26th EPS Conf. on Contr. Fusion and Plasma Physics, Maastricht, 14 - 18 June 1999 ECA Vol.23J (1999) 1585 - 1588

TALIF Measurement of Dissociation Degree in Pulsed Planar Microwave Hydrogen Plasma

H. Lange[1], X.R. Duan [2]

[1] *Institute of Low Temperature Plasma Physics, Friedrich-Ludwig-Jahn-Str. 19, 17489 Greifswald, Germany*
[2] *Southwestern Institute of Physics, P.O.Box 432, 610041 Chengdu, Sichuan, P.R. China*

By means of two-photon-absorption laser induced fluorescence (TALIF) at 205 nm the temporal dependences of the absolute atomic hydrogen density were measured in the pulsed planar microwave plasma source. The microwave discharge was operated at 2.45 GHz, the modulation frequency is 500 Hz with 1:1 duty cycle, its power is 0.6 kW in lower power phase and 3.6 kW in higher power phase. The absolute density of atomic hydrogen was obtained by calibrating with NO_2 titration in a flow tube reactor. The dissociation degree was determined under different discharge pressures.

INTRODUCTION

Microwave plasmas are of great interest in the field of plasma processing and basic research because they can supply considerable concentrations of activated neutral particles. Hydrogen is one of the most common reactive gases added to these plasmas, and it is believed to play an important role(1). Determination of the absolute atomic hydrogen density is very important to understand the plasma-induced processes and to determine the degree of the dissociation for modelling the reaction kinetics in these plasmas.

Laser-induced fluorescence(LIF) is a sensitive and non-perturbing probe for the detection of atoms and molecules, especially it can detect atoms in the regions of the plasma where the temperature is too low to excite appreciable emission. For atomic hydrogen direct resonance fluorescence measurement is inconvenient because vacuum-UV source and optics are required. Two-photon absorption laser induced fluorescence, however, allows operation with excitation in the UV, and detection in the visible, it was successfully used in a few kinds of experiments, such as in the glow discharge(2), arc discharge(3)etc.

There are a lot of excitation schemes for measuring atomic hydrogen by LIF(4). In our work the excitation scheme with two photons at 205 nm was used. The excitation and fluorescence processes are shown in Fig. 1. H atoms are excited from the ground level 1s ^{2}S to the excited level 3d ^{2}D by absorbing two photons with 205 nm. Fluorescence is observed at 656.3 nm because of n=3 to n=2 radiation decay.

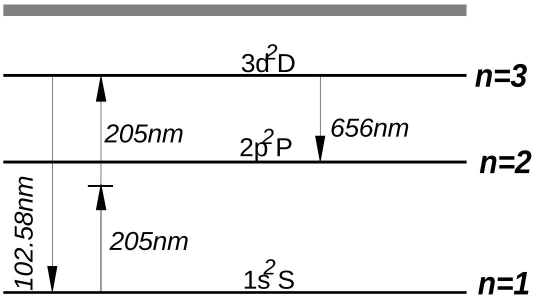


Fig.1: Two-photon excitation scheme for atomic hydrogen

EXPERIMENTAL SETUP

1. Experimental setup

The experimental setup is shown in Fig. 2. The pulsed tunable laser system consists of a Nd:YAG laser , a dye laser and a frequency conversion system. The laser energy is about 800 mJ/pulse at 532 nm and pulse duration is 5 ns (FWHM) with 10 Hz repetition frequency. 532 nm laser is used to pump rhodamine 640 in the dye laser and about 90 mJ laser radiation at 615 nm is obtained. The 205 nm radiation is generated by tripling the dye frequency 615 nm (ω_0). In a KD*P crystal the dye laser frequency is doubled to provide radiation at 307.5 nm ($\omega_{1/2}$). Both dye fundamental and second harmonic are mixed in a BBO crystal producing the sum frequency $\omega_0 + \omega_{1/2} = \omega_{1/3}$ at 205 nm with a maximum energy of 1.8 mJ/pulse.

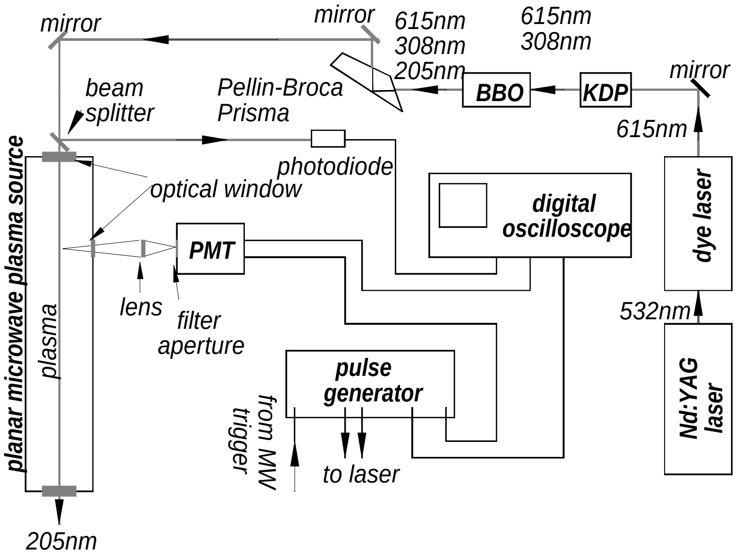


Fig. 2: Laser system and experimental setup

The 205 nm laser beam was transferred in the planar microwave plasma source$^{(5)}$ parallel to the direction of the optical axis. The fluorescence was observed perpendicular to the laser beam by using a gated photomutiplier R928. An interference filter at 656 nm with 1nm FWHM was placed before the slit to reduce other emission from plasma. The output from photomultiplier was digitalized by a 1 GHz digital real-time oscilloscope (Tektronix TDS 680B). In our experiments, an unfocused laser beam was used. Its advantage is, that the reactive volume of the 205 nm laser with the atomic hydrogen can be ensured to be the same during the experiments and calibration.

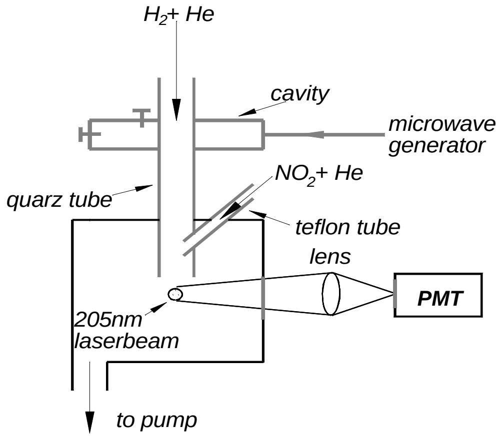
The experimental conditions were as follows: the microwave frequency 2.45 GHz, flow of the hydrogen 50 sccm, the modulation frequency of the microwave power 500 Hz with duty cycle 1:1. The power of the microwave was 3.6 kW in the high power phase and 0.6 kW in the low power phase.

2. Absolute calibration in a flow tube reactor

LIF signal must be calibrated to determine the absolute densities. In our experiment, the calibration of the atomic hydrogen fluorescence against the atomic hydrogen density was made in a special flow tube reactor (Fig. 3). A microwave discharge creates atomic hydrogen in this reactor. To obtain an expected intensity of LIF signal the gas flow and microwave power are adjusted. According to the reaction$^{(6)}$:

$$NO_2 + H \rightarrow NO + OH \qquad (1)$$

the atomic hydrogen is destroyed by NO_2. If NO_2 increases to a certain amount, the atomic hydrogen would be extinguished, and the fluorescence signal is dissapeared. Fig. 4 shows the NO_2 titration results. From the measured flow rate of NO_2 the absolute atomic hydrogen density can be calculated.



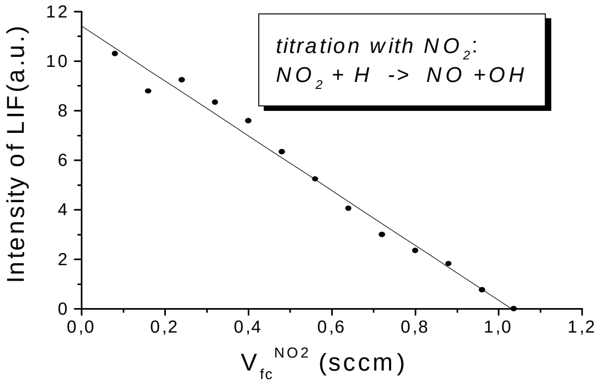


Fig. 3: Flow tube reactor

Fig. 4: Results of NO_2 titration

RESULTS AND DISCUSSIONS

Fig. 5 shows the experimental results of the atomic hydrogen density at 0.55 mbar, 1.1 mbar, 2 mbar and 4 mbar. At the beginning of the high power phase, the atomic hydrogen density increases rapidly to a maximum value because of the high power pulse. At a pressure of 4 mbar the density of the atomic hydrogen is nearly unchanged in this phase. By lowing the gas pressure the density at the beginning of the high power phase decreases rapidly with the time.
This time behaviour of the atomic hydrogen density at lower pressure can be explained with the gas temperature volume effect. At lower pressure the difference of the gas temperature T_{gas} between two power phases is great. In Fig. 6 the atomic hydrogen density and the inverted gas temperature (T_{gas} measured by emission spectroscopy$^{(7)}$) at 0.55 mbar are plotted against the time. At the first 200 μs of the high power phase, the gas temperature increases dramatically, from about 500 K to about 1000 K, therefore the density decreases rapidly because the gas pressure is fixed. According to the measurements, the time constant for $1/T_{gas}$ is nearly the same as the decay time of the measured atomic hydrogen density in high power phase.

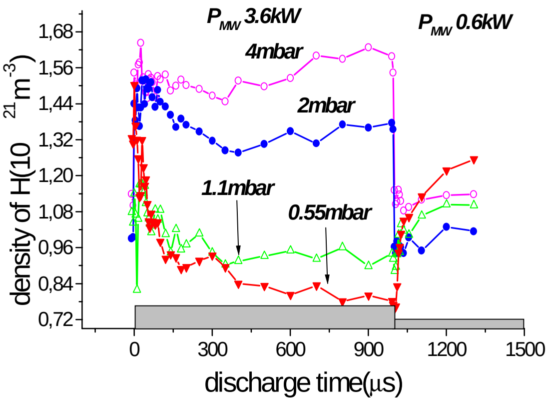


Fig. 5: Ttime dependance of atomic hydrogen density

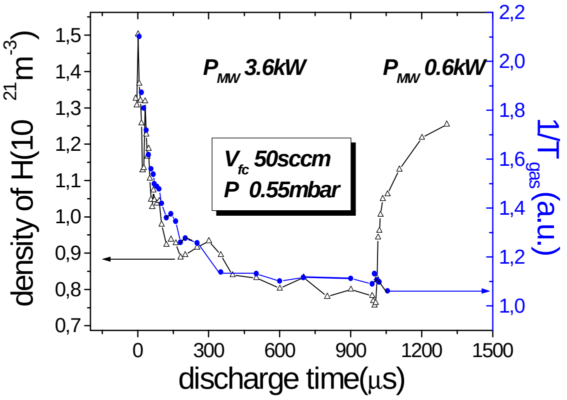


Fig.6 : Ttime dependence of gas temperature

According to the measured atomic hydrogen density, the dissociation degree was determined (see Fig.7). In low power phase (t=1500 µs), the dissociation degree is estimated to be about 7 % at 0.55 mbar and 2 % at 2 mbar, in high power phase (t=500 µs), it is about 16.5 % and 5 % respectively. The difference of the dissociation between the lower pressure and higher pressure could be explained with an effect of the electron temperature. Obviously, in the high power phase there is more atomic hydrogen at 2 mbar than at 0.55 mbar, but according to the measurement, in the higher power phase the H_α spontaneous emission is smaller at 2 mbar than at 0.55 mbar (see Fig. 8). One possible reason is, that the electron temperature at 0.55 mbar is much more higher, this results in more dissociation, excitation and ionization.

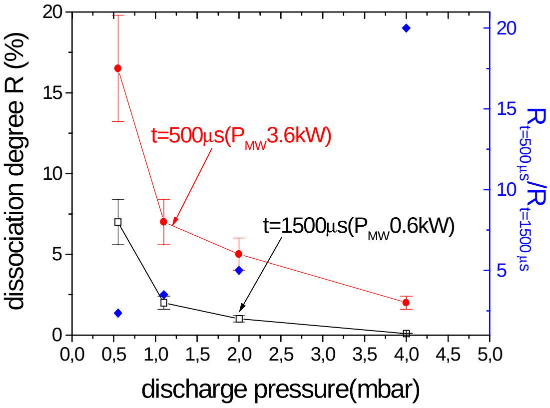


Fig. 7: Dissociation degree

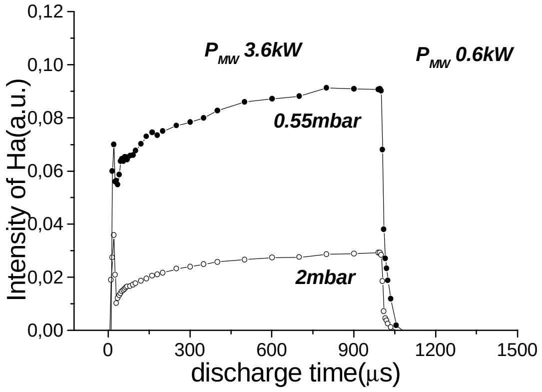


Fig. 8: Spontaneous emission of H_α

CONCLUSION

The temporal dependence of the absolute atomic hydrogen density was measured in pulsed planar microwave plasma source by means of TALIF. The dissociation degree was obtained. For better interpretation and understanding of the experimental results and processes in the plasmas, measurements of the gas and electron temperature and the spatial distribution of the atomic hydrogen density are necessary.

REFERENCES

(1) D.P.Dowling, et.al., Diamond and Related Materials **3** (1994) 702.
(2) J.Amorim, G.Baravian, and G. Sultan, Appl. Phys. Lett. **68** (1996)1915.
(3) S.C. Snyder, et.al., Physical Review E. **52** (1995)2999.
(4) U. Czarnetzki, K. Miyazaki, T. Kajiwara, and K.Muraoka, M. Maeda, H.F. Döbele, J. Opt. Sci. Am. B, **11**(1994) 2155.
(5) Andreas Ohl, Microwave Discharges: Fundamental and Application, Edited by C.M. Ferreira and M.Moisan, Plenum Press, New York, 1993, P205.
(6) D.W.Setser, Reactive Intermediates in the Gas Phase, Academic Press, New York,1979.
(7) N. Lang, et.al. Workshop on Frontiers in Low Temperature Plasma Diagnostics III, 15 - 19 February 1999, Saillon (Valais),Switzerland. P.II.13.