

## Progress in the modeling of H<sup>-</sup> and D<sup>-</sup> ion sources

C. Gorse

Department of Chemistry, University - Centro di Studio per la Chimica dei Plasmi  
del CNR, Bari, Italy

**Abstract** - We present a self-consistent approach for solving at the same time the vibrational kinetics, the electron kinetics and the plasma chemistry in magnetic multicusp sources and compare, for H<sub>2</sub> plasmas, our theoretical results with the available experimental ones for different quantities (vibrational distribution, electron density, atomic and negative ion concentrations). Moreover for the same experimental conditions discussed for H<sub>2</sub> we report the results derived applying our model to the description of a D<sup>-</sup> volume source. In both cases we used quite recent data to describe the relevant interactions occurring in H<sub>2</sub>, D<sub>2</sub> plasmas.

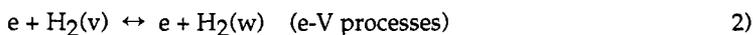
### INTRODUCTION

Large interest has been recently devoted to the production of negative H<sup>-</sup>/D<sup>-</sup> beams in magnetic multicusp sources due to their wide use for technological applications (refs. 1-4). Optimization of these sources requires a complete understanding of kinetic processes occurring in H<sub>2</sub>/D<sub>2</sub> plasmas. New and accurate experimental data for vibrational distributions (N<sub>v</sub>) (REMPI method at FOM (ref. 5) - Amsterdam -, VUV absorption spectroscopy at Berkeley (ref. 6)), for negative ion concentration (photodetachment at FOM (ref. 5) and Ecole Polytechnique - Paris (ref. 7)-), for electron energy distribution functions (eedf) (probes at Belfast and Dublin (ref. 8)) encouraged a large computational effort aimed to a better understanding of the microscopic processes originating in such devices (refs. 9-11)

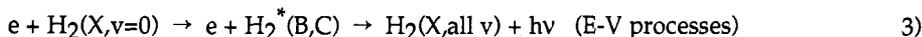
Dissociative attachment processes from vibrationally excited molecules are considered as responsible for the formation of negative ions in volume sources



Cross sections (ref. 12) relative to elementary processes 1, involve low energy electrons, and strongly increase with the vibrational level *v* so that to describe the kinetics of the ion (H<sup>-</sup>/D<sup>-</sup>) one should first study the vibrational distribution of the parent molecules. In multicusp sources operated at low pressures (p~mtorr) electrons are produced by the heated filaments (discharge current I<sub>d</sub>~10A) and accelerated over the applied discharge voltage (V<sub>d</sub>~100V). To reduce the electron losses the metallic walls of the container are equipped with permanent magnets in a linecusp configuration generating a cusp field of about 0.1T. In such a configuration the confined primary electrons undergo many collisions with the filling gas molecules transferring them their energy. In the earlier stage of the discharge, on the first levels the vibrational quanta are mainly introduced by slow electrons (ε<5eV)



while fast electrons (ε>12eV) are thought to efficiently populate the intermediate and high levels



Vibrationally excited molecules can then be dissociated, ionized, deactivated by atoms... Later on, when the electron energy distribution function already reached a quasi-stationary state, the container walls play an important role in the molecular, atomic and ionic distribution evolutions, in particular processes



will be of fundamental importance in the energy rearrangement. From this simplified scheme it clearly results that the vibrational kinetics has to be coupled with the plasmachemistry. Furthermore, keeping in mind that the electronic rate coefficients depend on the electron energy distribution function, it follows that the heavy particle kinetics must also be coupled with the electron kinetics. The previously developed computing code (refs. 3-4) has been run anew to simulate the properties of  $H^-$  and  $D^-$  ion sources making use of new extended sets of cross sections for both electrons and heavy particles.

### METHOD OF CALCULATION

The vibrational kinetics is numerically derived solving a system of vibrational master equations coupled to the dissociation kinetics and to the Boltzmann equation for the electrons. In implicit form we can write

$$\left(\frac{dN_v}{dt}\right) = \left(\frac{dN_v}{dt}\right)_{e-V} + \left(\frac{dN_v}{dt}\right)_{E-V} + \left(\frac{dN_v}{dt}\right)_{V-V} + \left(\frac{dN_v}{dt}\right)_{V-T} + \left(\frac{dN_v}{dt}\right)_{e-D} + \left(\frac{dN_v}{dt}\right)_{e-I} \quad a)$$

$$+ \left(\frac{dN_v}{dt}\right)_{e-da} + \left(\frac{dN_v}{dt}\right)_{e-E} + \left(\frac{dN_v}{dt}\right)_{wall}$$

where the different relaxation terms respectively represent the energy exchanges in processes e-V, E-V, V-V (vibration-vibration), V-T (vibration-translation in molecule-molecule or molecule-atom interactions), e-D (dissociation), e-I (ionization), e-da (dissociative attachment), e-E (electronic excitation), wall (particle-surface area interaction). The dissociation kinetics considers beside the production of atoms in both ground and excited states by direct electronic mechanism (ref. 13) as well as by dissociative attachment, their production in molecular reactions (ref. 3) such that



while atomic recombination on the wall is accounted as a loss term. In the negative ion kinetics, dissociative attachment from all the vibrational levels contribute as a gain term while ionic and atomic recombinations represent the losses.

The electron kinetics is derived from a time dependent solution of the Boltzmann equation in the two-term approximation (ref. 14)

$$\frac{\partial n(\epsilon, t)}{\partial t} = \left(\frac{\partial J_{el}}{\partial \epsilon}\right)_{e-M} + \left(\frac{\partial J_{el}}{\partial \epsilon}\right)_{e-e} + In + Ion + Sup + S - L \quad b)$$

where  $n(\epsilon, t)$  represents the number density of electrons with energy between  $\epsilon$  and  $\epsilon + d\epsilon$ , on the right hand side the different terms represent the flux of electrons along the energy axis driven by elastic (electron-molecule/atom, electron-electron) collisions, inelastic, ionization and vibrational superelastic collisions. The source term, S, represents the number density of electrons emitted by the filaments and assumed to be uniformly distributed in the whole plasma volume (ref. 4). The loss term (L) includes the total losses of electrons by diffusion toward the walls and by gas phase recombination (ref. 4). It is worth noting that, since in our model the positive ion kinetics ( $H^+$ ,  $H_2^+$ ,  $H_3^+$ ) is not selfconsistently coupled to the other kinetics, the experimental measured plasma potential value ( $V_p$ ) is utilized in all the calculations.

### INPUT DATA

#### Electronic cross sections

Formation of negative ions mainly occurs in dissociative attachment processes involving high vibrational levels. A crucial point in this modeling is the knowledge of electronic cross sections involving vibrational excited levels of  $H_2(D_2)$  molecules. Recently the entire set of cross sections (electronic excitation, dissociation, ionization, dissociative ionization) from each vibrational level has been improved ( $H_2$ ) (ref. 15) or newly calculated ( $D_2$ ) (ref. 16) according to the classical Gryzinski approximation. Agreement between experiment and theory is now enhanced (see for instance fig. 4a of ref. 15), the simple Gryzinski method seems to be adequate (see fig. 3 of ref. 15) to obtain, in realistic computing times, reliable excitation cross sections from all the vibrationally excited levels. The excitation cross sections to  $H_2^*(B, C)$  from vibrational level  $v=0$  calculated in the Gryzinski method

together with the decay probabilities reported by Hiskes ( $H_2$ ) or with the decaying cross sections reported in ref. 16 ( $D_2$ ) have been introduced to simulate the E-V processes. For e-V processes we use both experimental (ref. 17) and theoretical (ref. 12) cross sections for transitions  $0 \rightarrow w$  ( $0 < w < 6$ ) and a scaling law for those processes starting from level  $v > 0$  in  $H_2$  description and theoretical cross sections reported in ref. 12 for  $D_2$ .

### Heavy particle collisions

It has been experimentally shown that in multicusp sources the translational temperature of atoms is much larger than the rotational molecular one (ref. 18). Multiquantum V-T processes for the atomic deactivation of vibrationally excited molecules have been revised, the new values obtained for  $T_H=500K$  and  $T_{H_2}=4000K$  ( $T_H$  and  $T_{H_2}$  are respectively the atomic and molecular temperatures) are introduced in this modeling (ref. 19). As for the V-T molecule-molecule rate coefficients a more accurate set, generated on the basis of a semi-classical approach, is now available (ref. 20).

### Wall interactions

The vibrational deactivation of  $H_2$  on the walls has been inserted according to the data reported in ref. 21, in this representation highly excited molecules are completely deactivated after only a few collisions. Production of  $H_2(v \neq 0)$  by Auger neutralization of  $H_2^+$  molecules on the walls is written in the formalism suggested by Hiskes (ref. 22) where impinging  $H_2^+$  ions are converted in  $H_2(v)$  molecules with an equal probability on each vibrational level. Vibrational excitation of molecule can also be attributed to recombinative desorption of hydrogen atoms on the cold walls of the discharge chamber, for this process we use the probabilities of populating  $H_2(v)$  molecules reported by Hall (ref. 23). The  $H_2$ -wall interaction treatment has been identically reversed to the  $D_2$ -wall description.

## RESULTS

### Effect of the new cross sections ( $H_2$ discharge)

It is worth noting that new excitation cross sections to B and C states are now a factor of two smaller than those previously reported by Hiskes and used in refs.3-4. On the contrary the new dissociation cross sections are about twice as large as the old ones, moreover the e-D processes have been enriched by dissociative ionization as well as by dissociation leading to excited atomic hydrogen. The new V-T molecule-molecule rate coefficients are higher than the old ones (ref. 24) up to one order of magnitude for the higher vibrational levels. All these modifications lead to a lowering of the densities of vibrationally excited molecules (see fig. 1 where experimental (ref. 5) and theoretical  $N_V$  distributions are represented for the condition:  $p=4.5\text{mtorr}$ ,  $I_d=10A$ ,  $V_d=115V$ ,  $V_p=2.9V$ ). Figure 2

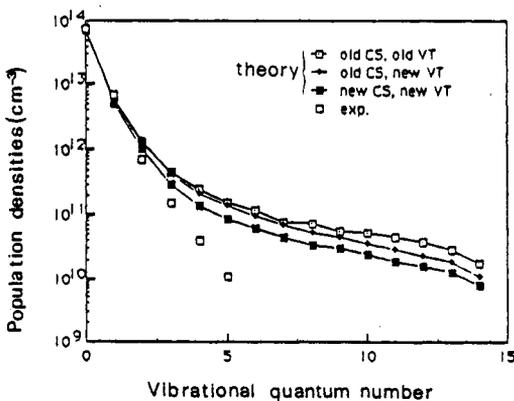


Fig. 1  $N_V$  distributions (CS: electronic cross sections, VT: heavy particle collisions)

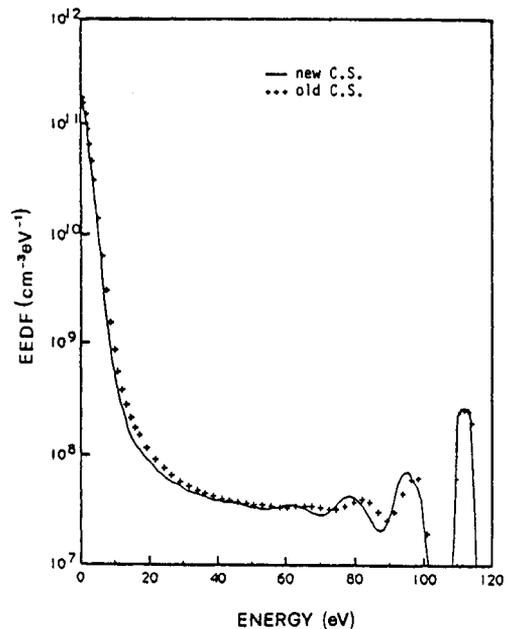


Fig. 2 Stationary eedf calculated with old and new electronic cross sections (same conditions as fig. 1)

reports a comparison of the stationary eedfs obtained with the old and the new sets of cross sections: we note a slight decrease of electron density in the range between 10 and 30 eV. The shape of the eddf is characteristic of a discharge sustained by a beam of energetic electrons.

**Comparison with experimental data (H<sub>2</sub>)**

Following the experimental work by Eenshuistra and Hopman (ref. 5), we study the effects of changing the current intensity or the filling pressure for a constant applied voltage ( $V_d=115V$ ) in the source. Figures 3a-d compare experimental and theoretical values for electron density ( $n_e$ ), bulk electron temperature ( $T_e$ ), atom ([H]) and negative ion ([H<sup>-</sup>]) densities as a function of the current intensity. Save for the electron density the theory seems to be able to quantitatively describe the variations of the measured quantities. We get to the same conclusion when looking at their dependence on the filling pressure (fig. 4 reports the evolution of [H<sup>-</sup>] versus the pressure). Both experimental and theoretical results show that to increase the density of negative ions in low pressure discharges it is necessary to raise the current density and, to some extent, the filling pressure.

**Application of the modeling to D-source**

As well supplied new sets of cross sections for both electrons and heavy particles interactions are now available for deuterium we extend the previous approach to the prediction of D<sup>-</sup> concentration in a multicusp source identical to the one used by Eenshuistra (ref. 5). We report respectively on fig. 5 and fig. 6 the calculated eedfs and vibrational distributions for H<sub>2</sub> and D<sub>2</sub>. The isotopic effect is small both for electron and vibrational distributions. But we note that the plateau of the vibrational distribution is higher in D<sub>2</sub> than in H<sub>2</sub>, nevertheless the concentrations of negative ions versus the current or the pressure (○ symbols on figs. 3 and 4) are lower in D<sub>2</sub> than in H<sub>2</sub> sources. It is worth noting that our calculations have been obtained assuming the same plasma potential value for H<sub>2</sub> and D<sub>2</sub>, Mullan et al (ref. 8) report, for the extractor region of a tandem source, measured  $V_p$  values higher in D<sub>2</sub> than in H<sub>2</sub>. Crosses on fig. 4 correspond to new [D<sup>-</sup>] densities obtained scaling the plasma potential in agreement with the experimental data of ref. 8. In these conditions we see that negative ions densities are very similar for H<sub>2</sub> and D<sub>2</sub> sources.

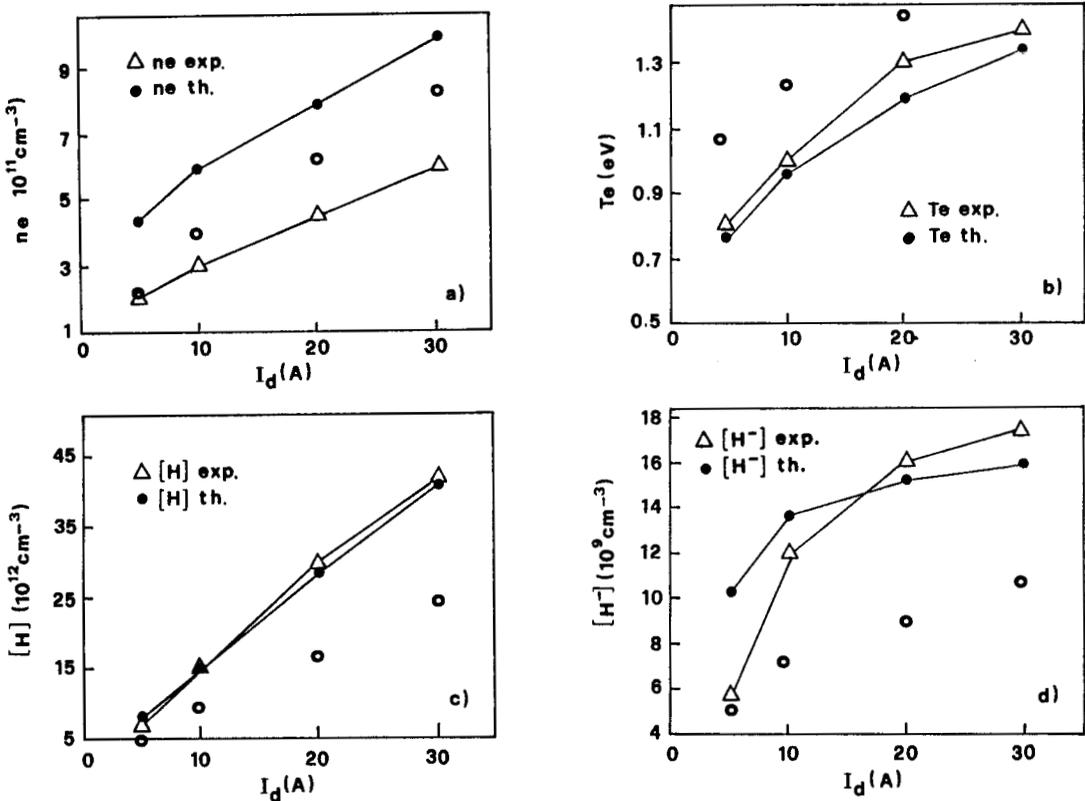


Fig. 3 a-d A comparison between theoretical and experimental quantities ( $p=7.5$  mtorr,  $V_d=115V$ ) (○: results for  $D_2$  discharges)

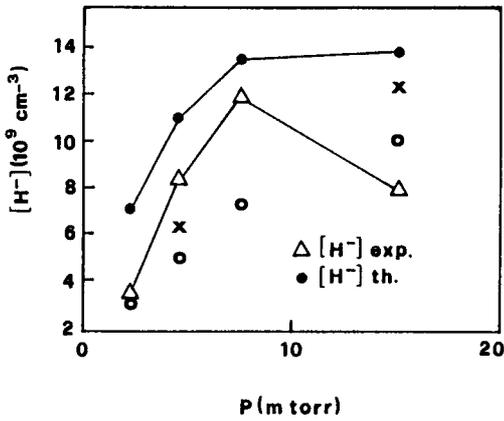


Fig. 4 A comparison between theoretical and experimental  $H^-$  ( $D^-$ ) densities ( $I_d=10A$ ,  $V_d=115V$ )

( $\circ$ :  $D^-$  calculated with the same  $V_p$  values for both  $H_2$  and  $D_2$ ;  $\times$ :  $D^-$  calculated modifying the  $V_p$  values for  $D_2$  as explained in the text)

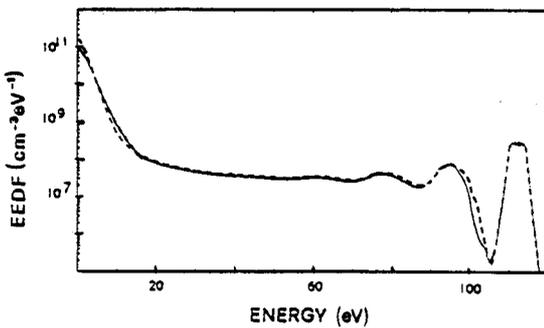


Fig. 5 A comparison of stationary eedfs for  $H_2$  and  $D_2$  (full line:  $D_2$ , dashed line:  $H_2$ )(same conditions as in fig. 1)

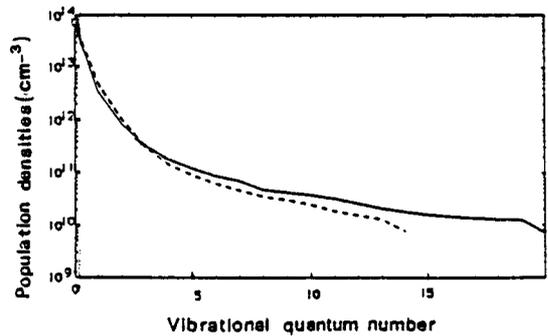


Fig. 6 A comparison of  $N_v$  distributions for  $H_2$  and  $D_2$  (full line:  $D_2$ , dashed line:  $H_2$ ) (same conditions as in fig. 1)

### CONCLUSIONS

We have seen that the proposed model with the new sets of cross sections gives theoretical results in satisfactory agreement with experimental ones. Nevertheless we must remind that the interactions between excited particles and metallic surface are far from being completely understood. In our modeling recombination of molecular ions on the walls (process 5) has been treated assuming arbitrarily the  $H_2^+$  density to be one third of the electron density. Decreasing to zero the  $H_2^+$  concentration lead to a consequent lowering of the vibrational distribution bringing it towards the experimental values (ref. 11). In this condition the plateau over the high vibrational levels decreases with the relative consequences on the  $H^-$  concentrations. As a second example we would like to mention that recent calculations do not show any deactivation of an excited  $H_2$  molecule colliding with a clean copper surface (ref. 25) in contradiction with previous data reporting a complete deactivation of  $H_2(v)$  on an iron surface. These considerations clearly show that a better understanding and description of the chemical processes originating on the walls is now necessary to improve our modeling.

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