### Les molécules H<sub>2</sub>(v") formées en surface dans les sources d'ions négatifs

M. Bacal, A.A. Ivanov Jr, J. Breton, Ecole Polytechnique, France

M. Glass-Maujean, Université Paris-VI, France

M. Nishiura, National Institute for Fusion Science, Japon

M. Sasao, Tohoku University, Japon

M. Wada, Doshisha University, Japon

20-23 Mai 2007, GDR ARCHES

Observatoire de Paris

### $H_2(v'')$ are the precursors of the negative ions in plasmas

Two important mechanisms for the production of  $H_2(v'')$  in electronmolecule collisions in the plasma volume have been identified:

a) radiative decay from singlet electronic states excited by collisions with energetic electrons
 (E-V);

b) collisions of ground state molecules with low energy electrons, through the  $H_2^-$  resonance (e-V).

A new mechanism due to recombinative desorption of molecules from surfaces is proposed, following experiments indicating the role of surfaces on negative ion density.

#### Production of vibrationally excited molecules

Vibrationally excited molecules are strongly related to H<sup>-</sup> ion production. They are produced by

- e-V
  - $H_2 + e (slow) \rightarrow H_2(X^1 \Sigma_g^+, v'') + e$ followed by the dissociative attachment of Eq. (1)
    (4)
- E-V

$$H_2 + e \text{ (fast)} \rightarrow H_2(B^{1}\Sigma_u^+, C^{1}\Pi_u) \rightarrow H_2(X^{1}\Sigma_g^+, v'') + \mathbf{hv} \quad (5)$$

Cascade

The cascade transitions are 10 -20 % of the total emission cross section of  $H_2(B \ {}^1\Sigma_u^+)$  from the calculated cross section.  $H_2(X \ {}^1\Sigma_{\sigma}^+, v=0) + e \rightarrow H_2(E, F \ {}^1\Sigma_{\sigma}^+)$ 

$$\xrightarrow{} H_2(B \stackrel{1}{\Sigma_u^+}) \xrightarrow{} H_2(X \stackrel{1}{\Sigma_g^+}, v'') + \frac{h\nu}{6}$$

Recombinative desorption(RD)

 $H + H + wall \rightarrow H_2(X^{1}\Sigma_g^{+}, v^{\prime\prime} = 1, 2)$ followed by the E-V excitation of the  $X^{1}\Sigma_g^{+}$  state with the low  $v^{\prime\prime}$ :  $H_2(X^{1}\Sigma_g^{+}, v^{\prime\prime} = 1, 2) + e \text{ (fast)} \rightarrow H_2(B^{1}\Sigma_u^{+}, C^{1}\Pi_u) \rightarrow$  $H_2(X^{1}\Sigma_g^{+}, v^{\prime\prime} \ge 1, 2) + hv$ (7)

### Production of negative ions(H<sup>-</sup>)

#### Volume production

• Dissociative attachment(DA)  $H_2(X^{1}\Sigma_g^{+}, v'') + e(slow) \rightarrow H^{-} + H$  $H_2(C^{3}\Pi_u) + e(slow) \rightarrow H^{-} + H(2p)$ 



#### How physics of volume production fits the facts ?

A recent diagnostics effort at the University Duisburg-Essen (courtesy of Dr T. Mosbach) allows to compare directly :

a) the H<sup>-</sup> ion density measured by laser induced photodetachment (full squares)

b) the H<sup>-</sup> ion density calculated from measured rovibronic population distribution of the hydrogen ground state molecule (open squares)

Here the population of high vibrational states is determined with the aid of Laser-induced fluorescence (LIF) spectroscopy in the vacuum ultraviolet range. The vibrational temperature of the lower states is determined by means of optical emission spectroscopy.

The EEDF is measured with a Langmuir probe system.

The measurements are effected in a magnetic multipole plasma source, with 2 tungsten filaments, equivalent to the driver of an H- tandem source, for a discharge voltage of 100 V and a hydrogen pressure 1.5 Pa.



Les parois et l'électrode plasma contribuent à la production en volume en augmentant la densité de leurs précurseurs, les molécules  $H_2(v'')$ , par désorption recombinative :

$$H^* + H^* + paroi \to H_2(v'') + paroi$$
 (DR)

Si la production des  $H_2(v'')$  dans le plasma a été étudiée et optimisée, ce n'est pas le cas de la production de ces molécules en surface.

L'idée de cette proposition est d'augmenter considérablement la p roduction en volume par l'adjonction de la production en surface des  $H_2(v'')$ .

### Réactions de désorption recombinative

Il y a quelques années l'on invoquait les réactions Langmuir–Hinshelwood ou Eley-Rideal pour expliquer la DR.

Une troisième réaction est actuellement considérée, celle des **atomes chauds**. Dans le cas des métaux, cette réaction peut être beaucoup plus rapide que les deux autres.



### Qu'apportent les réactions d'atomes chauds?

- Les réactions des atomes chauds peuvent conduire à la formation de molécules très excitées si les atomes piégés réagissent avant d'avoir dissipé une part importante de leur énergie dans le substrat.
- Le même métal peut avoir un comportement différent en fonction de sa structure.

A-t-on observé l'effet de la désorption recombinative?

- A l'Université d'Eindhoven (Pays Bas)
- Dans des sources d'ions pour le LINAC JPARC au KEK et JAEA (Japon)
- Dans les sources d'ions H- à DESY (Allemagne)
- A l'Ecole Polytechnique

### Vibrational excitation by recombinative desorption: results of Hall et al, Phys. Rev. Lett.(1988)



### Experimental set-up Camembert III at Ecole Polytechnique, Palaiseau, France







### PRINCIPLES OF NEGATIVE ION PHOTODETACHMENT DIAGNOSTICS

- indirect method
- selectively converts  $H^-$  into H + electron
- the particles thus produced (H or electron) are then detected
- we chose to detect the photodetached electrons with a positively biased Langmuir probe.

One laser -> density of  $H^-$  ions

Two lasers -> temperature of H<sup>-</sup> ions, (from the time evolution of the H<sup>-</sup> density)

### Laser photodetachment diagnostic for measuring N- and T-





### Typical probe signals in two-laser-beam photodetachment technique

Probe signal from two laser shots for different time delays





2 lasers

### Negative ion temperature and population fractions



## Single and double temperature fit comparison

Double temperature fit:

$$\frac{n^-(r=0,t)}{n_0^-} = \alpha \exp(-\left(\frac{R_L}{v_{th\alpha}^- t}\right)^2) + \beta \exp(-\left(\frac{R_L}{v_{th\beta}^- t}\right)^2), \alpha + \beta = 1$$



8 W filaments, pure H2,

2 mTorr, 50 V - 5 A, 1991



### Methods used for modifying the Wall surface

In order to change the wall material, we used the following three methods:

- depositing a thin film of a new material by operating longer the filaments made of the new material or by flasing them;
- depositing a fresh film of the existing already material, by flashing the existing filaments
- adding to hydrogen an inert gas, such as argon.

Effect of covering the walls of Camembert III with Tantalum, Tungsten and Cesium upon the extracted negative ion and electron currents.



### Effect of wall surface condition on the negative ion density

- The negative ion density increases by 60-90% when evaporating a fresh tantalum film.
- This is done by overheating the tantalum filaments for a few minutes.
- Fresh tantalum deposited on the wall enhances the H- ion density (also N-/Ne) probably due to enhanced H<sub>2</sub>(v'') formation on the wall.



### Effect of wall condition on the negative ion temperature

We found that a fresh tantalum film also leads to the enhancement of the temperature of the hot negative ion population measured in the central region

(2.5 eV versus 1.25 eV).

We explain this by the presence of sheaths accelerating the negative ions formed near the wall on their way toward the source center.

### Time Dependence of N<sub>-</sub>/N<sub>e</sub> in H<sub>2</sub>-Argon Plasma

- The decay of the negative ion density in the presence of argon additive is accelerated when the argon pressure is enhanced.
- The initial  $N_N_e$  is not recovered by simply stopping the argon additive flow. The initial  $N_N_e$  is not recovered by simply stopping the argon additive flow.



### Camembert III modified for plasma production by ECR with microwaves at 2.45 GHz





### Extraction experiments with tantalum evaporation in ECR driven Camembert III.



10

### Experimental set-up at NIFS, Toki, Japan

#### Diagnostic tools

- Langmuir probe for electron density and temperature
- Laser photodetachment technique for H<sup>-</sup> density
- VUV spectrometer for vibrationally excited hydrogen molecules



### Typical VUV spectrum in the ion source.



- The B-X transitions and the C-X transitions are indicated.
- Next page shows the Ar mixture experiments for the enhancement of H<sup>-</sup> ions, but ...



# Evidence that $H_2(v''=1)$ molecules serve as initial species in F-C transitions to B-state

• Here we present a small part of the *Lyman system which is B-X transitions* 

• The transitions observed with Tantalu walls may be initiated from molecules with v'' > 0. The presence of intense structures (3 10), (3-11), (4-11), (4-12), (5-10), (5-12) indicates that vibrationally excited molecules v''=1 possibly serve as initial species in the Franck-Condon transition to the B state, because the B(v'=3, 4 and 5) states are promoted by v''= 1.



### CONCLUSION

Nous proposons de rechercher un matériau à fort rendement de désorption recombinative, qui puisse servir comme électrode plasma, ou même couvrir les parois de la souce.

### Objectif et méthodes (2/3)

Le dispositif où cette étude serait faite peut représenter

\*un plasma multipolaire généré par RF ou microondes, afin d'éviter le transport de matière du filament.

\*une source d'atomes telle que le 'cascaded arc', où on optimiserait le flux d'atomes.

Il faut pouvoir traiter le m atériau (chauffage, bombardement par ions positifs) afin de modifier la structure de sa surface. Première expérience proposée: le matériau étudié peut être chauffé ou bombardé par des ions positifs, N- étant mesuré par photodétachement

