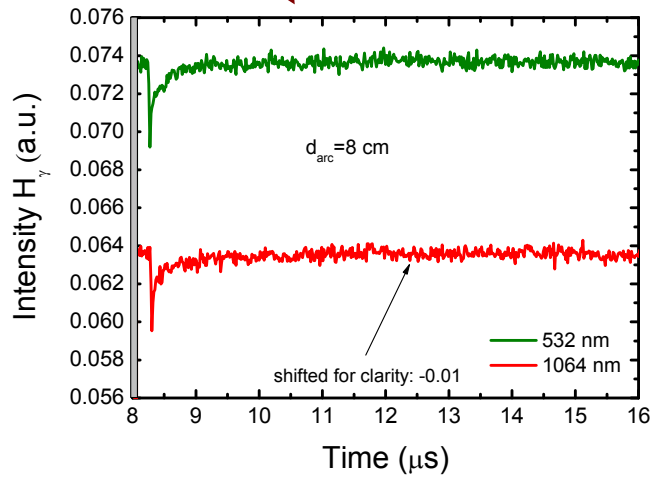
Laser: 1064 and 532 nm
Detection on Hβ
P_{laser} = 100 mJ

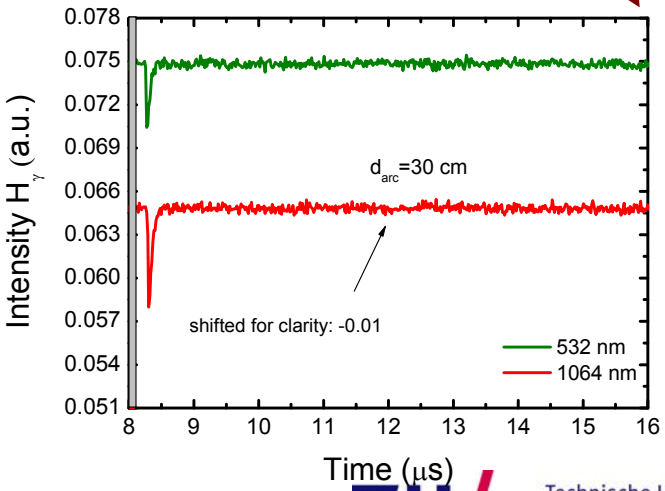
Experiments (Axial photo-detachment p=5)





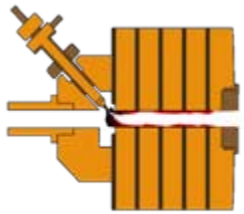






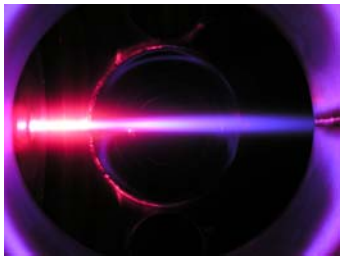
Laser: 1064 and 532 nm
Detection on Hγ
P_{laser} = 100 mJ

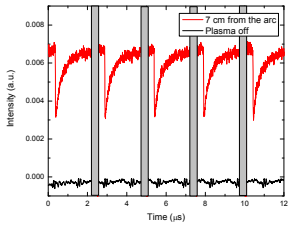
Conclusions



The cascaded arc is an efficient $H_2^{r,v}$ $D_2^{r,v}$ and $HD^{r,v}$ source

The red to blue transition in the plasma expansion indicates H^-





Indirect proof of presence of H^- through decrease in Balmer-series emission after detachment (but: photo-ionization might be important, and modelling is necessary)

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