Studies of hydrogen interaction with fusion relevant materials

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1. Introduction

- Plasma wall interaction in fusion reactor
- Hydrogen molecules in edge plasma

2. Vibrational spectroscopy of hydrogen molecules

- Experimental method
- Some results

3. In situ measurements of hydrogen distribution on the surface and near surface bulk by ERDA

- Experimental method
- Some results

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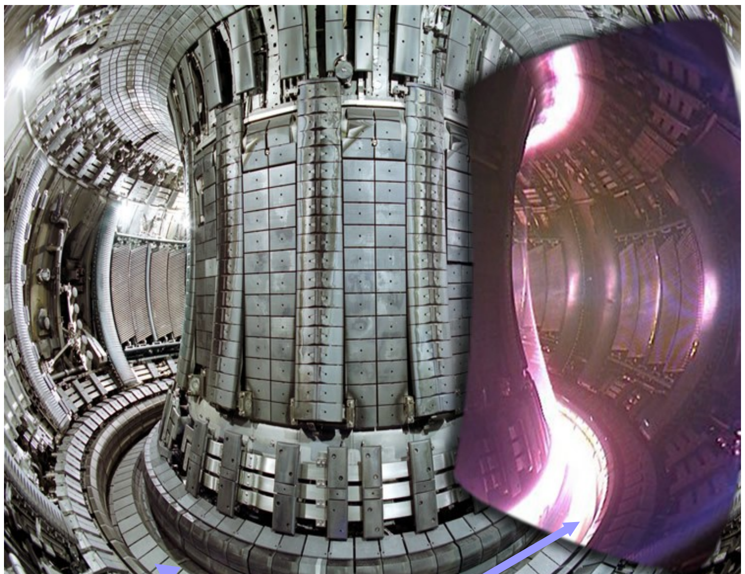
1. Introduction

1.1 Plasma wall interaction in fusion reactor

Plasma wall interaction is an important issue within current very dynamic fusion relevant research in Europe due to the ITER project.

Fusion research in EU is coordinated by EFDA (European Fusion Development Agreement) - **http://www.efda.org/**

Special EU task force for coordinating research in the field of fusion relevant PWI is organized by EFDA - **http://www.efda-taskforce-pwi.org/**



DIVERTOR, region where plasma is intentionally directed towards special targets in order to control plasma stability. It is also the pumping port for excess fuel and helium what is also needed for plasma stability.

Key issues regarding current activities on PWI – most of these involve to some extent hydrogen (H, D, T) interaction with surfaces

Special topics:

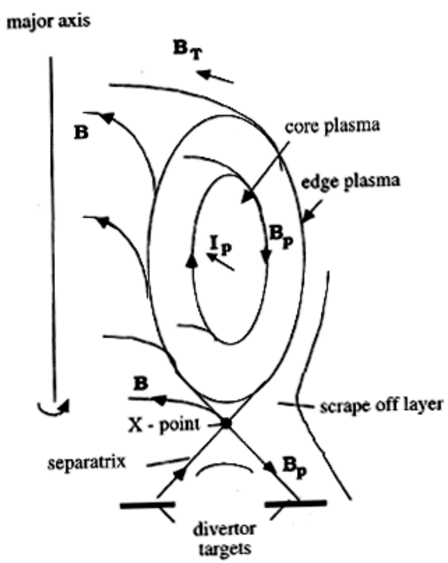
- Chemical Erosion and Transport
- High-Z Materials
- ITER Material mix – Be, W & C (ITER-like Wall experiment in JET)
- Transient Heat Loads
- Gas Balance and Fuel Retention
- Fuel Removal Methods
- Dust in Fusion Devices

But also:

- Edge plasma modeling
- Edge plasma and SOL physics
- PWI relevant diagnostics

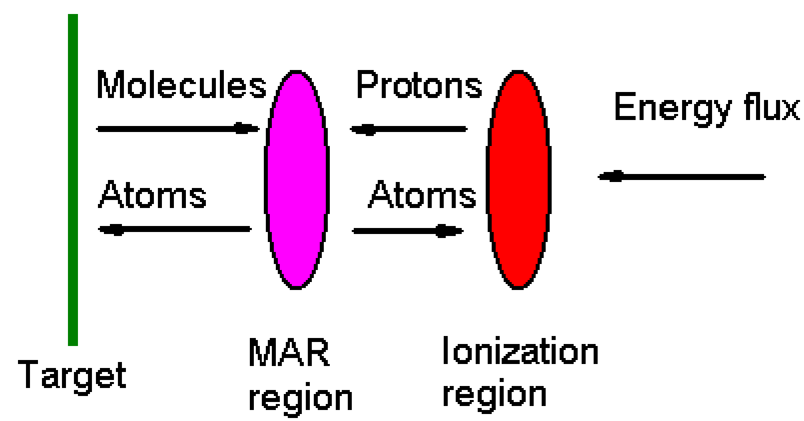
1.2 Hydrogen molecules in edge plasma

Neutral hydrogen molecules are present in near wall plasma and are mainly formed by wall-neutralization and recombination



Neutral atoms and molecules are very important for divertor plasma

- radiation from plasma
- energy load to target plates
- plasma detachment

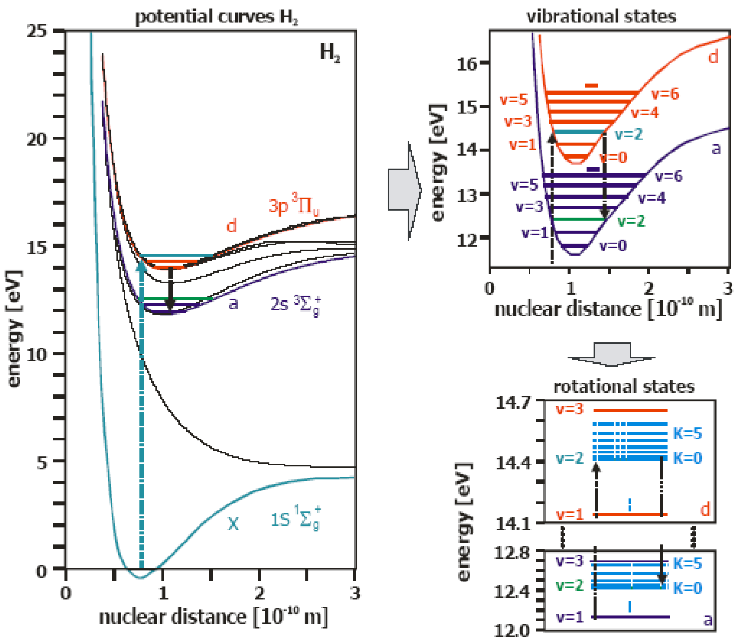


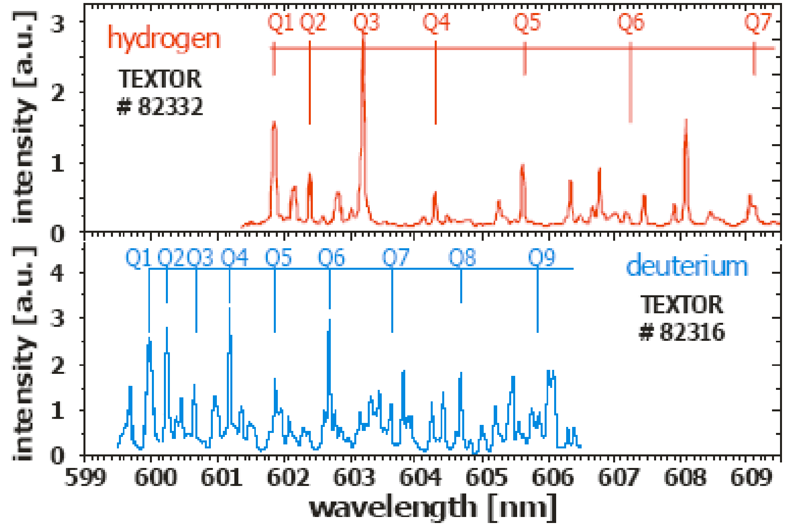
In divertor: $\mathbf{T_e}$ from 100-300 eV to < eV ;

n 10^{13} cm^{-3} – 10^{14} cm^{-3} - 10^{15} cm^{-3}

From Krasheninnikov, 2002

Optical spectroscopy of hydrogen molecules in fusion plasma





Diagnostic technique based on Fulcher band emission developed by U. Fanz et al.

It was shown that hydrogen molecules in edge plasma are ro-vibrationally excited

Vibrational/rotational temperatures in TEXTOR:

H_2	7140 K / 1078 K
HD	5950 K / 932 K
D_2	5270 K / 850 K

From Brezinsek *et al.*,2003

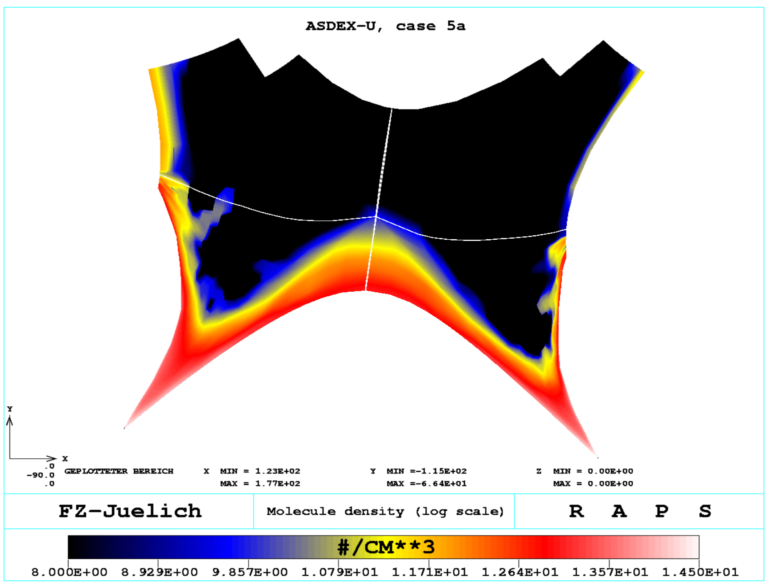
Self sustained neutral cushion?
B2-EIRENE simulation: MAR or MAD?

$p + H_2(v) \rightarrow H + H_2^+, \; e + H_2^+ \rightarrow H + H^* \rightarrow H + H$ (MAR)

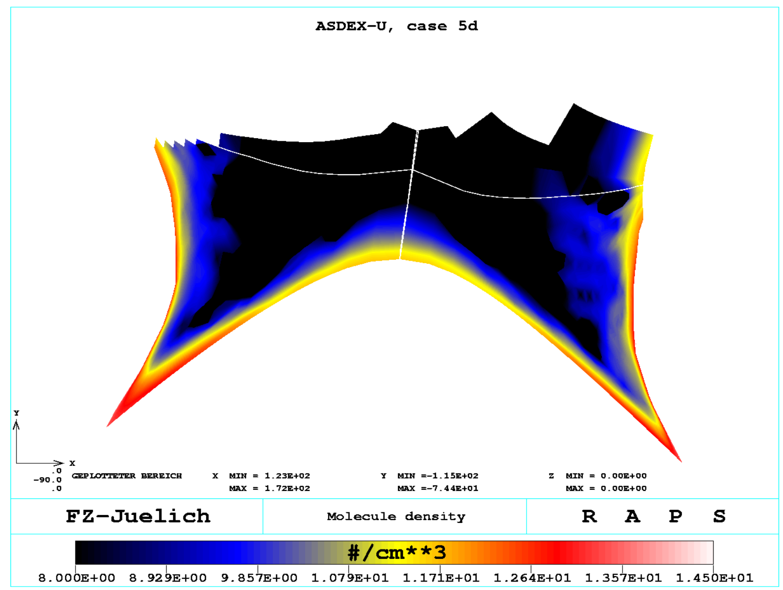
$p + H_2(v) \rightarrow H + H_2^+, \; e + H_2^+ \rightarrow H + H^* \rightarrow H + H^+ + e$ (MAD)

H_2 density field in Divertor:
Neutral gas "cushion" is strongly reduced by H_2 + p ion conversion

only $H_2(v=0)$



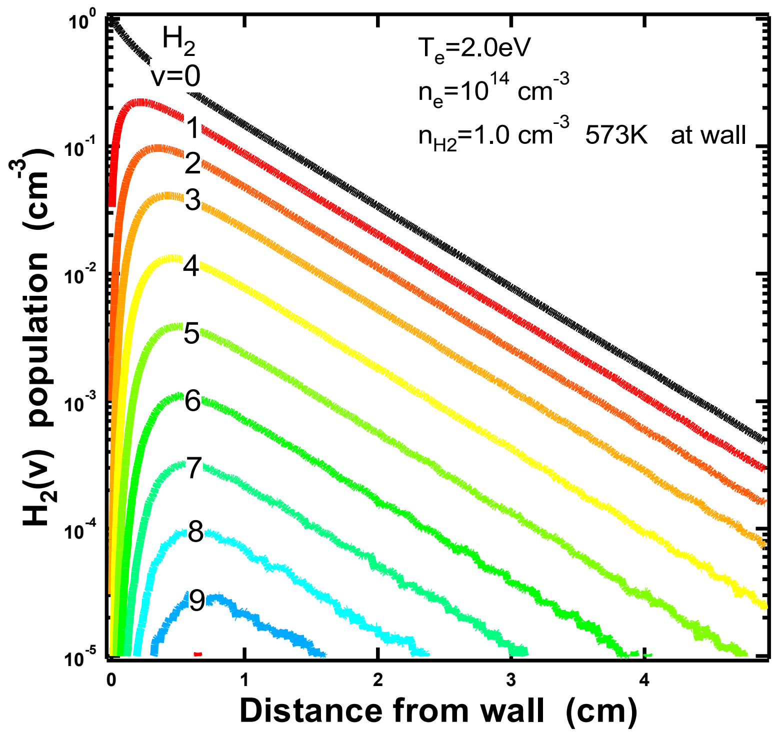
$H_2(v)$ included



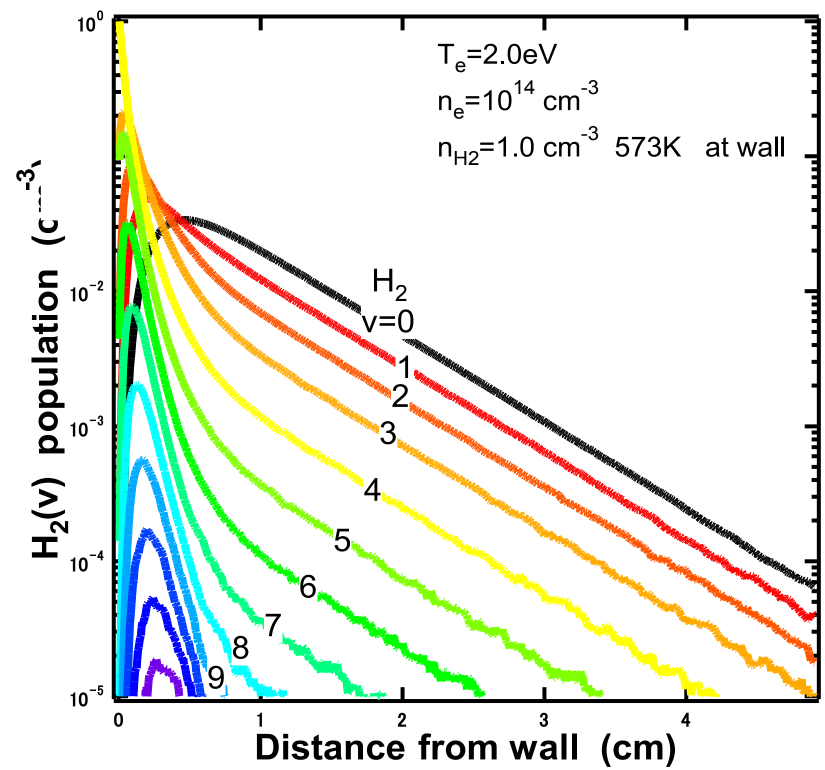
Kotov, Reiter in Sawada, 1st FZJ-JSI meeting on PWI, 28-29 September 2005

Sensitivity to surface produced vibr. excitation

1-D Monte-Carlo Model of Neutral-Particle Transport (K. Sawada)



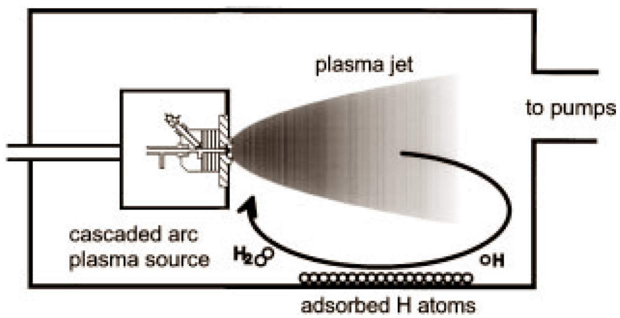
All H_2 from wall: v=0

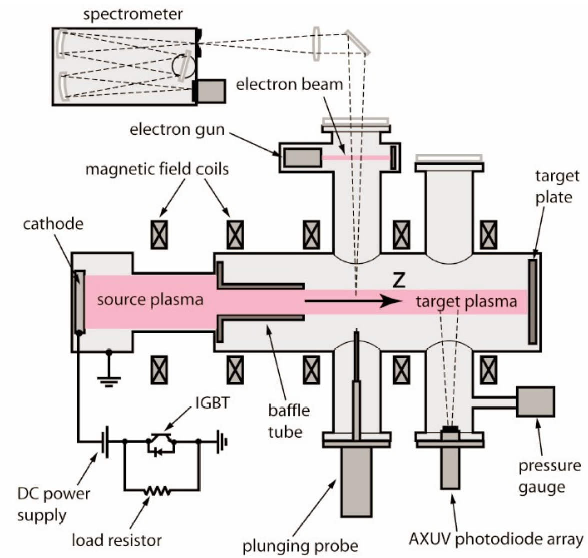


All H_2 from wall: v=4

Kotov et al., *ibid.*, 2005

Plasma cooling by hydrogen molecules

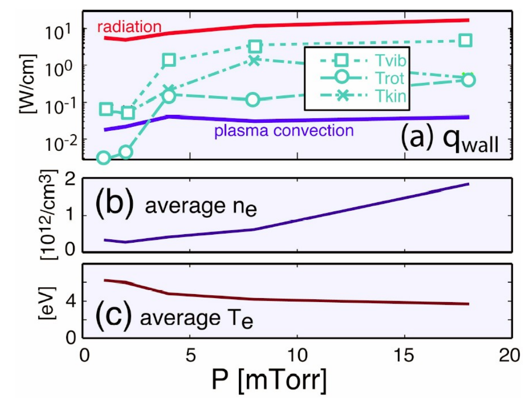




Influence of hydrogen molecules produced by wall-recombination on properties of expanding thermal arc plasma

From: Meulenbroeks et al., PRL, 76 (1996) 1840

More recently, experiments at PISCES-A have shown high importance of hydrogen molecules on plasma cooling. Values of 4500 K, 700 K and 600K were determined for T_{vib}, T_{rot} and T_{kin} respectively.



From: Hollmann et al., 33rd EPS 2006, P4.172

Hydrogen molecules (different isotopologues) that are vibrationally excited are important for PWI and edge plasma:

- "v" dependence of different binary CSs in plasma and therefore influence on the macroscopic phenomena such as divertor plasma detachment
- Contribution to the surface processes at the wall (e.g. chemical erosion, retention)
- Plasma cooling by neutral molecules

Sources of VEH molecules: edge plasma, desorption and recombination at plasma facing components and remote surfaces

From here on we will mainly present some results and current activities on the project P2 of SFA:

Interaction of vibrationally excited hydrogen with fusion relevant materials

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[3] Faculty of Electrotechnic, University of Ljubljana

[4] University Nova Gorica

Main goal:

Providing quantitative data on processes with vibrationally excited hydrogen molecules needed for modelling edge plasma and PWI and search for specific phenomena with these molecules.

Processes of interest:

- **- Vibrational distribution of molecules released from surfaces due to thermal desorption and recombinative desorption for different surface conditions (temperature, composition, impurities).**
  - Ratio of atomic to molecular species released from surface and its variation with surface parameters.

2. **Interaction of vibrationally excited molecules with plasma-facing materials:**
   - Change of vibrational distribution caused by interaction with surfaces,
   - Transfer of vibrational energy to the wall and its effects on erosion yields, and
   - Wall sticking probability for excited molecules.

3. **Binary collisions (volume processes, spectroscopy).**

Isotope effect in above processes is of key importance!

2. Vibrational spectroscopy of hydrogen molecules

2.1 Experimental method

Diagnostic technique is based on the detection of negative ions produced by the dissociative electron attachment (DEA) in hydrogen through the "4 eV" resonance state:

$$AB\,(X\,^1\Sigma_g^+, v) + e \rightarrow AB^-\,(X\,^2\Sigma_u^+) \rightarrow A^- + B$$

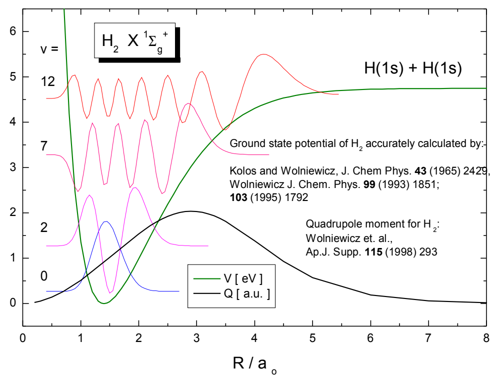
where A and B stands for any of hydrogen isotopes, H, D or T – only H and D are of our present experimental interest.

Method was originally developed in DIAM, Université Pierre et Marie Curie, Paris. Long-time collaboration with colleagues from DIAM: Dick Hall, Catherine Schermann, Francoise Pichou, Michel Landau and later Laurent Philippe and Eric Humbert and others is gratefully acknowledged as well as the loan of original experimental equipment for H2(v) spectroscopy to us by UPMC and CNRS.

4 eV DEA:

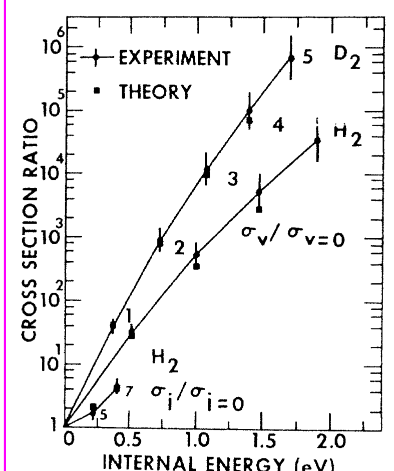
$$e + H_2 (X\ ^1\Sigma_g^+) \rightarrow H_2^-(X\ ^2\Sigma_u^+) \rightarrow H + H^-$$

- Very strong rise of CS with v,R excitation (CSs up to 10^{-15} cm^2 for high v)
- Vertical threshold → production of low-energy ions and displacement of thresholds
- Pronounced isotope effect for low v
- Extensively studied theoretically (benchmark case) but only few experimental studies

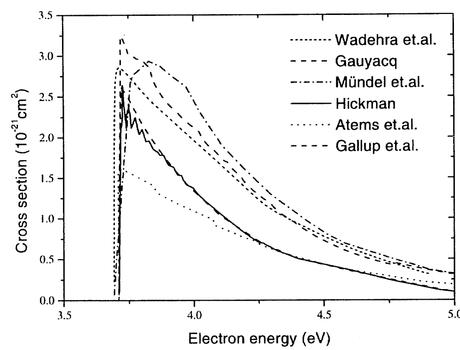


	Th. E.	CS [10^{-21} cm ^{2}]
H_2	3.71 eV	1.8
HD	3.75 eV	0.14
D_2	3.78 eV	0.009

EA_H = 0.754 eV



Alan and Wong, 1978

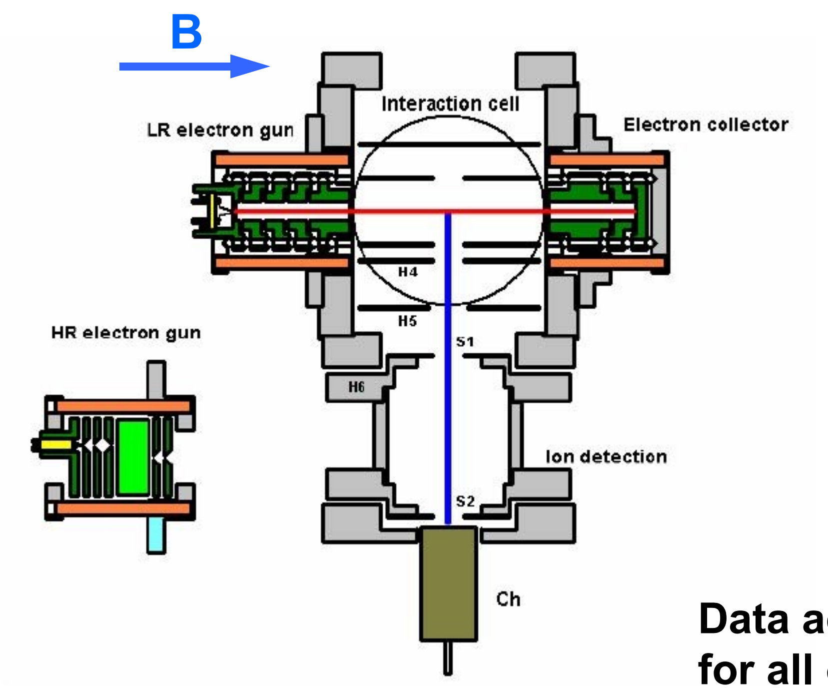


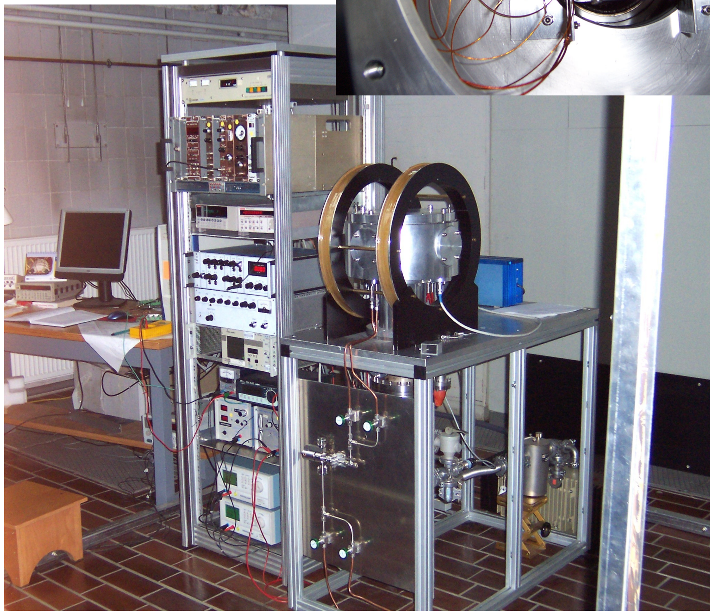
Theoretical CSs for H^-/H_2 - from Drexel et al., 2001

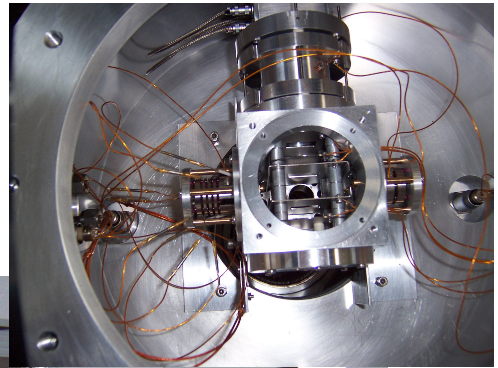
For experimental data evaluation we use the most recent and elaborate theoretical cross sections from Horáček et al. 2004

New experimental set-up

In a new set-up the guiding magnetic field for incident electron beam and ion extraction is used



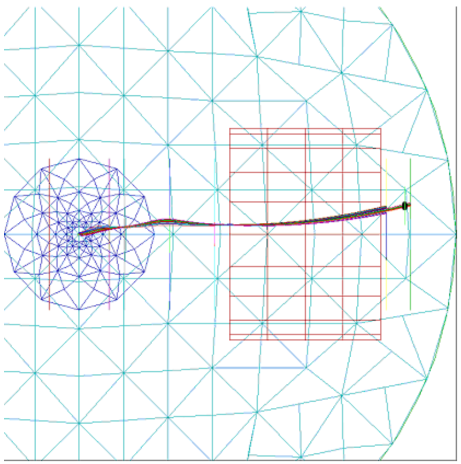




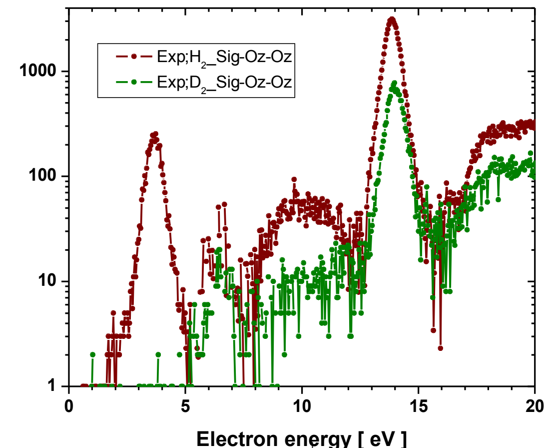
Data acquisition and control of experimental parameters for all our experiments are performed by special programs developed in LabView environment.

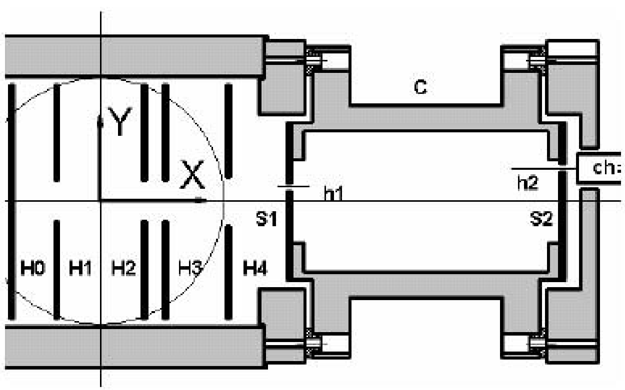
Original method for light (H^-, D^-) ion extraction was developed. This is of importance also for mass spectroscopy in general.

Main experimental raw result is ion (H^- or D^-) yield dependence on electron energy:

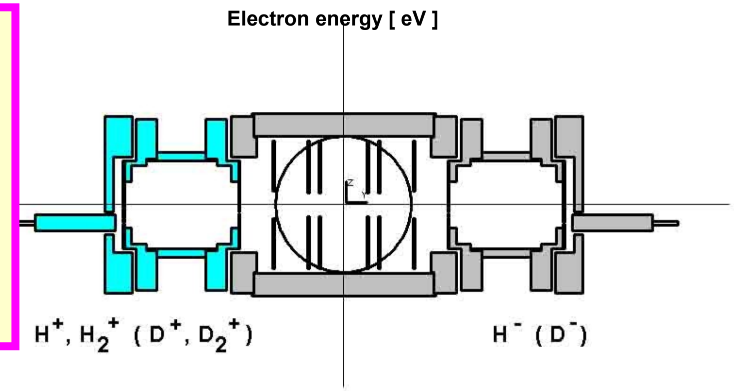


Efficient zero-energy ion collection.
Sufficient mass selectivity.

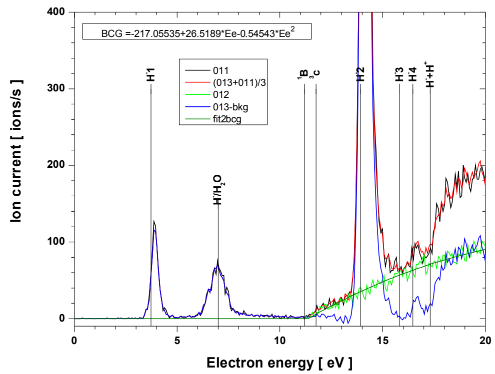


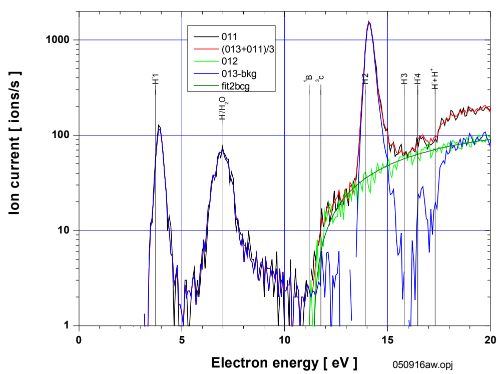


Planned to be added in 2007: Branch for positive ions that will allow determination of H/H_2 fraction determination simultaneously with vibrational spectroscopy.

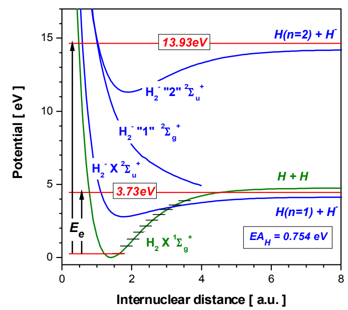


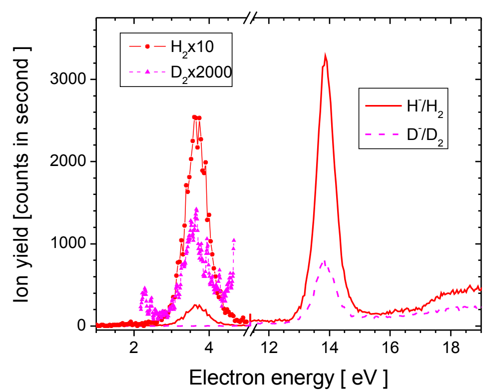
DEA with cold molecules





Overall ion spectrum (background!)





Isotope effect in DEA

14 eV process

Hydrogen isotopomer	Rapp et al. (1965) [10^{-24} m²]	Proposed [10^{-24} m²]
H_2	2.1	1.5
HD	1.5	1.0
D_2	1.0	0.44

4 eV process

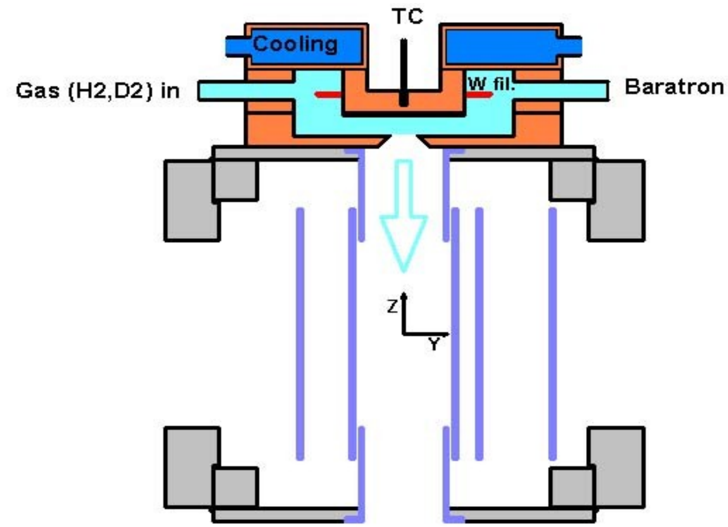
Hydrogen isotopomer	Sch&Asu (1967) [10^{-26} m²]	Proposed [10^{-26} m²]
H_2	16	12
HD	2.1	-
D_2	0.08	0.04

Proposed new CS for 4 eV and 14 eV DEA

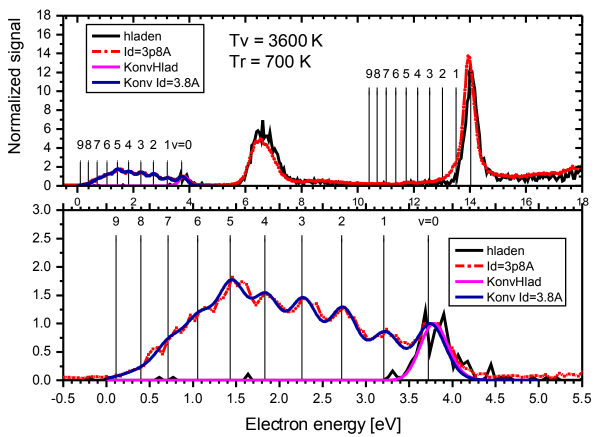
2.2 Some results

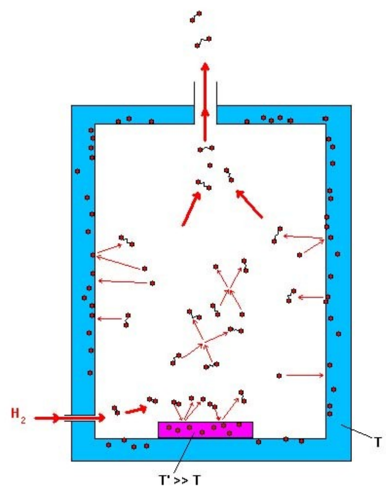
Source of $H_2(v)$ and $D_2(v)$

- Recombination of hydrogen atoms on surfaces of different fusion relevant materials (W, C, Ta)
- Hydrogen molecules dissociate on tungsten filament
- Vibrationally excited molecules are produced by recombination

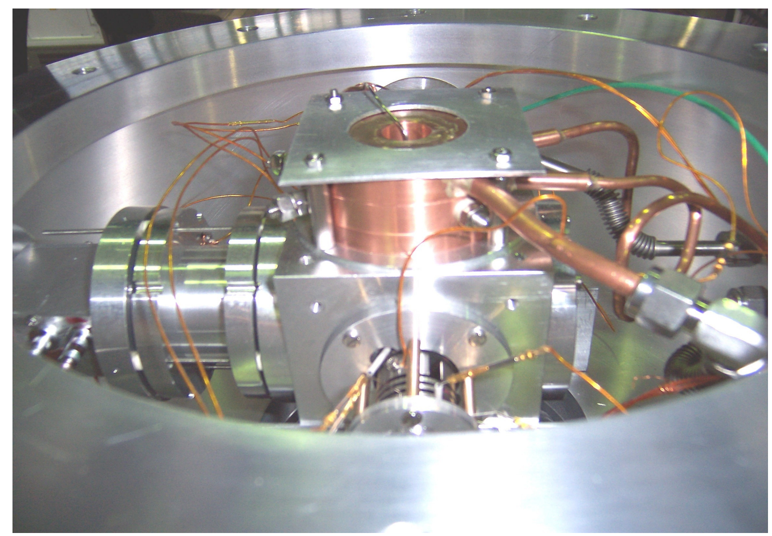


H_2/W





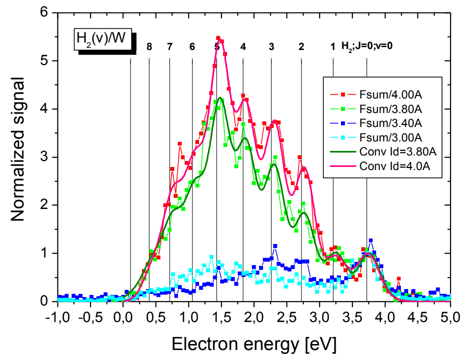
Modeling is of key importance for extracting numerical values for particular processes.



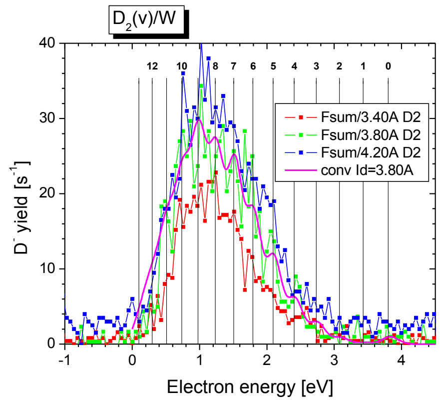
Production of vibrationally excited molecules by recombination on W

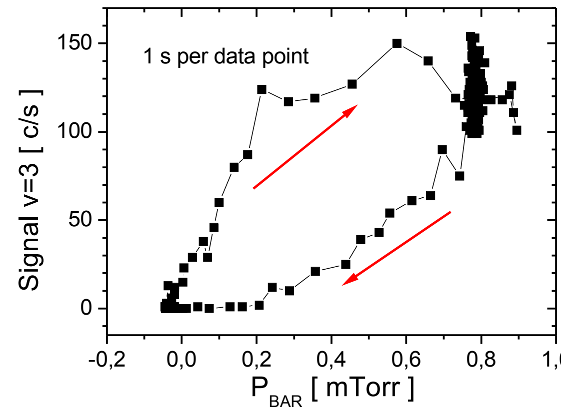
Pressure and temperature dependence of $H_2(v)$ production on W

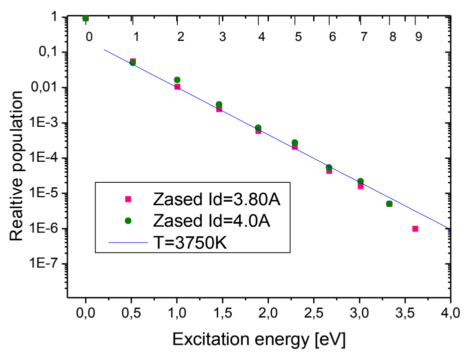
H_2

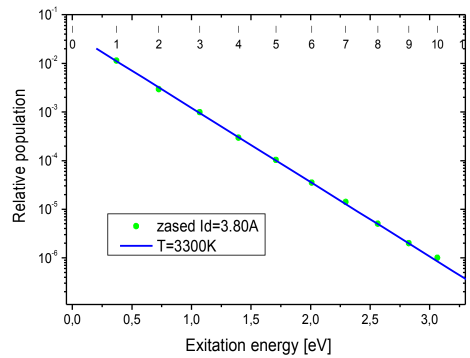


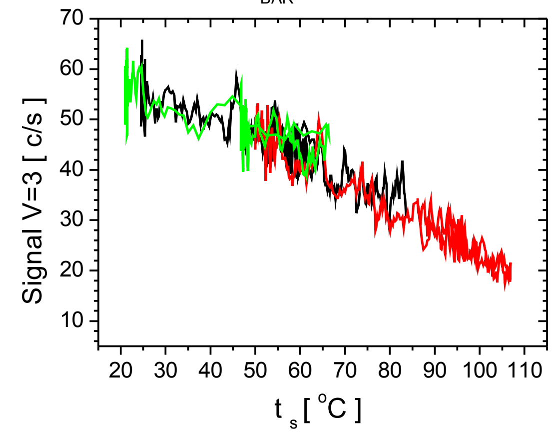
D_2





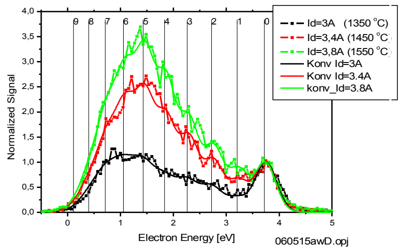




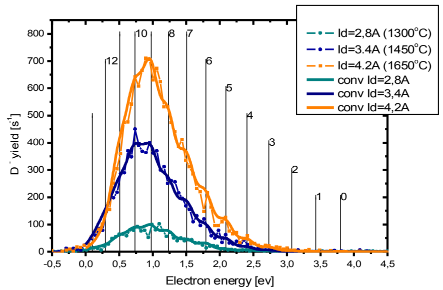


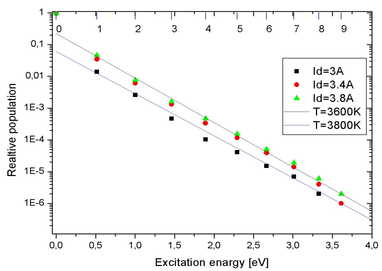
Ta

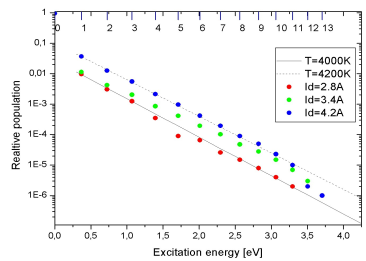
H_2



D_2



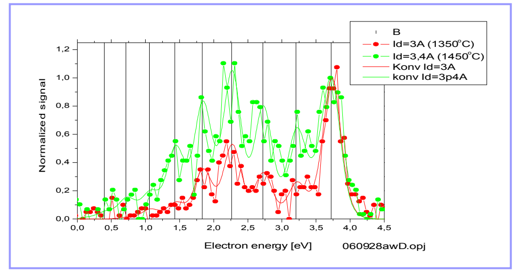


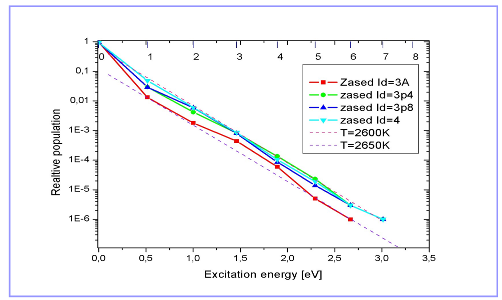


Our new results are qualitatively consistent with previous measurements in Paris

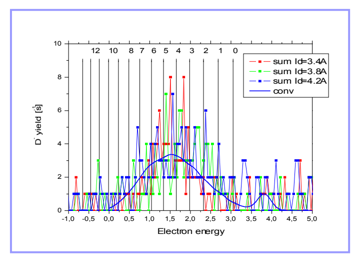
Cu

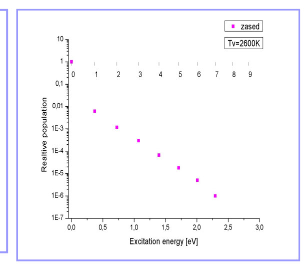
H_2



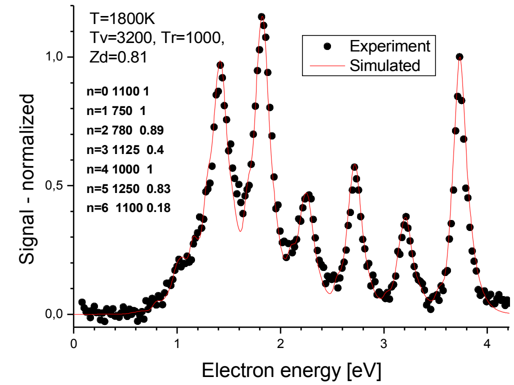


D_2 Almost negligible D^- production





Older results obtained at the first vibrational spectrometer in DIAM at Université Pierre et Marie Curie, Paris (in 90-ties) in collaboration with C. Schermann, R. I. Hall et al. (e.g. C. Schermann, F. Pichou, M. Landau, I. Čadež, R. I. Hall, *J. Chem. Phys*. 1994, *101*, 8152).



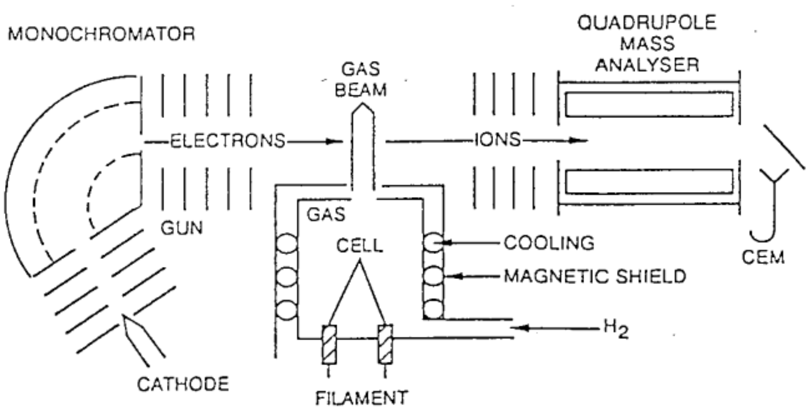
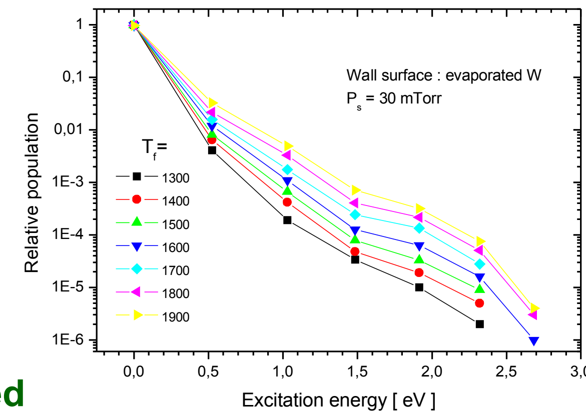


Figure 1. Schematics of the experimental set-up for determination of the vibrational population distribution in hydrogen.[14]

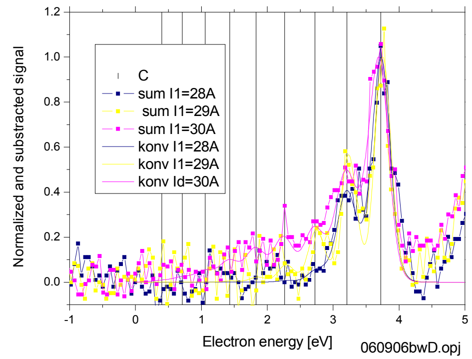


Previous measurements were performed with in situ evaporated tungsten. Older electrostatic experiment had better energy resolution but new one with guiding magnetic field has better e-beam control at very low energy.

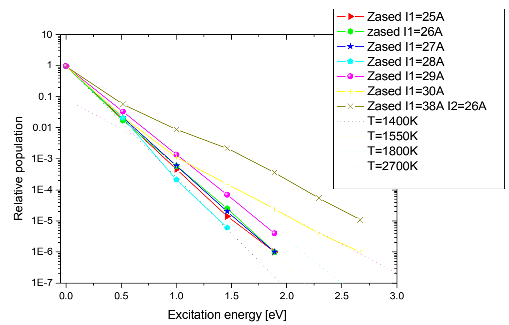
$H_2(v)$ from a hot W capillary

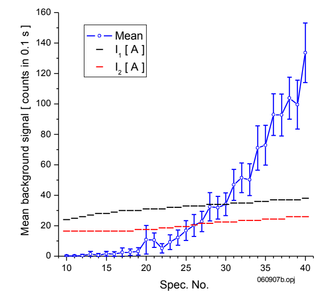
- **Vibrationally excited molecules from hydrogen atoms source:**

Experiment performed in collaboration with IPP, Garching (Th. Schwarz-Selinger)



Some problems appear due to the insufficient pumping speed of our vacuum system – improvements in progress



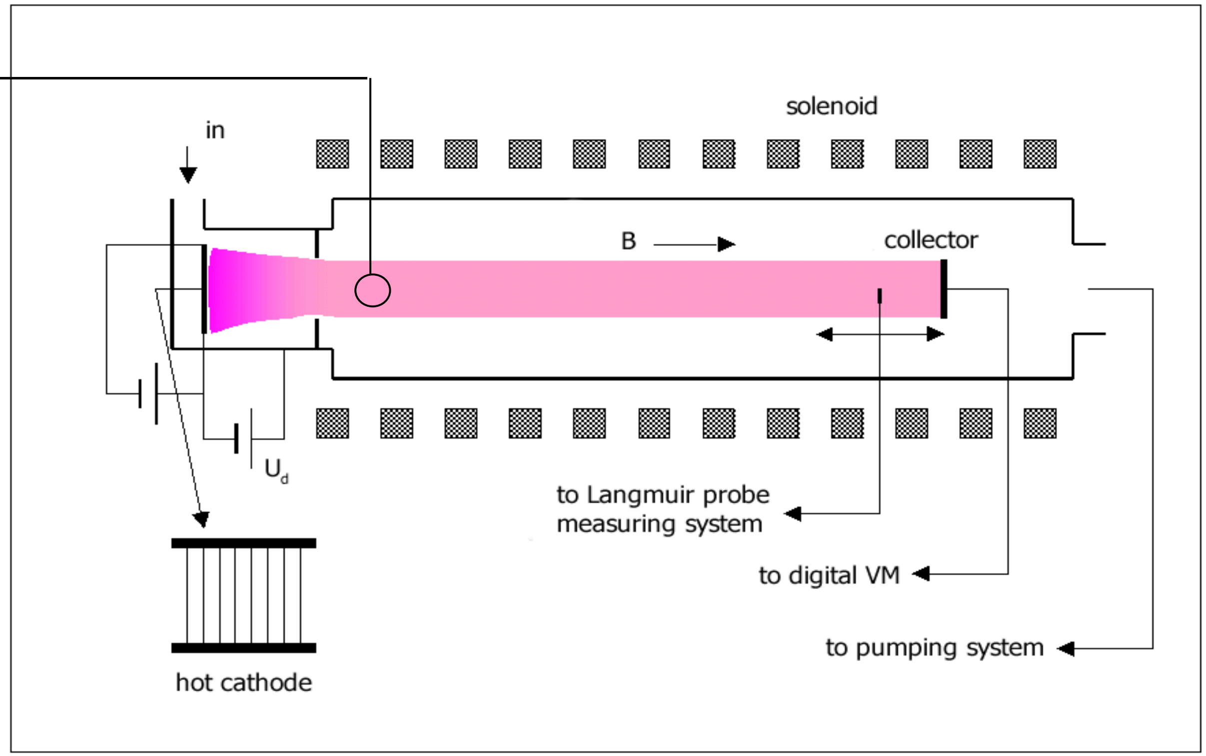


spec.#	I_1/A	I_2/A
10	24	16.5
11	25	16.5
12	26	16.5
13	27	16.5
14	28	16.5
15	28	16.5
16	29	16.5
17	30	16.5
18	30	16.5
19	30	17.5
20	31	17.5
21	31	17.5
22	31	18.5
23	32	18.5
24	32	19.5
25	33	19.5
26	33	20.5
27	33	21.5
28	34	21.5
29	34	22.5
30	34	22.5
31	35	22.5
32	35	23.5
33	35	23.5
34	36	23.5
35	36	24.5
36	37	24.5
37	37	24.5
38	37	26
39	37	26
40	38	26

Vibrational temperatures roughly correspond to the capillary temperature but some still unexplained nose appears on higher temperatures – secondary processes due to electron emission or metastable production?

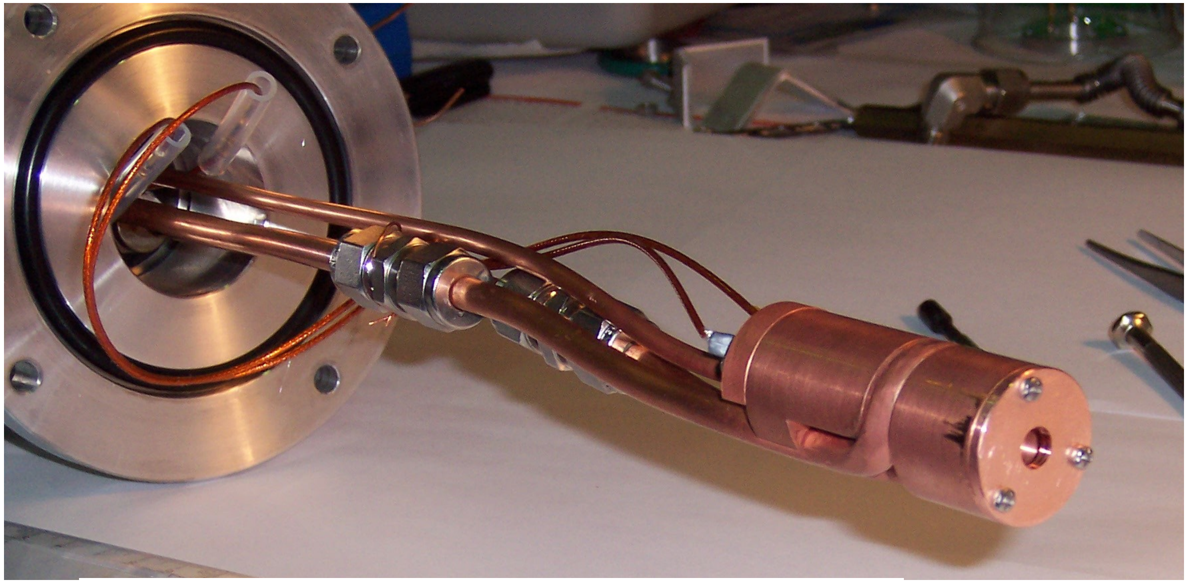
Influence of surface created vibrationally excited hydrogen molecules on optical emission from plasma

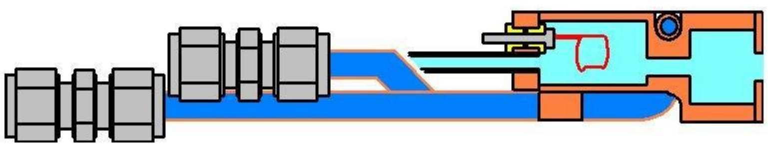
Linear magnetised plasma machine (LMPM) at JSI

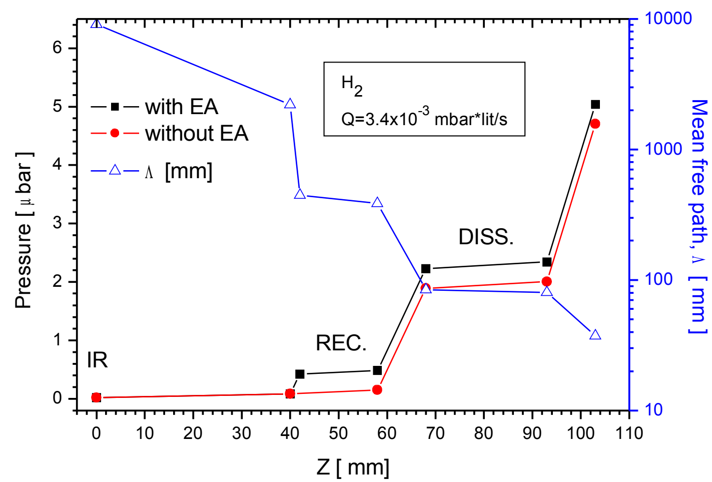


Main goal is to check to what extent wall created VEH molecules influence plasma emission in Fulcher band region.

Experiments performed in collaboration with Forschungszentrum Jülich (S.Brezinsek)



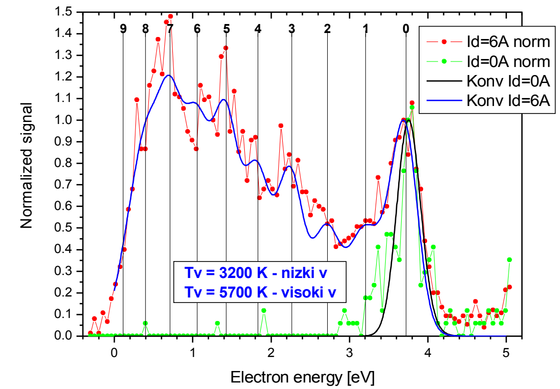


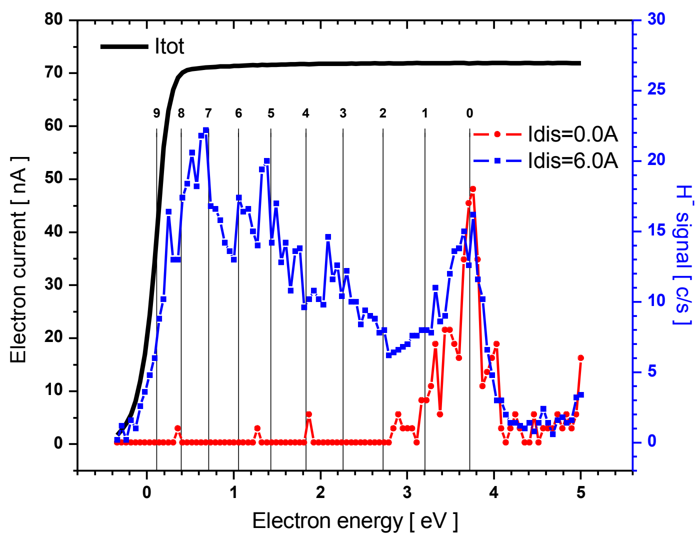


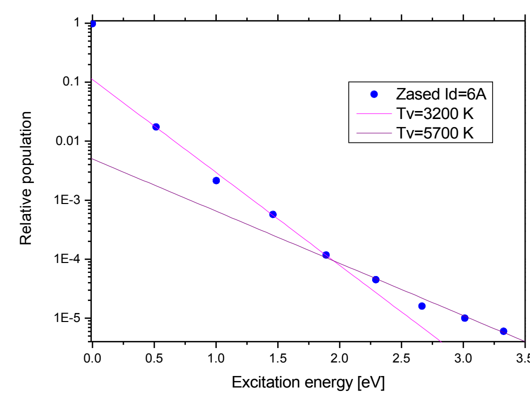
First chamber: ∅ 14 x 25 mm
Filament: ∅ 0.3 mm W
Channel: ∅ 4 x 10 mm
Second chamber: ∅ 16 x 15 mm
Exit Aperture: ∅ 0.3 mm
Material: OFHC cupper

Performances of the $H_2(v)$ source as determined by DTVE-B:

Hydrogen flow*: 3.4x10^{-3} mbar*lit/s
Filament temperature*: 2100 K
Vibrational temperature:
3200 K for v = 1 to 4
5700 K for v = 4 to 8

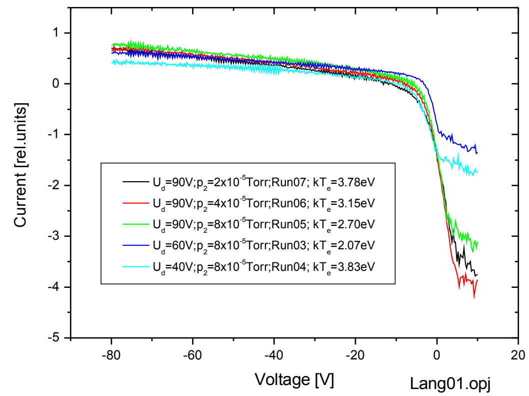




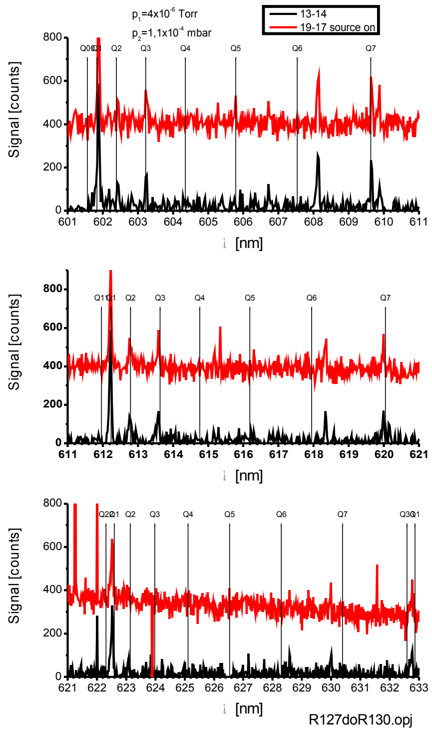


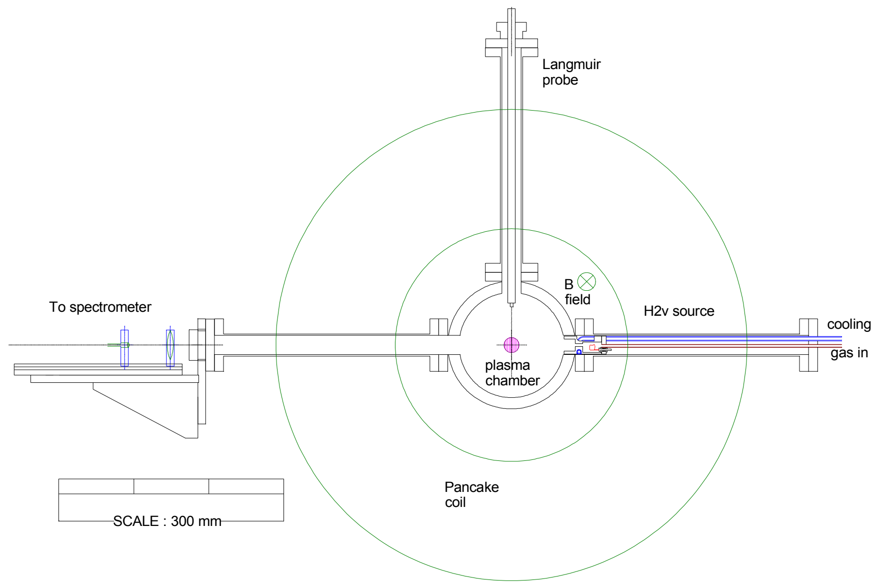
First measurements with H_2(v) source

First spectra were observed.
Experimental modifications are in progress:
- Optical fiber in vacuum
- 90° source mounting
- Reduction of pumping speed
- Inert gas + hydrogen

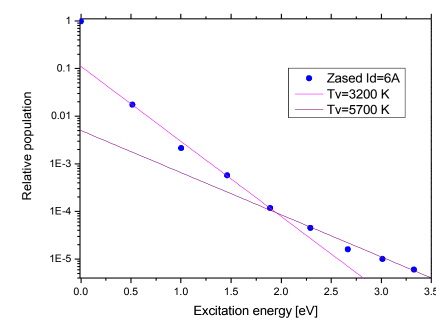


Langmuir probe diagnostics of plasma conditions





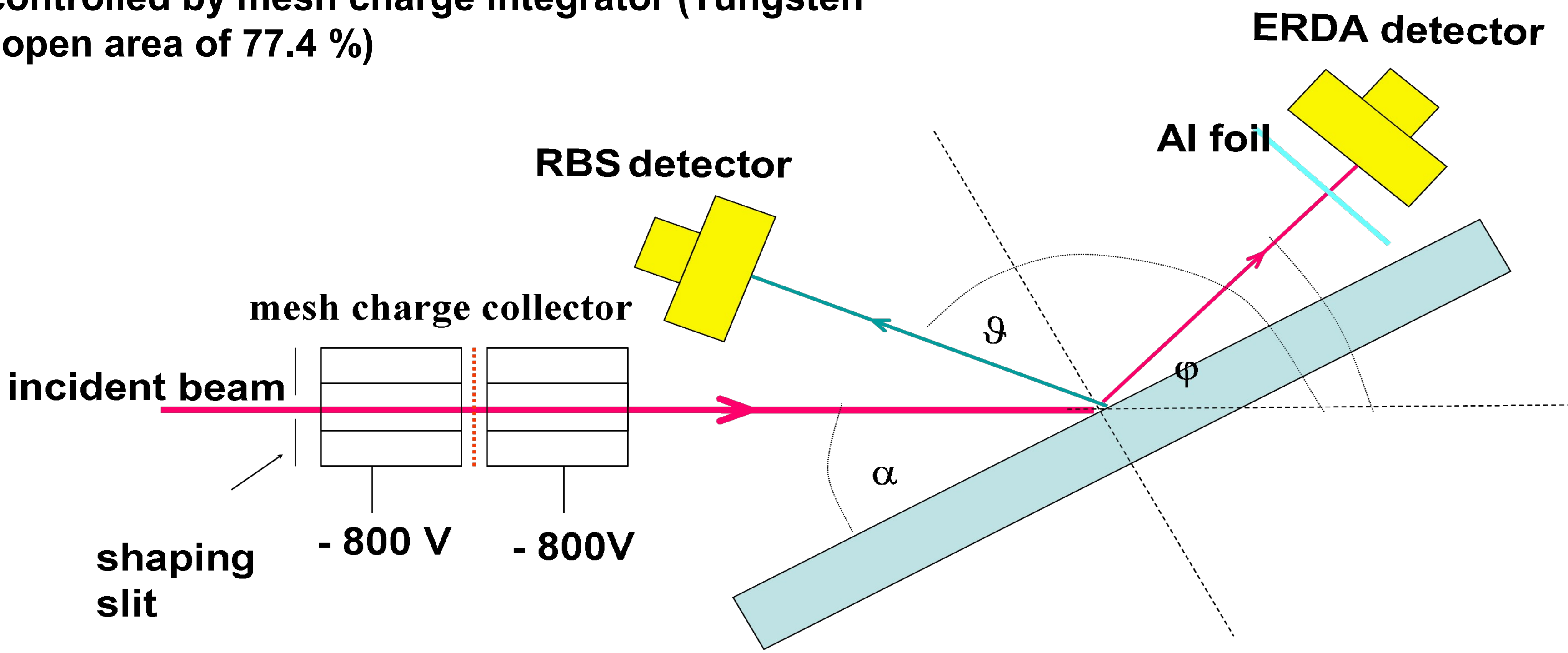
Performances of the H_2(v) source have to be improved!



3. In situ measurements of hydrogen distribution on the surface and near surface bulk by ERDA

3.1 Experimental method

Beam: 4230 keV $^{7}Li^{2+}$, Sample tilted 75°
RBS detector at 160°, ERDA detector at 30°
ERDA detector equipped with 11 μm Al foil
Dose controlled by mesh charge integrator (Tungsten mesh, open area of 77.4 %)



TOF ERDA is also available

ERDA for H and D with ^{7}Li ions, P. Pelicon et al., NIM B 227, 591 (2005)

2 MV HVEE Tandem accelerator ”Tandetron”

4 beam lines:

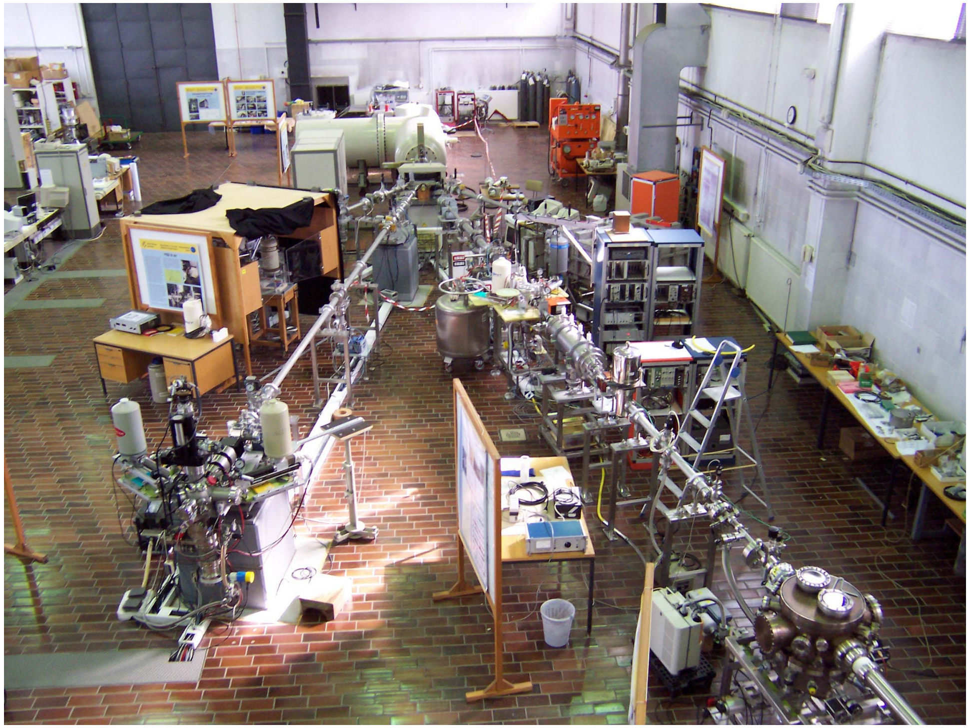
- External beam (PIXE, NRA)

- Micro-beam (PIXE/RBS/STIM/SE)

- PIXE/RBS and ERDA/TOF-ERDA

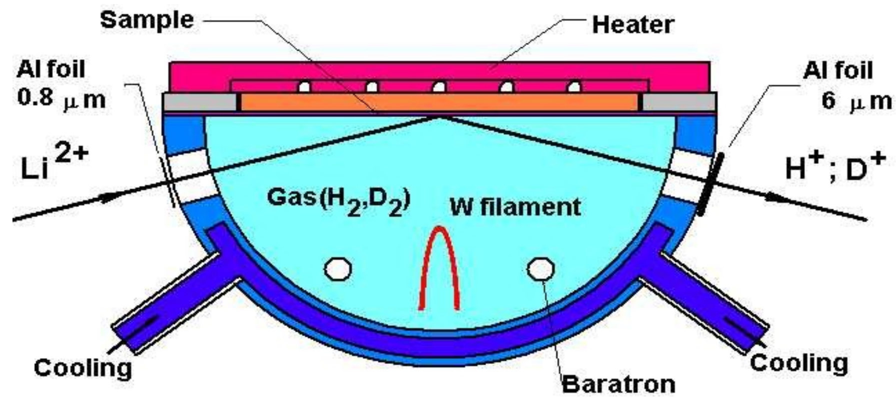
- High resolution X-ray spectrometer (atomic physics),

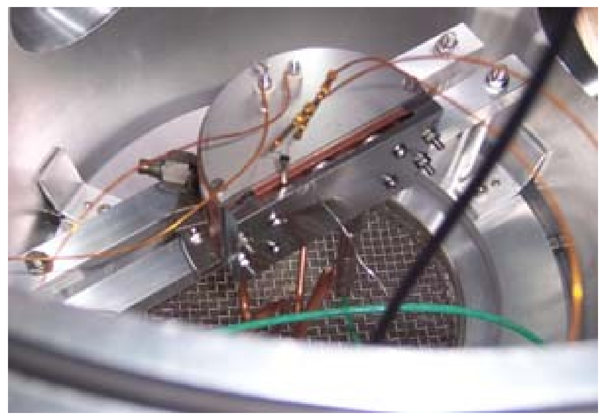
and two separate smaller experiments: electron coincidence spectrometer and hydrogen vibrational spectrometer.



Hydrogen exposure cell

- Samples exposure to controlled neutral hydrogen atmosphere
  - **rate of dissociation**
  - **vibrational distribution of molecules**
- Depth profile of H and D determined by ERDA at JSI 2MV tandem accelerator
- Main interest:
  - H and D depth profile in W, C and other materials
  - interaction of neutral particles (H_2, H, D_2, D) with materials
- Project goal are quantitative data for edge plasma modeling

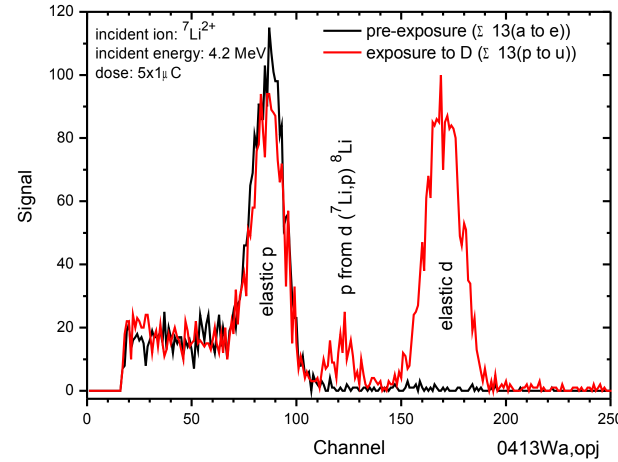




3.2 Results

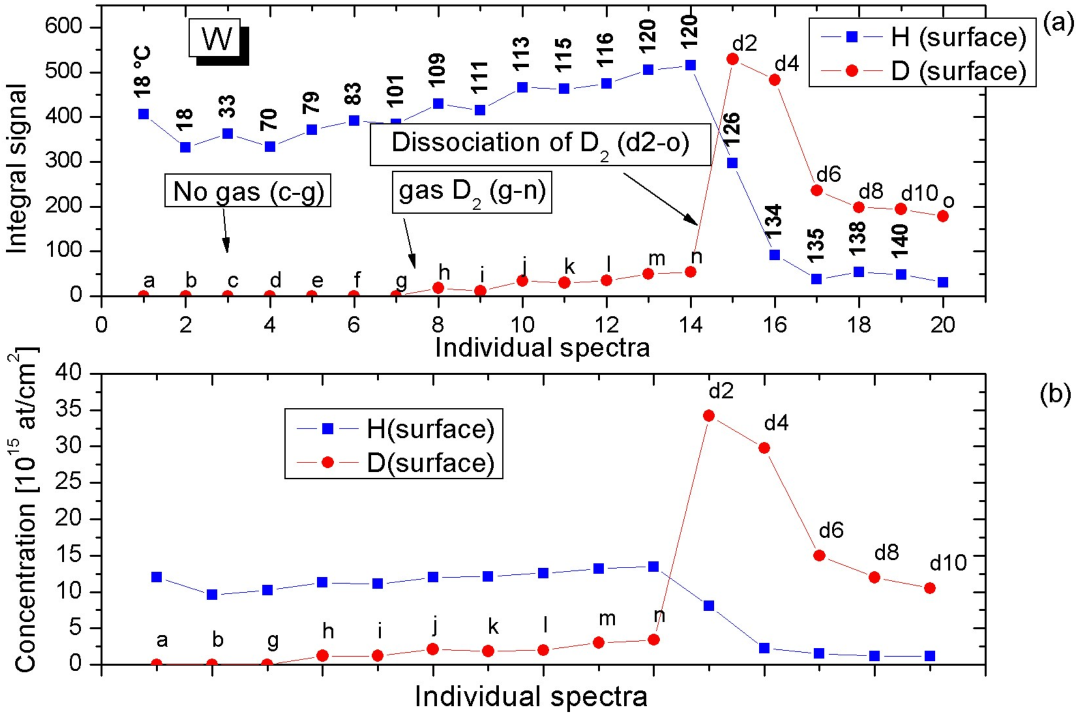
By changing conditions in HEC we are getting experimental evidence on the following processes:

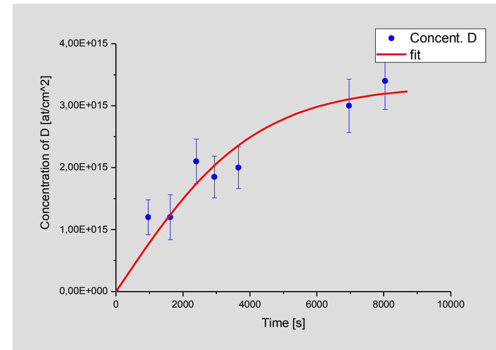
- Chemisorbtion – sample exposed to H_2 or D_2
- Adsorbtion of D (H) on the surface
- Abstraction of H (D) by D (H)
- Desorbtion of HD, H_2 and D_2 molecules
- Diffusion surface-bulk



Absolute concentration evaluation from experimental spectra is performed by SIMNRA (M. Mayer)

Due to low solubility for tungsten hydrogen is mainly present at the surface. However, hydrogen content in the below surface bulk depends on the exposure history of the sample – very slow process.





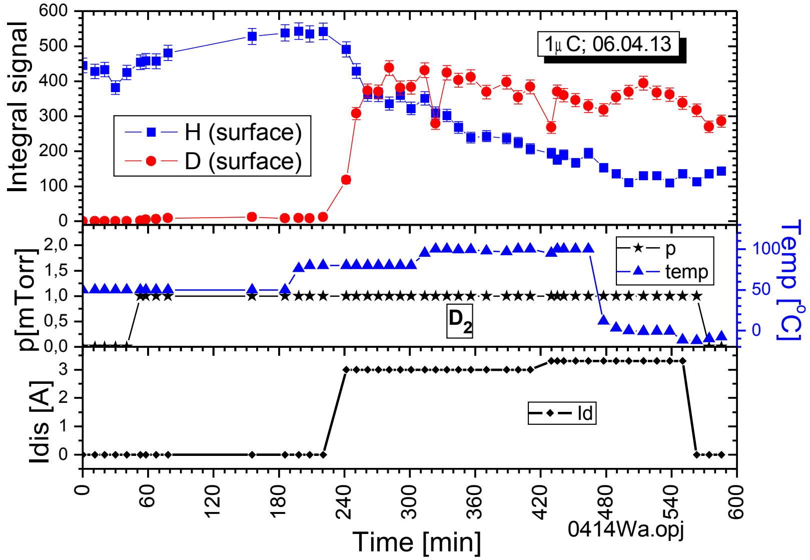
$$\theta(t) = \sqrt{\frac{S\gamma}{k^{\theta}}}\left(\tanh(2\sqrt{S\gamma k^{\theta}}\, t + C_1)\right)$$

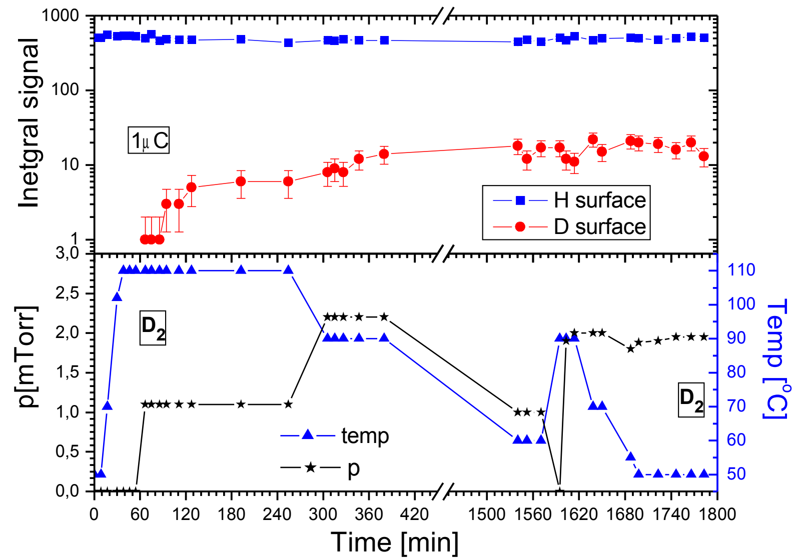
$S = 2.1*10^{-7} \pm 0.6*10^{-7}$

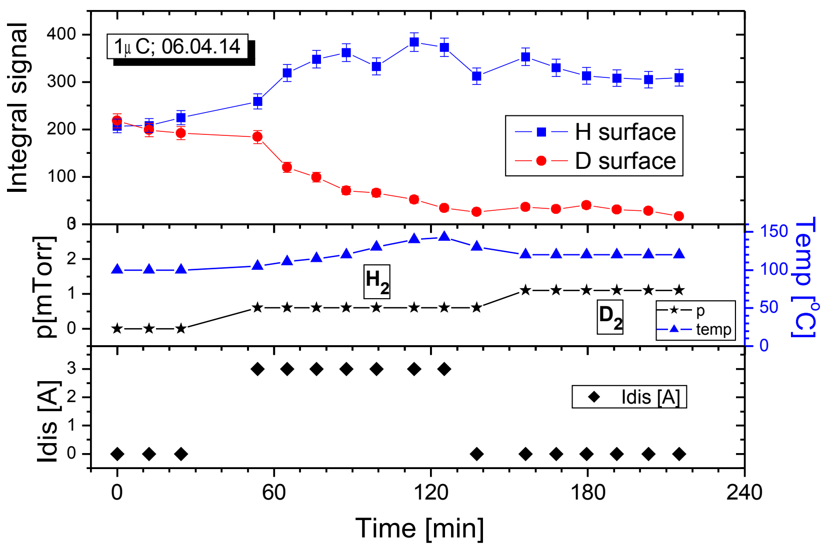
$k^{\theta} = 3.6*10^{-20}$ cm^2/s $\pm\ 2.4*10^{-20}$ cm^2/s

Detailed study of time evolution of surface H and D concentration in two HEC arrangements and for some different state of the sample was performed.

S.Markelj at al., NIM B, 2007a, in press



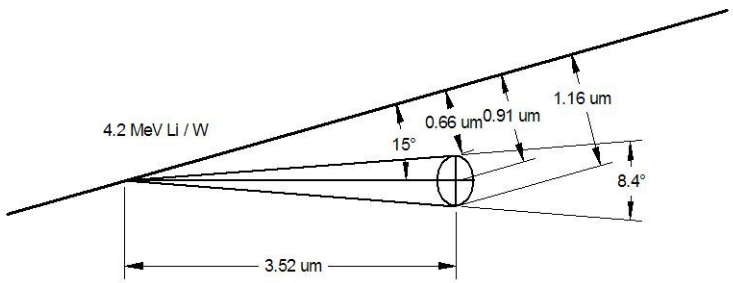




Few more examples obtained with DD version of HEC – W - sample surface is not exposed to dissociation filament

Problems related to the present status of studies with HEC

“Nondestructive” IBA method
ERDA is not that nondestructive:
- sample heating
- projectile implanting
- build-up of vacuum oil deposit



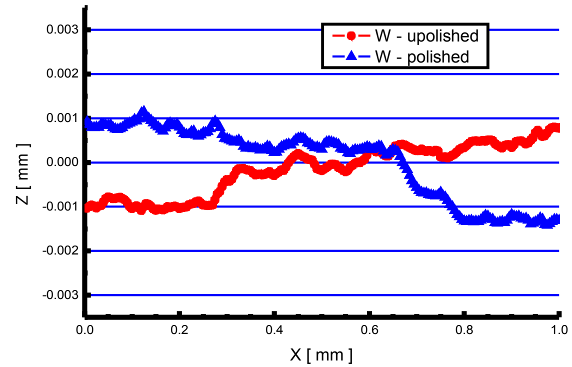
power deposited on the target under present beam conditions (5nA of 4.2MeV $^{7}Li^{2+}$ over 4mm x 4mm):

10 mW in 14 mm^3 !

Number of implanted ^{7}Li during continuous 1 h irradiation:

5.6×10^{13} ($n_W = 6.3 \times 10^{16}$ at/mm^3)

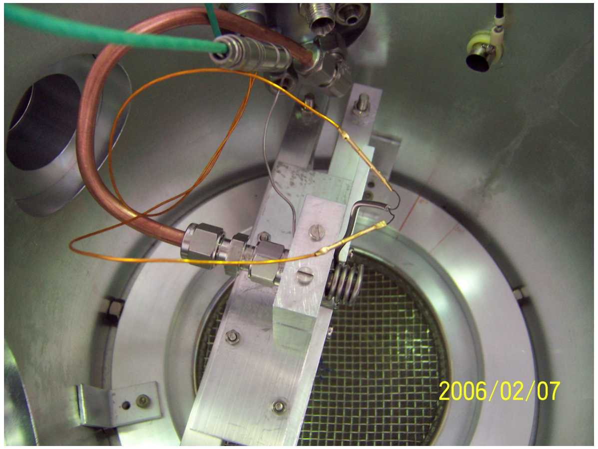
Surface roughness



Needed elastic CS for ^{7}Li on D scattering is not available – for the time being we can only use Rutherford CS for quantitative determination of D.

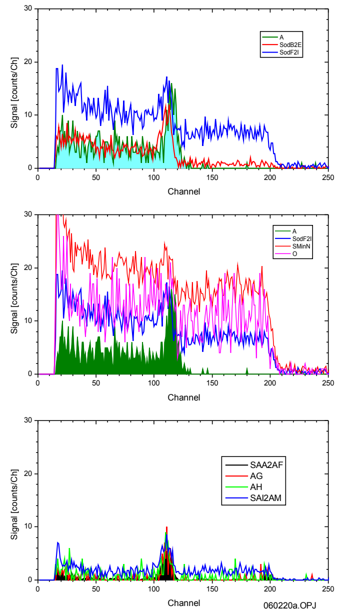
Contribution of nuclear reaction $^{7}Li(d,p)^{8}Li$ to the evaluation of D-depth profile.

Hydrogen permeation through Pd (and Ta)

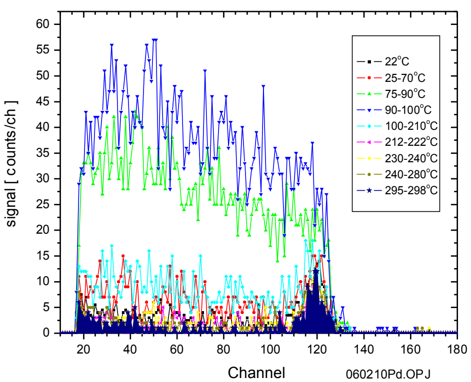


Z ERDO

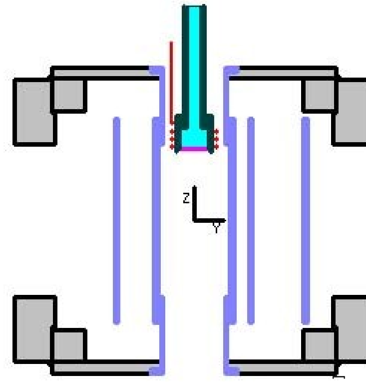
D/Pd



H/Pd

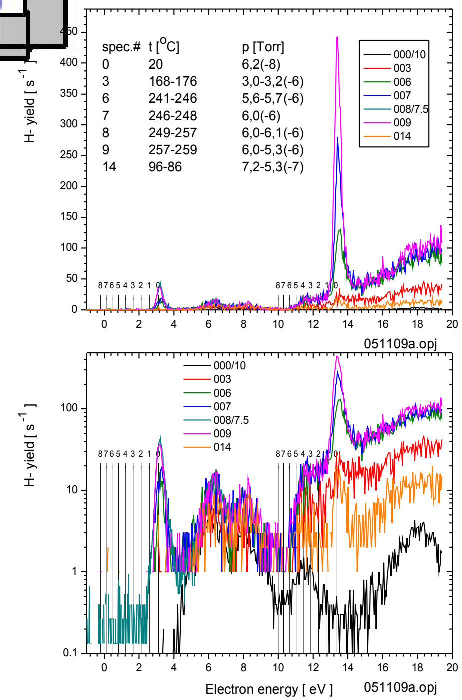


S. Markelj at al., NIM B, 2007b, in press



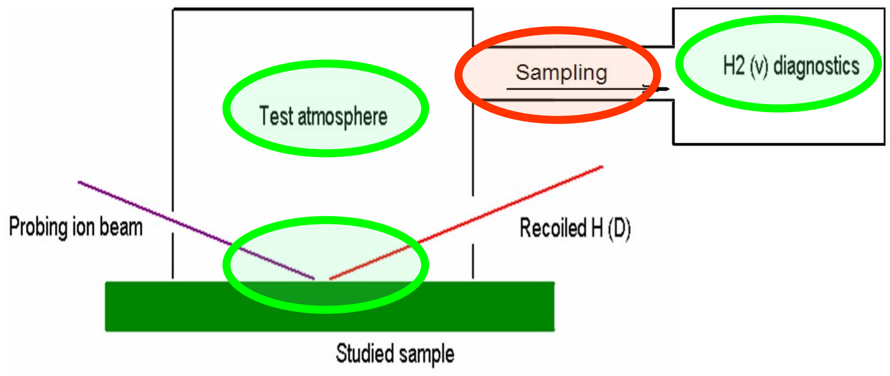
Z DTVE

We have not observed any vibrational excitation of H_2 molecules created by recombination after permeation through Pd and PdAg alloy



4. Concluding remarks

Our final experimental goal is to integrate IBA surface analysis with vibrational spectroscopy:



- Vibrational spectroscopy (can and will be used in some other experiments as well)
- developed and available (…)
- Atom/molecule ratio (+ion branch). – to be developed and tested
- Test atmosphere – partially dissociated neutral hydrogen gas or low temperature hydrogen plasma. – partially available but still to work
- In situ and in real time H and D depth profiling by Li ERDA. – developed and available (…)
- Sampling. – to be developed and tested

- **There are interesting and important phenomena with neutral vibrationally excited hydrogen molecules in fusion edge plasma. Processes involving neutrals are going to be even more important for ITER divertor due to the size scaling. Some of these problems are also important for astrophysics and technological plasmas.**

- **With new experimental methods we are starting to study some particular processes with the main goal to acquiring quantitative data needed for modelling.**

- **Active international collaboration is of crucial importance for efficient and prompt search for the appropriate answers. We are looking forward to collaborating with interested laboratories.**

- **Other subjects in the field of PWI that we are dealing with in SFA are application of IBA methods, hydrogen retention, plasma cleaning and development of new materials.**