

Absolute calibration of Xe/O cross-section ratio for TALIF in nanosecond capillary discharge: optimization of the discharge parameters

Z. Shu^{(*)1}, N. A. Popov², S. M. Starikovskaia¹

¹ *Laboratory of Plasma Physics (CNRS, Ecole Polytechnique, Univ. Paris-Sud, Observatoire de Paris, Sorbonne Université, l'Institut Polytechnique de Paris), Ecole Polytechnique, route de Saclay, 91128 Palaiseau, France*

² *Skobeltsyn Institute of Nuclear Physics, Moscow State University, Moscow, 119991, Leninsky gory, Russia*

(*) zhan.shu@lpp.polytechnique.fr

Atomic species are readily produced in reactive media, such as discharge or combustion environments. Absolute measurements of the density of these reactive atoms are essential for the validation of models of plasma in molecular gases. Two-photon absorption laser-induced fluorescence (TALIF), offers a well-suited technique for detecting the densities of main atomic species, such as atomic O, H, and N, with sub-millimeter spatial resolution and high temporal resolution. However, high accuracy is difficult to achieve due to precise calibration of all the components of the diagnostic facility.

O-TALIF was initially developed in the 1980s as a gas discharge diagnostics tool. In 2005, Niemi *et al* suggested a calibration method for determining absolute atomic oxygen densities, based on a comparative measurement with a noble gas xenon that exhibits a two-photon resonance close to the transition of atomic oxygen [1]. In that paper, the atomic oxygen density was determined by titration and a “kit” for O-TALIF measurements was provided, including radiative lifetimes, room temperature quenching coefficients, and the ratio of the two-photon absorption cross-sections $\sigma_{\text{Xe}}^{(2)}/\sigma_{\text{O}}^{(2)} = 1.9 \pm 20\%$. Although it has been widely used, the cross-section ratio was only measured by Niemi’s group. Recently, more and more papers have been dedicated to comparing different techniques to calibrate the Xe/O cross-section ratio. Drag *et al* [2] directly measured the two-photon absorption cross-section for the Xe transition, and found a value of about one-half of that implied by Niemi’s measurement. By the use of this new ratio, Wubs *et al* [3] obtained an absolute O-atom density 25% smaller compared to the THz absorption spectroscopy results on the same capacitively coupled radio frequency oxygen discharge.

Our latest work [4] proposes a novel approach to calibrate the two-photon absorption cross-section ratio $\sigma_{\text{Xe}}^{(2)}/\sigma_{\text{O}}^{(2)}$ by using a nanosecond discharge with 100% dissociation of molecular oxygen, and so with a known ‘reference’ density of oxygen atoms ($[\text{O}]_{\text{ref}} = 2 \times [\text{O}_2]$). The discharge was initiated between two pin electrodes, with 53 mm of inter-electrode distance, and in the capillary tube with 2 mm of internal diameter. High-voltage pulses, with 9 kV of amplitude, 30 ns of FWHM and 10 Hz of repetitive frequency, were delivered to the electrodes via coaxial cables. A gas mixture of nitrogen plus a few percent of oxygen flowed through the capillary at 27 mbar with a total flow rate of 50 sccm so that each discharge was initiated in the new gas portion. Voltage at the electrodes, electrical current in the plasma, longitudinal electric field and energy delivered to the gas were measured by the technique of back current shunt and capacitive probe, giving the value of a few hundred Td of the reduced electric field and about 1 eV/molecule of the specific deposited energy. Additionally, the radial distribution of emission of excited nitrogen molecules and gas temperature in the discharge and afterglow were obtained experimentally. A detailed 1D numerical modeling was developed to analyze the main reactions and confirm the complete dissociation of O₂ in the afterglow (5 μs to 25 μs after ignition). Therefore, by comparing the data from TALIF technique in the afterglow and Xe calibration with the ‘reference’ density of O-atoms, the two-photon absorption cross-section ratio was calculated as:

$$\sigma_{\text{Xe}}^{(2)} / \sum_{J'} \sigma_{\text{O}(J \rightarrow J')}^{(2)} = 1.8 (\pm 0.2), \quad (1)$$

where J is the angular momentum quantum number of the ground-state oxygen atom level, and J' is the angular momentum quantum number of the upper fine structure level of atomic oxygen.

In this work, we are aiming to design a cell that is closed with known concentration of oxygen under tens of mbar. By using this portable device, similar nanosecond discharge could be applied to

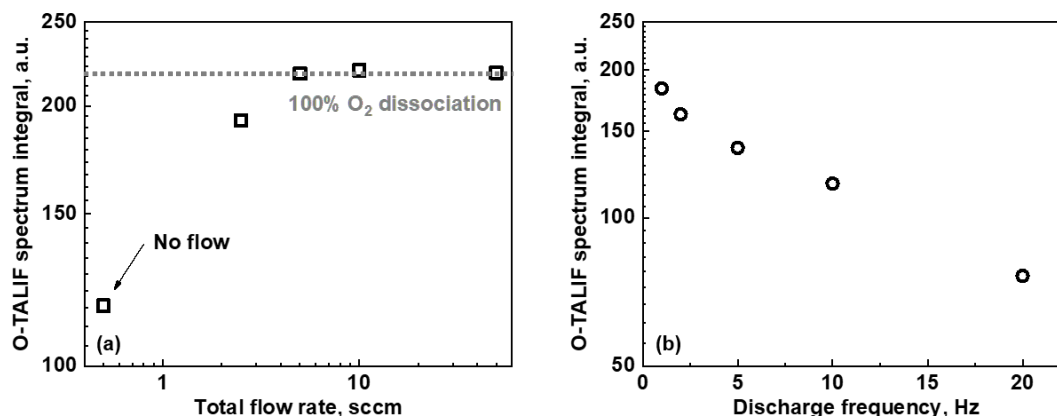


Fig. 1: (a) O-TALIF signal with different total flow rate of $N_2 + 5\% O_2$ mixture at $5 \mu s$, laser and discharge pulse repetition frequency of 10 Hz. (b) O-TALIF signal with discharge frequency in static $N_2 + 5\% O_2$ mixture at $5 \mu s$ and laser pulse frequency of 10 Hz.

help calibrate the parameters in more diagnostic methods such as cross-section ratio in picosecond or femtosecond TALIF. An attempt was made for the nanosecond discharge in the same capillary tube to verify the feasibility of the new device by changing the flow rate of $N_2: O_2$ mixture and the repetition frequency of the discharge pulses. Fig.1(a) shows that O-TALIF signal maintains the level of 100% O_2 dissociation at 5 sccm minimum, which is coherent with the calculated flow rate to keep the gas renewal under 10 Hz discharge frequency in the existing capillary tube. However, the signal decreases when the flow rate reduces to 2.5 sccm or 0 sccm (static state). This may be caused by the disturbance of long-lived species, like nitric oxide, accumulated after multiple discharges. In that case, it is reasonable to check the signal again with lower discharge and acquisition frequencies. Fig.1(b) gives the increasing trend of the O-TALIF signal with lower discharge frequency. When the frequency decreases to 1 Hz, the signal reaches almost 80% of the intensity where O_2 is fully dissociated.

Further work will be focused on two paths. One is to try to get the highest possible O-TALIF signal in $N_2: O_2$ mixtures and analyze the gas composition during and after each discharge. In other words, this is to keep the same dissociation fraction for different applications. The other way is to try different gas mixtures like $Ar + O_2$ to achieve full oxygen dissociation and avoid disturbance terms.

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