Vacuum ultraviolet emission from microwave driven hydrogen plasmas

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1. Introduction

Hydrogen discharges, in particular discharges driven by surface waves at microwave frequencies, have the potential to become alternative sources of vacuum ultraviolet (VUV) radiation. VUV radiation falls in the region of the electromagnetic spectrum with wavelengths below 200 nm. Atomic and molecular hydrogen emissions in this spectral range correspond to photon energies high enough to break most organic chemical bonds, initiate surface reactions and modify surface properties [1, 2]. The optimization of these sources requires deep insight of the mechanisms occurring inside the plasma. However, results concerning VUV emissions from discharges operating at microwave frequencies are quite scarce. In this work, we present an experimental investigation of VUV radiation emitted by hydrogen discharges operating at microwave frequencies and low-pressure conditions (0.1 – 0.4 mbar). Emission spectroscopy in the VUV region is used to identify and investigate the excited hydrogen species emitting at these wavelengths as a function of the operational conditions. To complement these results, optical emission spectroscopy in the visible is used to determine the plasma electron density and the temperatures of the gas/excited species.

2. Plasma source and experimental conditions

A classical surface-wave-sustained discharge at 2.45 GHz, with a waveguide-surfatron based setup as the field applicator [3] has been used as plasma source [4]. The experimental setup is illustrated in Figure 1. The discharge is produced inside a quartz tube with inner/outer radii equal to 0.15/ 0.25 cm. The gas flow and the pressure is regulated with mass flow controllers and three pressure sensors. The microwave power delivered to the launcher has been varied between 80 and 300 W. The VUV radiation emitted by the plasma is collected by a Horiba Jobin-Yvon Plane Grating Monograph (PGM), which is directly coupled to the discharge tube, thus granting unobstructed line-of-sight over the axis. The PGM is equipped with one of two plane gratings (1800 or 500 gr/mm) allowing the detection of wavelengths from 8 to 125 nm. The VUV photons are then directly collected by a Silicon photodiode detector. To assure a good signal-to-noise response, the light path inside the spectrometer is kept at low pressures 10^{-4} - 10^{-5} mbar by a turbopump. All the spectra are background corrected. The visible radiation has been collected by an optical fiber positioned perpendicularly to the discharge tube. The optical fiber transmits the visible radiation to the entrance slit of a Jobin-YvonSpex 1250M spectrometer, coupled to a CCD detector. The Jobin-YvonSpex 1250M has a spectral resolution of 0.06 Å. Optical emission spectroscopy of the H_β line (486.1 nm) and of the Fulcher-α Q-branch transitions $\left[d^3\Pi_u \rightarrow a^3\Sigma_g^+\right]$ (600 – 616 nm) was performed to determine basic plasma parameters, such as the plasma electron density and the gas/excited species temperatures [5].

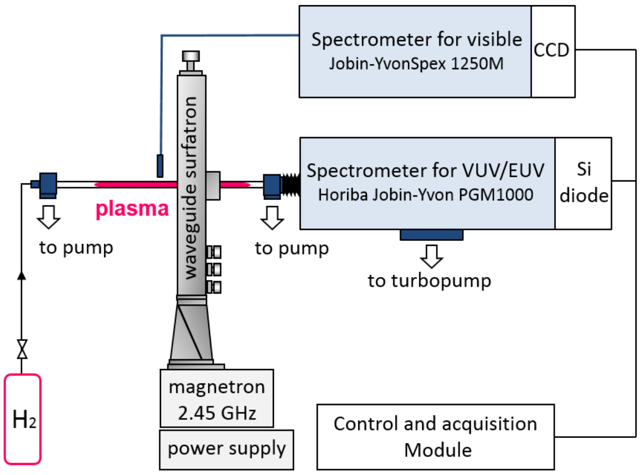

Figure 1. Experimental setup for spectroscopy in the visible and VUV spectral ranges.

3. Results and discussion

Experimental spectra of hydrogen measured between 8 – 125 nm show the presence of atomic and molecular emissions as seen in Figure 2. The atomic emissions correspond to transitions of the hydrogen Lyman series. The most intense spectral lines correspond to Lyman-α (n=2 → n=1) and Lyman-β (n=3 → n=1) transitions emitting at 121.6 nm and 102.6 nm, with respective energies 10.2 eV and 12.1 eV. Other spectral lines from the Lyman series are also observed at 97.3 nm and 93.8 nm, corresponding to Lyman-γ (n=4 → n=1) and Lyman-ε (n=6 → n=1) transitions. The atomic Lyman series comprises transitions up to level n=12. However, the contribution of the full Lyman series to the spectrum cannot be distinguished from the hydrogen molecular emissions. The observed molecular bands correspond to Lyman H_2 ($B^1\Sigma_u^+$ - $X^1\Sigma_g^+$) and Werner H_2 ($C^1\Pi_u$ - $X^1\Sigma_g^+$) transitions detected in the 80 - 125 nm spectral range.

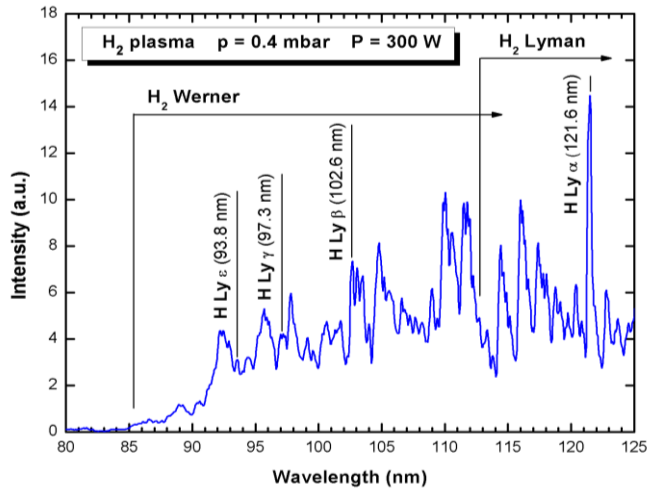

Figure 2. Experimental spectrum of hydrogen in the VUV spectral range.

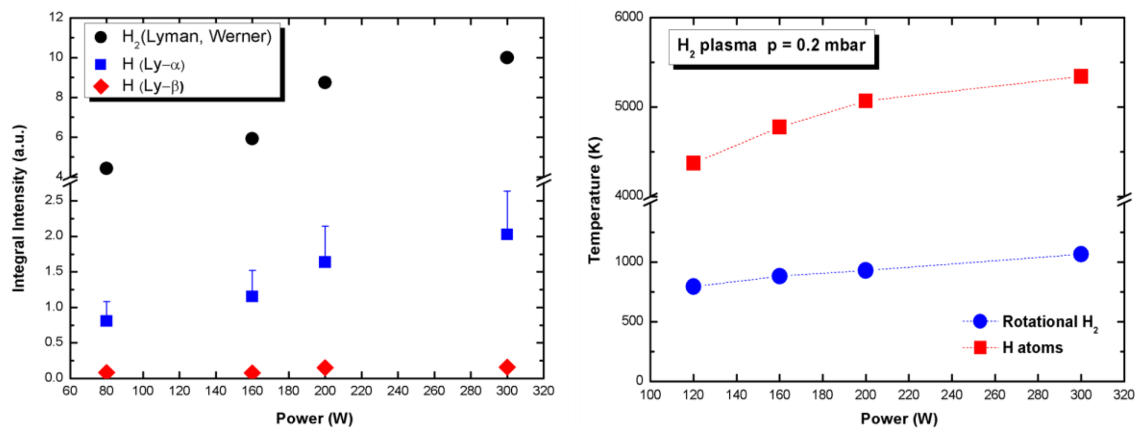

Figure 3. (a) Intensity of the Lyman-α, β atomic lines and of the Lyman/Werner molecular bands of hydrogen, as a function of power delivered to the discharge (constant pressure 0.2 mbar). (b) Rotational temperature of hydrogen molecules and kinetic temperature of hydrogen atoms as a function of microwave power.

The relative intensity of the atomic and molecular hydrogen emissions has been investigated as a function of microwave power delivered to the plasma. The integral intensities of the Lyman-α, β lines and of the Werner and Lyman molecular bands are calculated for different powers applied to the discharge at constant pressure 0.2 mbar and the results are shown in Figure 3. The effect of self-absorption on the lines intensity, represented by the bars, was estimated assuming a hydrogen dissociation degree of 50% in the discharge and calculating the axial evolution of the absorbing H atoms density due to axial transport, diffusion, and wall reassociation [6]. The intensity of the molecular bands increases linearly from 100 to 200 W. Both the Lyman-α and Lyman-β line intensities increase by a factor of two when increasing the power from 80 W to 300 W. This is due to the increase of the plasma electron density as the power delivered to the discharge rises, since at low-pressure conditions the main creation mechanism of the excited Lyman-α, β states is by electron impact excitation. Values of the electron density are estimated to be of the order of 10^{12} cm^{-3} at the launcher position, decreasing by one order of magnitude towards the end of the plasma column. As shown in figure 3.b, the kinetic temperature of the hydrogen atoms (~5000 K) is found to be much higher than the rotational temperature of the hydrogen molecules (~900 K). This indicates that the hydrogen atoms and the gas are not in thermal equilibrium and the reason for this is connected with the main creation processes of excited atoms as observed in previous works [5].

4. Conclusions

Emission spectroscopy was used as a diagnostic, both in the VUV and in the visible regions of the electromagnetic spectrum to study the spectral features of hydrogen microwave driven discharges. It has been shown that hydrogen microwave plasmas have strong and energetic emissions in the 80 – 125 nm range, originating from atomic Lyman-α, β transitions and molecular Werner/Lyman bands. The intensity of these spectral emissions strongly rises as the microwave power is increased. This is due to the strong increase of the plasma electron density with the power delivered to the discharge. As in previous works, it has been found that the excited hydrogen atoms are not in thermal equilibrium with the background gas.

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