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TALIF INVESTIGATION AND MODELLING OF THE ABSOLUTE N(^{4}S) DENSITY IN A N_2-CH_4 LATE AFTERGLOW

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Introduction

The purpose of the present work is the experimental and numerical study of the absolute ground-state nitrogen atoms density N(^{4}S) in the late afterglow of a pure N_2 flowing microwave discharge in which different amounts of CH_4 have been injected at 25 cm downstream from the nitrogen discharge. The absolute N(^{4}S) concentrations have been measured using Two-photon Absorption Laser-Induced Fluorescence (TALIF), while a detailed kinetic model has been developed to simulate both the discharge and the post-discharge regions. Theoretical predictions are then compared to experimental measurements.

Experiment

The experimental set-up is presented in Fig. 1. The characteristics of the microwave plasma source with its post-discharge as well as the laser system used to generate the UV photons that probe N(^{4}S) atoms have been detailed elsewhere [1]. In brief, we just remind here that the nitrogen microwave plasma discharge is generated in a quartz tube through a surfatron device connected to a microwave generator at 2.45 GHz with a maximum output of 300 W. The quartz tube (8 mm external diameter, 6 mm internal diameter and 50 cm of length) is cooled by an air gas flow and connected to a post discharge reactor of 20 cm in diameter. The CH_4 amounts are injected into late afterglow, at 25 cm downstream from the nitrogen discharge (Fig.1).

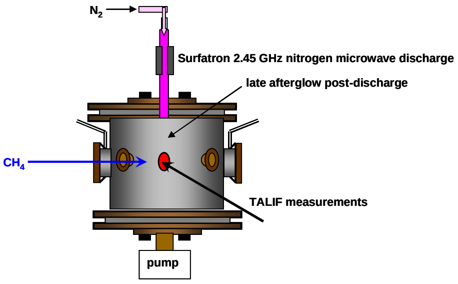

Fig. 1: Experimental set-up.

For TALIF measurements, a Q-switched Nd:YAG laser (repetition rate=10 Hz, pulse-width duration=8 ns) with second harmonic generation at 532 nm is used to pump the dye laser system of RhB/Rh101 mixture in ethanol. The output which maximum of efficiency is observed at 615 nm is doubled and tripled using (KDP) and (BBO) crystals generating UV photons at 206.7 and

204.3 nm and with a maximum energy of 4 mJ. These wavelengths permit the two photons excitation of nitrogen (N ($2p^3\ ^4S_{3/2}$) → N ($3p\ ^4S_{3/2}$) transition) as well as krypton (Kr ($4p^6\ ^1S_0$) → Kr (5p' $[3/2]_2$) transition) which is used as a reference for TALIF calibration [2]. The fluorescence signals are observed in the 742 and 746 nm region and at 826.3 nm corresponding to N ($3p\ ^4S_{3/2}$) → N ($3s\ ^4P_{1/2,3/2,5/2}$) and Kr (5p' $[3/2]_2$) →Kr (5s' $[1/2]_1$) transitions respectively. The photons are imaged perpendicularly to the laser beam by a lens system to the detector. Under these conditions, it is then possible to determine the absolute nitrogen atom density [1].

Kinetic model and results

The present model is based on a previous work [3], where we start from a self-consistent determination of the active species concentrations in the pure N_2 discharge. The obtained results are then considered as initial conditions in a system of coupled time-varying kinetic master equations, in the near afterglow where only N_2 exists. Then we consider the introduction of methane into the post-discharge, at 25 cm downstream from the end of the discharge (see Fig. 1). At this point, we solve a system of time-varying kinetic rate balance equations for a N_2-CH_4 mixture.

Fig. 2 shows the dependence of the absolute N (^4S) atom density as a function of the methane mixing ratio, calculated by the numerical model as well as those measured for gas pressures of 2933, 2133 and 1600 Pa. The comparison between the two studies shows a similar behavior even though theoretical predictions are always higher by about a factor of 3. The addition of CH_4 in the late afterglow induces no significant decrease of the N (^4S) density up to a methane mixing ratio of 15%. Our simulations show that besides the three body recombination $N+N+N_2 \rightarrow N_2(B)+N_2$, the nitrogen atoms are mainly destroyed in the mixture by collisions with CH_3, H_2CN and NH through the mechanisms $N+CH_3 \rightarrow H_2CN+H$, $N+H_2CN \rightarrow HCN+H$ and $N+NH \rightarrow N_2+H$, respectively. As CH_4 is added into the post-discharge, the depopulating rates of these three mechanisms become larger, leading then to a decrease of [N(^4S)].

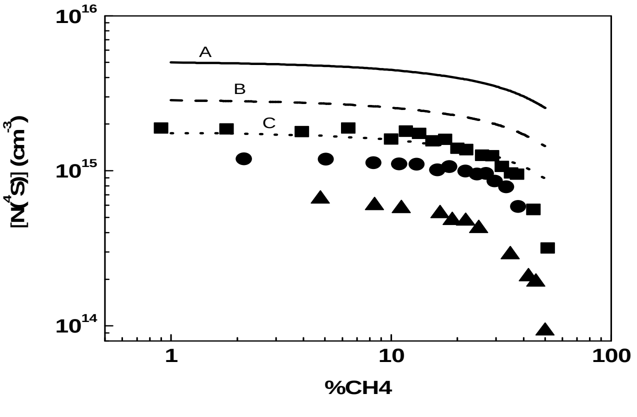

Fig. 2: Measured (symbols) and theoretical (curves) absolute densities of N(^4S) atoms in N_2/CH_4 mixture as a function of the CH_4 mixing ratio injected in the afterglow for a N_2 flow rate of 500, 200 and 100 sccm corresponding to pressures of 2933 Pa(■, curve A); 2133 Pa (●, curve B) and 1600 Pa (▲, curve C), respectively. The microwave discharge power is 100 W.

Reference

[1] Et Es-sebbar, Y. Benilan, A. Jolly and M. C Gazeau, 2009 *J. Phys. D: Applied Physics* **42** 135206
[2] K. Niemi, V. S. von der Gathen and H. F. Döbele, 2001 *J. Phys. D: Applied Physics* **34** 2330-2335
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