MODIFICATION OF THE GROUND STATE VIBRATIONAL POPULATION OF MOLECULAR HYDROGEN AND DEUTERIUM BY DIFFERENT WALL MATERIALS IN LOW TEMPERATURE PLASMAS

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Abstract. Among others, graphite, tungsten and high-grade steel are proposed as wall materials in fusion experiments. In cold divertor plasmas, hydrogen molecules which result from recombining atoms at the walls affect the energy balance of the plasma if they are vibrationally excited. For this paper, the vibrational population of H_2 and D_2 in their electronic ground state was investigated in a microwave generated low temperature plasma with special respect to different wall materials which were exposed to the plasma. Especially for graphite strongly enhanced vibrational populations were measured. With respect to divertor plasmas this can considerably enhance the contribution of the hydrogen molecules to the volume recombination.

1. Introduction

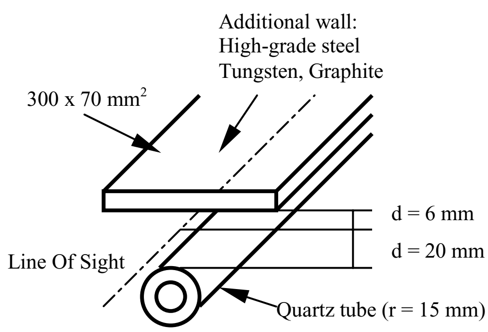
Vibrationally excited H_2 and D_2 molecules play a role in the energy balance of divertor plasmas via dissociation, ionization and recombination processes [1]. This is mainly due to the energetically large separation of the vibrational levels of these light molecules. A population of higher levels can obviously lead to a reduction of threshold energies for several plasma reactions. The processes responsible for the vibrational population are roughly divided in three types. Electron impact excitation or de-excitation, reactions at the walls and molecule – molecule collisions. The latter is of minor importance in divertor plasmas but can dominate in technical plasmas. A more detailed discussion of these processes is given in [2].

In fusion experiments the molecules result from recombining atoms at the divertor tiles and then penetrate into the plasma. The vibrational population is on the one hand due to interaction with the plasma but may on the other hand also depend on the material and the temperature of the divertor tiles. Such wall effects were investigated, using a spectroscopic method, in a microwave generated low temperature plasma by exposing different materials to H_2 (D_2) / helium plasmas. Measurements were carried out for graphite, tungsten and high-grade steel as these are candidates for wall materials in fusion experiments. Especially graphite is of interest as here not only surface reactions themselves but also hydrocarbons, formed in the plasma by erosion products, may be responsible for the resulting vibrational population of the hydrogen molecules. In that respect the results presented here support comparative investigations of H_2 and CH_4 plasmas presented in [3].

2. Experimental Setup and Diagnostic method

Measurements were carried out in a microwave generated low temperature plasma based on the principle of the Duo Plasmaline [4]. Electron temperatures and densities range from 2.4 to 3.7 eV and 0.3 to 2 x 10^{17} m^{-3}, respectively, dependent on the pressure (6 to 100 Pa), the percentage of helium and the presence of an optional wall material. Gas temperatures range from 550 K at low pressure to 750 K at high pressure. The plasma is of cylindrical symmetry which provides axially homogeneous and radially variable plasma parameters. The vessel is a cylinder of 700 mm length and 420 mm diameter. The plasma column is about 300 mm long and varies in radius from about 30 mm to vessel dimensions, depending mainly on the pressure. As the plasma is investigated near the axis, the large vessel dimensions reduce the effects of the vessel wall material (high-grade steel) on the observed plasma volume. However, the Duo Plasmaline principle contains a quartz tube in the central axis of the plasma. This means that there is always a quartz wall in the plasma which must be kept in mind when discussing the effects of additionally exposed materials.

Fig. 1: Geometry of the experimental setup

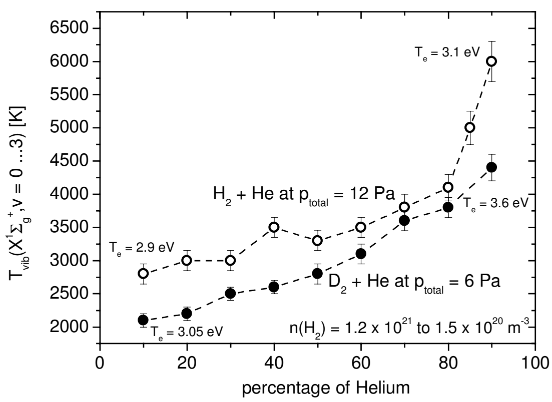


The spectroscopic method for determination of the ground state vibrational population is based on the measurement of the radiation of the Fulcher transition and the application of the Franck – Condon principle. It was already established for H_2 and D_2 in laboratory plasmas [5] as well as in fusion edge plasmas [1]. Beneath Franck – Condon factors, branching ratios, effective lifetimes, vibrationally resolved excitation rate coefficients as well as quenching corrections are considered for the interpretation of the radiation. It has to be mentioned that this method is sensitive for the first 4 (5) ground state vibrational levels of H_2 (D_2). For these levels the vibrational distribution is characterized by a vibrational temperature $T_{vib}(X^1\Sigma_g^+)$.
The tungsten, high-grade steel and graphite samples have a size of 300 x 70 mm^2 and were placed 26 mm above the central quartz tube. The line of sight used as standard for emission spectroscopy of the Fulcher bands runs parallel to the axis (homogeneous plasma) at a distance of 20 mm to the quartz tube and, therefore, 6 mm to the optionally exposed walls. Fig.1 illustrates the geometrical setup.

3. Results

From investigations in H_2 (D_2) plasmas with different admixtures of helium at low pressure (12 (6) Pa) and without additional surface information on the effect of heavy particle collisions was gained. The results of $T_{vib}(X^1\Sigma_g^+)$ are given in Fig. 2. H_2, as well as D_2, show a strong increase in vibrational excitation with increasing percentage of helium. T_e rises by about 0.5 eV being in the order of 3 eV. In the present low pressure discharge this does not cause the observed change in vibrational excitation as known from calculations with a collisional radiative model (in fact T_{vib} is

Fig. 2: $T_{vib}(X^1\Sigma_g^+)$ of H_2 and D_2 (without additional walls) in mixtures with helium.



inversely correlated with T_e) [2]. The rise in $T_{vib}(X^1\Sigma_g^+)$ is caused by the decrease in molecular density and the correlated decrease of depopulating molecule – molecule collisions.

Investigations with additional walls were carried out in mixtures with 90% helium. The pressure ranges from 6 to 100 Pa which provides molecular densities of 7×10^{19} to 1×10^{21} m^{-3}. The results of $T_{vib}(X^1\Sigma_g^+)$ are shown in Fig. 3 for H_2 and D_2, respectively. Both isotopomeres show similar dependencies on pressure and wall materials. $T_{vib}(X^1\Sigma_g^+)$ ranges from 4600 to 9000 K (H_2) and 3000 to 5300 K (D_2). Furthermore, they show similar vibrational excitations, which is manifested in lower temperatures for D_2 due to the lower energy separation of D_2 compared to H_2. Regarding the effect of exposing different materials to the plasma, enhanced vibrational populations for all materials compared to the plasma without additional surface were measured above p = 20 Pa. One has, however, to keep in mind that the plasma is in all cases in contact with the quartz tube. The measured effects give therefore a relative enhancement with respect to quartz. The strongest enhancement in $T_{vib}(X^1\Sigma_g^+)$ was measured with the graphite sample in the plasma. Here, not only surface reactions themselves may be responsible for the high excitations but these may also be caused by plasma reactions of hydrocarbons which are formed in the plasma by erosion products. From spectroscopic observation of the CH (CD) band at 431 nm a content of 1% of CH_4

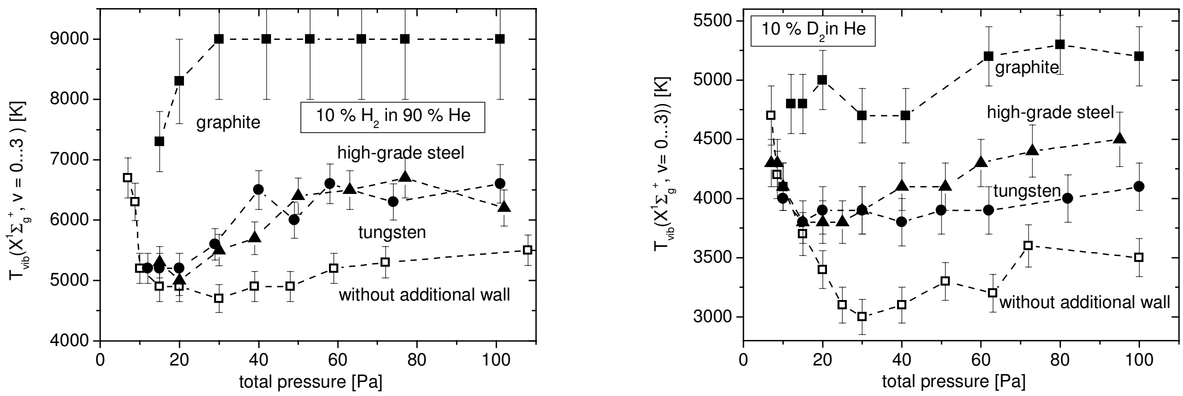


Fig. 3: $T_{vib}(X^1\Sigma_g^+)$ of H_2 (left) and D_2 (right) with and without additional walls.

(CD_4) in the 10% H_2 (D_2) / He plasma was estimated. The role of hydrocarbons was investigated in [3] by a comparison of H_2 with CH_4 plasmas in an ECR plasma source. The reaction channels of the hydrocarbons which lead to vibrationally excited hydrogen molecules are presented and discussed in [3].

Considering the dependence on pressure it has to be kept in mind that the measured $T_{vib}(X^1\Sigma_g^+)$ are the result of a mixture of the three types of population mechanisms mentioned before (plasma reactions, wall effects, heavy particle collisions). With the change of the pressure by more than one order of magnitude, their influence on the vibrational distributions in the observed plasma volume also changes as the mean free path of the molecules ranges from about 5 mm at low pressure to less than 0.3 mm at 100 Pa. At the lowest pressures and, therefore, low molecular densities the heavy particle collisions are negligible. With increasing density they become more important which leads to a decrease of $T_{vib}(X^1\Sigma_g^+)$. This is shown best in Fig. 3 by the results for the measurements without additional wall between 6 and about 30 Pa. Additionally, at these low pressures the whole plasma volume is large compared to the area of the quartz tube and the additional walls which lowers the influence of these surfaces on the measurements. On the other hand, at the highest pressures the influence of the quartz tube is assumed to vanish with respect to the line of sight used. This is estimated from the mean number of molecule – molecule collisions between the quartz tube and the distance of the line of sight (20 mm). At p = 6 Pa this number is less than one increasing to more than 100 at 100 Pa. However, for the additional walls at a distance of 6 mm to the line of sight this mean number only rises to about 10 at 100 Pa so that their influence is present in the observed plasma volume. In the range between low and high pressure the mentioned regimes overlap. In order to clarify these items further investigations are in progress including spatially resolved measurements and investigations on the effect of the gas temperature (and, therefore, the temperature of the walls) which increases by about 200 K with the pressure.

4. Conclusions

For dealing with hydrogen (deuterium) molecules in plasma edge codes it has to be known if the molecules start with some vibrational excitation at the walls or if they get their vibrational distribution by plasma processes alone. For the present paper a dependence of the ground state vibrational population of molecular hydrogen and deuterium on different wall materials was measured in a low temperature plasma. Especially graphite and, to a lower degree, tungsten and high-grade steel lead to a remarkable increase in vibrational excitation (e. g. H_2 without additional wall (5000 K): 70% in v = 0, 2% in v = 3; and with graphite (8000 - 9000 K): 48% in v = 0, 6.5% in v = 3). In case of graphite this is mainly due to the formation of hydrocarbons from erosion products in the plasma. Up to now the molecular assisted recombination is estimated to contribute in the order of some percent to the volume recombination of divertor plasmas. The measured wall effects may lead to a significant increase in this contribution. The presented results, therefore, suggest to implement the wall effects on the vibrational population of H_2 and D_2 molecules in plasma edge codes for a proper modelling of recycling regimes. Additionally, further investigations are needed to clarify in which way such an implementation can be done best.

References

[1] U. Fantz, D. Reiter, B. Heger, D. Coster, J. Nucl. Mater. 290 – 293 (2001) 367 – 373
[2] U. Fantz, B. Heger, D. Wünderlich, Plasma Phys. Contr. Fusion, in press
[3] U. Fantz, S. Meir, this conference
[4] E. Räuchle, J. Phys. IV France 8 (1998) Pr7-99
[5] U. Fantz, B. Heger, Plasma Phys. Contr. Fusion 40 (1998) 2023