Quantification of the VUV radiation from RF discharges in hydrogen

U. Fantz[1,2], S. Briefi[2], D. Rauner[1,2], D. Wünderlich[1]

[1] *Max-Planck-Institut für Plasmaphysik, Boltzmannstr. 2, D-85748 Garching, Germany*
[2] *AG Experimentelle Plasmaphysik, Universität Augsburg, Universitätsstr. 1, D-86159 Augsburg, Germany*

Hydrogen discharges emit intense radiation in a broad wavelength region in the VUV. In order to quantify this radiation VUV measurement in the wavelength region from 120 nm to 280 nm have been carried out using a RF discharge. The dominant transitions are the Werner band (C–X), the Lyman band (B–X), the continuum (a–b) as well as the L_α line from the hydrogen atom. It is shown that depending on the pressure, up to 20% of the RF power (600 W) is found in the VUV, whereas only about 2% are emitted in the VIS represented by the Balmer series emission (H_α - H_ε) and the Fulcher emission (d–a).

1. Introduction

Low temperature, low pressure plasmas with atomic and molecular hydrogen are often characterised by a faint pink colour originating from the Balmer line emission and the Fulcher band emission (d $^3\Pi_u$ – a $^3\Sigma_g^+$ transition of the molecule, Figure 1). This conveys the impression that the radiant power of hydrogen plasmas compared to the power coupled into the plasma is negligible. On the other hand it is well known that hydrogen molecules emit a strong continuum radiation (a $^3\Sigma_g^+$ – b $^3\Sigma_u^+$ transition) (see [1] and references therein) and have resonant transitions in the VUV range. Figure 1 gives an overview on the electronic states of molecular hydrogen and the optically allowed transitions within the first three principle quantum numbers. Most prominent in the singlet system are the Lyman band (B $^1\Sigma_u^+$ – X $^1\Sigma_g^+$ transition) and the Werner band (C $^1\Pi_u$ – X $^1\Sigma_g^+$ transition). A band system consists of several vibrational bands and their rotational lines.

In order to quantify the VUV radiation of hydrogen plasmas a spectroscopic system with high spectral resolution is applied which has been absolutely calibrated from 120 nm to 280 nm. Thus, the radiant power of the atomic L_α line, Werner and Lyman band as well as the continuum radiation in this wavelength region can be obtained and compared to the radiation of the Balmer lines and the Fulcher band in the VIS: For the latter another dedicated spectroscopic system is also available. The radiant power is compared to the RF power used to generate the hydrogen plasma at pressures of 1, 3 and 6 Pa for two types of RF discharges: the ICP mode and a Helicon setup.

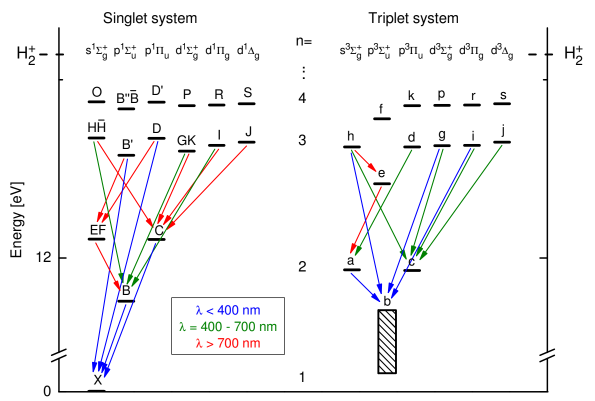

Figure 1: Energy level diagram of the hydrogen molecule with the optically allowed transitions for the main quantum numbers up to three.

2. Experiment and results

2.1. Experimental setup

The RF discharge (shown in Figure 2) consists of a quartz glass cylinder with a length of 40 cm and a diameter of 10 cm. The vacuum system is attached at the ends of the cylinder. Two types of RF antenna are used: a conventional RF coil with 5 windings (ICP mode) or the Nagoya-type III Helicon antenna which couples to the |m| = 1 Helicon modes [2] if an external magnetic field is applied. Via a matching network the antenna is connected to an RF generator (600 W power) which operates at a frequency of 13.56 MHz. An external magnetic field parallel to the cylindrical axis of the discharge vessel can be applied by using a pair of Helmholtz coils generating the maximum field strength of 12 mT.

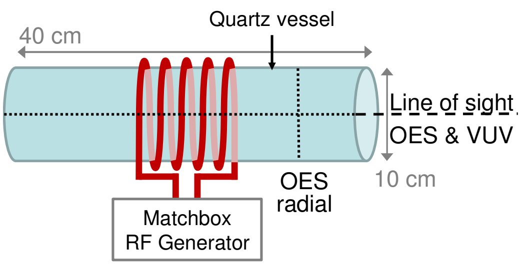

Figure 2: Sketch of the experimental setup.

The flexible experimental setup allows for comparing plasmas operated in ICP mode or with Helicon antenna with or without magnetic field [3].

The spectroscopic investigations are carried out using an axial line-of sight (radially centred) as indicated in figure 2. The Balmer line emission and the molecular Fulcher band emission (d-a transition) is measured by Optical Emission Spectroscopy (OES) utilizing a high resolution spectrometer ($\lambda_{FWHM} \approx 20$ pm). The analysis of the absolute emission and of line ratios is supported by the collisional radiative model `Yacora` for hydrogen [4,5] such that the electron density, the electron temperature and the density ratio of atomic to molecular hydrogen n_H/n_{H2} is obtained. By applying Abel inversion, the radial emission profile is derived from lateral measurements [3].

The spectroscopic system for the VUV spectral range consists of a high resolution spectrometer (McPherson, 1 m focal length, $\lambda_{FWHM} \approx 35$ pm) and a solar-blind photomultiplier. The system is relatively calibrated in the wavelength range from 120 nm to 280 nm by using the branching ratio method applied to the vibrational bands of nitrogen molecule [6]. An absolute calibration is achieved by using helium lines (around 280 nm) which can be detected by both, the VUV and the OES system, the latter being calibrated by a deuterium lamp in this wavelength region.

2.2. VUV and VIS spectra

Figure 3 shows calibrated emission spectra of the RF discharge in ICP mode at a pressure of 3 Pa. The wavelength region of the individual transitions is indicated; due the strong overlap of vibrational bands and the corresponding rotational lines in the VUV the assignment was done only for the Fulcher emission in the VIS range. The continuum spectrum of the a–b transition is overlapped by the continuum emitted from the Lyman band [7] caused by transition of the v' = 9 level of the B $^1\Sigma_u^+$ state into the dissociation limit.

The L_α line has the highest intensity with a peak intensity of 1.3×10^{23} Ph/nm/s/m^3. The most intense molecular emission originates from the first electronic state of the molecule, the Lyman band to which a wavelength range of about 60 nm (130 – 190 nm) can be clearly assigned. The Werner band overlaps partly with the Lyman band and extends from the indicated region towards lower wavelengths. Here, the wavelength range from 120 nm to 130 nm is assigned to the Werner radiation. The continuum emission is much weaker

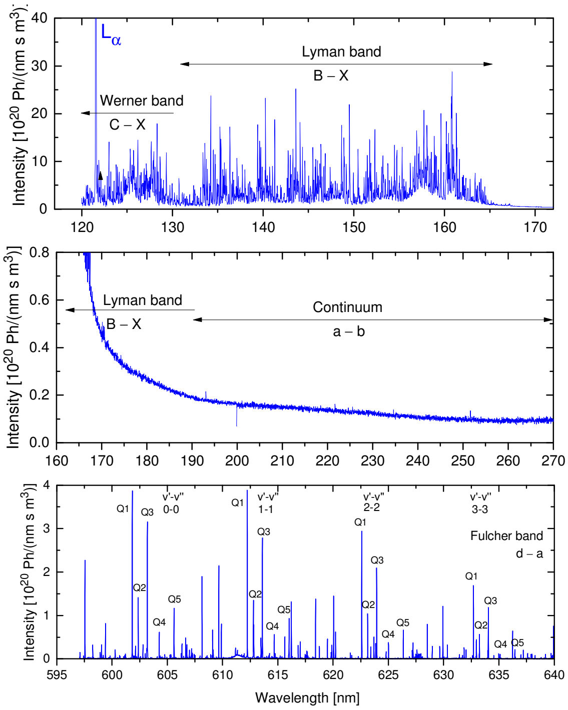

Figure 3: Example of the VUV and VIS spectra taken at a pressure of 3 Pa and 600 W RF power in the ICP mode.

than the Lyman and Werner bands. As the continuum is overlapped by the Lyman band the wavelength region from 190 nm to 280 nm has been chosen as representative for the continuum emission. Comparing the Lyman emission with the Fulcher emission, it becomes clear that the Fulcher band is much weaker. This is caused by the fact that this is a non-resonant transition and that the electron impact excitation from the ground state to the d $^3\Pi_u$ state is an optically forbidden transition resulting in a smaller excitation rate coefficient. The analysis of the vibrational bands and rotational lines is used to determine the vibrational and rotational temperature of the hydrogen molecule in the ground state, the latter representing the gas temperature [8]. The measured Fulcher emission of the first four diagonal vibrational transitions is extrapolated to the total Fulcher emission by using the procedure described in [8].

The continuum emission is simulated by using a corona model, i.e. balancing the electron impact excitation from the ground state with spontaneous emission applying vibrationally resolved spectral transition probabilities [7]. Figure 4 shows the continuum radiation for a certain set of plasma parameters used to fit the measured spectra. The shape of the continuum is determined predominantly by the vibrational population in the a $^3\Sigma_g^+$ state which is calculated from the vibrational population

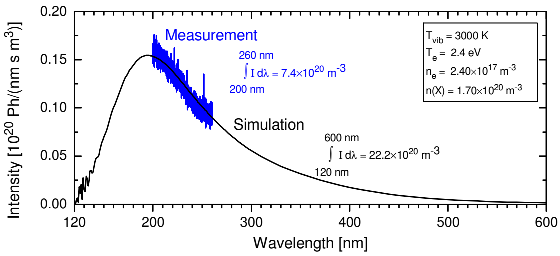

Figure 4: Simulation of the continuum radiation fitted to the measured radiation.

of the ground state using a vibrational temperature T_{vib}. Electron density n_e, electron temperature T_e and molecular hydrogen density n(X) predominantly determine the absolute intensity [7].

The continuum radiation ranges from 120 nm to 600 nm. The position of the peak around 200 nm strongly depends on the vibrational population in the excited state. Figure 4 demonstrates also that the measured continuum reflects only a part of the full continuum radiation. In particular, the lower wavelength range is not accessible by measurements as it is overlapped by the Lyman continuum (see Figure 3). In fact, the total integrated intensity is a factor of about three larger than the integrated intensity of the measurement. Ro-vibrationally resolved calculations of the Lyman band and the Werner band show, that the Lyman band is well represented by the measurements whereas for the Werner band only a small part can be measured as the band extends to wavelengths down to 90 nm [5].

2.3. Results

For determining the wavelength integrated radiant power of the individual bands the emission spectra are converted first into the spectral radiant flux density and integrated over the aforementioned wavelength regions (see also table 1). The L_α line and the Balmer lines are fitted by a Gaussian profile and then integrated. The L_α line is explicitly excluded from the Lyman band emission.

Measurements were carried out in the ICP mode at three pressures, whereas for the Helicon antenna measurements were carried out only at 1 Pa without and with additional axial magnetic field. For the latter the magnetic field was chosen such that the Helicon discharge operates in the low field peak [3]. The radial emission profiles obtained from radially measured intensities and the Abel inversion show a very flat emission profile for all cases with the exception for the Helicon discharge with magnetic field for which a deep hollow profile is obtained [3]. Thus, the integration of the radiant flux density over the discharge volume is well justified except for the Helicon discharge at the low field peak. In this case the radiant power has to be regarded as a lower limit. Table 1 summarizes the results.

Independent from the pressure and RF mode (ICP or Helicon), the largest radiant power originates from the Lyman band emission. The energy of the photons ranges from 9.5 eV to 6.5 eV. More than a factor of two smaller is the power emitted from the Werner band which is attributed to the fact that the Werner band is represented only within the measured 10 nm although the dominant part of the band system is between 90 nm and 130 nm. The radiant power from the atomic line radiation in the VUV, the L_α line with the photon energy of 10.2 eV, is despite the narrow wavelength region remarkably high compared to the molecular emission in this energy region (10.3 eV to 9.5 eV, Werner band). The continuum emission which covers about 100 nm spectroscopic range represents photon energies from 6.5 eV to 4.4 eV yields by far the lowest radiant power of the measured VUV radiation. In the visible range, the radiant power is very small which is mainly attributed to the low energy of the photons (Fulcher emission is at energies of about 2 eV).

Table 1: Radiant power for the individual emissions. For the molecular emission the following integrals are taken: C–X: 120-130 nm; B–X: 130-190 nm; a–b: 190-280 nm. VIS represents the Balmer lines (H_α-H_ε) and the total Fulcher emission (d–a). The power ratio denotes the sum of the measured radiant power to the RF power (600 W). ICP denotes measurements of discharges with the ICP antenna, H denotes the Helicon antenna; B denotes the axial magnetic field of 3.5 mT.

p [Pa]	L_α [W]	C–X [W]	B–X [W]	a–b [W]	VIS [W]	Power ratio
6^{ICP}	27.7	21.7	58.5	3.0	1.7	18%
3^{ICP}	24.0	20.1	53.9	2.7	1.6	17%
1^{ICP}	7.7	11.1	29.8	1.1	0.7	8%
1^{H}	6.3	14.7	38.2	1.1	0.8	10%
$1^{H,B}$	7.9	19.3	48.8	2.0	0.7	13%

In the ICP mode, the radiant powers of the individual emissions decrease with pressure with a pronounced decrease in the step from 3 Pa to 1 Pa. In particular the L_α line decreases strongly, a factor of about three is obtained whereas the molecular radiation is reduced by a factor of less than two. This indicates that the density ratio of atoms to molecules decreases, i.e. the degree of dissociation is lower at 1 Pa.

From the analysis of the Balmer lines and the Fulcher emission the plasma parameters are derived. Table 2 summarizes the results. As already indicated by the VUV data, the density ratio decreases strongly when the pressure is reduced from 3 Pa to 1 Pa. Furthermore, the electron density and the gas temperature decrease slightly, whereas the electron temperatures increase with lower pressure as can be expected from a simple ionisation balance.

Table 2: Plasma parameters obtained from the analysis of the Balmer line radiation and the Fulcher band emission. The estimated error is ± 10% for T_e, ±20% for n_e and the density ratio atoms to molecules and ±50 K for T_{gas}. n_0 denotes the particle density of the neutrals using the relation $p = n_0 k_B T_{gas}$.

p [Pa]	T_e [eV]	n_e [m^{-3}]	n_H/n_{H2}	T_{gas} [K]	n_0 [m^{-3}]
6^{ICP}	2.0	2.7×10^{17}	29%	880	4.9×10^{20}
3^{ICP}	2.4	2.4×10^{17}	32%	850	2.6×10^{20}
1^{ICP}	3.0	1.8×10^{17}	21%	750	9.7×10^{19}

As summarized in Table 1 the sum of all measured radiant powers represents about 10% to 20% of the power used to generate the plasma. The amount decreases with decreasing pressure dominantly determined by the reduction of the particle density of the neutrals which can be not compensated by the increasing electron temperature.

The change of the ICP antenna for the Helicon antenna results in an increased radiant power particularly for the Lyman band. The application of the axial magnetic field results in an increase of all emissions but again the Lyman band is stronger influenced. Compared to the ICP mode the power ratio is increased by a factor of 1.5. It should be kept in mind that this represents a lower limit only, as the radial emission profile is strongly hollow.

3. Conclusions

RF discharges at 600 W power were investigated with the aim to quantify the radiant power in the VUV range and to determine the contribution of the individual electronic transitions of the molecule as well as those of the atoms. The analysis of the wavelength region from 120 nm to 280 nm showed that the first resonant transition of the hydrogen molecule, i.e. the Lyman band contributes most: depending on the pressure up to 50 W power is radiated. In contrast, the continuum radiation covering the largest wavelength range (about 100 nm) contributes with about 3 W at maximum only. The trends are almost independent on the type of antenna used for the plasma generation, however by applying a magnetic field with a strength of 3.5 mT and thus operating in the low-field peak of a Helicon discharge, the radiant power of all transitions observed increases but the Lyman band increases stronger than the other transitions. The contribution of the radiation in the VIS (Balmer lines and Fulcher emission) is at 2 W at maximum and almost negligible compared to the Lyman band.

The ratio of the radiant power (VUV and VIS) to the RF power achieves values of up to about 20% decreasing with pressure to 10%. In other words, 120 W (or 60 W) power is emitted by the measured radiation.

On the one hand this huge amount of power should be considered in plasma modelling as it has consequences on the power balance. On the other hand the energy of the photos is very high, e.g. photons from the Lyman band cover the energy range from 6.5 eV to 9.5 eV. Depending on the quantum efficiency of surfaces photoelectron emission might become a relevant process in the plasma surface interaction, influencing the plasma sheath properties. First investigations have been started to determine the relevance of the VUV emission for the production of negative hydrogen ions at caesiated surfaces [5].

4. References

[1] B.P. Lavrov, A.S. Melnikov, M. Käning, J. Röpcke, *Phys. Rev. E* **59** (1999) 3526.

[2] F.F. Chen, Phys. Plasmas **3** (1996) 1783.

[3] D. Rauner, S. Briefi, U. Fantz, *Proc. ICPIG XXXII, Iași, (2015).*

[4] D. Wünderlich, S. Dietrich, U. Fantz, *J. Quant. Spec. Radiat. Transfer* **110** (2009) 62.

[5] D. Wünderlich, U. Fantz, S. Briefi, D. Rauner, NNBI Team, *Proc. ICPIG XXXII, Iași (2015).*

[6] M. J. Mumma, J. Opt. Soc. Am. **62** (1972) 1459.

[7] U. Fantz, B. Schalk, K. Behringer, *New J. Phys.* **2** (2000) 7.1.

[8] U. Fantz, B. Heger, *Plasma Phys. Control. Fusion* **40** (1998) 2023.