# Fs-TALIF for Low Pressure Interfacial Plasmas

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**Plasma-surface interaction has been an important area of research for many aerospace applications such as hypersonic re-entry vehicles, design of plasma discharge systems for flow control, supersonic combustors, plasma propulsion ion thrusters, and thin film recoating of mirrors on satellites. Therefore, there is a need to advance the fundamental knowledge of plasma-surface interaction using advanced laser diagnostics. This will allow for the study of plasma-generated species with high spatial and temporal resolutions nearby solid surfaces. This work presents a study on low-pressure ($5 - 160$ mtorr) interfacial plasmas using femtosecond two-photon absorption laser-induced fluorescence (fs-TALIF). The experimental setup consists of a low-pressure plasma produced by a radio-frequency power source. The saturation of the ground state is investigated by varying the pulse energy, and the relationship between laser intensity and fs-TALIF signal is found to be linear up to $5\ \mu$J. The excited state lifetime is measured at different pressures and power levels, it is observed to be around $\tau = 84.6$ ns. The study also examines the effect of plasma power on the fs-TALIF and plasma emission signals. It is observed that higher power levels result in higher signal intensity up to a certain pressure, beyond which the signal declines. The observed trends suggest that the power supplied to the plasma could be a limiting factor in the fs-TALIF and plasma emission signals. Further work will focus on obtaining absolute density values through krypton calibration and investigating spatial and temporal measurements of plasma-surface interactions.**

## I. Introduction

Plasma surface interactions have been a critical area of research for many aerospace applications such as hypersonic re-entry vehicles in various planetary atmospheres [1], design of plasma discharge systems for flow control and drag reduction [2], supersonic combustors [3], space-craft solar panels [4], plasma propulsion [5], ion thrusters [6], thin film recoating of ultraviolet (UV) mirrors on satellites, materials processing, microfabrication [7], and plasma-catalysis [8].

Plasma generated non-equilibrium atomic and molecular species interact with a solid surface in a complex manner. In Earth's atmosphere, various species such as electronically excited $O_2*$, $N_2^*$ molecules, atoms, and ions such as $O_2^-$, $O_2^+$, $O^+$, $O^-$, $O^*$ are formed in the plasma which are highly reactive and corrosive. In rarefied atmospheres, the ions can reach high kinetic energies, easily reacting and damaging solid surfaces. Advanced quantitative laser diagnostics with high spatial and temporal resolution will be highly useful to understand the physics of plasma-surface interactions. [7, 9–11]

Currently various methods have been used to study plasma-surface interactions. Thermoreflectance methods include Time Domain Thermoreflectance (TDTR) and Frequency Domain Thermoreflectance (FDTR) to measure surface temperature changes [9]. They work on the principle of change in surface emissivity with temperature. These require complex scanning methods to generate spatial mapping and required long integration times which can compromise on the required time resolution.

For low pressure plasmas, only certain optical techniques can be used. At low pressures, the signals obtained from Raman scattering are either weak or negligible [12]. To increase the signal level, the laser power can be increased and focused to the point of interest. However, such high laser intensities can damage the surfaces that are being probed. One dimensional spatial resolution using Raman scattering has been obtained in high-pressure combustion environments only. Optical Emission Spectroscopy (OES) has been used to give abundant information about plasmas [13]. However, OES is a line-of-sight signal collection method and does not achieve the required spatial resolution.

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Laser Induced Fluorescence (LIF) methods are highly sensitive laser diagnostic tools for quantitative 2D measurements of atomic species, especially at low pressures. In LIF, atoms or molecules are electronically excited using a well-defined resonant laser wavelength, which is characteristic of the atom or molecule of interest. The molecule de-excites by emitting photos of a certain wavelength. Using an appropriate optical band pass filters, these photons (fluorescence) can be collected to obtain spatial and temporal information. However, some atoms such as $H$, $O$, $N$ require deep UV lasers (< 200 nm) for a single photon fluorescence excitation. Nevertheless, deep UV light is difficult to generate and propagate through atmospheric air, which readily absorbs and scatters such a short wavelength of light. Another way is to excite the atoms with two photons, each having about half the energy required from the previous single photon. This process is known as Two Photon Absorption Laser Induced Fluorescence (TALIF) [14, 15].

This study presents the lifetime of the $N$ atom excited stated ($3\,p\ {}^4\mathrm{S}_{3/2}$) for different radio frequency (rf) supplied powers and pressures (5 – 160 mtorr) using femtosecond (fs) TALIF. This work also examines the effect of plasma power on the fs-TALIF and plasma emission signals. Lastly, the final manuscript will also examine the plasma-surface interaction with high spatial and temporal resolution as well as absolute density, obtained through krypton calibration.

## II. Experimental Setup

Figure 1 shows the diagram for the experimental setup. Fig. 2 shows the actual experimental setup used for nitrogen fs-TALIF. The setup consists on an low pressure plasma being produced by a AG0613 T&C Inc. rf power source. The rf source presents a frequency of 13.56 MHz and its supplied power can be changed in a range between 15 – 35 W. An AIT-600R rf matching network was employed between the source and the load (chamber) in order to match the variable impedance of the two components. The Spectra-Physics Solstice Ace femtosecond laser tuned at 826.8 nm was used with a fourth harmonic generator to excite the atomic nitrogen ground state ($2\,\mathrm{p}^3\ {}^4\mathrm{S}_{3/2}$) into its excited state ($3\,p\ {}^4\mathrm{S}_{3/2}$) through a two photon transition ($2x$206.6 nm). The laser presents an initial energy per pulse of 3.7 mJ/pulse and it has a repetition rate of 1 kHz. The photons from the transitions $3\,p\ {}^4\mathrm{S}_{3/2} \rightarrow 3\,s\ {}^4\mathrm{P}_{1/2,3/2,5/2}$ were collected using a band-pass filter centered at 750 nm with a bandwidth of ±40 nm. A pco.edge 5.5 and a Stanford Computer Optics Quantum Leap intensifier were used to obtain the TALIF signal.

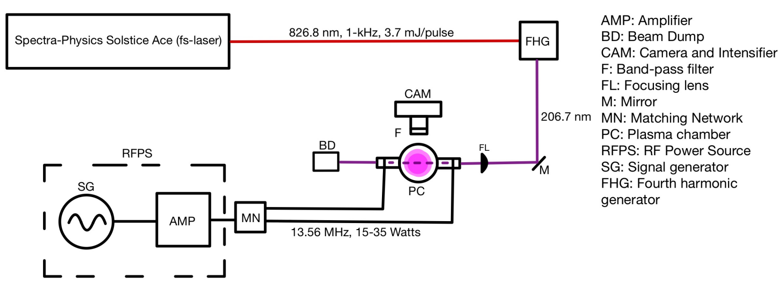



**Fig. 1 Experimental setup diagram.**

A variable attenuator (not shown in Fig. 1) was used to vary the pulse energy in order to check for the saturation of the ground state. In addition, the camera delay and gate width were varied to obtain the lifetime of the ground state at various pressures (7 – 160 mtorr) and power levels (15 – 35 W).

## III. Results and Discussion

Firstly, the saturation of the ground state ($2\,\mathrm{p}^3\ {}^4\mathrm{S}_{3/2}$) as a function of laser intensity was obtained. The chamber pressure was set at 7 mtorr and the plasma was ignited with a forward power of 25 W. Then, a variable attenuator was used to vary the pulse energy from 0.6 – 10 $\mu$J. Subsequently, the fs-TALIF intensity was plotted as a function of laser intensity ($\mu J^2$) as shown in Fig. 3. The fs-TALIF has arbitrary units. It is worth mentioning that the focal point was avoided when analyzing the TALIF signal due to saturation concerns. It is possible to observe from Fig. 3 that the laser intensity and nitrogen TALIF signal have a linear relation from $0 - 25\mu J^2$. After $25\mu J^2$ saturation of the ground state starts to develop a nontrivial role in the collected signal. Therefore, for proper data acquisition not surpassing this value

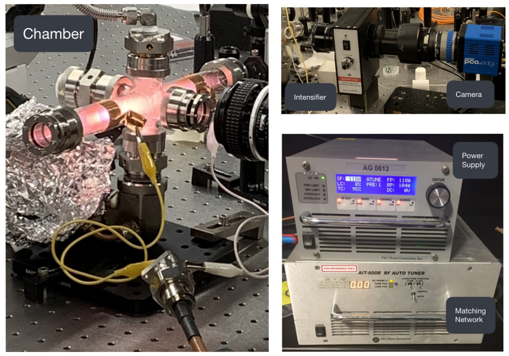



**Fig. 2 Experimental setup.**

is of extreme importance. Thus, a value of 5 $\mu$J ($25\mu J^2$) for the laser energy was chosen to maximize the TALIF signal obtained without causing saturation.

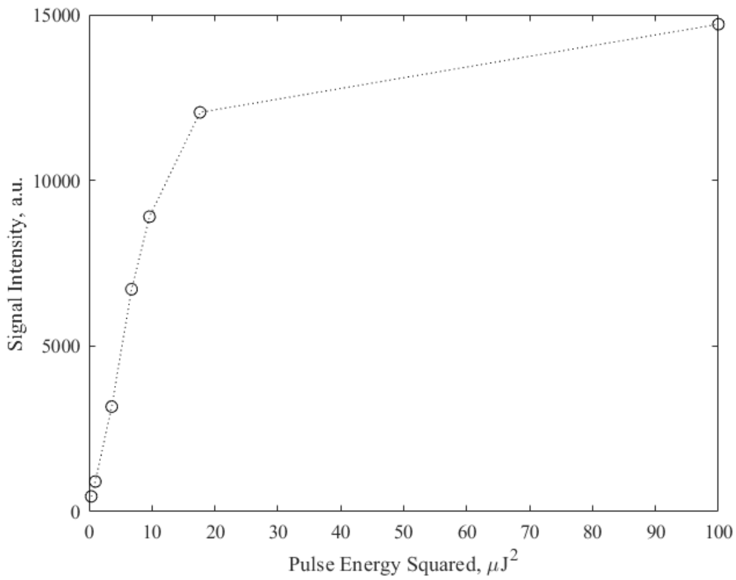



**Fig. 3 Nitrogen TALIF signal intensity as a function of laser intensity at a pressure of 7 mtorr, power of 25 W, and gate width of 1 ns.**

The lifetime of 7 mtorr atomic nitrogen as function of gate delay was obtained for a plasma power of 25 W, as shown in Fig. 4. An exponential fit was employed to obtain the excited state lifetime. The lifetime constant turned out to be $\tau = 84.6$ ns with 95 % confidence intervals of $\pm 21.3$ ns. This value shows good agreement with the weighted average of the $3p\,^4S_{3/2} \rightarrow 3s\,^4P_{1/2,3/2,5/2}$ lifetimes presented in [16].

The nitrogen lifetimes for three different supplied powers were obtained. The values studied were 15 W,25 W, and 35 W and at each power level different pressures were considered. The exponential decay was integrated over the time domain by increasing the gate width, this was performed in order to minimize the noise in using a shorter gate width and changing the time delay instead. This change in approach resulted in going from a exponential decay ($S_dec(t) = Ae^{-t/\tau}$) to an asymptotic growth fit ($\int_a^b S_{dec}(t)\,dt = A(1 - e^{-t/\tau})$). It is worth mentioning that the focal point was avoided during data processing to prevent obtaining data from a saturated ground state. The plasma emission far away from the laser path (fs-TALIF signal) was also considered. This was done to establish a relation between the signal obtained and plasma pressure for different supplied powers.

Fig. 5.a shows the nitrogen TALIF signal intensity as a function of gate width for a power of 15 W while Fig. 5.b

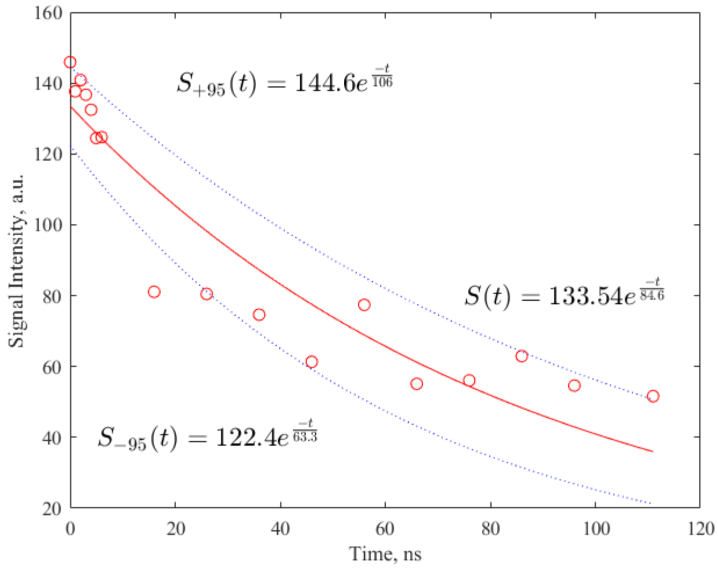



**Fig. 4 Nitrogen TALIF signal intensity as a function of gate delay at a pressure of 7 mtorr, power of 25 W, and gate width of 1 ns.**

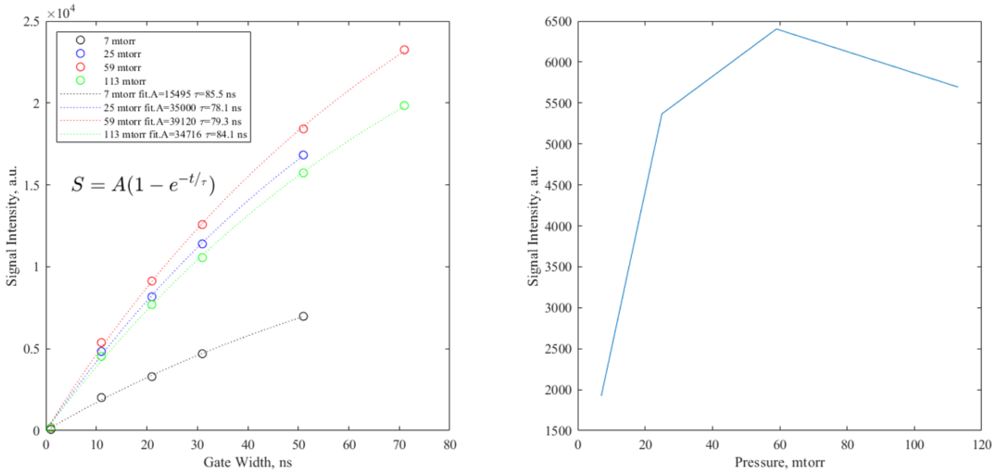
shows the plasma emission intensity as a function of pressure. The atomic nitrogen lifetime at a supplied power of 15 W turned out to be between $\tau_{15W}$ = 78.1 ns and $\tau_{15W}$ = 85.5 ns. As for the 25 W case, the lifetime was between $\tau_{25W}$ = 72.4 ns and $\tau_{25W}$ = 93.3 ns. Lastly, the nitrogen TALIF lifetime at 35 W was the case that presented the most variation (from $\tau_{35W}$ = 60.2 ns to $\tau_{35W}$ = 102 ns). These three set of values seem to be in the range observed by [16].

The signal intensity in Fig. 5.a seemed to have reached a peak at around 59 mtorr before declining at higher pressures. This trend was also found in the plasma emission intensity , see Fig. 5.b. As for the 25 W (Fig. 6) and 35 W (Fig. 7) cases, similar trends were detected. The nitrogen TALIF and plasma emission signals would rise and reach a peak at around 60 mtorr before declining at higher pressures. This indicates that there seems to be a relationship between the supplied power into the plasma and the TALIF signal. A connection between the two can be observed at the signal peak for each of the three cases (15 W,25 W, and 35 W), higher supplied powers have higher TALIF and plasma signal intensities overall. Therefore, it is possible that the power supplied to the plasma could be the limiting factor in the TALIF and plasma emission signals observed. This mechanism could work in the following way: for a constant supplied power (15 W) an increase in pressure will increase the TALIF signal obtained up to a certain point (aorund 60 mtorr). After this threshold is surpassed, the power supplied will not be enough to dissociate the additional nitrogen molecules, resulting in an stagnant signal. For higher pressures (> 60 mtorr) collisions could play a significant role in the recombination of nitrogen atoms, thus reducing the overall signal, as seen in Fig. 5, Fig. 6, and Fig. 7. These are only preliminary explanations, more work will be performed in order to answer this phenomena. Lastly, absolute density values will be obtained through krypton calibration. Furthermore, spatial and temporal measurements will also be obtained for nonuniform plasma-surface interactions.

## IV. Conclusion

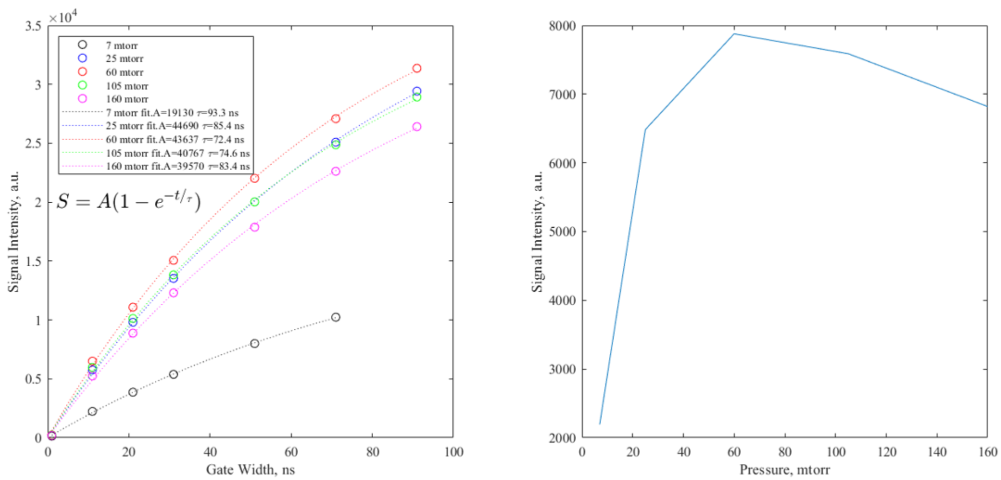
In conclusion, this paper presented the experimental setup and results of nitrogen fs-TALIF measurements for low-pressure plasmas. The experimental setup consisted of a low-pressure plasma produced by a rf-power source. A femtosecond laser was used to excite the atomic nitrogen ground state to its excited state, and the TALIF signal was collected and analyzed. The saturation of the ground state was investigated, and it was found that the laser intensity and nitrogen TALIF signal had a linear relation up to $25 \mu J^2$, beyond which saturation effects started to appear. A pulse energy of 5 $\mu$J was chosen to maximize the TALIF signal without causing significant saturation.

The lifetime of excited atomic nitrogen was measured at a pressure of 7 mtorr and a plasma power of 25 W. An exponential decay fit was employed to obtain the excited state lifetime, and it was found to be $\tau$ = 84.6 ns with 95 %




a) b)

**Fig. 5 a) Nitrogen TALIF signal intensity as a function of gate width at power of 15 W and various pressures. b) Plasma emission intensity as a function of pressure for a gate width of 51 ns.**

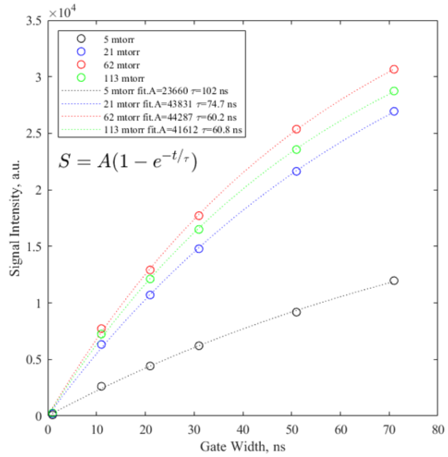



a) b)

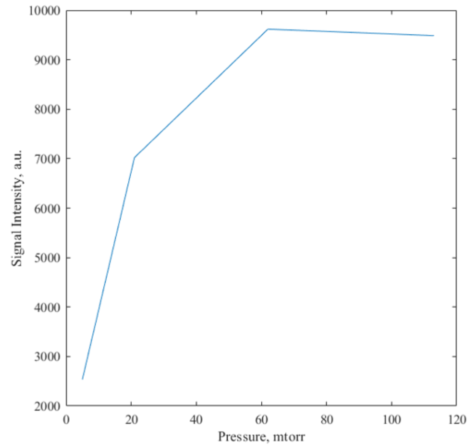
**Fig. 6 a) Nitrogen TALIF signal intensity as a function of gate width at power of 25 W and various pressures. b) Plasma emission intensity as a function of pressure for a gate width of 51 ns.**

confidence intervals of ±21.3 ns. Thus, showing good agreement with the literature. Additionally, the atomic nitrogen lifetimes were studied for different supplied powers (15 W, 25 W, and 35 W) and at various pressures. The TALIF signal and plasma emission intensities were analyzed as functions of gate width and pressure, respectively. It was observed that the TALIF and plasma signals reached peak values at around 60 mtorr before declining at higher pressures. This trend suggests a relationship between the supplied power and the TALIF and plasma signals, with higher power resulting in higher signal intensity. However, further research is needed to fully understand the underlying mechanisms and optimize the measurement technique.

Overall, the results of this study provide valuable insights into the behavior of low-pressure interfacial plasmas. Future work will focus on obtaining absolute density values through krypton calibration and investigating nonuniform plasma-surface interactions through spatial and temporal measurements.




a)




b)

**Fig. 7 a) Nitrogen TALIF signal intensity as a function of gate width at power of** 35 **W and various pressures. b) Plasma emission intensity as a function of pressure for a gate width of** 51 **ns.**

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