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Optimization of microwave Hydrogen plasma discharges to mimic Lyman α (121.6 nm) solar irradiations

Y. Benilan (1), M.-C. Gazeau (1), E.-T. Es-Sebbar (2), A. Jolly (1), E. Arzoumanian (1), N. Fray (1) and H. Cottin (1)

(1) LISA (Laboratoire Interuniversitaire des Systèmes atmosphériques), UMR CNRS 7583, Universités Paris-Est Créteil Val de Marne (UPEC) and Paris Denis Diderot, CNRS-UMR 7583, IPSL, 61, avenue du Général de Gaulle, 94010 Créteil Cedex, France
(2) King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia

Abstract

We present the results obtained in low pressure microwave plasmas sustained in flowing mixtures of H_2/He with the addition of Ar. We investigated experimentally their properties in terms of VUV emissions and photons fluxes using a VUV spectrometer. The spectra are dominated by Lyman alpha at 121.6 nm and H_2 band emissions around 160 nm. The addition of Ar in the H_2/He mixture largely affects the line emission. Lyman alpha emission increases by a factor of 2 in the presence of 20% Ar while the H_2 bands decreases. This is a way to improve the monochromaticity of the lamp. Chemical actinometry method is used to determine the photons flux. We measured the temporal evolution of CO production issued from CO_2 photolysis. A photon flux of 4.10^{15} s^{-1} is obtained for a power of 100 W. The photon flux varies linearly with the power at a rate of 6.10^{13} $s^{1}.W^{1}$. Those results are applied to studies on methane photolysis and help to explain mismatch between photochemical models and experiments.

1. Introduction

Plasma emissions are widely used as continuous vacuum ultraviolet photon sources in astrophysical photochemical studies. In particular, hydrogen plasma laboratory sources should nicely mimic the VUV photons coming from the Sun since those mainly concentrate in the Lyman alpha transition at 121.6 nm. Consequently, those irradiation sources are widely used in photochemical simulations of planetary atmosphere chemistry.

One of the most challenging objectives of such experiments is to retrieve accurate quantitative laboratory data allowing a reliable comparison with theoretical and/or observational ones. This task can only be achieved when the irradiation source delivers a well characterized radiation in terms of photon flux and wavelength dependency.

2. Study of light emission

In hydrogen plasma lamps, the emitted radiation mainly comes from excited H atoms that can emit spontaneously from n=2 to n=1 level after electron excitation. Nevertheless, part of the dissociated molecular hydrogen will recombined in an excited $^1\Sigma_u$ state which can spontaneously radiate to ground state emitting radiations around 160 nm. Such radiation not present in the solar spectrum decreases the spectral purity the lamp (Fig 1).

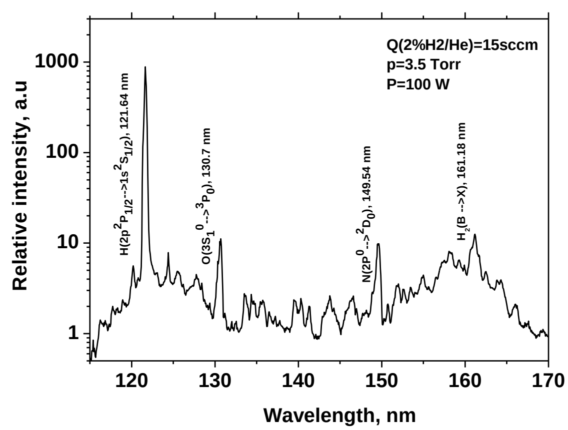

Figure 1: Typical spectrum between 115 and 170 nm at 0.6 nm resolution. The main features are identified (H, H_2, O and N). Experimental conditions: H_2/He(2/98), p=3.5 Torr, flow rate=15 sccm and P=100 W.

In our experiment, the plasma is generated in a quartz tube (i.d. 8 mm, e.d. 10 mm, length 20 cm) in which the flow of H_2 /He (/Ar) gas mixture is injected. The microwave excitation is coupled to the plasma through a McCarrol cavity (Opthos Instruments, Inc.) powered by a microwave generator (SAIREM-2.45 GHz-300W).

In order to optimize such lamp and to obtain the maximum flux at 121.6 nm, one can use heavy atoms such as Ar that are excited to metastable states or can be ionised inside the discharge. The excited Ar($^3P_2$) will then react with H_2 and form excited H. The reaction of Ar^+ ions with H_2 form ArH^+ that will then recombine with electrons and dissociate to create excited H atoms. So, those two processes tend to increase the intensity of Ly-α while the emission from H_2 is decreasing (Fig. 2).

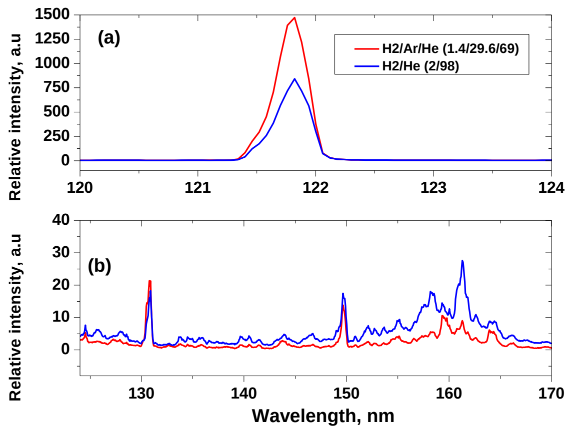

Fig 2: Variation of the VUV emission upon Ar addition in the H_2/He mixture, in the wavelength range (a) 120-124 nm and (b) 124-170 nm.

In our experiments, we use CO_2 photolysis as an actinometer to retrieve the absolute photon flux. For this, we quantify photon absorption by CO_2 using the absorption cross section of CO_2 with value at 121.6 nm taken from [3]. We follow the CO appearance using IRTF absorption. The temporal dependence of the CO density is determined by systematic comparison of experimental spectra with synthetic ones using spectroscopic parameters from the GEISA database [1]. We found a linear relation between the emitted photon flux and the power deliver to the discharge (Fig 3). A photon flux of 4.10^{15} s^{-1} is obtained for a power of 100 W. The photon flux varies linearly with the power at a rate of 6.10^{13} $s^{1}.W^{-1}$.

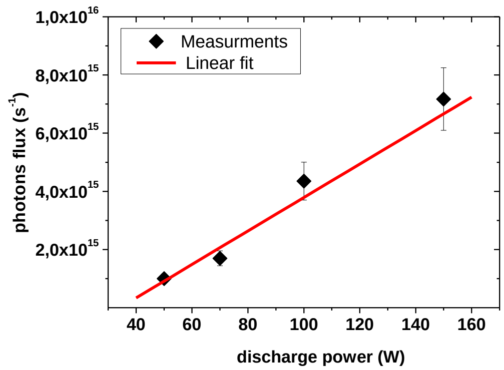

Fig. 3: Dependence of the photon flux on the discharge power (flow rate=15 sccm, 2% H_2/He/Ar, p=3.5 Torr)

3. Application to CH_4 photolysis

Methane photolysis at Lyman alpha is studied in the frame of a program dedicated to simulations of Titan's atmosphere [2]. Irradiations have been performed using a microwave He/H_2 (98/2) discharge lamp. The chemical evolution of the gas mixture resulting from the irradiations is monitored by FTIR. The determination of the chemical mechanism involved requires the comparison with a dedicated 0D kinetic model in which photolysis rates are fundamental parameters. Their values depend on the emission spectrum of the source. We will emphasize on the implications of this fact for the study of the methane photolysis at Lyman α and its comparison with the chemical mechanisms involved.

Acknowledgements

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