Reaction dynamics in a magnetized hydrogen plasma unraveled by optical spectroscopic techniques

Richard Engeln

Plasma and Materials Processing

PMP PLASMA & MATERIALS PROCESSING

TU/e Technische Universiteit Eindhoven University of Technology

IAEA technical meeting, March 2021 (webex)

Where innovation starts

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

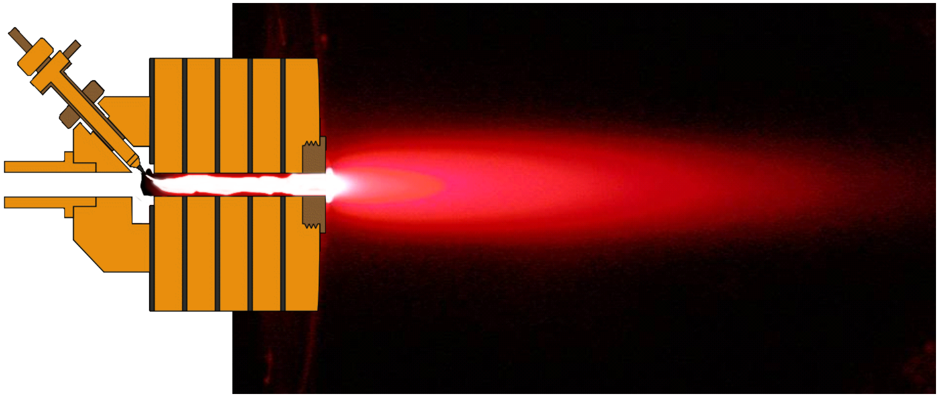
1. Use of H_2 gas in processing plasma application
- etching and cleaning
- passivation during deposition

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 $H_2^{r,v}$ at low T_e (around 1 eV)

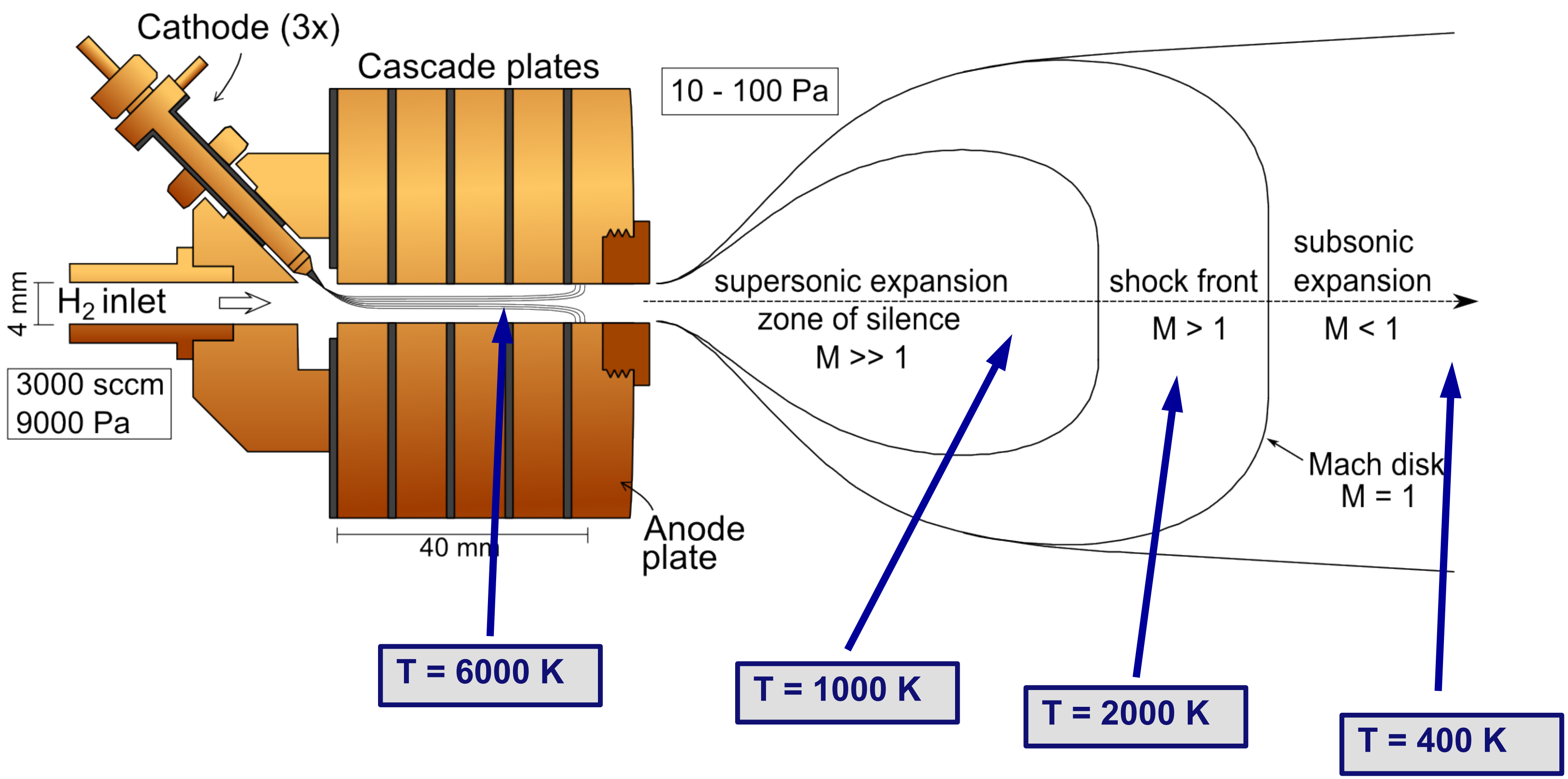
Plasma source and expansion



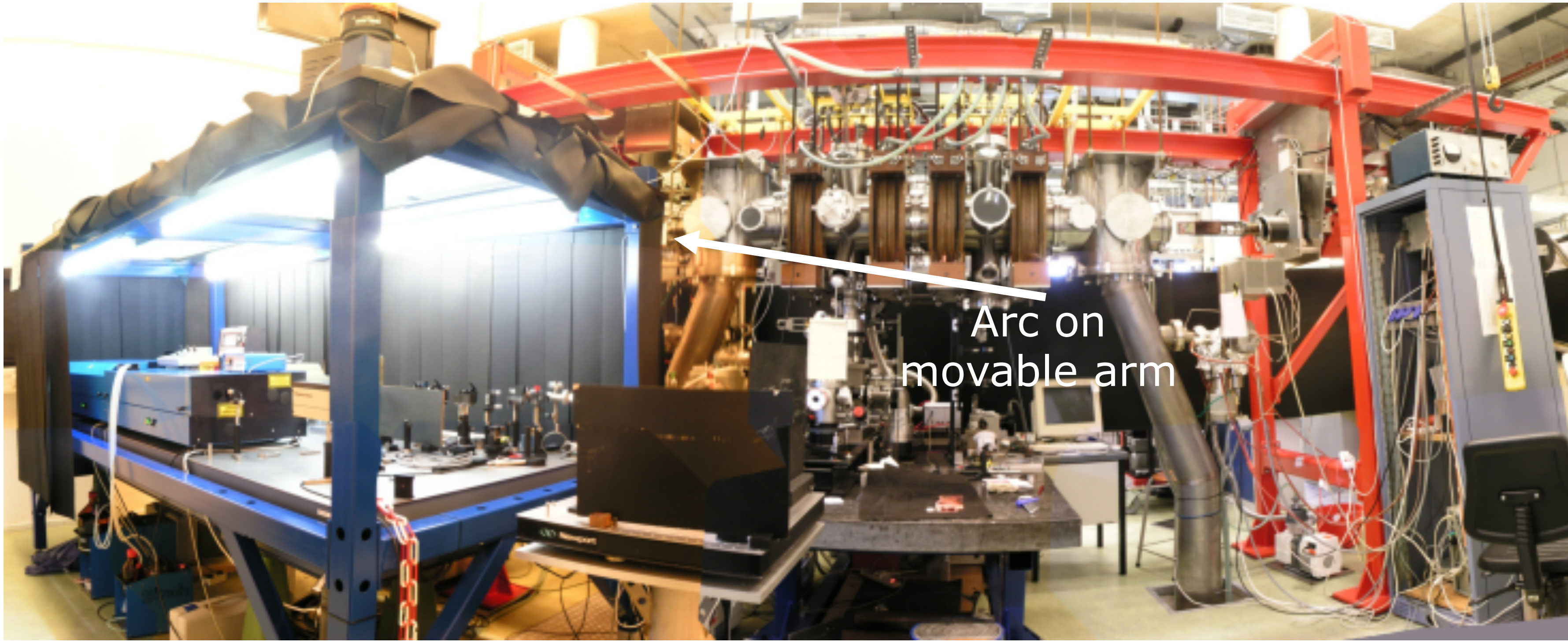
I	$= 40 – 60$ A
P	$= 5 – 10$ kW
Φ_{arc}	$= 3$ slm

p_{arc}	$= 0.2 \times 10^5$ Pa
p_{bg}	$= 100$ Pa

Plasma expansion



PLEXIS setup



Laser table
Nd:YAG
(450 mJ/shot @ 355 nm)
dye laser
(50 mJ/shot @ 460 nm)
(8 mJ/shot @ 230 nm)

Vacuum chamber
cylindrical (2m x 0.3m)
9 Pa / 3000 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. Optical emission and laser absorption spectroscopy

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

3. VUV-LIF

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

4. Langmuir double probe measurements

- n_e, T_e (for $z > z_{shock}$)

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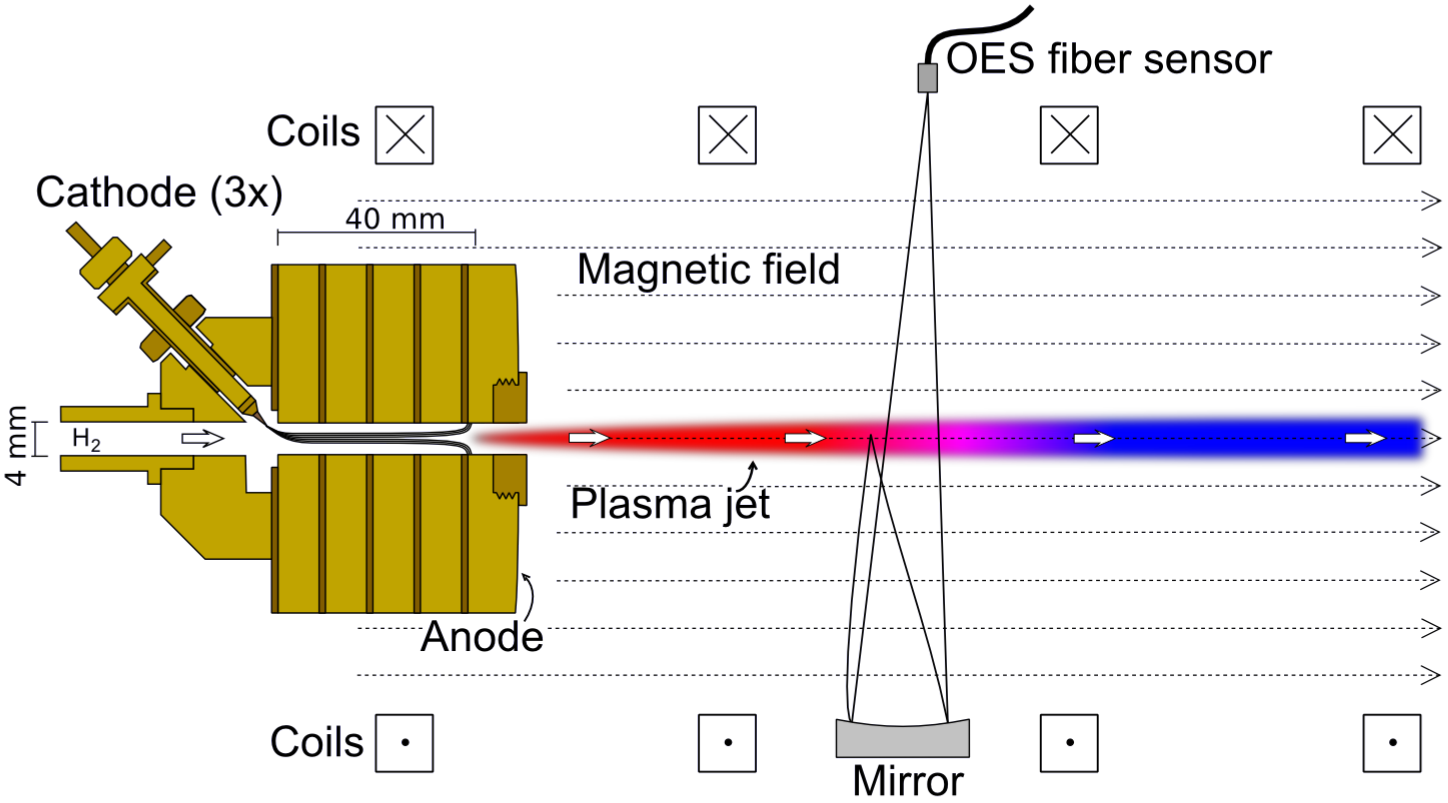
Effect of magnetic field on ETP



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

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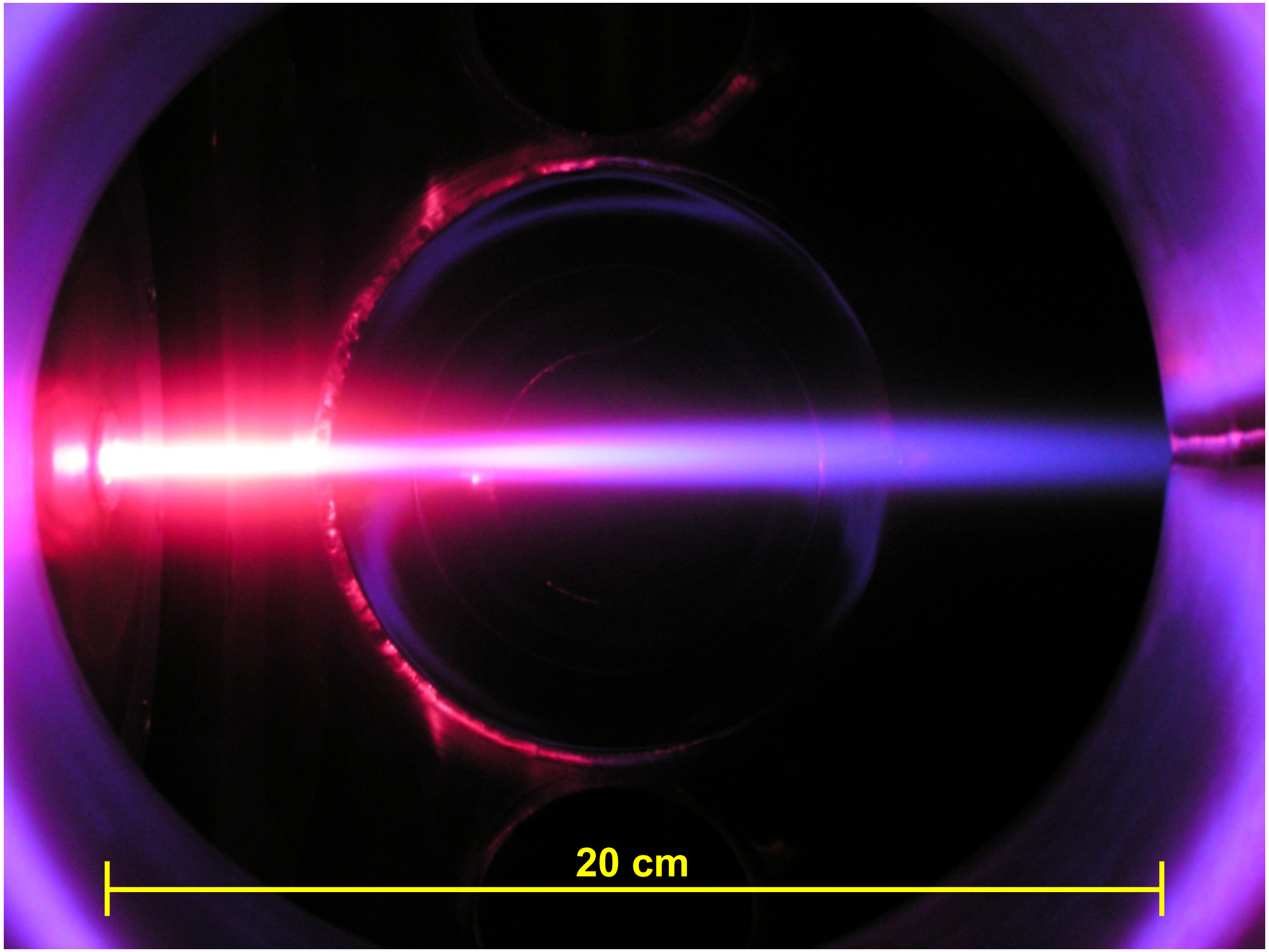
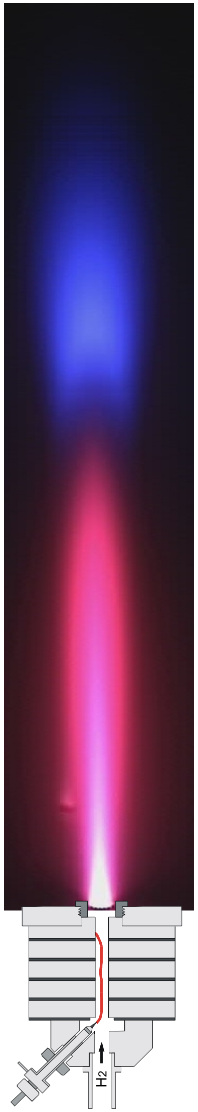
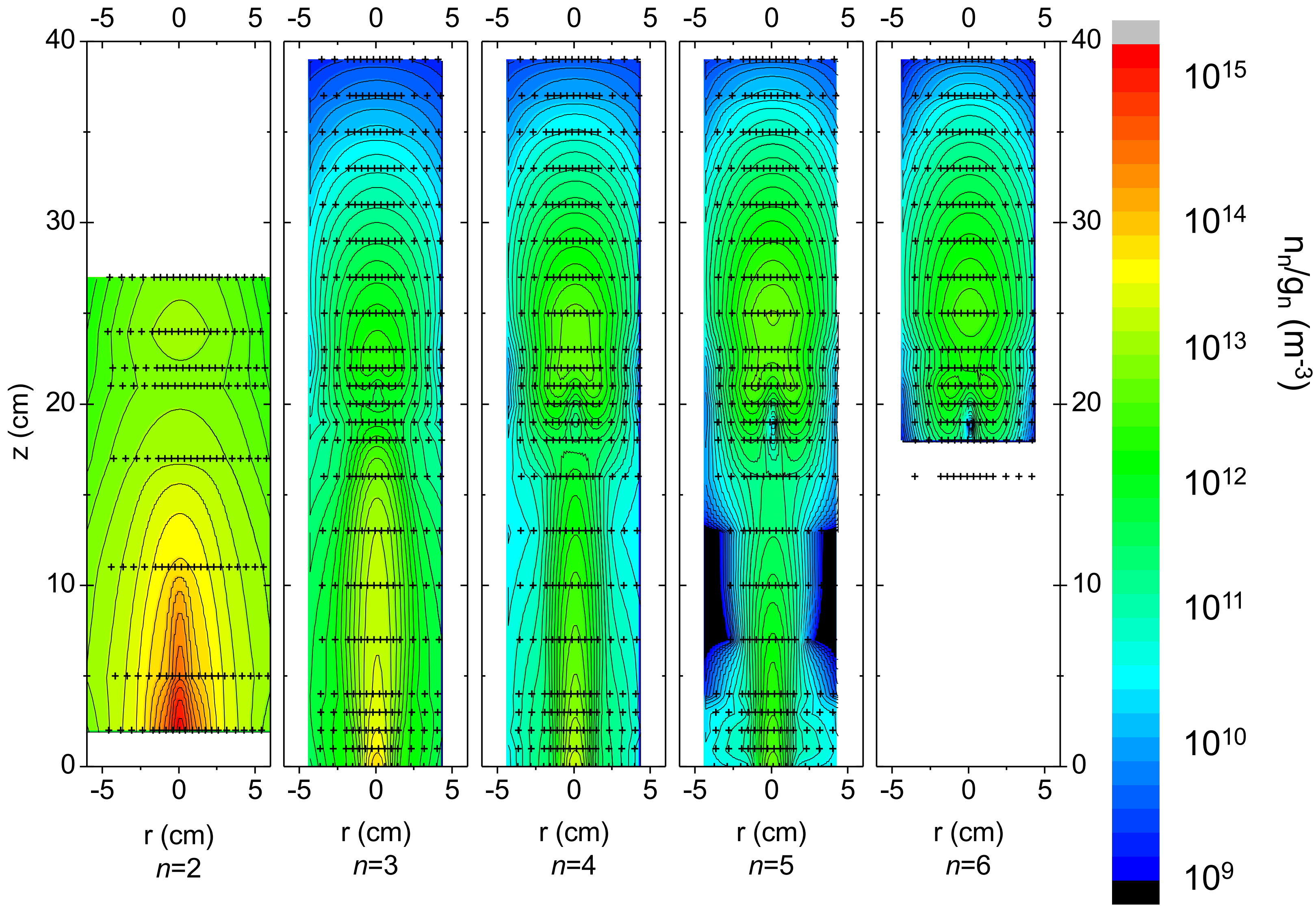
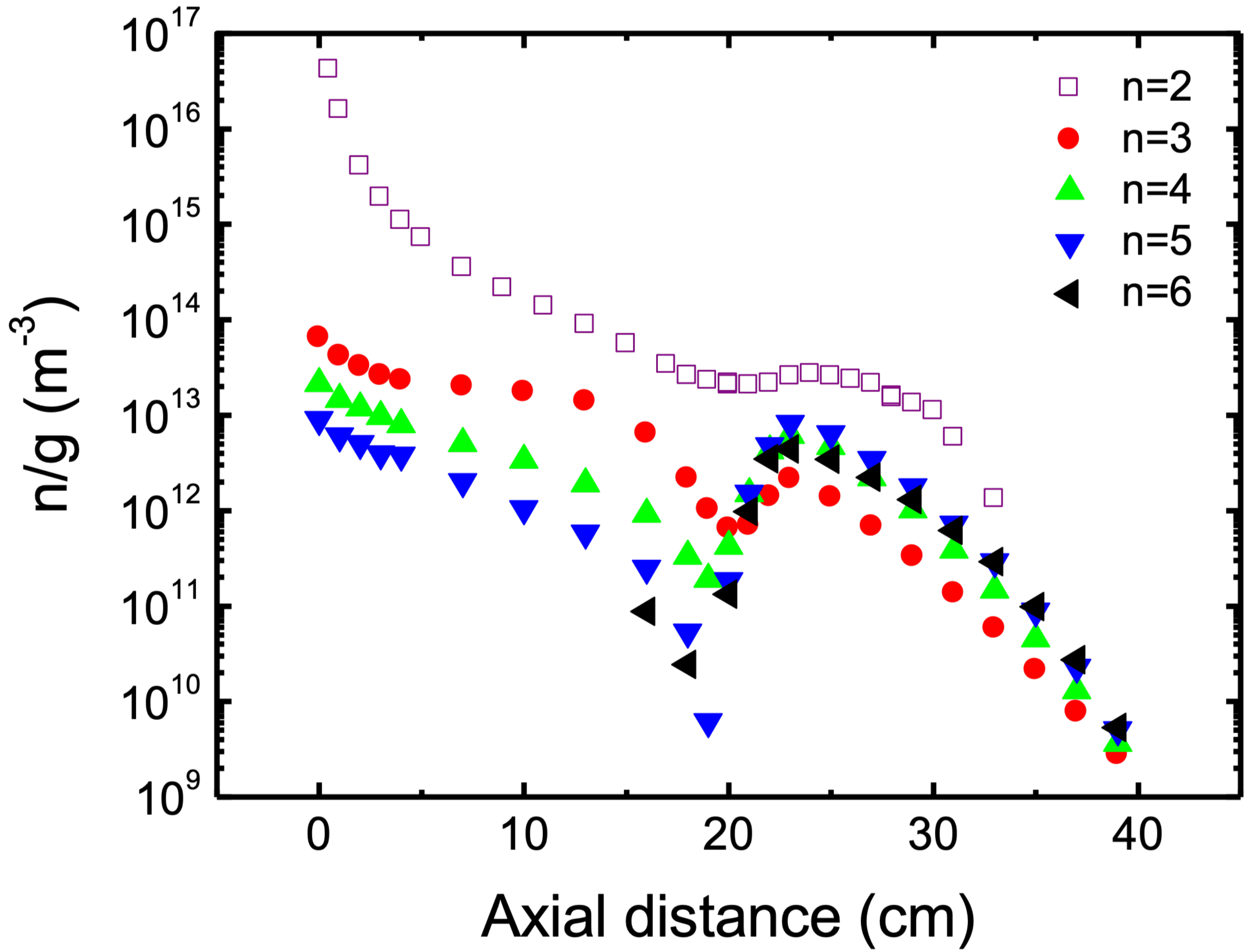


Abel inverted excited state densities

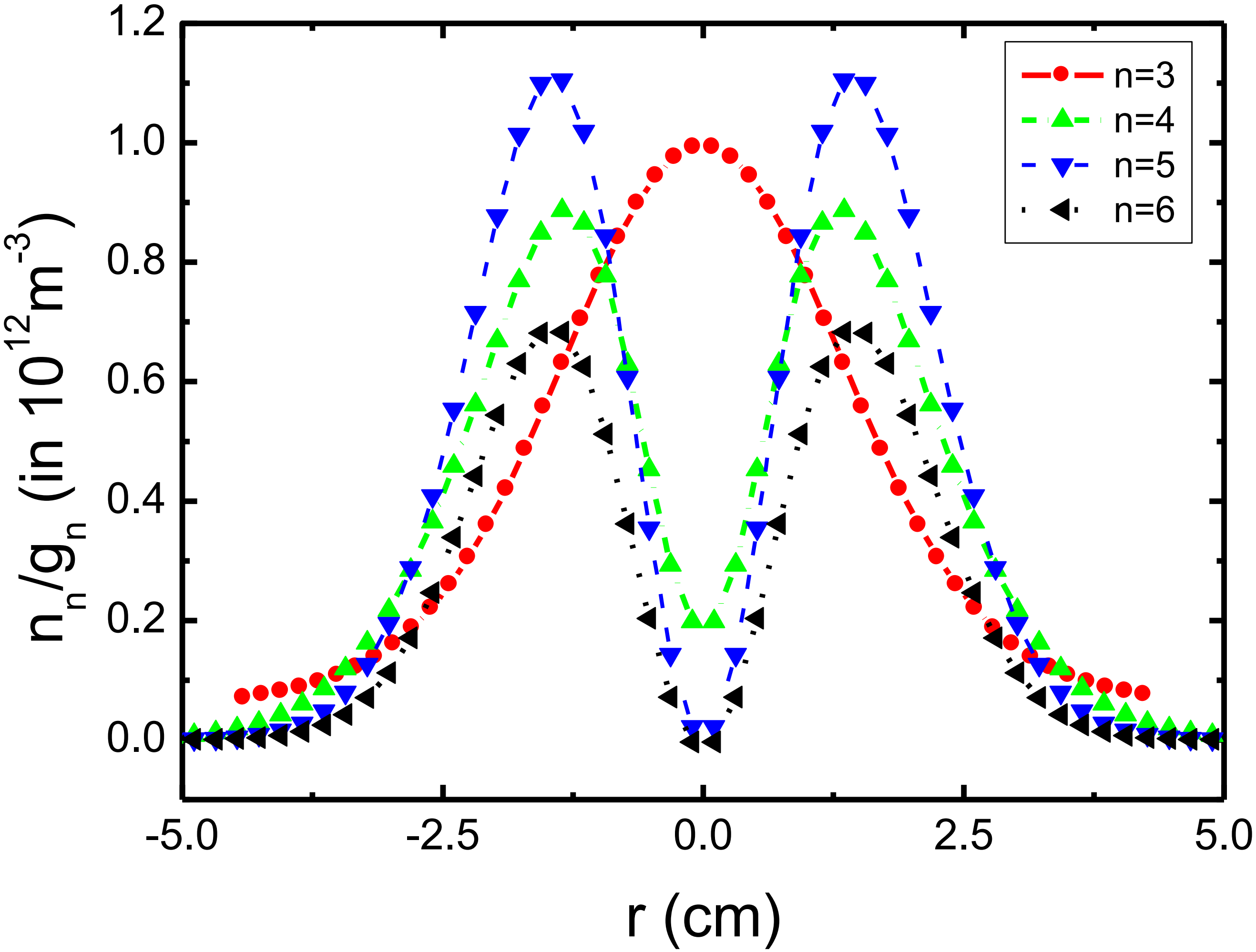




Axial distribution of excited H atoms (r = 0 cm)



Radial distribution of excited H atoms (z=19 cm)



Production mechanisms of excited atoms

Ion-ion recombination:

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

Molecular ion recombination:

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

$\mathit{H_3^+ + e \rightarrow H + H + H}$

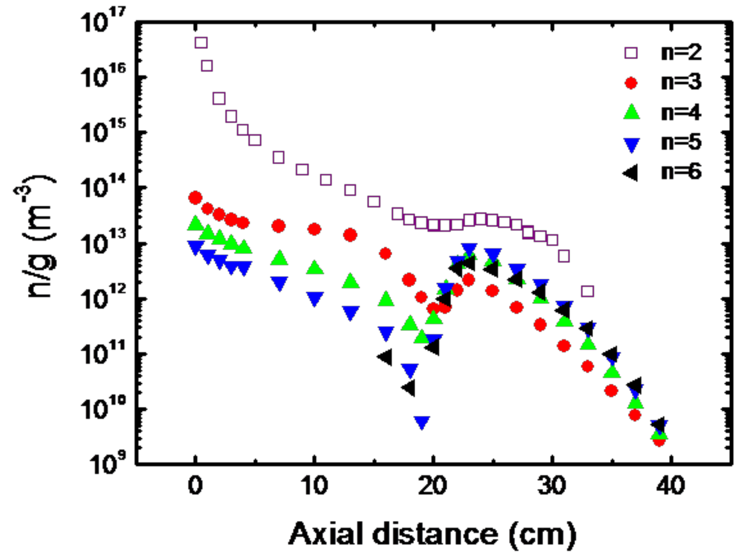
Mutual recombination (MAR):

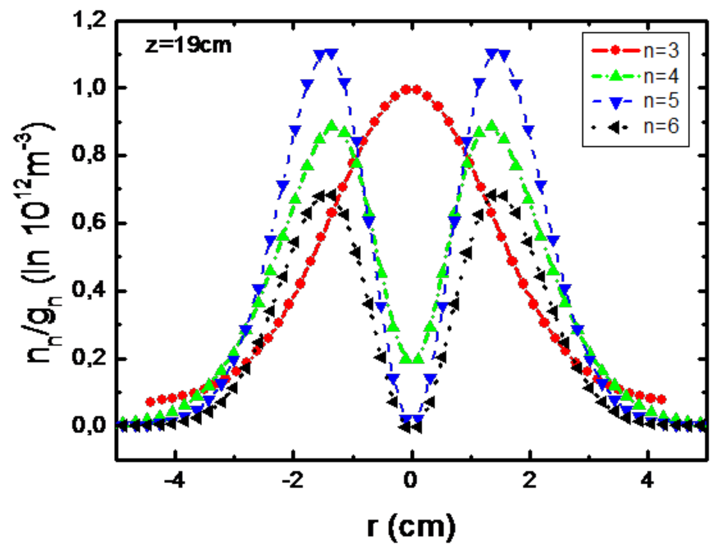
$H_2^+ + H^- \rightarrow H + H(n) \qquad (n \leq 5,6\ ?)$

$\mathit{H_3^+ + H^- \rightarrow H_2 + H + H(excitation(?))}$

$$\boxed{H_2^{r,v} + H^+ \rightarrow H_2^+ + H}$$

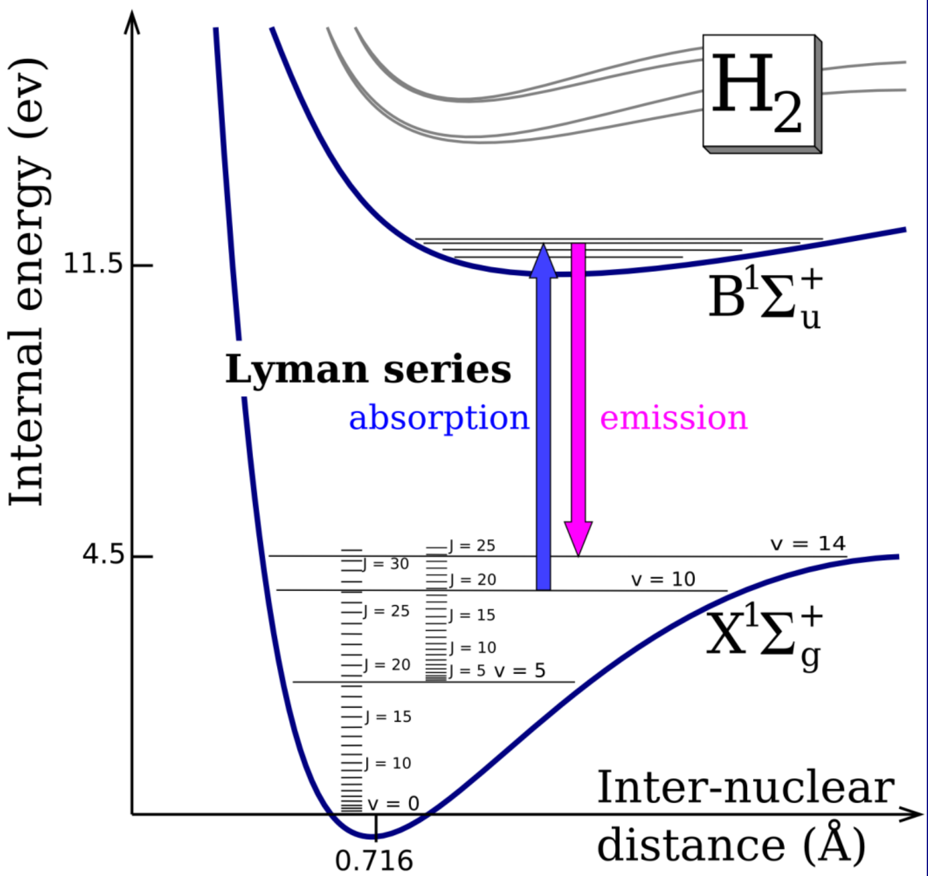
$$\boxed{H_2^{r,v} + e \rightarrow H^- + H}$$





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

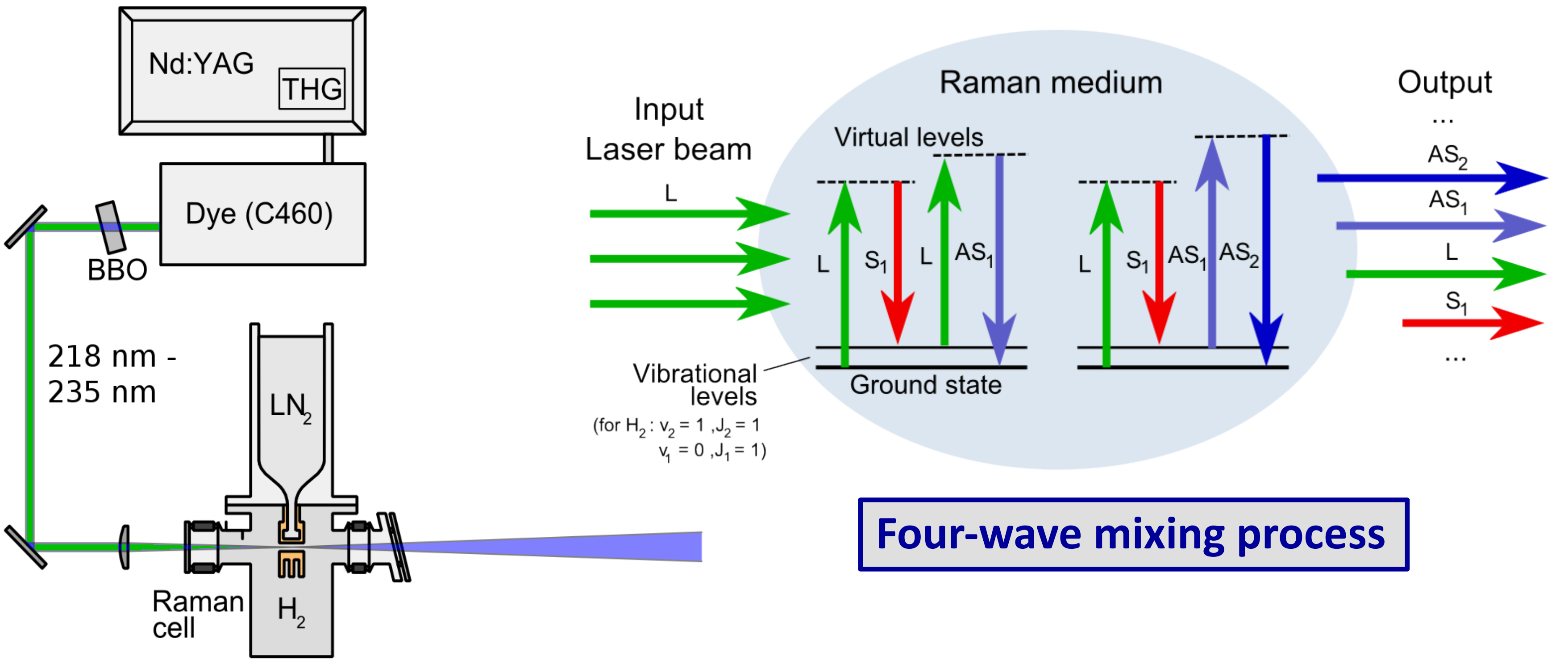
H_2 energy scheme



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 detection of $H_2^{r,v}$

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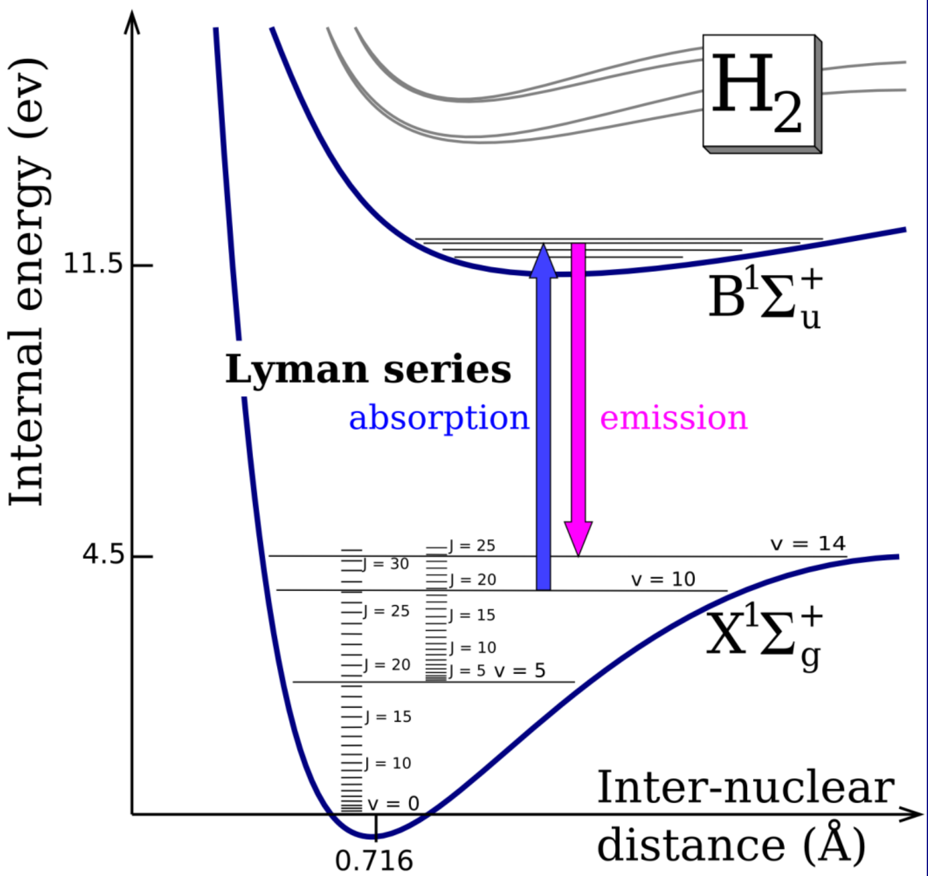

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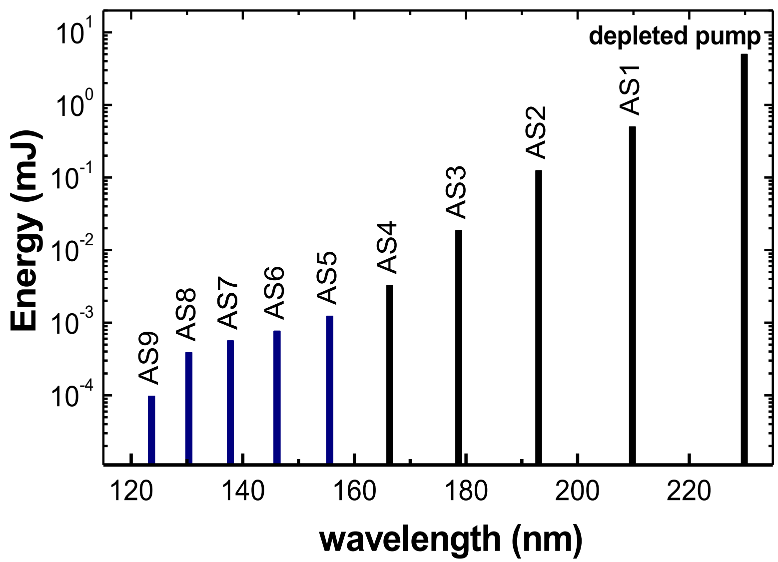

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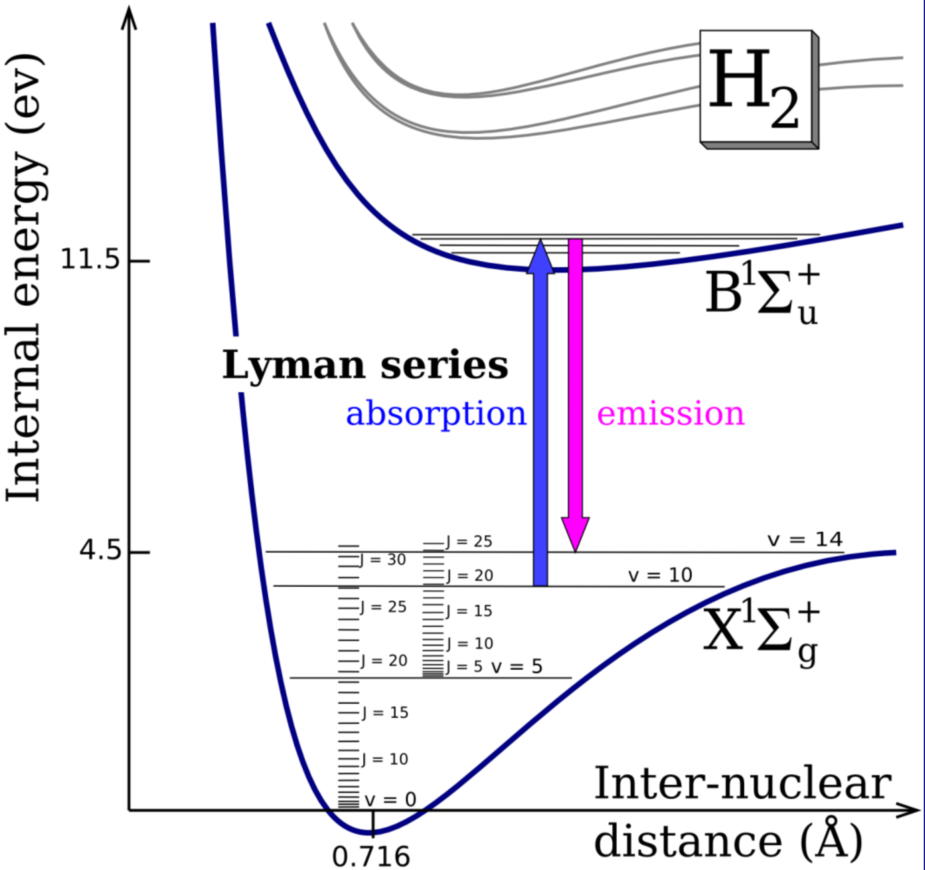
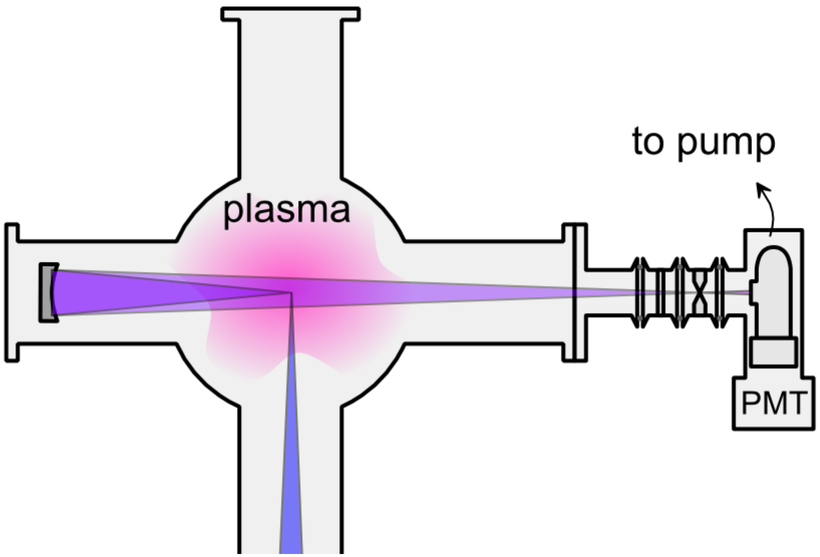
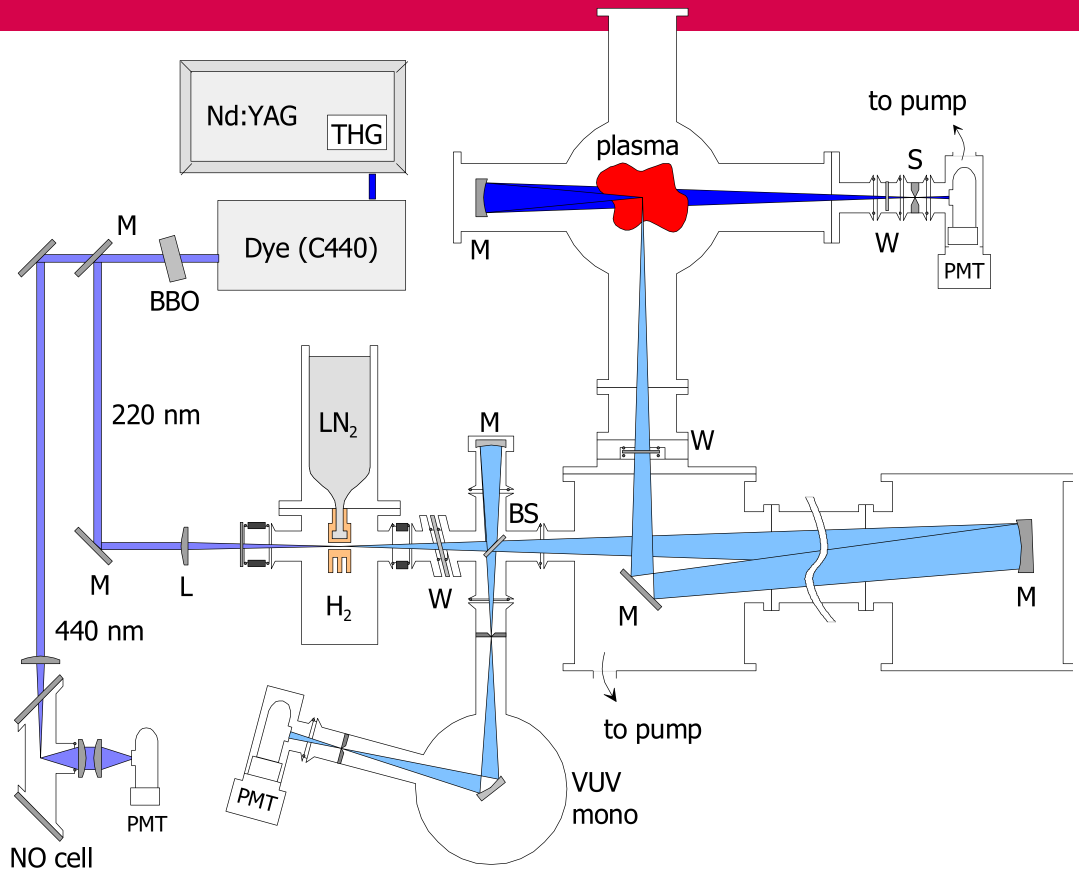


VUV-LIF detection scheme



VUV-LIF setup



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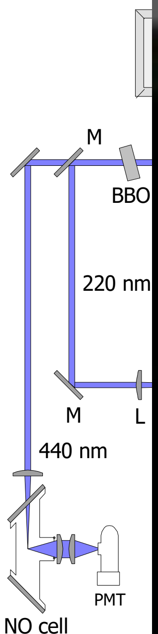
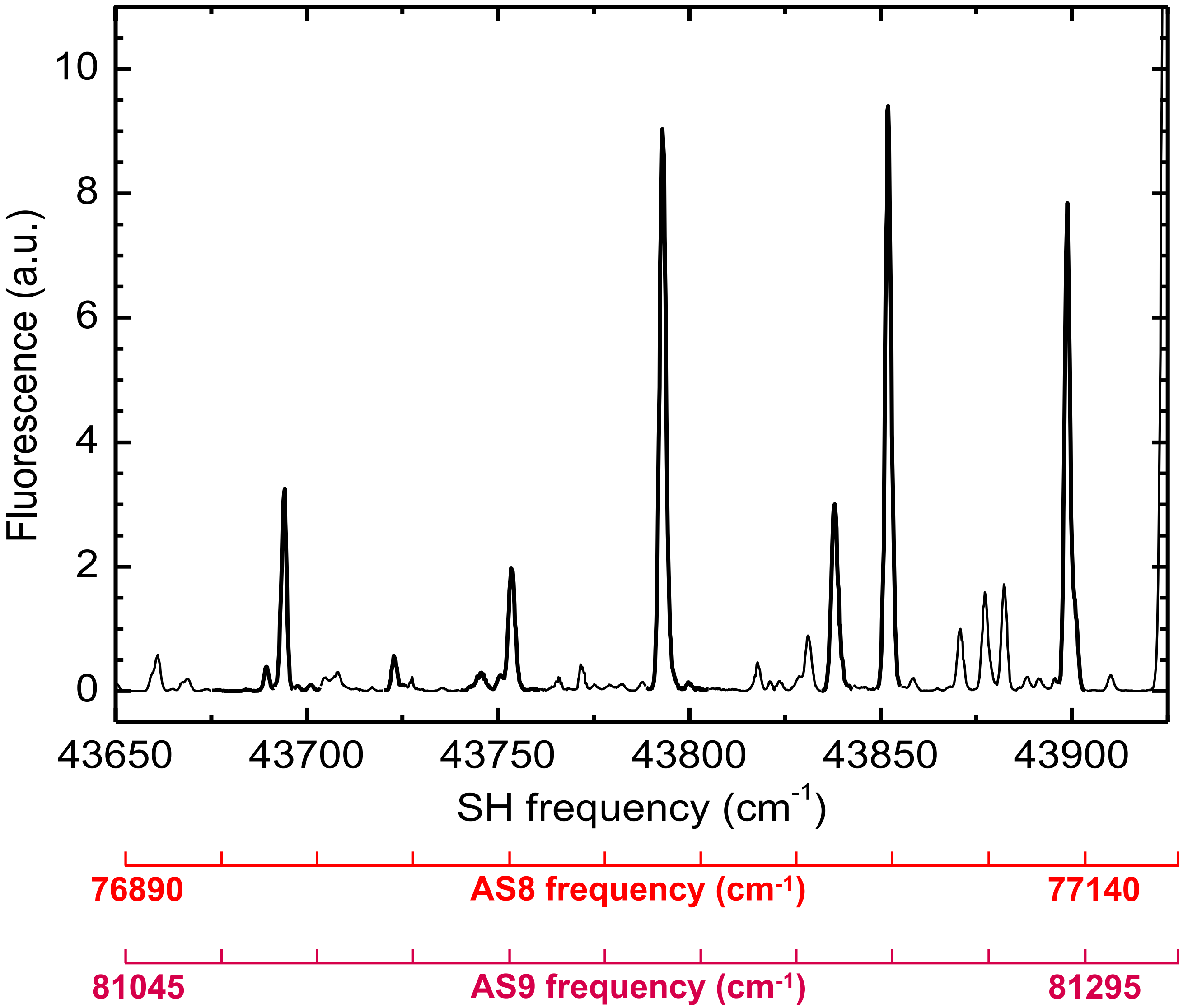
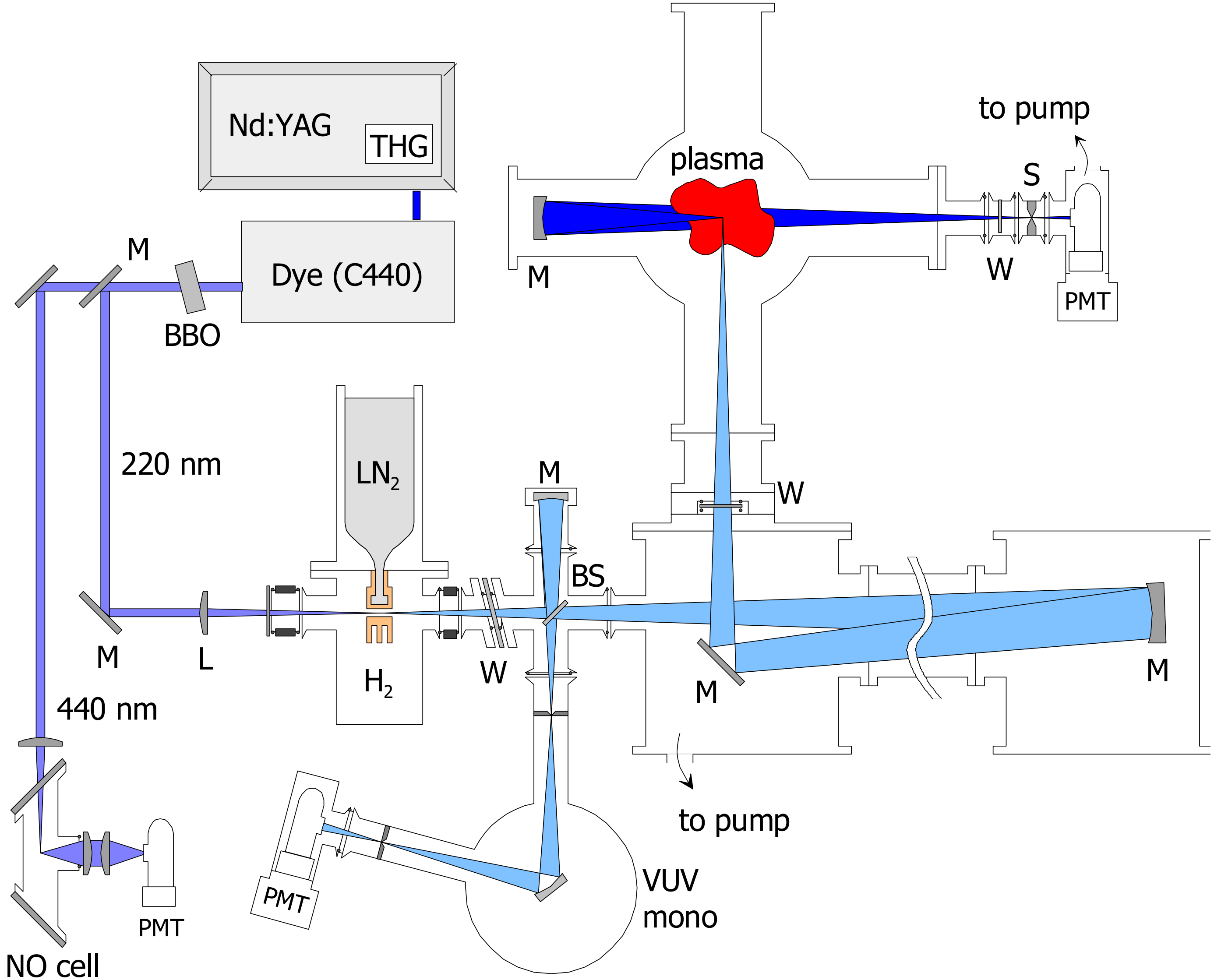


Measured H_2 Lyman spectrum



VUV LIF setup



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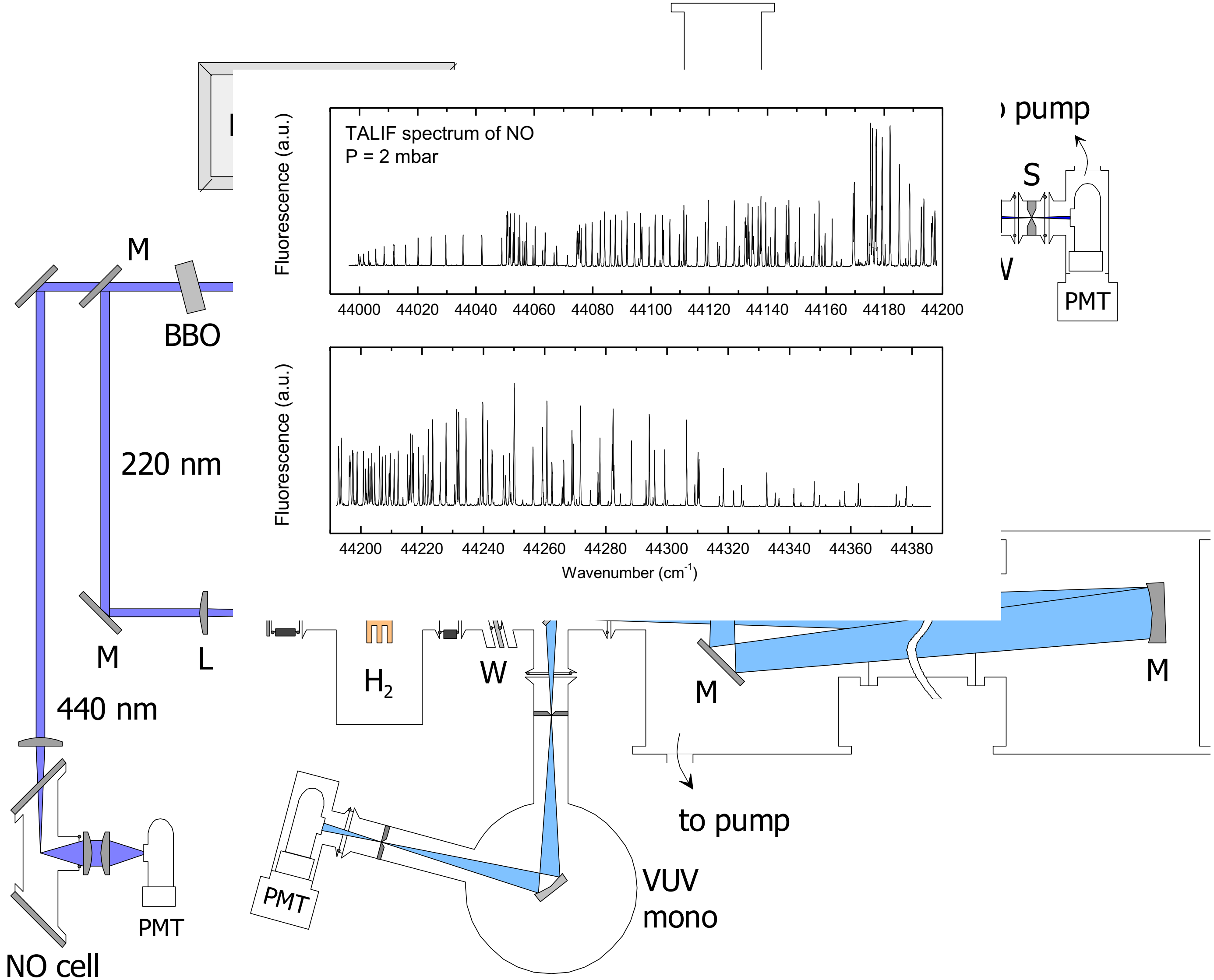
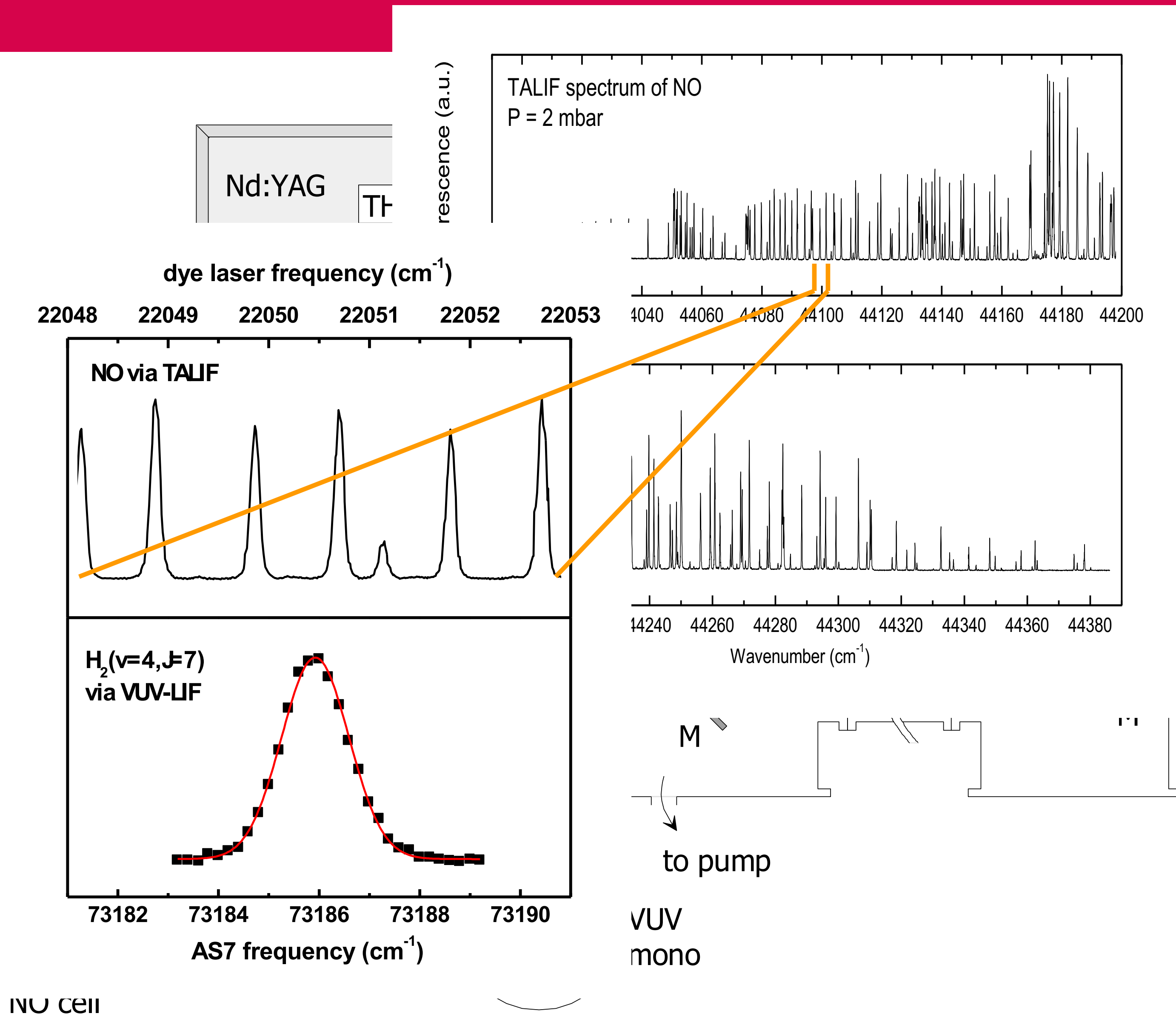
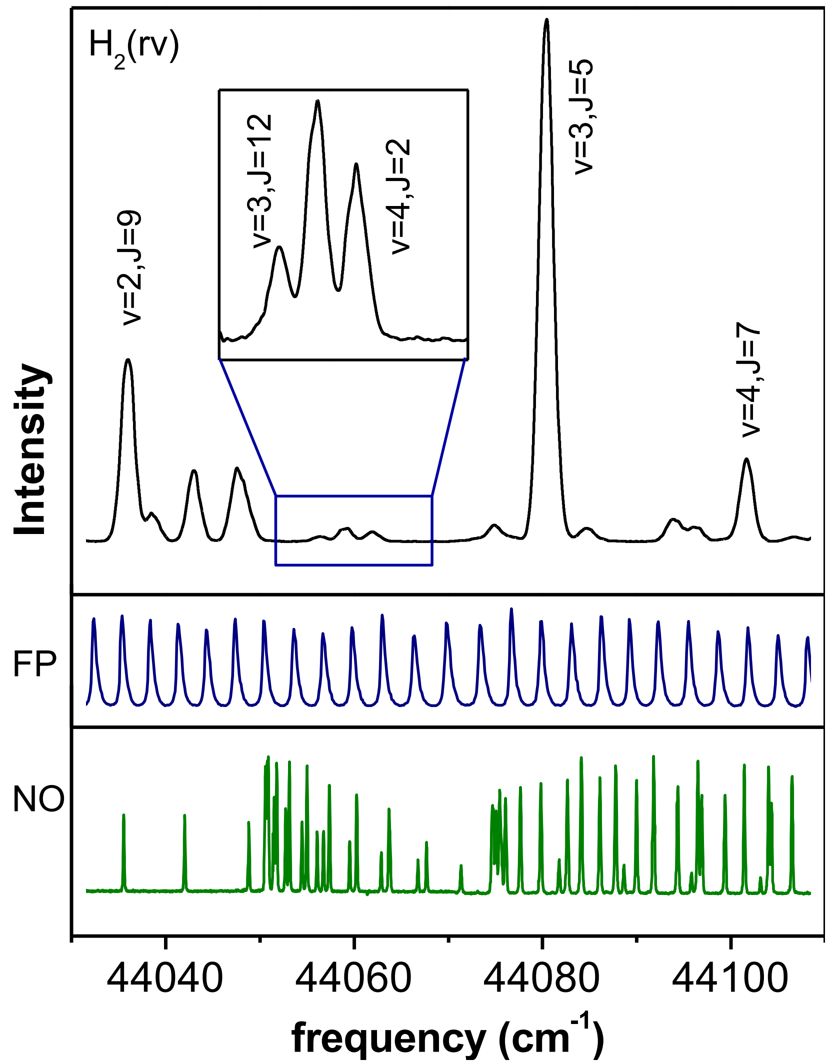


VUV-LIF spectroscopy



Measured H_2 Lyman spectrum



state-selective

spatially resolved

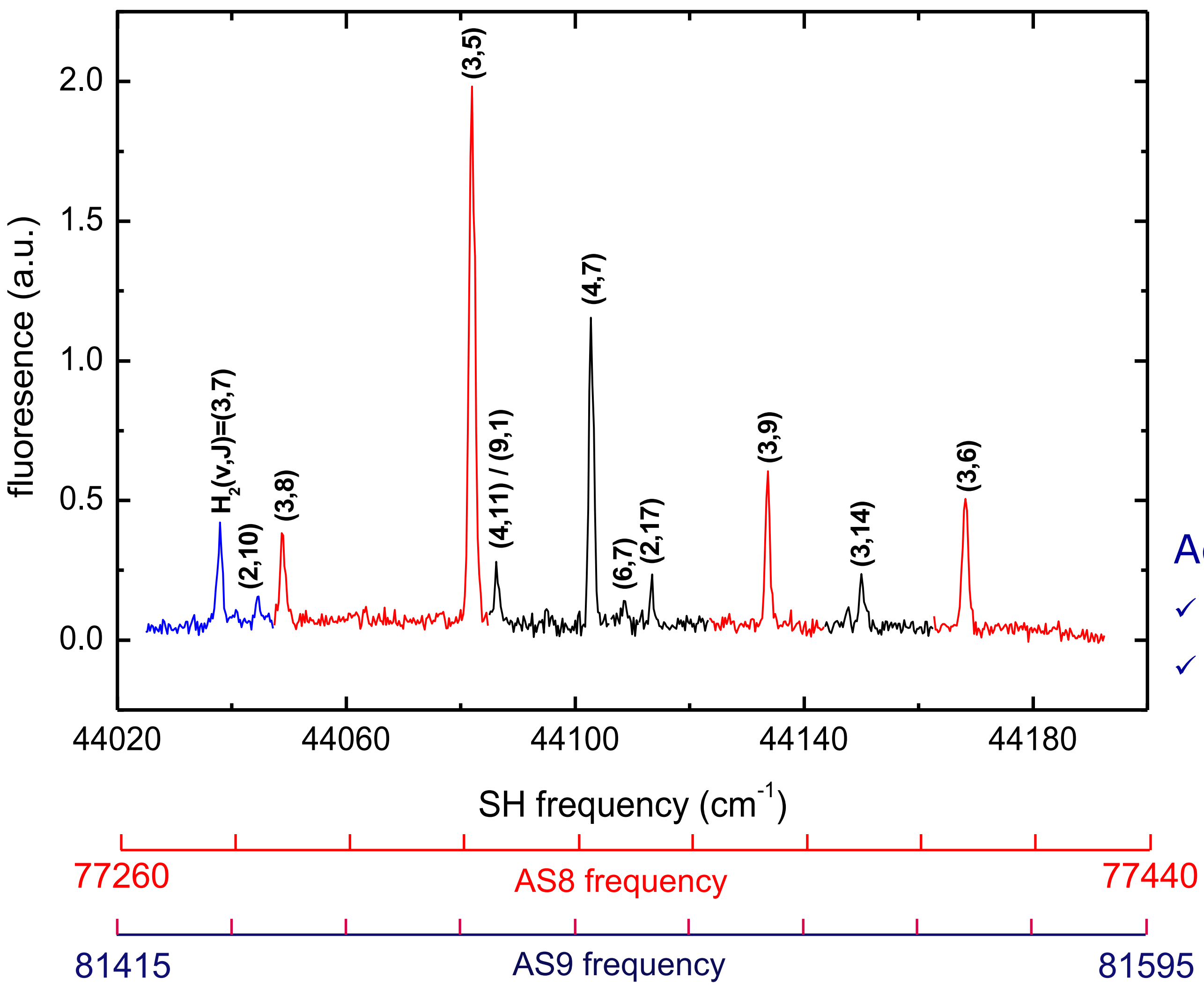
non-intrusive

dynamic range > 4 orders

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

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

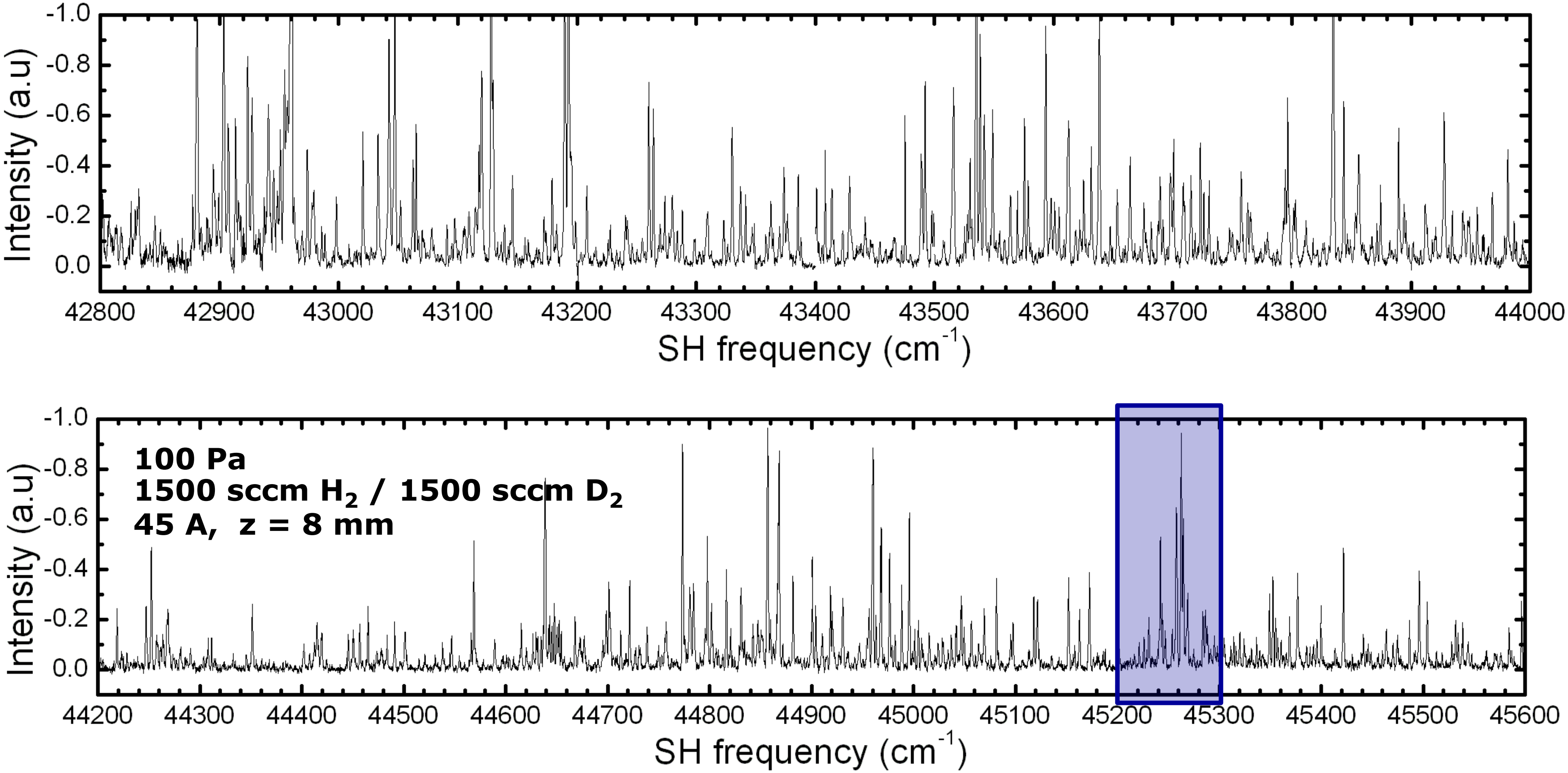
Measured H_2 Lyman spectrum



Advantages multiplexing:

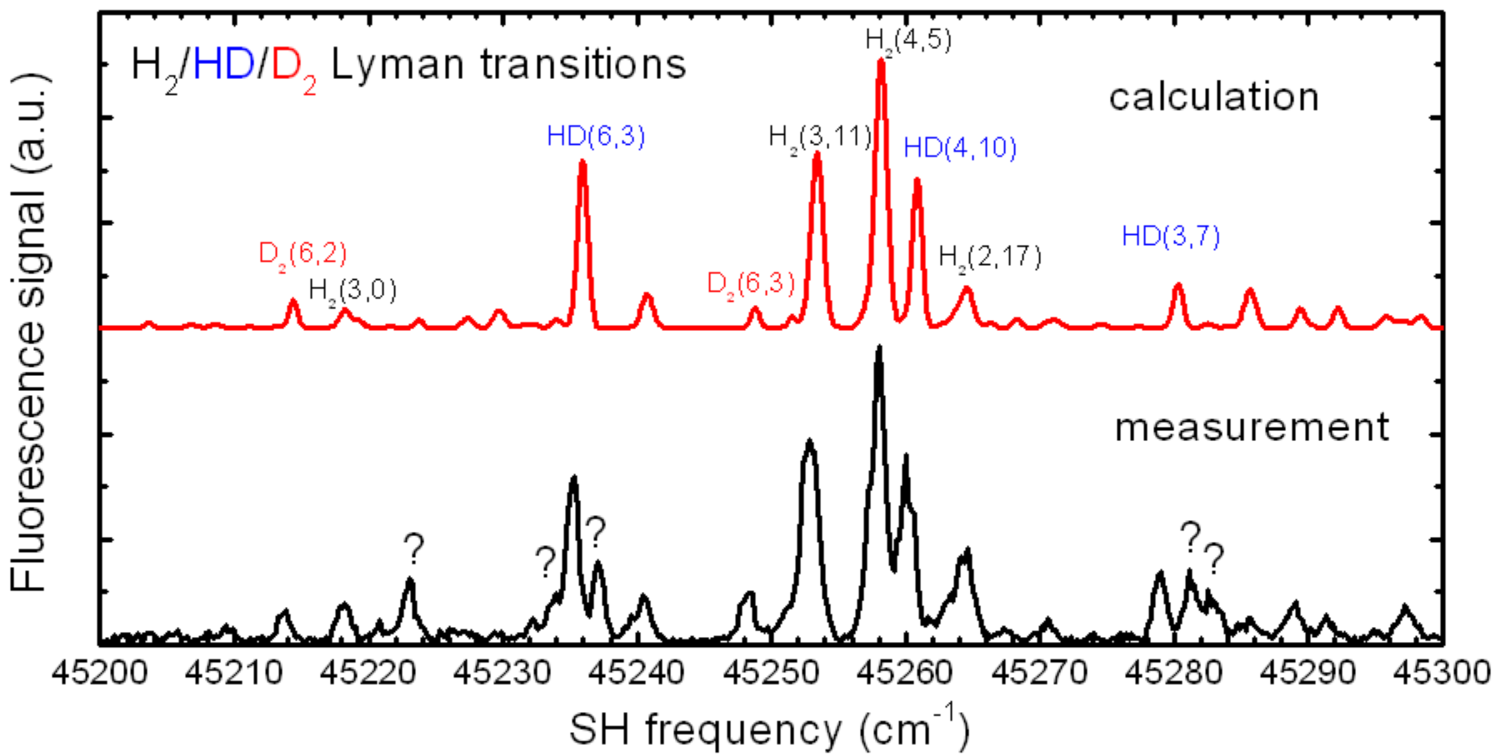
- ✓ **spectrum more dense**
- ✓ **efficient measurement**

Measured H_2/HD/D_2 Lyman spectra



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

Measured and calculated H_2/HD/D_2 Lyman spectrum



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

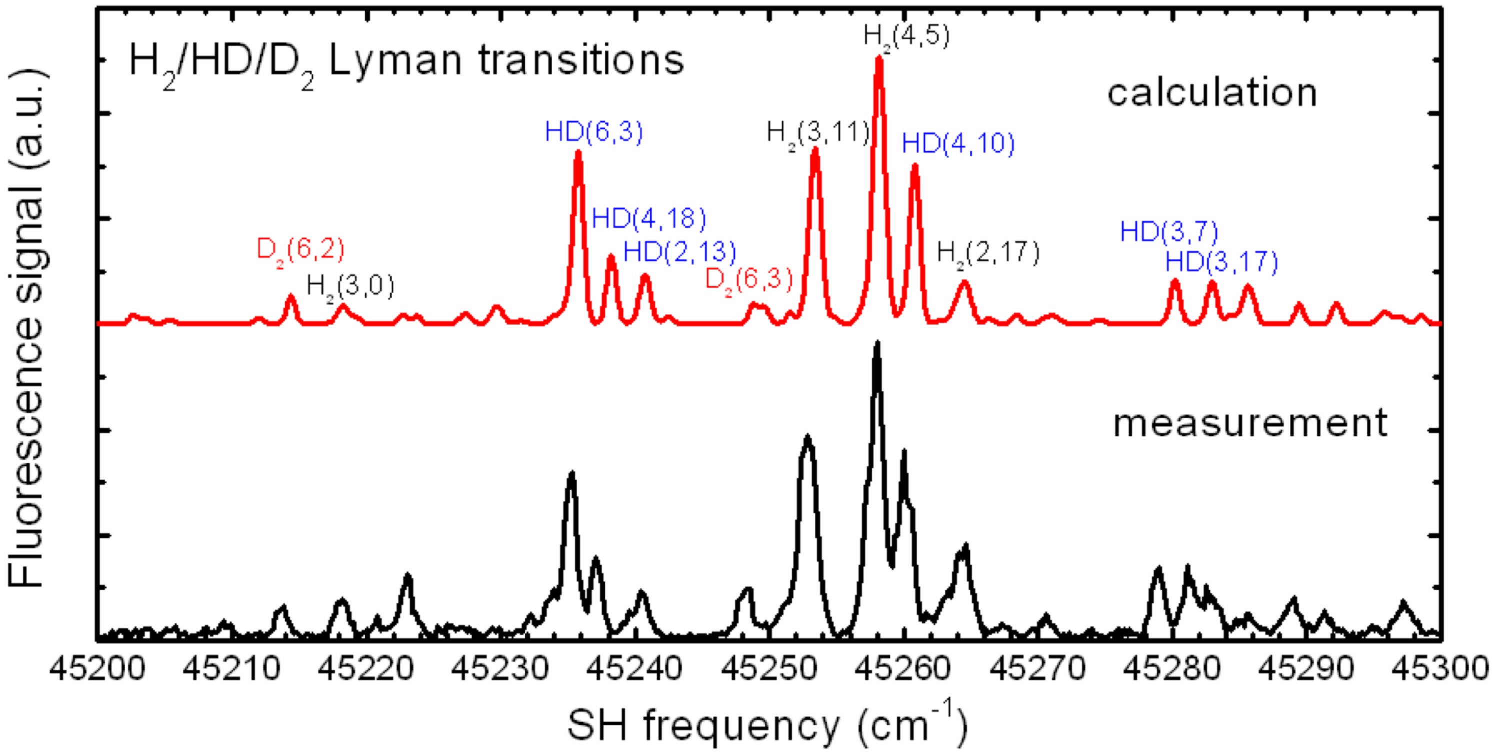
HD Lyman transitions J < 11

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

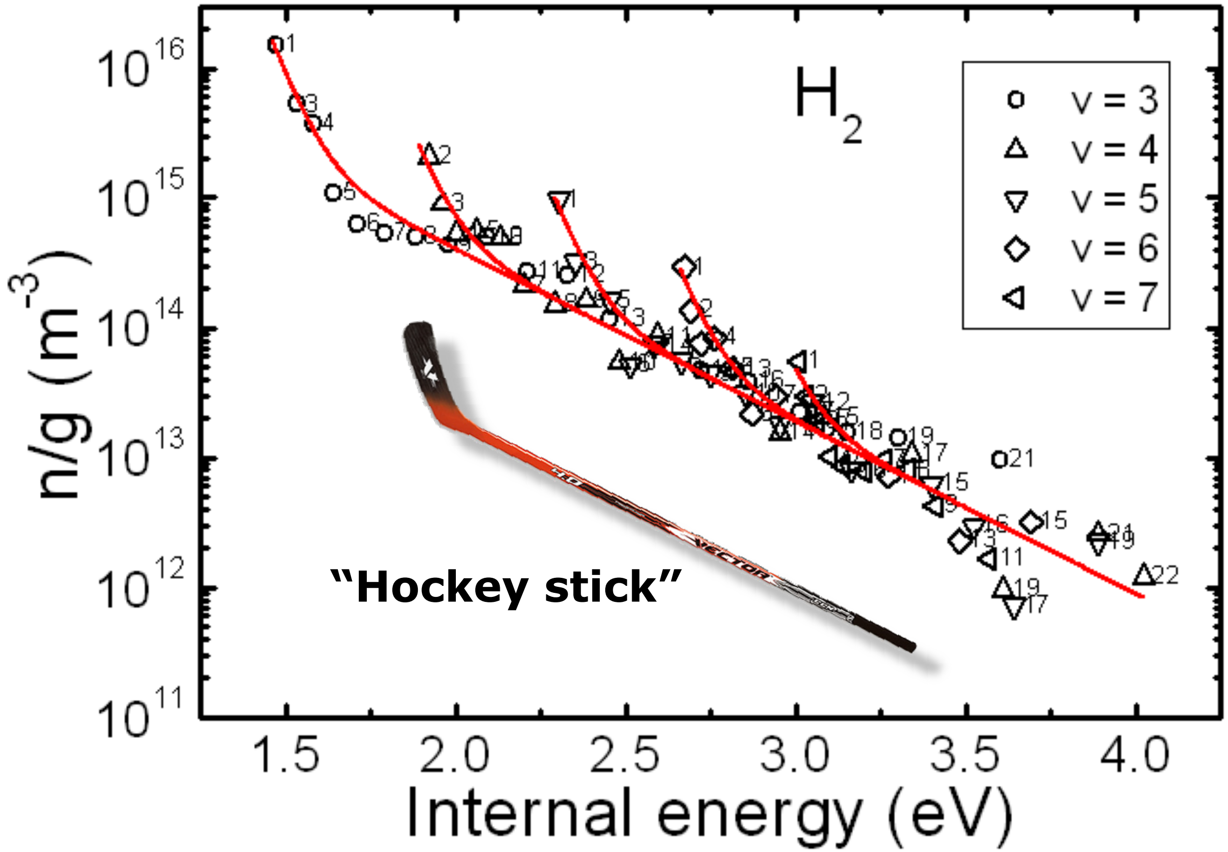
Measured and calculated $H_2/HD/D_2$ Lyman spectrum



New calculated Lyman transitions including higher rotational states (J > 10), in collaboration with Abgrall and Roueff

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

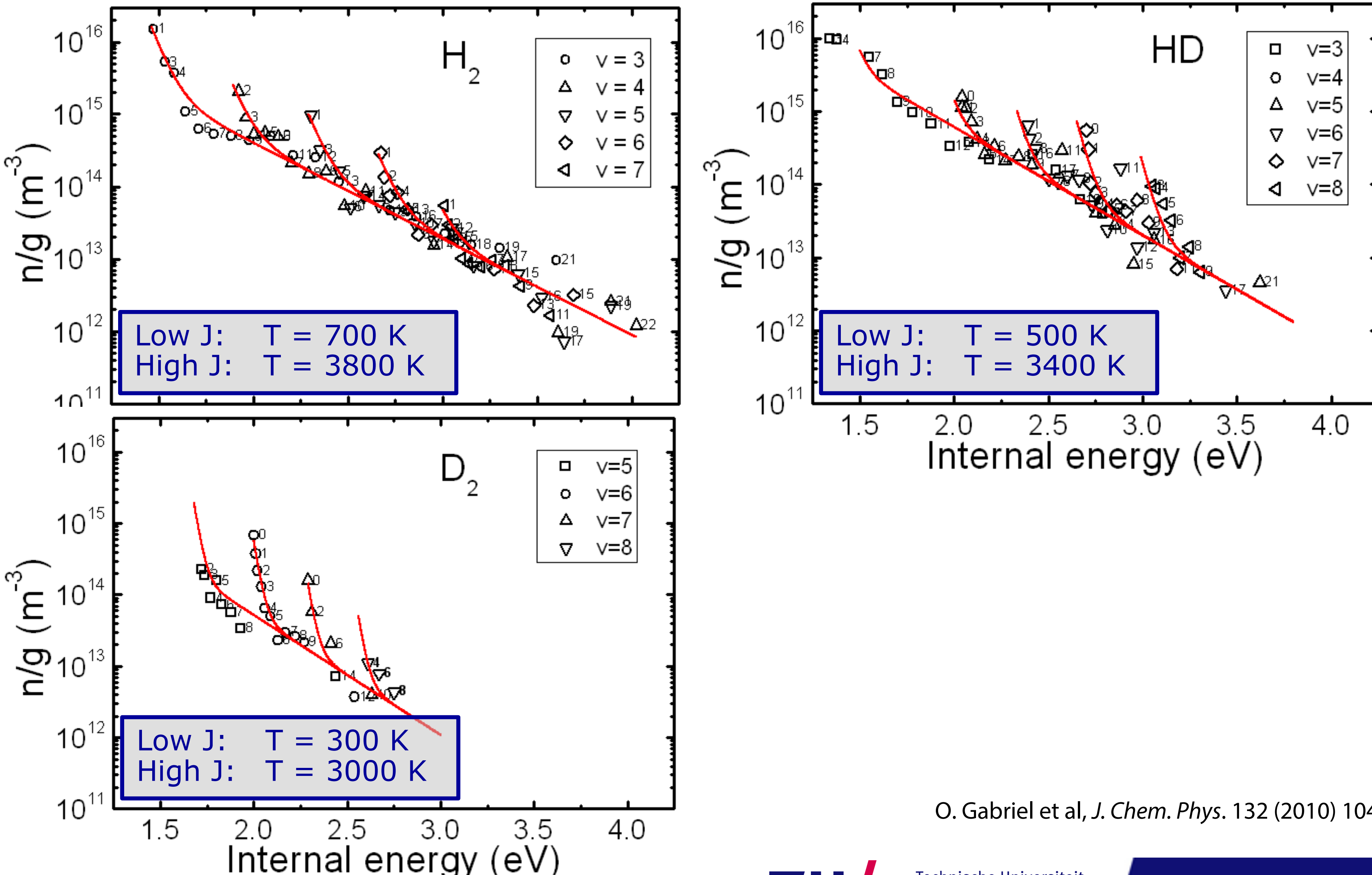
Non-Boltzmann distribution for H_2



700 K for low J

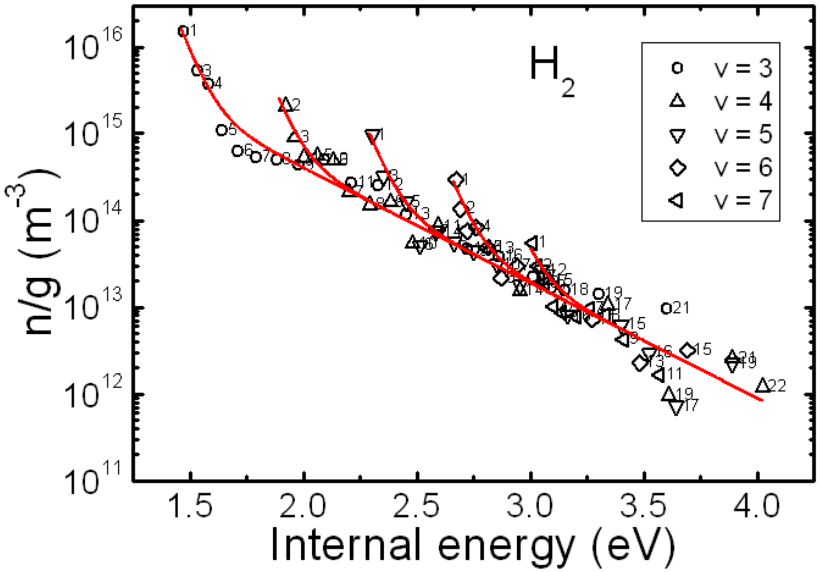
3800 K for high J

Non-Boltzmann distributions in H_2/D_2 jet



O. Gabriel et al, *J. Chem. Phys.* 132 (2010) 104305

Results on H^- production



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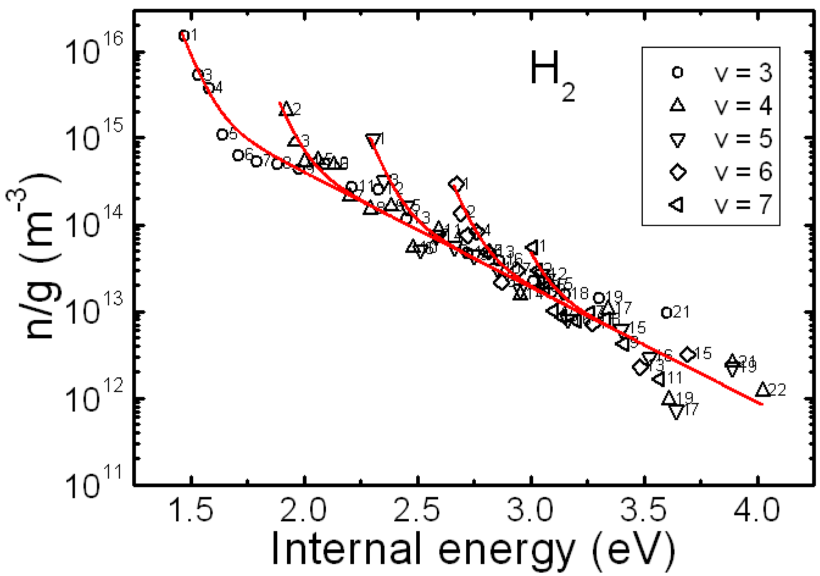

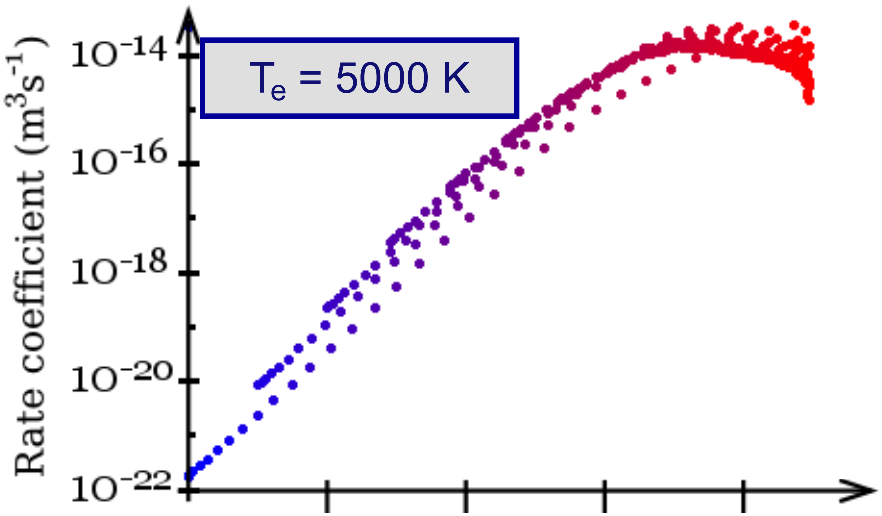

O. Gabriel et al, *J. Chem. Phys*. 132 (2010) 104305

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

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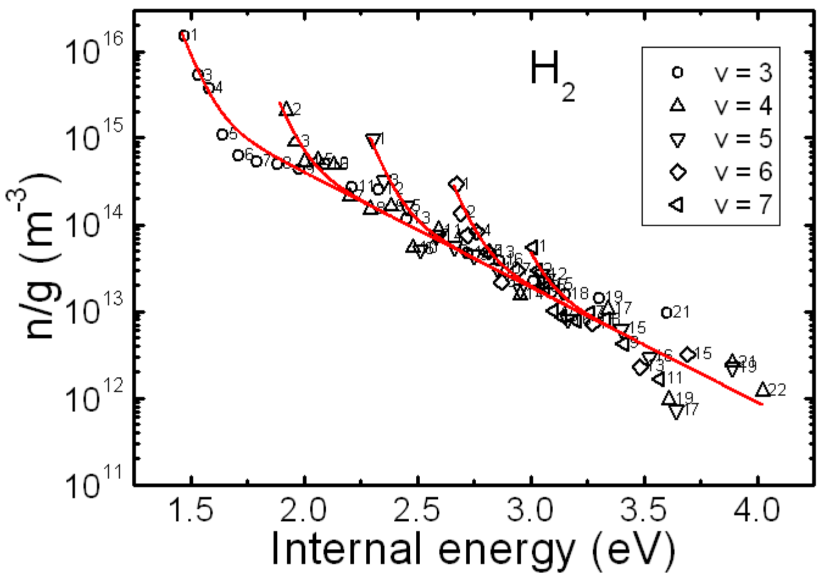
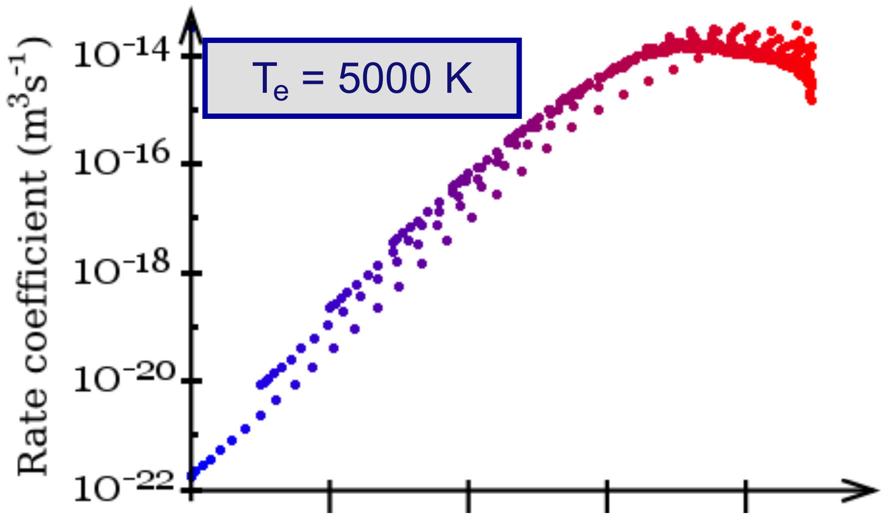
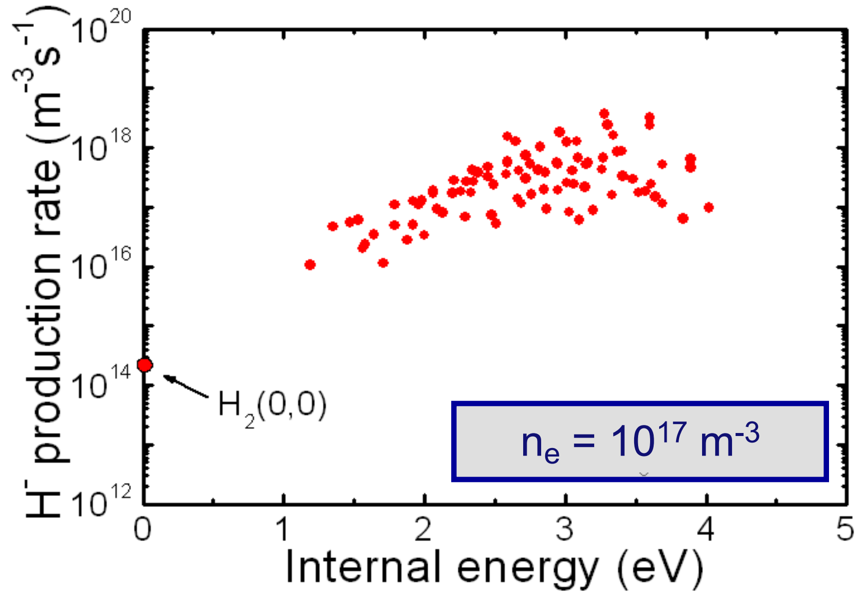




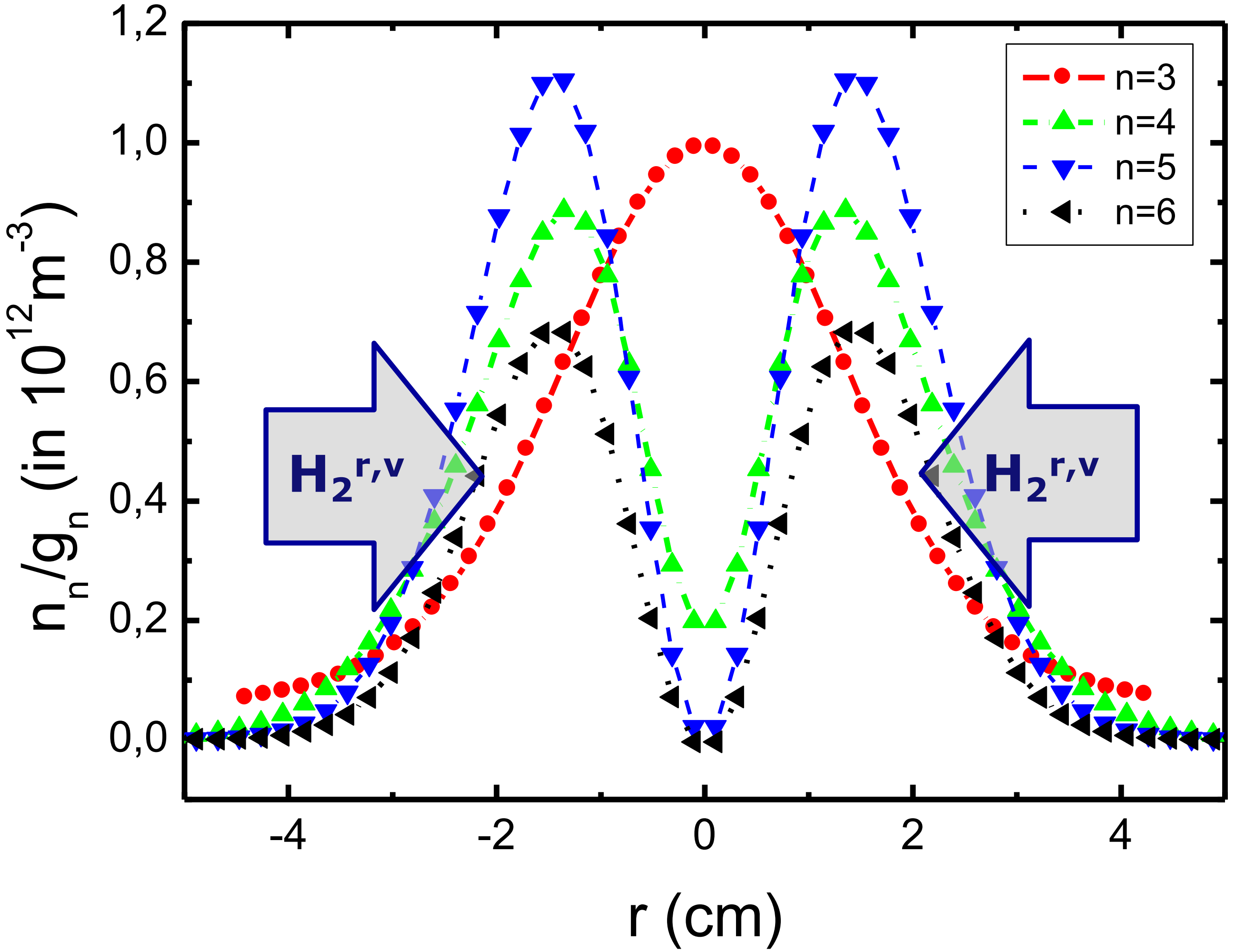


Production rate of H^- ions by dissociative attachment

O. Gabriel et al, *J. Chem. Phys*. 132 (2010) 104305

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

Radial distribution of excited H atoms (z=19 cm)



Production mechanisms of excited atoms

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Molecular ion recombination:

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$H_3^+ + e \rightarrow H + H + H$

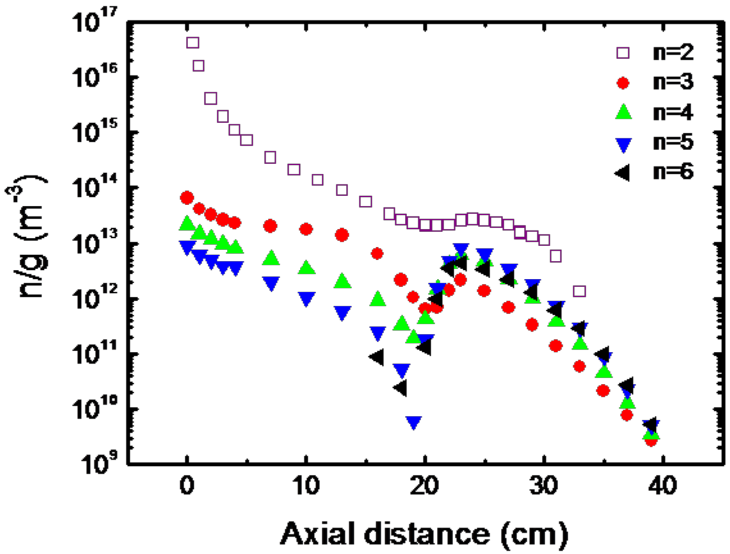
Mutual recombination (MAR):

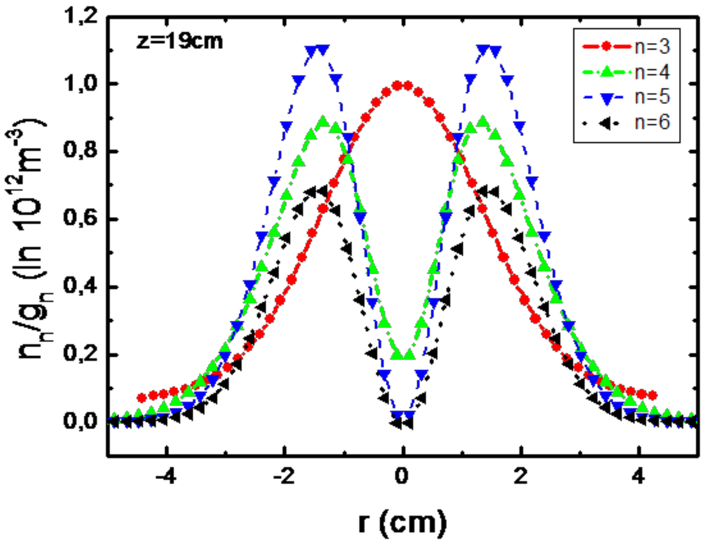
$H_2^+ + H^- \rightarrow H + H(n)$ (n ≤ 5,6 ?)

$H_3^+ + H^- \rightarrow H_2 + H + H(excitation(?))$

$H_2^{r,v} + H^+ \rightarrow H_2^+ + H$

$H_2^{r,v} + e \rightarrow H^- + H$





CR model (YACORA): processes included

$H(m) + e \rightarrow H(n>m) + e$ $H(m) + e \rightarrow H(n<m) + e$ $H(m) \rightarrow H(n<m) + h\nu$	H
$H^+ + e + e \rightarrow H(n) + e$ $H^+ + e \rightarrow H(n) + h\nu$	H^+
$H_2 + e \rightarrow H(n) + H(1) + e$	H_2
$H_2^+ + e \rightarrow H(n) + H^+ + e$ $H_2^+ + e \rightarrow H(n) + H(1)$	H_2^+
$H_3^+ + e \rightarrow H(n) + H_2$	H_3^+
$H^+ + H^- \rightarrow H(n) + H(1)$ $H_2^+ + H^- \rightarrow H(n) + H_2$	H^-

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$H(m) + e \rightarrow H(n<m) + e$

$H(m) \rightarrow H(n<m) + h\nu$

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

$H^+ + e \rightarrow H(n) + h\nu$

$H_2 + e \rightarrow H(n) + H(1) + e$

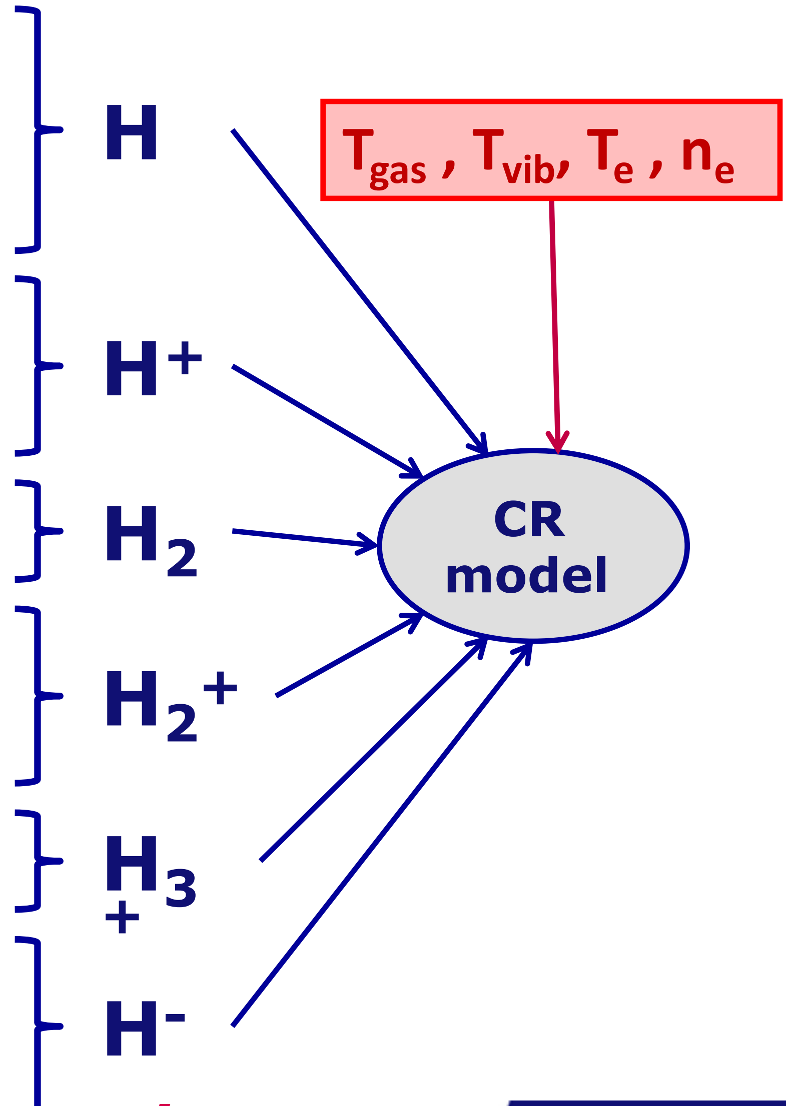
$H_2^+ + e \rightarrow H(n) + H^+ + e$

$H_2^+ + e \rightarrow H(n) + H(1)$

$H_3^+ + e \rightarrow H(n) + H_2$

$H^+ + H^- \rightarrow H(n) + H(1)$

$H_2^+ + H^- \rightarrow H(n) + H_2$



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$H(m) + e \rightarrow H(n<m) + e$
$H(m) \rightarrow H(n<m) + h\nu$

$H^+ + e + e \rightarrow H(n) + e$
$H^+ + e \rightarrow H(n) + h\nu$

$H_2 + e \rightarrow H(n) + H(1) + e$

$H_2^+ + e \rightarrow H(n) + H^+ + e$
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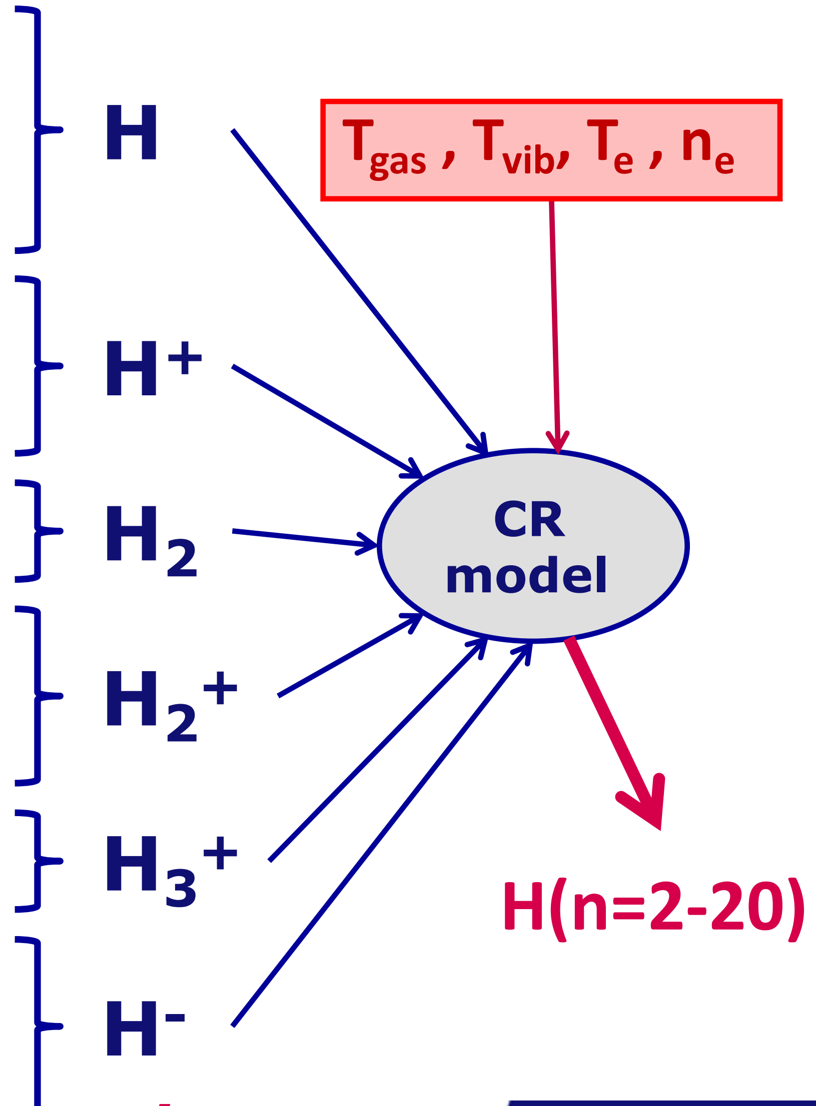
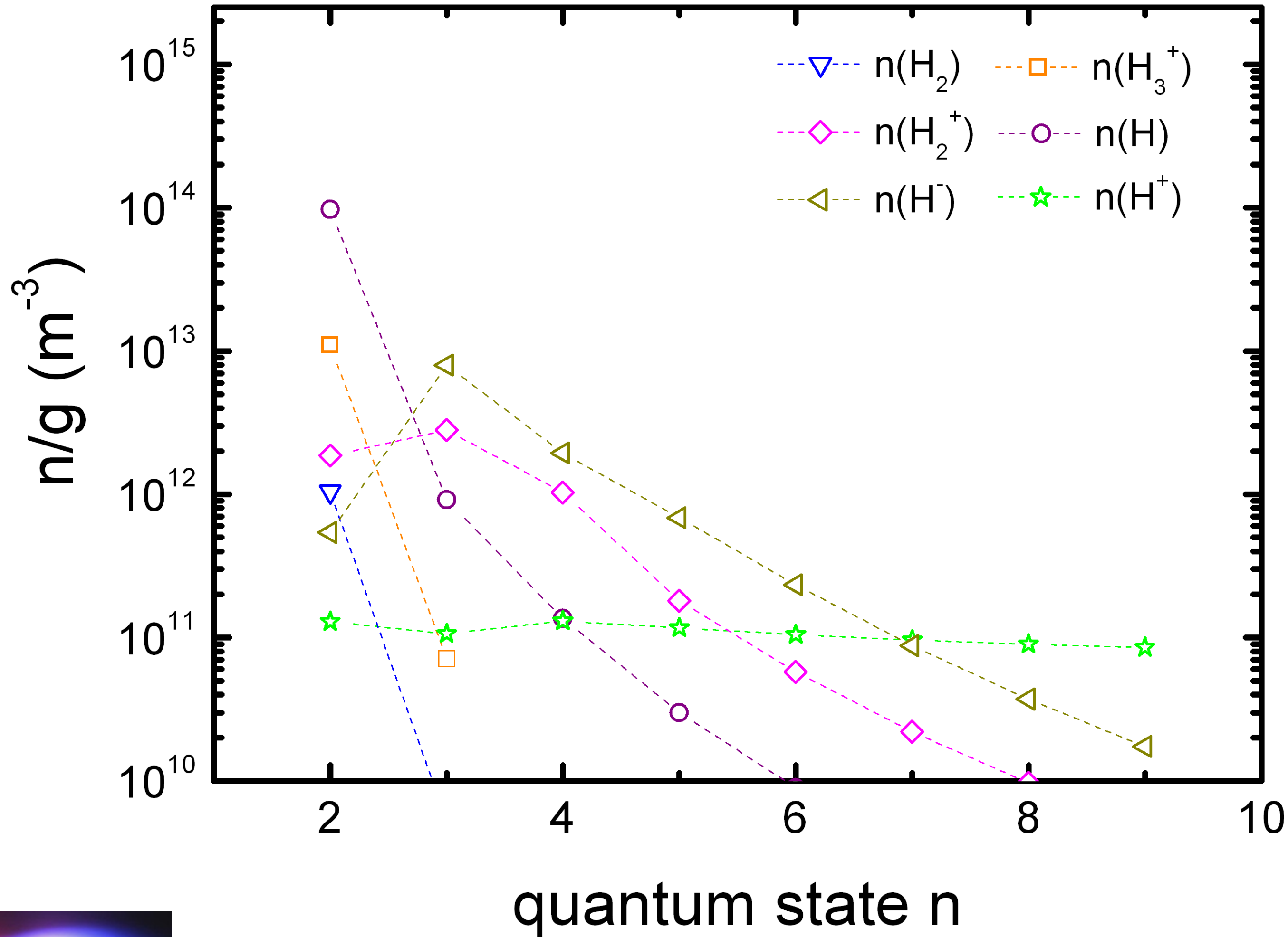
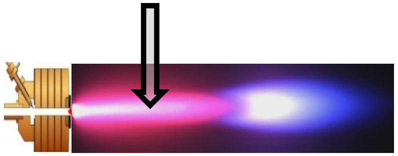


CRM results (z = 13 cm)





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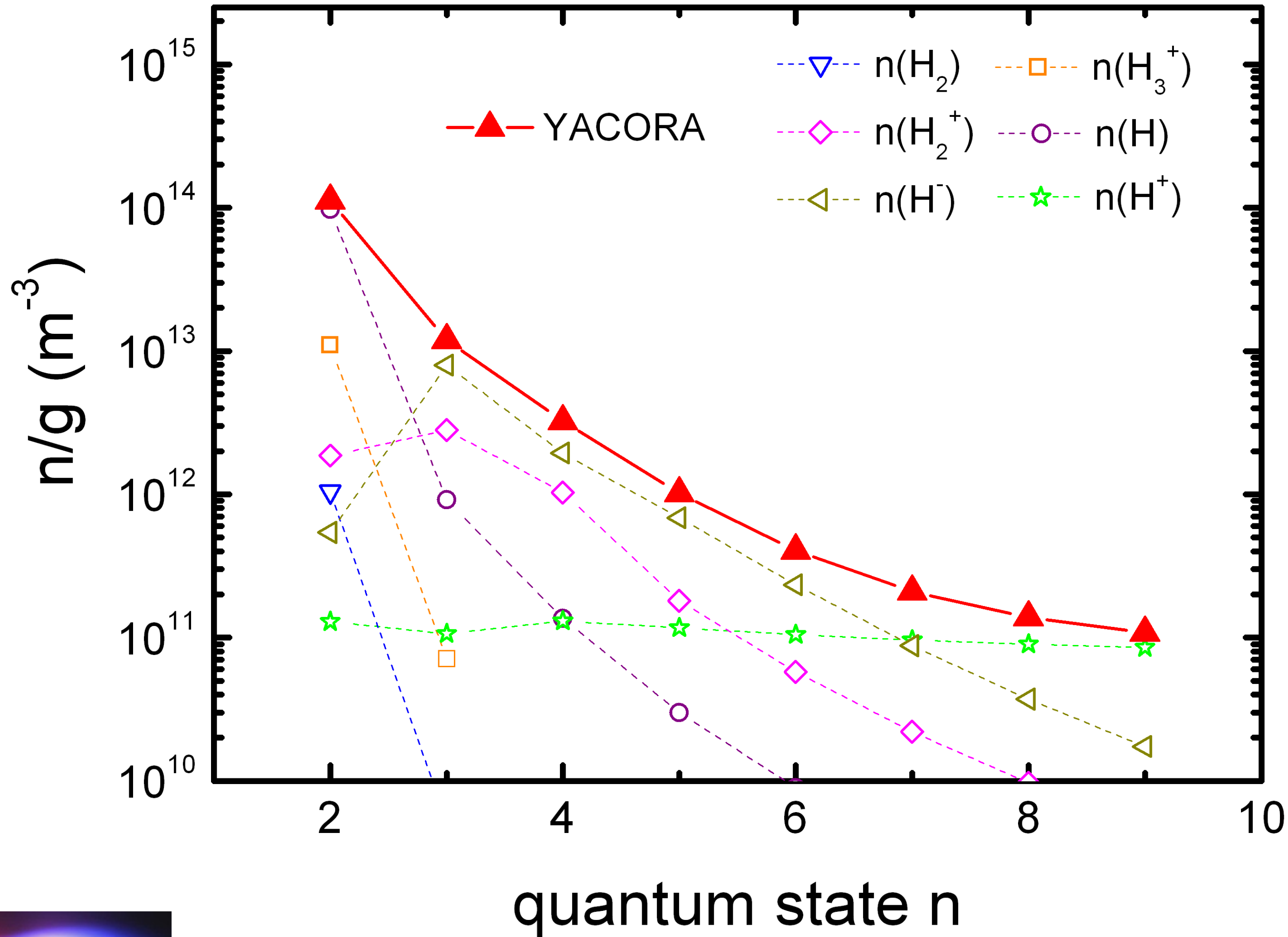
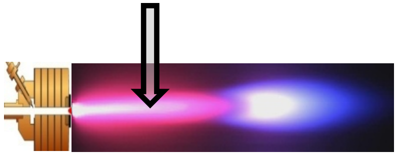
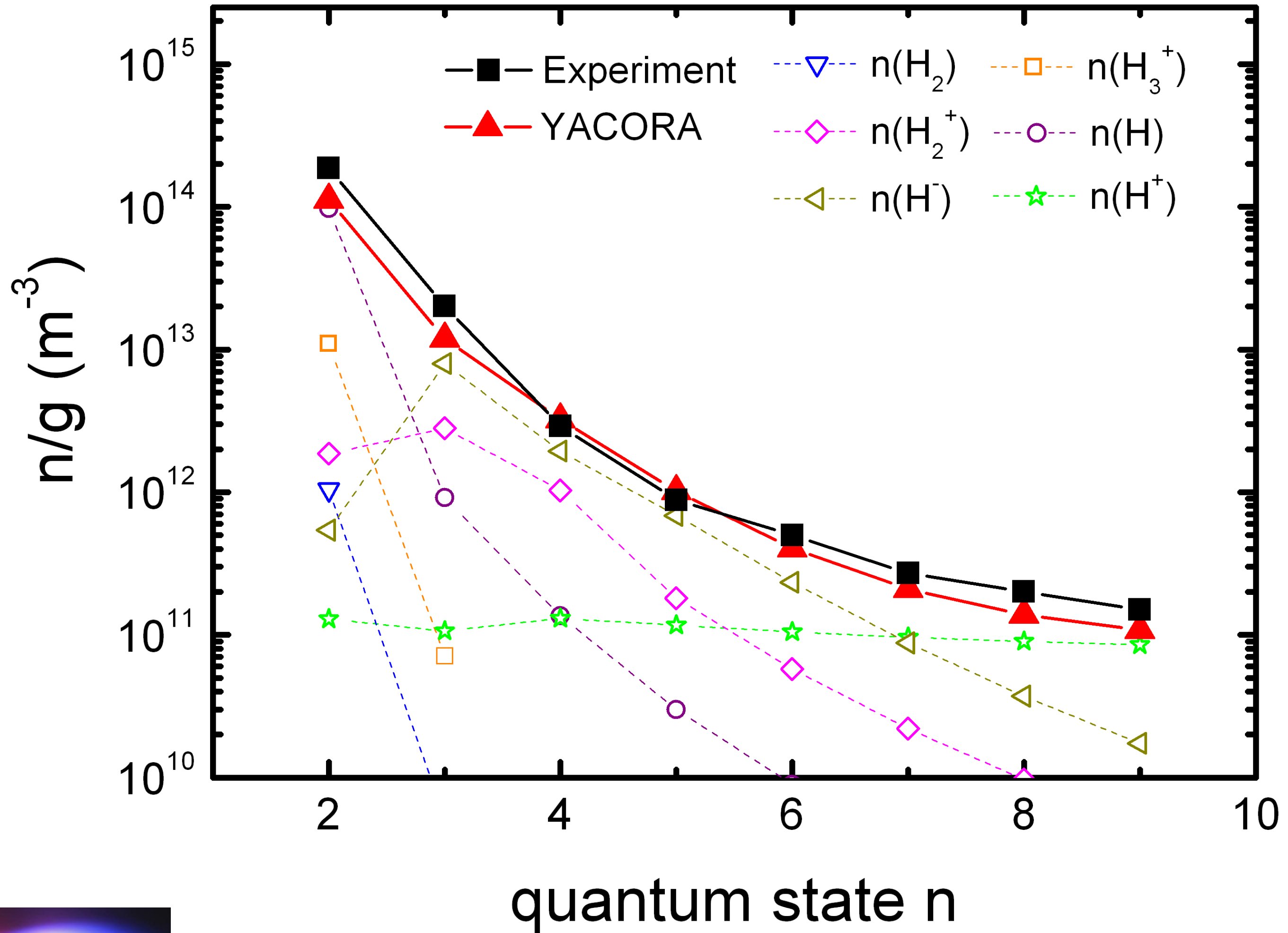
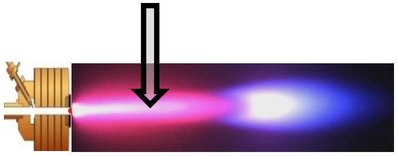


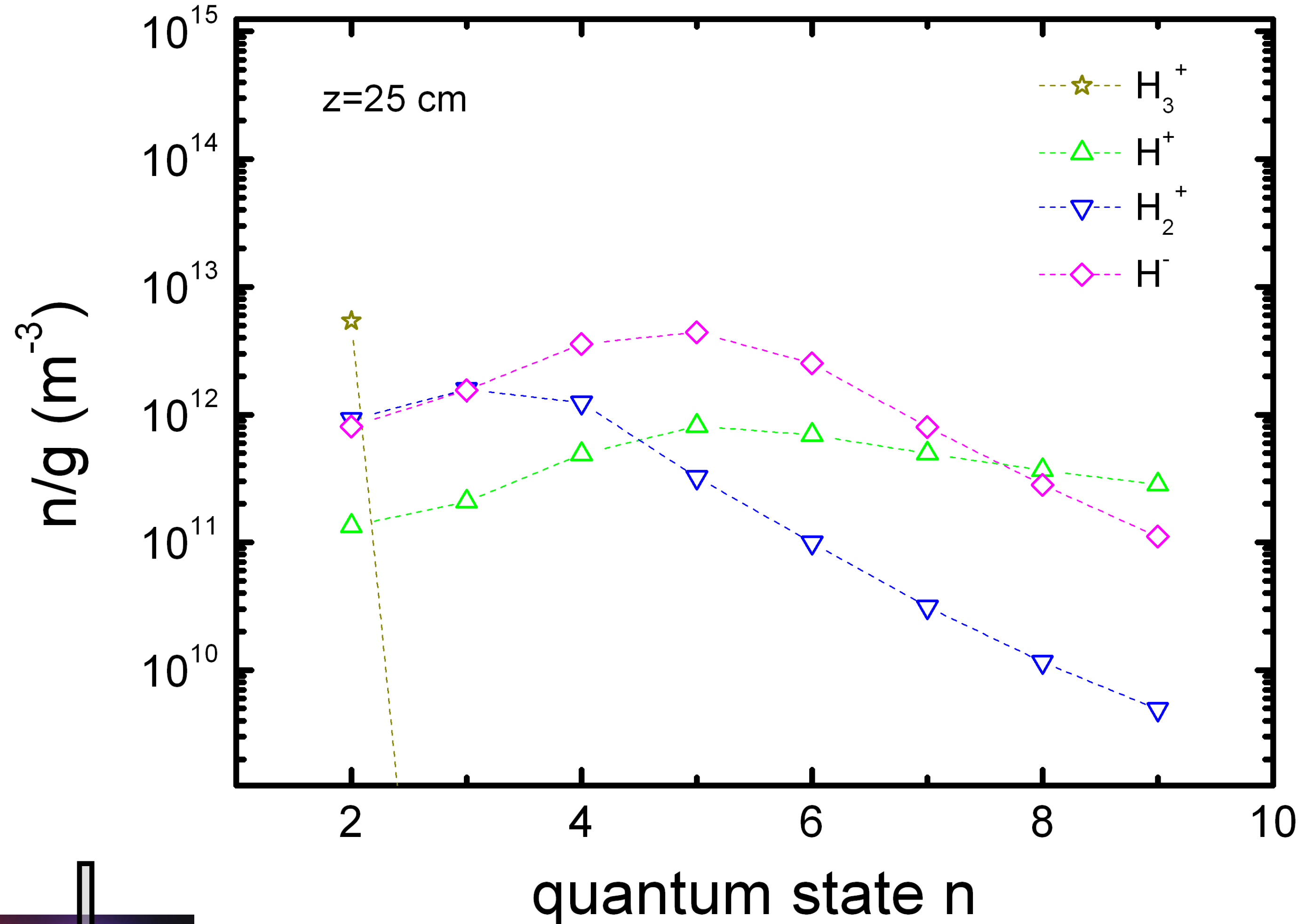


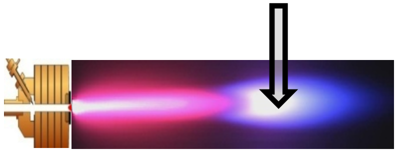
CRM results (z = 13 cm)





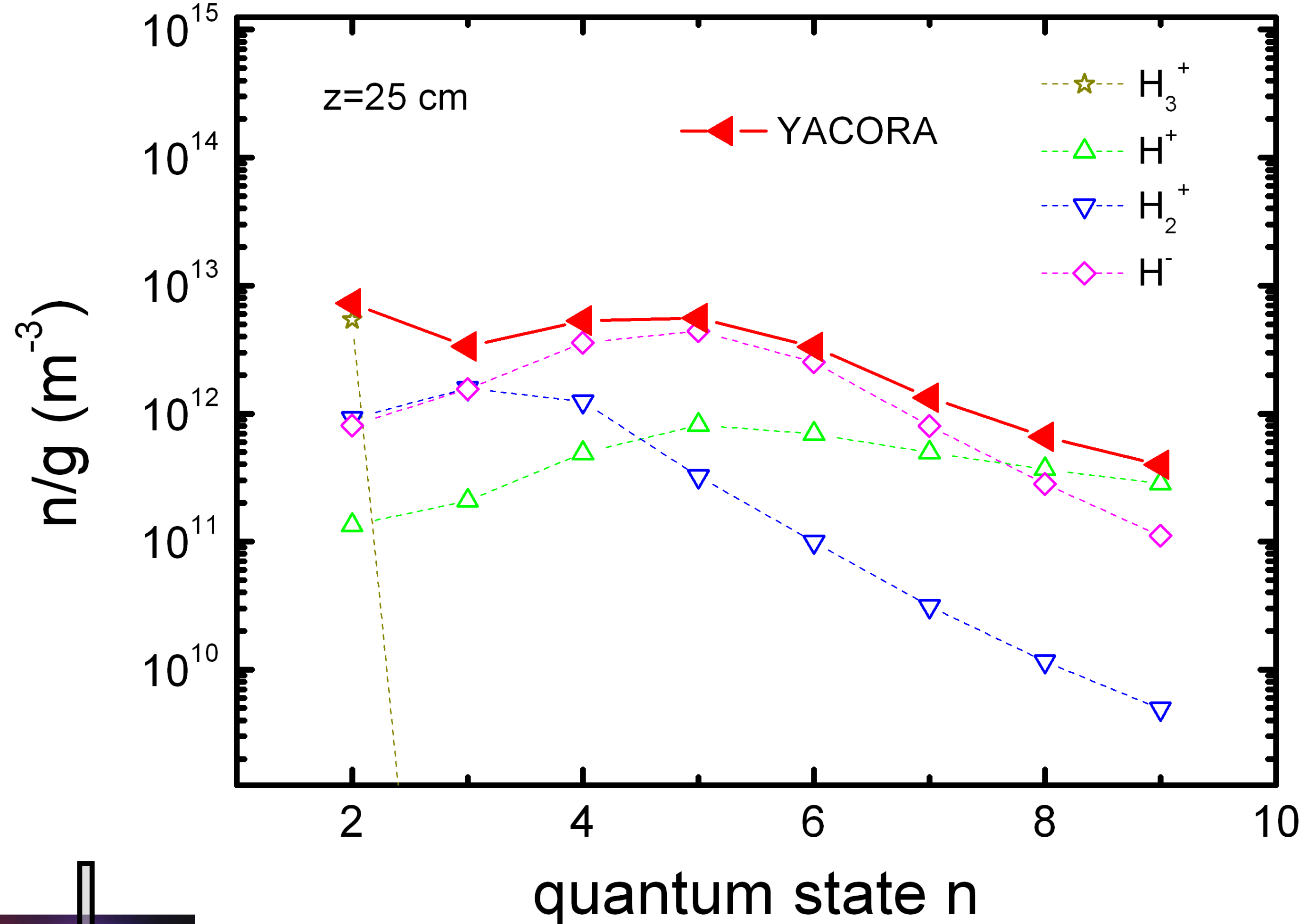
CRM results (z = 25 cm)

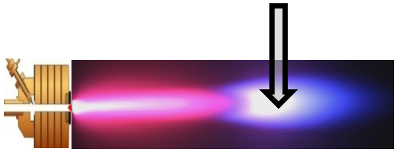




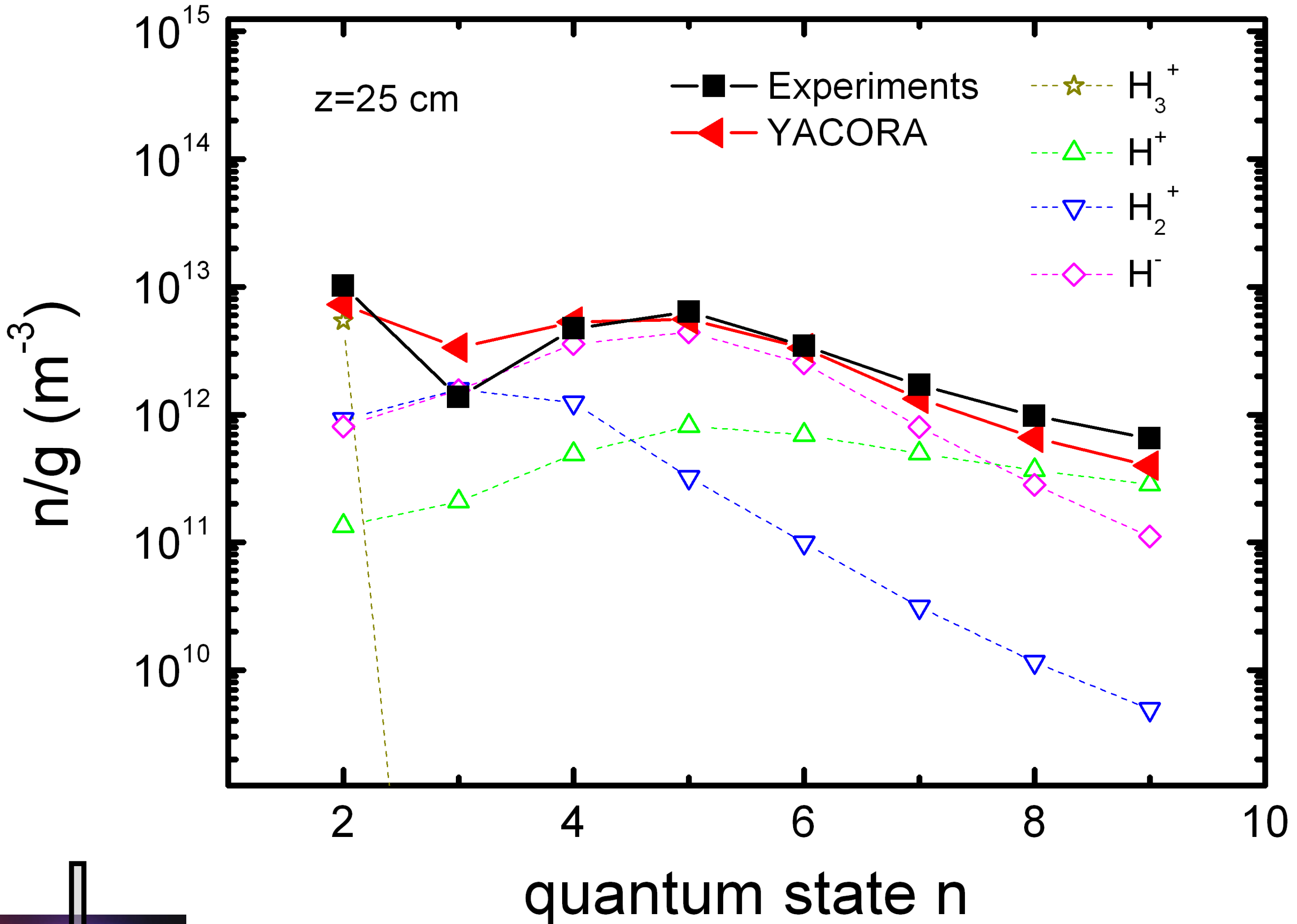
/ Plasma & Materials Processing

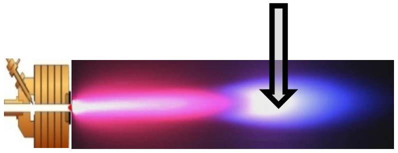
CRM results (z = 25 cm)





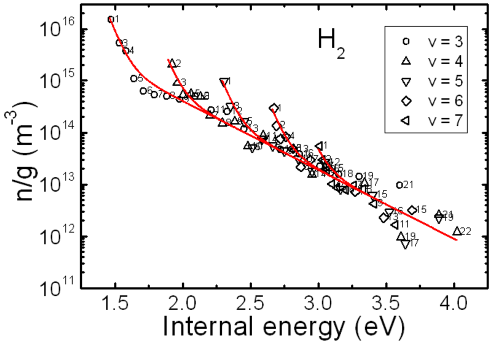
CRM results (z = 25 cm)

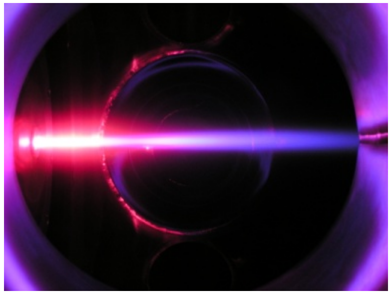




Conclusions

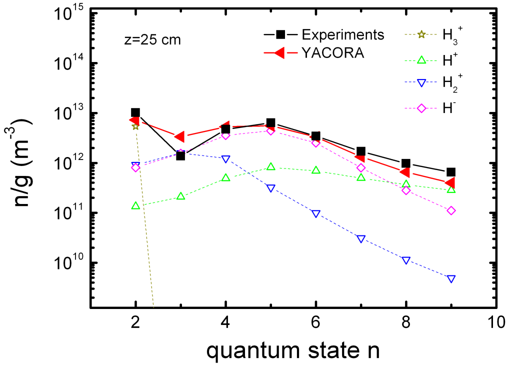
Presence of high ro-vibrationally excited hydrogen molecules proven by VUV-LIF





The red to blue transition in the expanding thermal plasma is explained by MAR processes

CRM results underpin the importance of MAR processes



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Reaction dynamics in a magnetized hydrogen plasma unraveled by optical spectroscopic techniques

Richard Engeln

Plasma and Materials Processing

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