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Vibrational excitation of hydrogen molecules released from surfaces

I. Čadež[1], S. Markelj[1], Z. Rupnik[1], S. Brezinsek[2]

[1]*Jožef Stefan Institute, EURATOM-MHST Association, SI-1000 Ljubljana, Slovenia*

[2]*Institut für Plasmaphysik, Forschungszentrum Jülich, EURATOM-Association, Trilateral Euregio Cluster, D-52425 Jülich, Germany*

Introduction

Neutral hydrogen is released from plasma-facing components in fusion devices during normal plasma operation. These particles – atoms and molecules - enter the edge plasma and take part in complex reaction scenarios determining plasma-wall interaction. Detailed modelling by taking into account reactions between all present species is needed for understanding and predicting tokamak plasma edge characteristics. Presence of atoms and molecules in the divertor region becomes even more important when modelling edge plasma of ITER because of its large dimensions [1].

Neutral molecules are important for detachment of plasma from target plates in divertor due to their large cross sections for recombination of atomic ions and also for other reactive collisions [2]. Optical spectroscopy of edge plasmas has revealed abundant presence of hydrogen molecules that are vibrationally and rotationally excited [3][4]. Internal excitation of molecules influences molecular reaction rates and this is the reason for theoretical analysis of the role of vibrationally excited molecules in edge plasma. Theoretical cross sections do exist for various binary volume collisions of vibrationally excited hydrogen molecules (VEHM) [5] but there is a lack of corresponding experimental results.

Surface processes are important sources and sinks of VEHMs and present knowledge of corresponding rates is rather poor. Besides complexity of elementary surface processes such as e.g. atom recombination, vibrational relaxation and thermal desorption one has also to consider surface impurities and morphology. For this reason experimental data are even more needed in order to check the applicability of theoretical estimates. VEHM production by surface processes is important not only at the plasma-facing sides in tokamaks but also at remote areas normally not exposed to plasma as it is the vacuum vessel itself. Surface processes involving VEHMs are also of primary importance for negative ion sources [6].

Experiment

We have initiated systematic studies of surface processes that are leading to formation and relaxation of VEHMs on fusion relevant materials such as tungsten, tantalum etc. For this purpose we have developed a new experimental set-up for vibrational spectroscopy of hydrogen molecules [7] that is based on specific characteristics of low energy (< 4 eV) dissociative electron attachment (DEA) in H_2 (and all its isotopomers):

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

Vibrational population of target H_2 (or D_2) molecules are obtained by proper deconvolution of H^- (or D^-) ion current dependence on incident electron energy in the range from 0 to 5 eV. Theoretical cross sections for DEA in H_2 and D_2 [8] are used for spectra deconvolution.

Here we concentrate on tantalum and present the study of VEHM production by atom (H or D) recombination at the surface exposed to a low-pressure partially dissociated hydrogen atmosphere. Study was performed in the experimental arrangement as shown in figure 1. VEHM source is a compact pancake structure made of copper. It operates on same principle as that extensively used in previous similar studies [9]. Hydrogen gas is introduced into the cell where it is dissociated by a hot tungsten filament. Rate of dissociation is controlled by filament temperature i.e. by the heating current through the filament. VEHMs are produced by atom recombination on the cold wall and after number of volume and surface collisions they leave the cell through the exit orifice. Sample plate of 0.5 mm thick tantalum is mounted on the cold finger just in front of the exit orifice of the cell in order to provide for as direct as possible path to the detection zone for molecules created there. Hydrogen gas containing VEHMs that flows out of the source is intercepted by probing electron beam and negative ions created by DEA are collected by an extraction system and then detected by a channel electron multiplier. The distance between the exit orifice of the source and the probing electron beam is 4 cm.

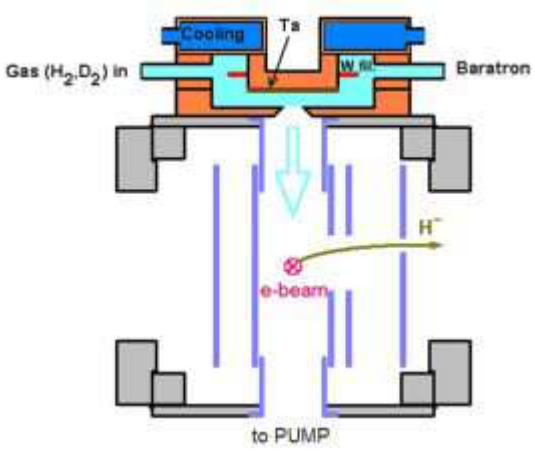

Figure 1. *Source of VEHMs used for the present study mounted at the top of cell for vibrational spectroscopy.*

Results

Recombination of H atoms at clean tantalum surface leads to production of higher vibrational states and the shape of vibrational distribution does not depend on dissociation filament temperature – higher temperature only increases percentage of excited molecules. However, the situation changes once the surface is not clean. Ion spectra of H^-/H_2 for three different dissociation filament temperatures are shown in figure 2 for tantalum after it has been exposed to the air (contamination with O_2, H_2O). Relative populations of vibrationally excited molecules for these spectra are shown in figure 3. For the lowest filament temperature vibrational distribution corresponds to Boltzmann distribution with vibrational temperature $T_v = 3000$ K. For higher filament temperature different distribution is observed: for the states v = 1, 2 and 3 vibrational temperature about 4000 K can be attributed but for the states from v = 4 to 8 vibrational temperature is higher, about 6000 K.

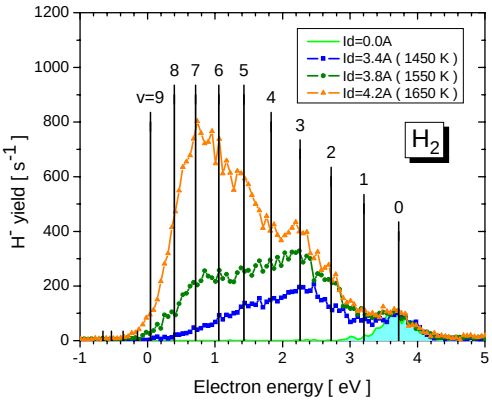

Figure 2. *Ion spectra of H^-/H_2 for three filament temperatures. Spectrum in green is obtained for the cold gas with no VEHM.*

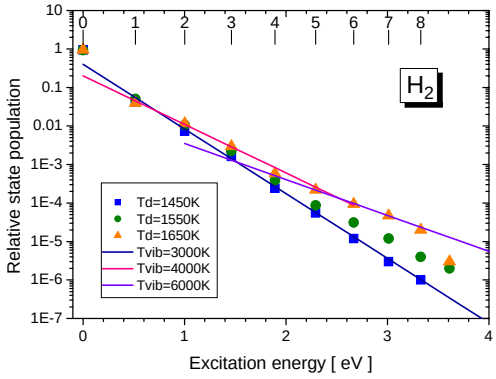

Figure 3. *Relative vibrational populations obtained by deconvolution of spectra from figure 2.*

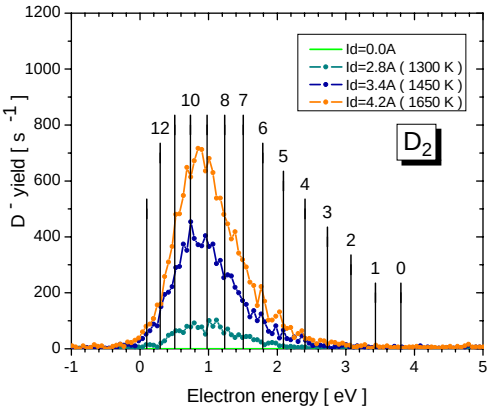

Figure 4. *Ion spectra D^-/D_2 for three filament temperatures.*

Ion spectra of D^-/D_2 acquired immediately after those shown in figure 2 are shown in figure 4. Complete absence of ion signal for lowest vibrational states is expected as DEA cross sections are very low due to the strong isotope effect. However, cross sections for DEA to vibrationally excited D_2 molecules for higher v become as big as in H_2, reaching values of the order of 10^{-15} cm^2 for the highest states. For deuterium vibrational distributions for different filament

currents are similar in shape showing no effect of contamination as described for H_2. Further studies are in progress in order to elucidate observed phenomenon that could be attributed to the isotope effect on reactions with impurities on the tantalum surface.

The described type of the source of VEHMs besides providing new data on atom recombination on surfaces will in future be used in an experiment for testing optical spectroscopic methods for determination of ground state vibrational distributions in plasmas.

In another recent experiment a permeation source was introduced in the cell for vibrational spectroscopy instead of the described VEHM source. Molecules formed by recombination of atoms transported to the surface by permeation through palladium membrane were found to be in the ground vibrational state. Study of hydrogen permeation through tantalum is in progress. These studies are accompanied by surface composition analysis and H and D depth profiling in the samples by use of ion beam analytical methods.

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