

Unravelling *in situ* atomic and molecular kinetics by LCIF and Thomson scattering

E. A. D. Carbone^(*), J. M. Palomares, S. Hübner, J.J.A.M. van der Mullen

*Department of Applied Physics, Eindhoven University of Technology,
P.O. Box 513, 5600 MB Eindhoven, The Netherlands*

^(*) e.a.d.carbone@tue.nl

The determination of the main excitation kinetics channels in the production of active species in a plasma is of main importance for the understanding and control of plasmas. In this contribution we will present the combination of two diagnostics methods, namely Laser (Collisional) Induced Fluorescence (LCIF) and Thomson scattering. The latter allows to measure locally the electron temperature and density meanwhile LCIF probes the excitation channels of selected atomic/molecular species with nanosecond time resolution after perturbation of the steady-state plasma with a nanosecond pulse tuned to an atomic transition.

Emission, absorption, fluorescence and scattering measurements are often used to determine steady state densities of selected species in the gas phase. The first two methods rely on a line-of-sight (and so on the assumption of a radial/axial profile of the plasma) meanwhile scattering and fluorescence methods allow the determination of the density/temperature within a 3D space provided adequate calibration. Very high temporal resolution can be also achieved depending of the laser used. Despite their high accuracy and complexity, these methods do not give however any direct information about the reaction channels in the plasma as only one parameter is measured at the same time. Power interruption measurements is another diagnostic method which gives a global plasma response but no direct information about selective excitation kinetics channels. This technique is also limited by the rise/decay time of the power supply which, in the case of microwave plasmas, is typically about a few microseconds.

In this contribution we will present a different approach to probe plasma kinetics with high spatial resolution ($\sim 50\mu\text{m}$) and nanosecond time resolution using the combination of two of the aforementioned diagnostic tools. Thomson scattering (for the determination of local electron density and temperature values) and nanosecond time resolved laser (collisional) induced fluorescence (LCIF) are performed on an intermediate pressure argon microwave plasma (with molecular admixtures).

A microwave plasma source belonging to the category of surface wave discharges (SWD) is investigated by means of the combination of these two methods. The excitation channels from electrons or heavy particles kinetics could be separated while resolving spatially the plasma and changing the plasma conditions such as the pressure and/or gas composition.

Thomson scattering (TS) is a non-intrusive and local method which allows the determination of the electron temperature and density (n_e , T_e) provided that not too high laser fluencies are used [1]. The plasma source used in this study was previously characterized both axially and radially by TS [2, 3] for different pressures of argon. Due to the small radius of the plasma (3 mm) the electron density follows a Bessel profile and the electron temperature is almost flat across the radius in a rather large pressure range (2-80 mbar) [3].

The time resolved L(C)IF experiments were performed with a dye laser pumped by a Nd-YAG laser. The pulse rep-rate of the YAG laser can be as high as 5kHz, with a maximum energy per pulse of 4 mJ; the pulse-duration is about 8 ns. The dye laser was fed with a pyridine-1 dye to get a tunable wavelength in the range 690-720 nm. In this wavelength range the Argon $1s_5$ metastable state (In Paschen's notation) was pumped by the laser towards a $2p_x$ state which could relax radiatively. The collisional induced fluorescence from nearby levels was monitored while tuning a spectrometer to a corresponding radiative transition of that level (see figure for more details and a schematic response of the atomic system).

One of the main properties of the SWD plasma used in this study is that the electron density varies (almost) linearly along the plasma column and does not oscillate with the E field. The electron creation

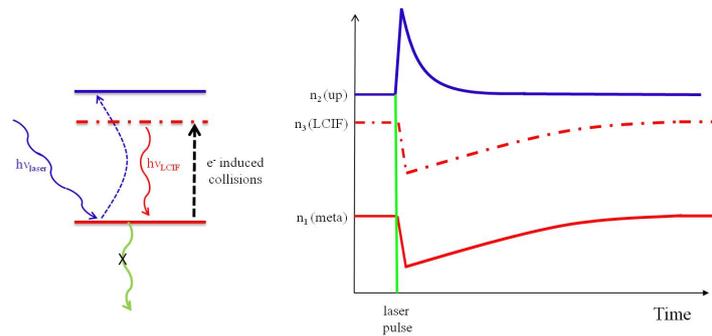


Fig. 1: Schematic of the excitation used to probe argon metastable kinetics as a function of electron density and temperature. The radiative level n_3 is strongly coupled to the metastable level n_1 and its nanosecond time resolved response follows the one of the metastable state which was depopulated by the laser pulse.

temperature remains on the other hand approximatively constant both radially [3] and axially [4]. For one plasma condition, this SWD is then an ideal candidate to probe the effects of the electron induced excitation processes and decouple them from heavy particles excitation transfers and/or quenching while resolving spatially the plasma.

The kinetics of argon metastables were probed as a function of the electron density and the pressure. A transition between two excitation regimes is found for a pressure of about 10 mbar indicating a production channel of argon metastables from dissociative recombination of Ar_2^+ in the high pressure limit [5] which overcome production channels from electron excitation only.

Small admixtures of nitrogen were added to study the effects of excitation transfer between Argon excited states and nitrogen molecules. A direct excitation transfer between Ar 4s state and N_2 in the ground state could be measured. This mechanism is known in the litterature [6], but to the best of our knowledge, it is the first time that this reaction was measured *in situ* in the plasma phase.

Argon-hydrogen plasmas were also investigated in the same way and atomic hydrogen was found to be sensitive both to Argon 4s and 4p states. Positive and negative responses of the Balmer serie were measured subsequent to the laser pulse who was disturbing only the Argon system. Some excitation transfer between Argon 4s and hydrogen atoms/molecules are known and used in collisional radiative models for Ar/H_2 but also excitation transfers between Argon 4p and atomic hydrogen are reported here.

Based on the results presented here, the combination of LCIF and TS is assessed and demonstrated as a powerful and very sensitive tool to unravel *in situ* known, as well as unknown, electron excitation kinetics as well as excitation transfer between heavy particles in the plasma phase.

References

- [1] E.A.D. Carbone, J.M. Palomares, S. Hübner, E. Iordanova and J.J.A.M. van der Mullen *JINST* **7** (2012) C01016.
- [2] J.M. Palomares, E. Iordanova, E.M. van Veldhuizen, L. Baede, A. Gamero, A. Sola and J.J.A.M. van der Mullen *Spectrochim. Acta At. Spectrosc* **B65** (2010) 225-233.
- [3] E.A.D. Carbone, S. Hübner, J.M. Palomares and J.J.A.M. van der Mullen *J. Phys. D : Appl. Phys.* *submitted* (2012).
- [4] N. de Vries, E. Iordanova, A. Hartgers, E. M. van Veldhuizen, M. J. van der Donker and J. J. A. M. van der Mullen *J. Phys. D : Appl. Phys* **39** (2006) 4194-4203
- [5] S. Hübner, J. M. Palomares, E. A. D. Carbone and J. J. A. M. van der Mullen *J. Phys. D : Appl. Phys* **45** (2012) 055203
- [6] M. Touzeau and D. Pagnon *Chem. Phys. Lett.* **53** (1978) 355-360