Spatial and Temporal Investigation of Atomic Oxygen Generation from ns-Pulsed Plasma using TAPLIF

J. Ravelid[1] , J. Sun[1], V. Kornienko[1], Y. Bao[1] E. Kristensson[1] and A. Ehn[1]

[1] *Division of Combustion Physics, Lund University, Lund, Sweden*

Abstract: Imaging of atomic oxygen has been performed on a pin-to-pin electrode, high voltage ns plasma in pure oxygen, using a two-photon laser excitation scheme. Spatial distributions and temporal dynamics are studied by delaying the laser excitation relative to the plasma. Single-shot imaging capabilities are shown and the lifetime of both plasma emission and atomic oxygen distribution are estimated. Furthermore, a new strategy for supressing plasma emission is demonstrated.

Keywords: Plasma, nanosecond pulsed plasma discharge, atomic oxygen, fs-TALIF, SLIPI, single-shot

1. Introduction

Atomic oxygen (O) is a highly reactive species and plays a central part in many chemical processes, including plasma-assisted processes [1,2,3]. As such, it is of vital interest for both experimentalists and modellers when trying to predict the behaviour of plasma-based systems and experimental models. Despite this interest, the available information on the temporal dynamics and spatial distribution of O is very sparse, mainly due to the inherent difficulty of such measurements.

So far, 0D (point) and 1D (line) measurements have been performed [4]. However, due to the very low excitation probability in the currently used excitation scheme, no single-shot 2D imaging of O has been achieved until now.

Furthermore, a challenge that often affect such measurements is the inherently strong plasma emission, that limits how early in the plasma formation process laser-induced information can be detected [5].

Hence, this work focuses on developing a method to distinguish between laser-induced light and plasma emission to carry out single-shot Two-photon Atomic Planar Laser-Induced Fluorescence (TAPLIF).

2. Experimental Setup and Method

A schematic of the setup, including the plasma generation and the optical diagnostics measurement, is displayed in Figure 1.

Oxygen gas is provided through a grounded porous plug with a diameter of 60 mm. A pin electrode is situated in the middle of the porous plug. A second pin electrode, located 30 mm above the porous plug, is connected to an ns-pulse high-voltage generator. The power supply is capable of generating pulses up to 200 kV, with a pulse duration of 4-6 ns. This configuration yields a ns plasma discharge in oxygen.

In the current work, the plasma generator is run at 30 kV and 1 Hz, to guarantee that consecutive events are independent.

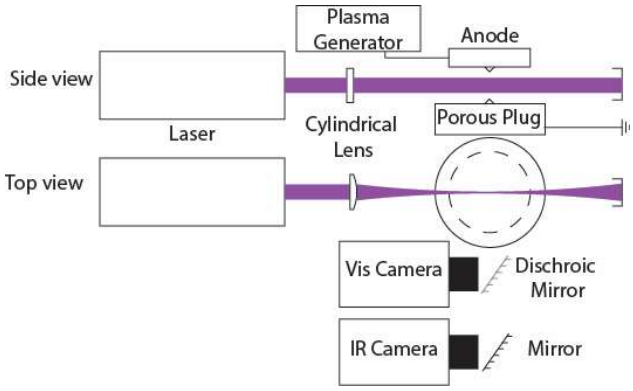

Figure 1: A schematic of the setup. Laser pulses are formed into a 2D sheet that transects the plasma volume in the oxygen flow. The induced fluorescence is detected by a gated ICMOS camera sensitive in the near infrared (NIR), while the plasma emission is obtained in the visible range by another ICMOS camera sensitive in the visible range (VIS).

O is detected TAPLIF and the excitation scheme is displayed in Figure 2. Laser pulses, with a duration of 40 fs, wavelength of 226 nm and pulse energy of 200 µJ, are generated using an Ekspla UltraFlux FT405-LLC. These pulses are formed into 2D sheets using a cylindrical lens (fused silica, f = 500 mm) The excited oxygen atoms emit fluorescence at 845 nm.

Since the laser pulses are formed into a 2D sheet, the fluorescence yields spatial information on the distribution of oxygen atoms in a cross-section through the volume of the discharge. The time delay between plasma and laser pulses is varied between 0 and 200 µs, in order to study the temporal dynamics of the oxygen atom distribution in the discharge.

To disqualify events where no plasma channel is formed between the pins, the plasma is also imaged using a camera with optimized spectral sensitivity in the visible range. For all cases, the gate width is set to 3 ns for both cameras.

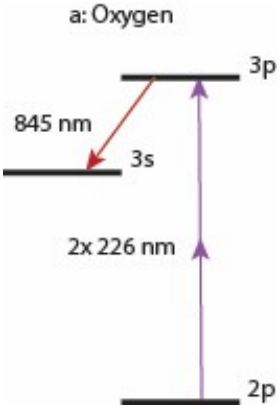

Figure 2: The two-photon excitation scheme used to excite and detect O.

A Diffractive Optical Element (DOE) is introduced in the beam path, to induce a spatial intensity modulation in the laser. The experimental arrangement for achieving structured illumination is illustrated in the new schematic in Figure 3. The DOE splits the laser beam into two diverging components. These components are then overlapped at a slight angle in the plasma volume, using a cylindrical lens. As the two beam components interfere in the probe volume, the resulting laser sheet is intensity-modulated in the vertical direction

Consequently, the laser-induced fluorescence signal from the O will take on a similar intensity modulation. The plasma emission will, however, lack this modulation, since it is unaffected by the laser light. This allows for the distinction between, and separation of, plasma emission and laser-induced fluorescence. The separation of signals is achieved by performing Fourier domain filtering around the first order Fourier component that arises from this striped pattern, as described in [6]. This post-filtering data is henceforth referred to as Fourier-filtered data.

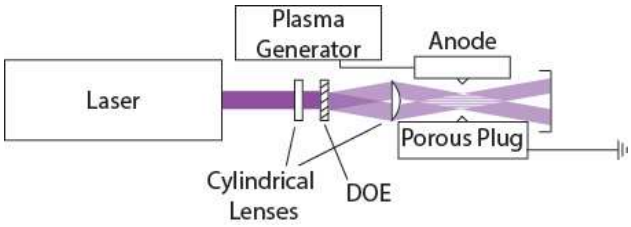

Figure 3: A side-view of the measurement setup altered for reduction of plasma emission detection. A DOE and another cylindrical lens give rise to a periodic pattern in the probe volume, allowing the distinction between oxygen signal and plasma emission.

3. Results

In Figure 4, the top row displays the single-shot data from the NIR camera for delays of 0-5 µs, with 1 µs increments. The bottom row shows the oxygen signal isolated in the method described earlier under each corresponding raw data image. Each pair of images is individually normalized. Note the different color bar magnitudes between the rows ([0 1] vs. [0 0.2]).

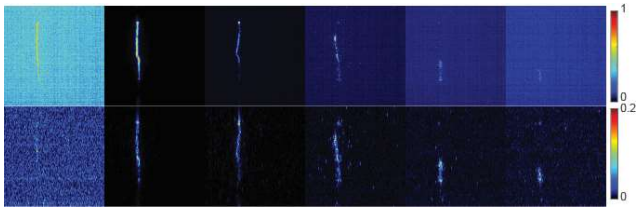

Figure 4: Single-shot images for delays between 0 and 5 µs. The top row shows raw data and the bottom row shows Fourier filtered data. Note the different color bar magnitudes.

Comparing the raw data and the Fourier-filtered data, it is clear that the Fourier filtering increases signal-to-noise ratio. Despite being single-shot, the contrast is high enough to distinguish signal-of-interest from the background noise, even for images with a longer delay (and thus lower signal strength). It also successfully distinguishes oxygen signal from plasma emission.

Furthermore, by fitting exponential functions to the means of the data in each image, lifetimes τ (the time at which the signal has decreased to $1/e$ of the initial signal) is found to be 1.05 ± 0.3 µs for the plasma emission and 45 ± 5 µs for the O distributions.

Figure 5 displays the images at delays between 30 and 130 µs, after applying the Fourier filtering. Each image is the sum of 100 measurements. It can be observed that the O fluorescence is clearly distinguishable from noise until after 110 µs after the plasma.

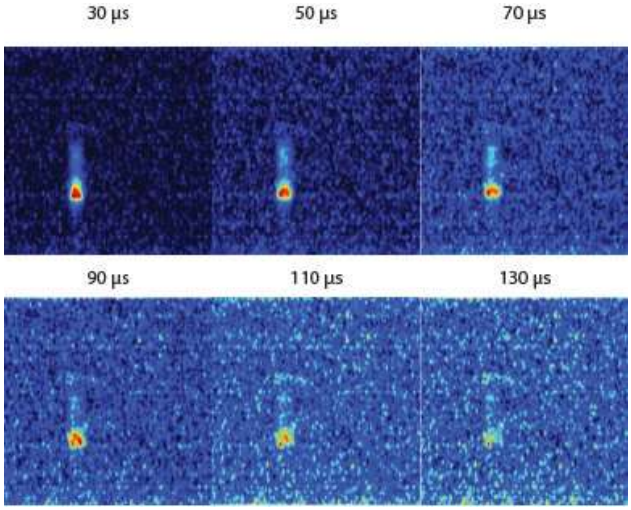

Figure 5: Each image is a sum of 100 images of the Fourier filtered data at delays between 30 and 130 µs. Even with the sum of 100 images, the signal is barely distinguishable from background after 130 µs.

4. Conclusion

In this work we have successfully recorded 2D single-shot images of the O distributions that arises from a ns-duration high voltage plasma, generated in a pin-to-pin

electrode configuration. Additionally, a novel method for suppressing both background and plasma emission for TAPLIF, using structured illumination and Fourier-domain filtering, is proposed, to significantly improve measurement accuracy, as well as signal-to-noise ratio.

Moreover, this data can be used to estimate the lifetimes of both the plasma emission and the O distributions.

5. Acknowledgements

This work is supported by ERC (852394, 803634), Swedish Research Council (2021-04506, 2019-05183), The Royal Physiographic Society as well as Knut & Alice Wallenberg Foundation (2019.0084).

6. References

[1] Zangouei, M., Haynes, B.S,
The Role of Atomic Oxygen and Ozone in the Plasma and Post-plasma Catalytic Removal of N_2O.
Plasma Chem Plasma Process 39, 89–108 (2019).
https://doi.org/10.1007/s11090-018-9926-y

[2] J. Benedikt, M. Mokhtar Hefny, A. Shaw, B. R. Buckley, F. Iza, S. Schaˆkermann J. E. Bandow,
The fate of plasma-generated oxygen atoms in aqueous solutions: non-equilibrium atmospheric pressure plasmas as an efficient source of atomic O(aq),
Phys. Chem. Chem. Phys., 20, 12037-12042 (2018).
https://10.1039/c8cp00197a

[3] W. Sun, M. Uddi, S. H. Won, T. Ombrello, C. Carter, Y. Ju,
Kinetic effects of non-equilibrium plasma-assisted methane oxidation on diffusion flame extinction limits,
Combust. Flame, 159(1), 221-229 (2012).
https://doi.org/10.1016/j.combustflame.2011.07.008

[4] M. Uddi, N. Jiang, E. Mintusov
Atomic Oxygen Measurements in Air and Air/Fuel Nanosecond Pulse Discharges by Two Photon Laser Induced Flourescence
Proceedings of the Combustion Institute 32(1):929-936
https://doi.org/10.1016/j.proci.2008.06.049.

[5] T. Freegarde, G. Hancock.
A Guide to Laser-Induced Fluorescence Diagnostics in Plasmas.
Journal de Physique IV Proceedings, 07(C4), C4-15 (1997).
https://doi.org/10.1051/jp4:1997403

[6] M. Gong, H. Kim, J. Larsson, T. Methling, M. Aldén, E. Kristensson, C. Brackmann, T. Eschrich, M. Jäger, W. Kiefer, and A. Ehn,
Fiber-based stray light suppression in spectroscopy using periodic shadowing,
Opt. Express 29, 7232-7246 (2021).
https://doi.org/10.1364/OE.410517