Two-Photon Absorption Laser Induced Fluorescence (TALIF) of Ground State Neutral Xenon

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Tad Wegner[1], Adam J. Friss[2], Ciprian Dumitrache[3], and Azer P. Yalin[4]
Colorado State University, Fort Collins, CO, 80523, USA

Two-photon absorption laser induced fluorescence (TALIF) is presented as a means to perform species specific density measurements of ground state neutral xenon. This method can be applied to electric propulsion systems to help characterize the role neutral particle interactions play in limiting the lifetime and performance of the device. We report TALIF measurements in a dilute xenon mixture (no plasma source) at densities as low as 2.7 x 10^{14} m^{-3} (256 laser shot average) with a detection limit of ~ 5 x 10^{15} m^{-3}. Analysis with a numeric model has shown that 2+1 photoionization is playing a role both as a signal loss mechanism and a source of noise. The results presented motivate further investigation into application to a plasma environment for future use on electric propulsion devices including Hall Effect thrusters.

Nomenclature

A_{2s}	=	decay rate from the excited state to the metastable state
$G^{(2)}$	=	photon statistic factor
$g(\Delta\nu)$	=	two-photon absorption line profile
h	=	Planck's constant
He	=	helium
I	=	laser pulse intensity
N_1^0	=	initial ground state population
N_2	=	excited state population density
n_{Xe}	=	neutral xenon density
OPO	=	optical parametric oscillator
PMT	=	photomultiplier tube
S/N	=	signal-to-noise ratio
T_2	=	total decay rate from the excited state
TALIF	=	two-photon absorption laser induced fluorescence
TPA	=	two-photon absorption
$W_{12}^{(2\nu)}$	=	two-photon absorption rate coefficient
$W_{21}^{(2\nu)}$	=	two-photon stimulated emission rate coefficient
$W_{23}^{(i)}$	=	photoionization rate coefficient
Xe	=	xenon
λ	=	wavelength
ν	=	frequency
σ	=	interaction cross-section

[1] Graduate Student, Department of Mechanical Engineering, Tad.Wegner@colostate.edu
[2] PhD, Department of Mechanical Engineering, Adam.Friss@colostate.edu
[3] PhD, Department of Mechanical Engineering, Ciprian@rams.colostate.edu
[4] Professor, Department of Mechanical Engineering, ayalin@engr.colostate.edu

I.Introduction

DEEP space exploration and satellite station keeping are facilitated by Electric Propulsion (EP) systems. Among these systems, Hall Effect Thrusters (HETs) stand as a focal point for researchers due to their high thrust efficiency. Research efforts seek to further improve mission capabilities by maximizing the device's lifetime and performance. The dynamics of neutral (xenon) particles in the plasma propellant are very important to understand as they significantly influence ionization processes, overall energy conversion, and interactions with surrounding wall materials. Among these neutral particle interactions, charge exchange reactions are of particular interest. Slow ions and fast neutrals resulting from these collisions can erode the channel walls, limiting thruster lifetime. The frequency with which charge exchange reactions occur increases with the density of neutral particles. In order to support further study of these reactions, and associated facility effects, a diagnostic method is required to quantify the density of neutral xenon (n_{Xe}) propellant. Laser Induced Fluorescence (LIF) is a non-invasive spectroscopic method previously used to perform velocimetry measurements in various EP devices[1,2,3]. Using LIF for xenon density measurements would require a deep ultraviolet laser source with wavelength ~125 nm based on the energy difference between the ground and low-lying excited states.

An alternative approach to probe transitions that are difficult to access with single-photon schemes is Two-photon Absorption Laser Induced Fluorescence (TALIF). TALIF has been used to perform density measurements in Ion Thrusters[4,5]. Eichhorn et. al. measured neutral xenon densities on the order of 10^{17} m^{-3} when performing TALIF on a radio frequency ion thruster (RIT-10). Crofton demonstrated the earliest use of TALIF on an ion thruster (T5 Mk3), reporting densities on the order of 10^{15} m^{-3}. Two-Photon Absorption (TPA) is a process by which an atom absorbs two photons to access an electron energy transition equaling the sum of the incident photon energies. With TPA, rather than directly probing ground state neutral xenon at λ ~ 125 nm, we instead use a beam with twice the wavelength as can be achieved with more readily available laser sources.

In section II, a physical understanding of LIF and TPA is introduced, followed by a mathematical model of the complete TALIF interaction. Section III describes the vacuum and optical setup to perform these experiments, and in section IV the measured spectra and subsequent signal analysis are presented. TALIF measurements presented here are at neutral densities lower than previously reported, with analysis showing that further improvements in signal-to-noise (S/N) and single-shot measurements are possible.

II.Theory

A. Laser Induced Fluorescence

Laser Induced Fluorescence is a diagnostic technique in which a collection of particles absorbs incident laser light to a desired energy level. The atoms in the excited state then undergo spontaneous emission down to the original state (in a resonant process) or some lower state (in a non-resonant process). The light from this emission, known as fluorescence, is then collected and imaged onto a photodetector, typically a photomultiplier tube (PMT). LIF is generally performed in one of two regimes, saturated or weak-field.

When a system of particles is saturated, the excited state population equals that of the initial state (neglecting degeneracy). The rate of gain of the excited state population matches its rate of loss, meaning the amount of light fluoresced is unaffected by a further increase in laser intensity. By removing intensity dependence in the fluorescence signal, any noise due to small laser power fluctuations is also removed. A challenge though is to obtain saturation at the spatial and temporal edges of the pulse.

In the weak-field or linear regime, the rate of loss far outweighs the rate of gain, so the excited state population will remain well below the initial state population. The fluorescence signal scales linearly with the particle density in the measurement volume, allowing direct measurement of species specific population densities. However, in order to calculate the population density, the rate coefficient for all gain and loss mechanisms needs to be known precisely. This challenge can be avoided by recording a calibration curve. A calibration curve can be developed by performing a set of TALIF experiments in a system where the population density is known. Then, when TALIF is used on a system with unknown density, the signal can be compared with the calibration data. To evaluate the feasibility of this approach, temperature variation effects will be investigated.

B. Two-Photon Absorption

Two-photon absorption is a non-linear optical process first theorized by Goeppert-Mayer in 1931[6].TPA is dependent on the square of incident light intensity, so it dominates over linear processes at high intensity. For this reason, it was not observed experimentally until 1961, soon after the invention of the laser[7]. TPA can be thought of as a variant of modulation spectroscopy. The electron's interaction with a single photon's electric field modulates the energy spectrum in a manner that allows the second photon to be absorbed[8]. The electron, which occupied a stationary state of the atom, enters a superposition of energy states of the new atom-photon system. In this superposition of states, often referred to as a virtual state, the electron can accept an additional photon to complete the absorption transition to the atom's excited energy state. The energy of the transition equals the sum of the energy of the two photons. Due to the time-energy uncertainty principle, the staying time of the electron in this superposition of states approaches zero[9]. This means the absorption of the second photon occurs effectively instantaneously with the first. The incredibly short lifetime of the virtual state further explains why high light intensities are necessary, as two photons must interact with the same electron on very short time scales.

C. Mathematical Model

In the weak-field regime, a simple rate equation can be used to model the TALIF interaction. Assuming the ground state population is approximately constant (well below saturation) and the pressure is low enough that collisional excitation and quenching can be neglected, the rate equation for the excited state population is given by Eq. 1.

$$\frac{dN_2}{dt} = W_{12}^{(2\nu)} N_1^0 - \left(W_{23}^{(i)} + W_{21}^{(2\nu)} + T_2\right) N_2 \tag{1}$$

where the gain coefficient is the two-photon absorption rate and the loss coefficients are the photoionization, two-photon stimulated emission, and spontaneous decay rates respectively. Here we are neglecting amplified stimulated emission along the beam path, so the two-photon absorption and stimulated emission coefficients are considered equal. A schematic of these gain and loss terms is given in Fig. 1. In our scheme, fluorescence is collected when atoms in the excited state decay down to metastable state s via spontaneous emission. This is represented by A_{2s}. For the 252 nm absorption line, fluorescence is collected at 823.2 nm[10]. The spontaneous decay rate has been reported[11] and can be calculated from experimental data. The two-photon absorption and photoionization rate coefficients can be calculated from Eq. 2 and Eq. 3 respectively[12]:

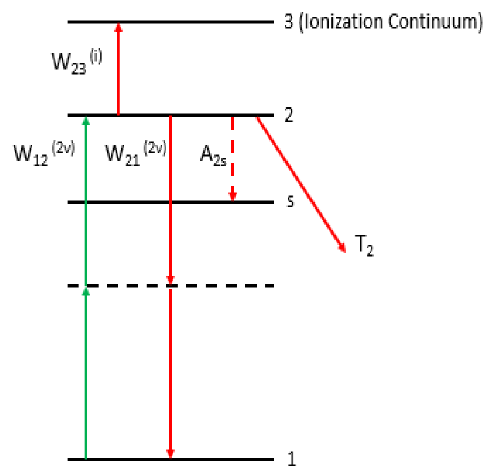


Figure 1. TALIF scheme. *The states 1,2,3, and s represent the ground state, excited state, ionization continuum, and metastable state respectively. Green arrows are used for gain terms and red arrows are used for loss terms.*

$$W_{12}^{(2\nu)} = \sigma_{12}(I/(h\nu_L))^2 \tag{2}$$
$$W_{23}^{(i)} = \sigma_i(I/(h\nu_L)) \tag{3}$$

where σ_{12} and σ_i are the effective cross section for two-photon absorption and the total cross section for photoionization, I is the incident beam intensity, and ν_L is the incident laser frequency. Because the temporal intensity profile of the laser pulse is not constant, the gain and loss coefficient terms should be treated as time dependent functions. This adds further complexity to the solution to Eq. 1, requiring a numerical approach. The effective cross section for TPA can be written in terms of its total cross section in Eq. 4[13]:

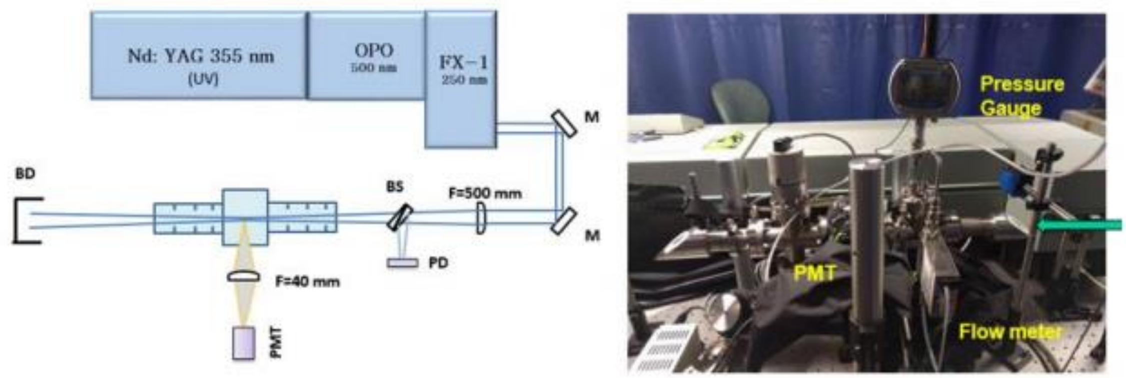
$$\sigma_{12} = G^{(2)} \sigma_0^{(2)} g(\Delta\nu) \tag{4}$$

where, $G^{(2)}$ is the photon statistic factor, which is often given a value of 2[14]. σ_0 is the total cross section and $g(\Delta\nu)$ is the normalized line profile of TPA. The product of the first two terms has been reported for the 252 nm absorption line[10], and the line profile can be treated as a convolution of two single-photon spectral profiles and the atom's absorption line profile at the two-photon resonance frequency. This is given by Eq. 5[14].

$$g(\Delta\nu) = g_A(2\nu - \nu_A) * g_l(\nu - \nu_l) * g_l(\nu - \nu_l) \quad (5)$$

III.Experimental Setup

The experimental setup used for TALIF experiments is shown schematically in Fig. 2-a. The third harmonic of an Nd:YAG laser (Continuum Powerlite 8010) at λ = 355 nm was used to pump an optical parametric oscillator (Sunlite EX). The OPO allows one to access a broad range of wavelengths $\lambda = 445 - 1750$ nm with an output power of E ~ 30 mJ. The output from the OPO is subsequently passed through a frequency doubler (Continuum FX-1) in order to reach the UV wavelengths required for TALIF. The choice of the 252 nm transition was motivated by the high branching ratio[10] along with a better PMT quantum efficiency at 823 nm compared to 828 nm. The laser output from the frequency doubler is steered with a pair of dielectric mirrors and focused using a plano-convex lens (f=500 mm) inside of a vacuum cell. The TALIF signal is collected perpendicular to the optical axis using a plano-convex lens (f=40 mm) and imaged 1-to-1 onto the detection array of a PMT (Hamamatsu R3896). Additionally, the shot-to-shot laser energy is monitored using a photodiode (DET-10A). The PMT signal is collected using an oscilloscope (Tektronix TDS5034B) and the results are post-processed using MATLAB.



a.) b.)

Figure 2. a.) Optical layout used for TALIF experiments. b.) Image showing the vacuum chamber and fluorescence collection setup

The vacuum chamber used (Fig. 2-b) was filled with a mixture of He and Xe (containing 10 ppm of Xe) and the xenon number density was set by varying the chamber pressure. For pressures lower than 1 mbar (corresponding to $n_{Xe} = 2.7 \times 10^{17}\ m^{-3}$), the chamber was converted into a flow system using a mass flowmeter (Unit Instruments 7360) and the concentration of xenon was established inside the chamber by varying the gas mixture flowrate.

IV.Results

D. Spectra

Fig. 3 shows TALIF excitation spectra recorded as a function of laser wavelength for several xenon densities. These measurements were accumulated from 256 laser shots at each wavelength. The TALIF signal measurement at n_{Xe} = 2.7 x 10^{14} m^{-3} establishes a detection limit lower than that demonstrated in recent ion thruster TALIF experiments[4]. If this detection limit can be maintain, it is very adequate for expected Hall thruster conditions.

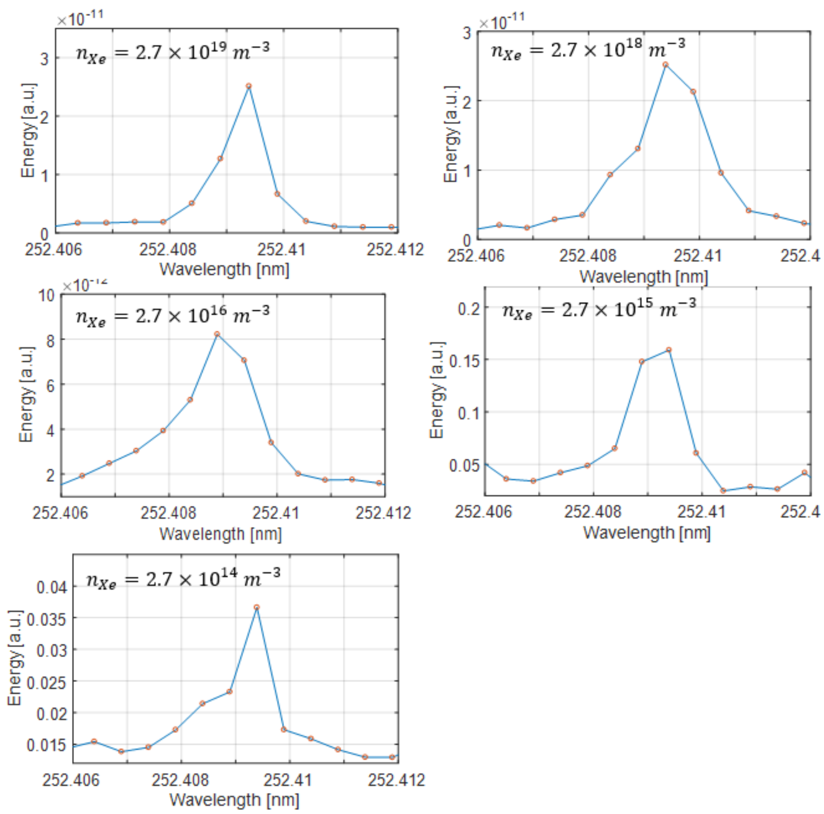
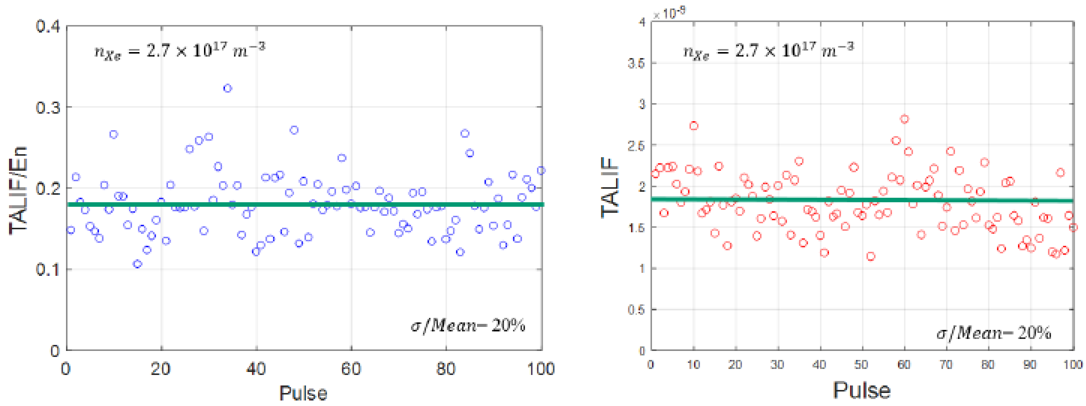


Figure 3. TALIF excitation scans at varying xenon densities. *The horizontal axis was recorded directly from the OPO. More recent trials using a wavelength meter (High Finesse WS5) recorded the peak at λ = 252.415 ± .003 nm.*

E. Noise Limitation and Single-Shot Detection

A preliminary study explored the ability to make single-shot laser measurements, which is preferred over accumulating multiple shots as it corresponds to a time resolution of 10 ns (i.e. the length of exciting laser pulse). Relatively high signal-to-noise single-shot TALIF was demonstrated at 2.7 x 10^{17} m^{-3} xenon density with a ±20% uncertainty. Statistical analysis of the data shown in Fig. 4 also shows comparable (fractional) standard deviation with and without power normalization which implies the variation in laser energy is not causing the variations in TALIF signal strength. Instead it is likely that the 2+1 photoionization loss mechanism is varying with the beam intensity profile. This variation is a stochastic process, matching the observed variation in TALIF signal. These measurements support the ability to measure with high temporal resolution (~10 ns) and spatial resolution (~5 mm in this case) at low xenon densities.

An estimate of the signal-to-noise ratio (S/N) for a single-shot measurement at the desired minimum concentration can be made as follows. The data of Fig. 4 show that the noise-to-signal ratio at a concentration of 2.7×10^{17} m^{-3} is 0.2, corresponding to a S/N of 5. In the shot noise limit, the S/N will scale as the square root of the number of photons. At a concentration of 5×10^{15} m^{-3} , the number of xenon atoms, and thus the number of fluorescence photons, is about



a.) b.)

Figure 4. a.) Scattered plot of normalized (by pulse energy) TALIF signal for a number density of 2.7 x 10^{17} m^{-3}. B.) Similar plot for the raw TALIF signal. The green line represents the average TALIF over 100 laser shots

50 times less, so the S/N should be about one seventh of what it is at 2.7×10^{17} m^{-3} . Thus, the S/N at 5×10^{15} m^{-3} approaches unity, and this represents the lower limit of detection for a single shot.

V.Conclusion

TALIF measurements at neutral xenon densities on the order of 10^{14} m^{-3} have been demonstrated, with potential for single-shot measurements above a calculated detection limit of 5 x 10^{15} m^{-3}. Potential improvements to signal strength include the use of a PMT with a higher quantum efficiency at our fluorescence wavelengths, and the use of a dye laser that is more energetically stable than the OPO used here. A new PMT could also open up the possibility of probing the 249.5 nm TALIF transition, which is known to have a higher cross section and branching ratio than the 252 nm line[10]. Further development of this work would involve performing the TALIF measurements on a barium oxide hollow cathode to evaluate how the presence of plasma affects the signal (e.g. luminous interference). Once application to a plasma environment has been demonstrated, a calibration method will be implemented to measure neutral xenon densities on a 1.5 kW Hall effect thruster under variations in radial position, axial position, chamber pressure and thruster current.

Acknowledgments

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