Two-Photon Spectroscopy on Neutral Xenon in the Plume of the Radio-Frequency Ion Thruster RIT-10

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Two-photon absorption laser induced fluorescence (TALIF) measurements of neutral xenon are presented in the plume of the radio-frequency ion thruster RIT-10. Two TALIF schemes including the $6p'[1/2]_0$ and $6p'[3/2]_2$ levels have been applied. Relative ground state densities have been measured in the cold gas flow and at several operating conditions of the thruster, including variation of the gas flow and the power of the radio-frequency generator.

I. Introduction

Laser-spectroscopy is a powerful tool for the non-intrusive detection of atoms and molecules in plasma flows.[1] Due to the large energy gap between the ground state and excited states of the investigated species, two-photon absorption laser-induced fluorescence (TALIF) spectroscopy is appropriate for ground state diagnostics of many atoms to circumvent excitation in the VUV. In plasma regimes, in which the depletion of the ground state is negligible small, the method thus allows the measurement of local particle number densities.

In the field of atmospheric reentry research, TALIF measurements are of interest to characterize high enthalpy plasma flows in plasma wind tunnel facilities. Recent studies included the measurement of particle densities and translational temperatures of atomic oxygen in inductively heated plasma.[2,3] Furthermore, atomic nitrogen has been investigated in the flow of arc discharges.[4,5] Moreover, rare gas calibration techniques for TALIF measurements have been developed.[6–8]

However, TALIF spectroscopy in xenon plasmas seems to have attracted less attention in the past. TALIF in xenon is of interest since, at present, xenon is the most promising propellant in ion thrusters. In the plume of ion thrusters, populations of neutral xenon can occur when ionization in the discharge is incomplete. Furthermore, neutrals can be involved in charge exchange processes with ions and finally produce a backflow.[9–11] TALIF measurements are potentially helpful to improve the understanding of this effect and to characterize thruster operation conditions.

Hitherto, ground state diagnostics in xenon plasmas concentrate on the neutral species, since excitation of ionized xenon requires either two-photon absorption at wavelengths <200 nm, or, at larger wavelengths, three-photon absorption schemes, which is both difficult to realize. A TALIF study in the plume of the T5

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ion thruster has been published by Crofton,[12] wherein radial and axial profiles of neutral xenon have been measured in the cold gas flow. Apart from ion thruster diagnostics, Aanesland et al[13] have measured the depletion of neutral xenon densities in a magnetized helicon plasma applying the TALIF technique.

At the Institut für Raumfahrtsysteme (IRS), most recent work was focused on two-photon spectroscopy on neutral xenon within a systematic study of different TALIF schemes in a cold gas cell, including some levels of the $6p$ and $6p'$ multiplets appropriate for xenon plasma diagnostics and higher energetic levels of $8p$ and $5f$ multiplets, which may be interesting for two-photon calibration procedures. Results have been published with respect to the determination of natural lifetimes.[14,15] Due to the low particle densities expected in the plume of ion thrusters such as the RIT-10 in the present work, the selection of the TALIF scheme deserves closer attention. In thin xenon plasmas, TALIF schemes of the $6p$ and $6p'$ multiplets seem to be most appropriate with respect to large two-photon absorption coefficients and large branching ratios of the selected florescence transitions.[8,16] However, quantitative ground state diagnostics of neutral xenon in a thin xenon plasma remains demanding since the TALIF process has low laser power density thresholds for competing phenomena such as resonant multi-photon ionization (REMPI) or amplified spontaneous emission (ASE) of the respective two-photon excited level.

In this paper, results from TALIF spectroscopy of neutral xenon in the plume of the RIT-10 ion thruster are reported. Measurements include relative particle density measurements in the cold gas flow and for several thruster operation conditions. Two different TALIF schemes are used involving levels of the $6p'$ multiplet. Results are discussed with regard to critical issues such as saturation effects of the TALIF processes and the contribution of xenon ambient pressure in the vacuum chamber to the measured signals.

II. Theory

The TALIF process can be modeled by solving a system of rate equations, which has been extensively discussed in literature.[6,7,17] Considering a model involving two-photon excitation from the ground state and fluorescence of the excited level, the fluorescence signal S can be written as

$$S = D\,\frac{E^2}{a^2}\,\frac{A_{21}}{A+Q}\,\frac{\hat{\sigma}^{(2)} n_0}{4\pi^2(\hbar\omega)^2}\int_{-\infty}^{\infty} F^2(t)dt. \tag{1}$$

Herein, E represents the energy of the laser pulse, a the beam waist at the detection area, n_0 the particle density, ω and $F(t)$ the frequency and the temporal profile of the laser beam respectively, A_{21} the Einstein coefficient for the observed fluorescence transition, A the radiative transition rate including all fluorescence channels of the two-photon excited state, Q the collisional deactivation rate and $\hat{\sigma}^{(2)}$ the two-photon absorption cross section. It is convenient to write the cross sections in terms of the fundamental quantity $\hat{\sigma}_0^{(2)}$

$$\hat{\sigma}^{(2)} = \hat{\sigma}_0^{(2)} g(\omega) G^{(2)} \tag{2}$$

that is independent of the line shape $g(\omega)$ of the two-photon excitation transition. Herein, $G^{(2)}$ is the two-photon statistical factor which accounts for temporal fluctuations of the laser intensity that are not resolved. The calibration constant D involves all factors with influence on the sensitivity of the detection system,

$$D = L\Omega\eta_t\eta_\Phi e K G_p \frac{G_a}{C}, \tag{3}$$

with η_t denoting the transmission of all windows and filters, η_Φ the quantum efficiency of the photomultiplier, G_p the photomultiplier tube gain, K including sensitivities of further analyzing instruments, G_a/C the ratio of preamplifier gain to preamplifier capacitance (in V/C), e the elementary charge (in C), L the effective length of the collection system (in cm) and Ω the solid angle (in sr) of the fluorescence collection lens. The units of S and D are V and V·cm·sr, respectively.

In the derivation of equation (1), processes competing with the fluorescence decay of the two-photon excited level have been neglected. If the power density of the laser pulse exceeds certain limits, REMPI and ASE can result in a deviation from the $S \propto E^2$ relation, which will be referred to as saturation in the following, and equation (1) becomes invalid. The saturation limit for the laser power density has to be determined for every two-photon transition.

In a recent publication, we investigated levels of the $6p$ and $6p'$ multiplets with respect to natural lifetimes, collisional deactivation coefficients and two-photon absorption cross sections in an experimental geometry

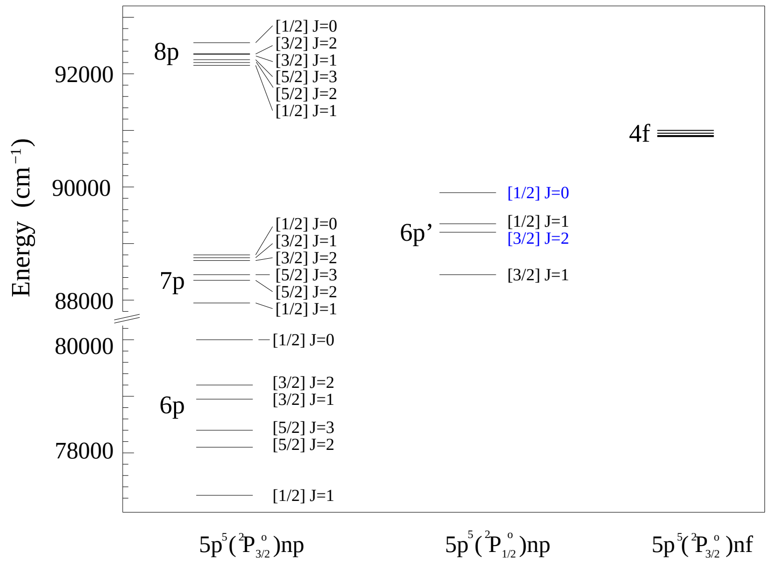


Figure 1. Partial Grotrian diagram of neutral xenon. Only levels of even parity are displayed. The highlighted levels have been involved in TALIF investigations in the plume of the RIT-10 thruster.

appropriate to perform TALIF spectroscopy in an ion thruster plume as part of a TALIF study in a static cold gas cell,[15] and discussed the results with respect to literature values. A partial Grotrian diagram of neutral xenon is shown in Fig. 1. TALIF schemes involving the $6p'[1/2]_0$ and $6p'[3/2]_2$ levels have been selected for neutral xenon diagnostics in the thruster plume, detecting fluorescence at 788.7 nm (branching ratio 0.62) and 834.7 nm (branching ratio 0.73, see[18]), respectively. Emphasis is put on the excitation scheme including the $6p'[1/2]_0$ level, as the intensity of the 788.7 nm line in the emission spectrum of the ion thruster plume has been found to be considerably weaker compared to emission at 834.7 nm for our ion thruster operation conditions. The two-photon absorption coefficient for the ground state excitation of the $6p'[1/2]_0$ level has been measured relative to the ground state excitation of the $6p'[3/2]_2$ level; we published[15] a two-photon absorption cross section ratio of ~ 1.

III. Experimental

Two-photon excitation is induced using the output of a pulsed Nd:YAG (Quantel Brillant B, pulsewidth 4.9 ns FWHM) pumped dye laser (Scanmate 2E). The laser system is tunable in the wavelength range between $\sim$ 435 to 454 nm using the laser dye Coumarin 120. Laser radiation is frequency doubled with a BBO crystal to provide the two-photon absorption wavelengths for the levels of interest. The output is focused (f=130 cm) in the vacuum chamber at the measurement position of interest in the ion thruster plume. Fluorescence radiation is collected using an achromatic lens (f=30 cm) at a distance of $\sim$ 120 cm to the measurement volume, and imaged onto a 100 μm slit placed in front of a gated photomultiplier (Hamamatsu 636-10). The respective fluorescence channel is selected using appropriate band pass filters (FWHM $\sim$ 10 nm) to minimize effects related to line blending. The data acquisition equipment consists of two Boxcar integrator channels (Stanford Research Systems SR250) and a computer interface (Stanford Research Systems SR245), controlled by a Labview program. Operations of pump and dye lasers are controlled by a trigger generator (Stanford Research SystemsDG535) in order to switch both the photomultiplier gate and the data acquisition with respect to the laser pulse. Fluorescence signal measurements are acquired using a 1 GHz oscilloscope (Gagescope 82G). Laser pulse energy measurement is performed using an energy monitor (Polytec RJP735).

The ion thruster is installed in a vacuum chamber of $\sim$ 5 m length and 2 m in diameter. The chamber is evacuated by two oil diffusion pumps with a pumping speed up to 50 000 l/s reaching a minimal pressure of 8×10^{-4} Pa.

The working principle of the RIT has been described in detail elsewhere.[19–21] The RIT-10 thruster of the present investigation consists of a two-grid system, applying voltages of 900-1100 V to the screen grid and -200 V to the acceleration grid within the operating conditions investigated in this work. The power of

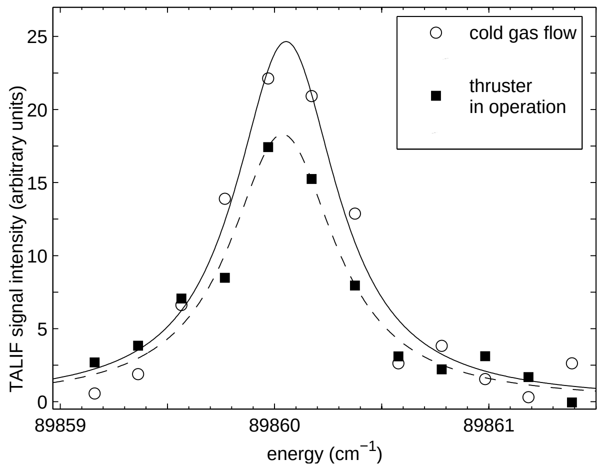


Figure 2. Spectral profile of the TALIF signal after excitation of the $6p'[1/2]_0$ level in the cold gas flow and when the thruster is operating; both measurements at xenon gas flow rate of 5.7 sccm. During operation, the screen grid voltage is 913 V at a radio-frequency generator power of 69.3 W.

the radio-frequency generator has been varied between 55 W and 110 W. Xenon flow rates between 2.1 sccm and 7.1 sccm have been investigated. In the present work, emphasis is put on the investigation of a varity of plasma conditions in the ion thruster plume aiming at the verification and improvement of the TALIF technique involving the excited levels mentioned above, rather than a complete characterization of a specific thruster operating condition.

IV. Results

All measurements have been carried out 12 cm downstream the acceleration grid on the central thruster axis. Each presented measurement is the result of a full spectral scan of 12-20 wavelength steps (6-10 pm in the frequency doubled regime). To obtain analyzable TALIF signal intensities, the averaged fluorescence of 500 laser pulses at each wavelength step is used.

Fig. 2 shows the absorption profiles after excitation of the $6p'[1/2]_0$ level at a xenon gas flow rate of 5.7 sccm, with fluorescence detected at 788.7 nm, both in the cold gas flow and during thruster operation, measured instantaneously one after another. Screen and acceleration grid voltages are 913 V and -200 V, respectively. The radio-frequency generator power is 69.3 W. At this operating condition, about 55% of the xenon atoms are ionized. When the thruster is turned on, the amplitude of the absorption line decreases, and the width of the profile increases slightly. Absorption lines have been fitted to a Voigt profile, assuming a Lorentzian contribution of the short-pulsed dye laser.

For a quantitative discussion of TALIF signal intensities with respect to particle number densities, saturation of TALIF process has to be excluded. Unfortunately, since the TALIF signal intensities are very weak due to the low particle densities in the thruster plume, it was not possible to vary the laser energy over an energy range wide enough required for a reliable check of the $S(E)$ relation. To allow for an interpretation of the data with regard to relative particle number densities, instead, at a constant laser pulse energy, the linearity between the fluorescence signal and the particle density $S \propto n_0$ was investigated. The result for the $6p'[1/2]_0$ level is shown on the left side of Fig. 3. Each step corresponds to an increase of the xenon particle density of $\sim 4 \times 10^{17}\mathrm{m}^{-3}$ in the vacuum chamber. Linear increase of the TALIF signal with the particle density has been found up to a xenon particle density of $\sim 2 \times 10^{18}\mathrm{m}^{-3}$. Collisional deactivation is assumed to be negligible at these low particle number densities, using deactivation coefficients for the respective excited levels from literature.[15,22,23] TALIF signal intensity in the cold gas flow is shown on the right side of Fig. 3. The signal has been measured directly before recording the data of Fig.3 (left). Comparing the TALIF signal intensities, we are confident that all measurements within this work are well in the linear regime.

It must be noted that the verification of $S \propto n_0$ shown on the left side of Fig. 3 is no strong evidence

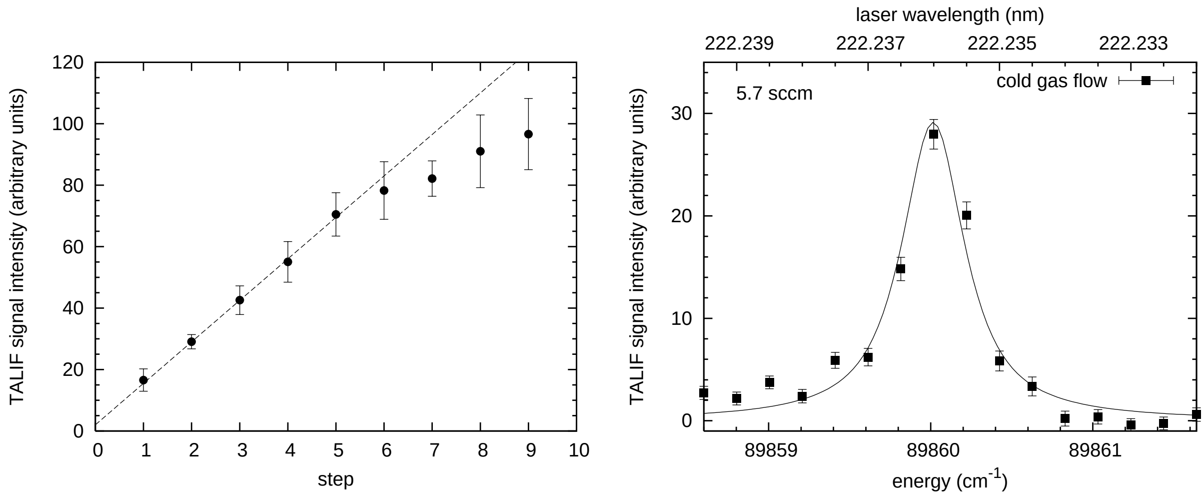


Figure 3. Left: Detected TALIF signal after excitation of the $6p'[1/2]_0$ level, in dependence of the xenon particle density in the vacuum chamber. Each step corresponds to an increase of the xenon particle density of $\sim 4\times10^{17}\mathrm{m}^{-3}$. Right: Spectral profile of the TALIF signal in the cold gas flow at xenon gas flow rate of 5.7 sccm. The excitation wavelength has not been corrected for internal laser offsets.

for the absence of saturation of the TALIF signal $S \propto E^2$. However, at fixed laser energies and $S \propto n_0$, measurements of relative particle densities are possible if the temperature of the investigated particle species does not vary from the cold gas conditions during the verification of $S \propto n_0$.

In Fig. 4, the measured TALIF signal intensity using excitation of the $6p'[1/2]_0$ and $6p'[3/2]_2$ levels is plotted over a variation of the xenon gas flow rate. Measurements have been performed in the cold gas flow at constant laser pulse energy. Signals are normalized to the intensity at 7.1 sccm. As expected, the TALIF signal increases with the mass flow rate.

In Fig. 5 the TALIF signal intensity is shown depending on the ratio of the beam current I_{beam} and the particle flow rate feeding the thruster $I_{tot} \equiv e\ \dot{m}/m_{Xe}$, wherein $\dot{m}/m_{Xe}$ denotes the xenon gas flow rate (in particles/sec) and e the elementary charge. I_{beam}/I_{tot} gives an estimate of the fraction of xenon atoms that are ionized after leaving the exit aperture. From a simplified point of view, the TALIF signal is expected to decrease when the fraction of ionized particles increases, although we note that a detailed knowledge of the particle beam density distribution in the plume at the respective ion thruster operation conditions is required for an accurate prediction. At fixed xenon gas flow rates of 5.7 sccm (Fig. 5, left) and 4.3 sccm (Fig. 5, right), the beam current was modified by variation of the power of the radio-frequency generator. All measurements use excitation of the $6p'[1/2]_0$ level, and TALIF signals are integrated over the spectral absorption profile. At 5.7 sccm, as can be seen in Fig. 5 (left), a decrease of the TALIF signal is observed at the position of the measurement volume when the beam current is increased.

Regarding the quantitative interpretation of the data, systematic error sources may arise for the case that the quadratic regime of the TALIF process is not reached: First, the line shape of the absorption profile might be broadened due to stronger saturation at the peak of the absorption line. Second, at gas temperatures larger than the $S \propto n_0$ relation is verified, Doppler broadening might lead to a decrease of ASE. As a result, the slope of the $S(n_0)$ (Fig. 3 left) may change. This effect is negligible, if either the spectral width of the laser pulse is significantly larger than the Doppler width, or the variation of the temperature is sufficiently small. However, the signal corruption by ASE is assumed to be unlikely at the low particle densities investigated here.

In addition, most probably, the xenon ambient pressure in the vacuum chamber, which was around 2.2×10^{-3} Pa at the 5.7 sccm operation condition, contributes to the TALIF signal of the flow species and diminishes the decrease of the signal intensities towards operation conditions with higher ionization yields. We speculate that in the present measurements the xenon ambient pressure was responsible for the rather ambiguous result in a measurement at the thruster operation condition at 4.3 sccm and a screen grid voltage of 1100 V (right side of Fig. 5). Here the decrease of the TALIF signal with larger beam currents seems to be diminished, however, more data is required at this operation condition. For future characterization of the plume of ion thrusters such as the RIT-10, the set-up needs to be improved, either by lowering the ambient

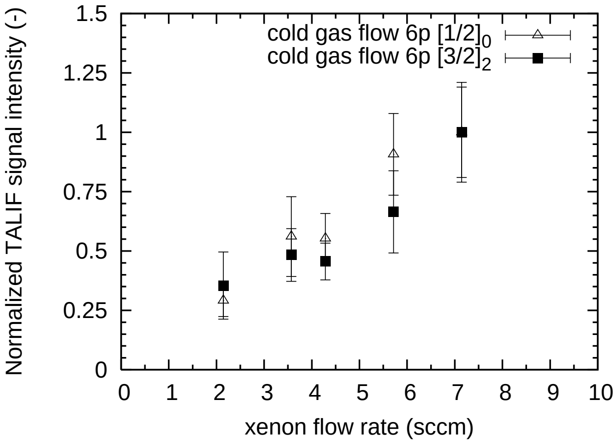
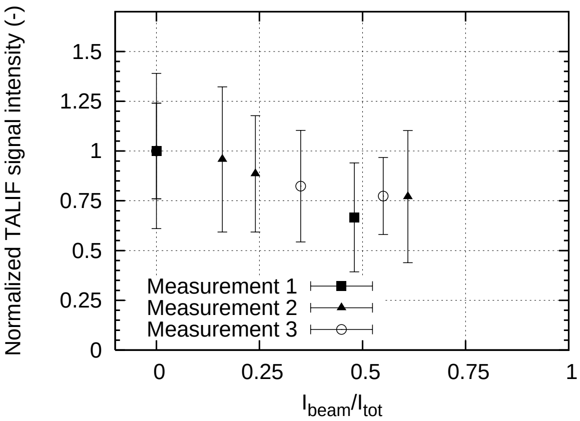


Figure 4. TALIF measurements using excitation of the $6p'[1/2]_0$ level (triangles) and of the $6p'[3/2]_2$ level (squares) in the cold gas flow of the RIT-10 thruster. Signals are normalized to the intensity at 7.1 sccm.



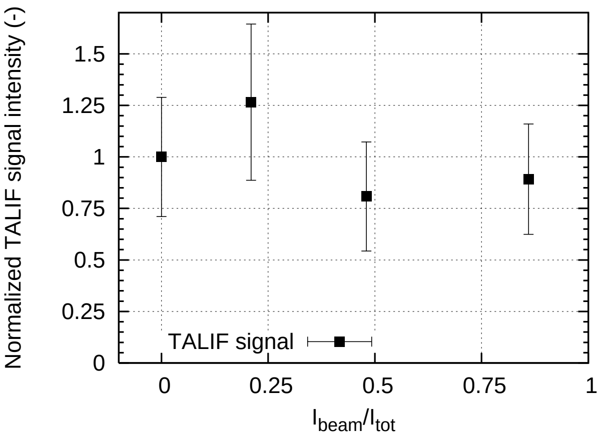


Figure 5. Measured TALIF signals as a function of the beam current I_{beam}, excitation of the $6p'[1/2]_0$ level. Thruster operation condition are 5.7 sccm at a screen grid voltage of 913 V (left) and 4.3 sccm at a screen grid voltage 1100 V (right). TALIF signals are normalized to the signal detected in the cold gas flow $I_{beam} = 0$.

pressure in the vacuum chamber or by increasing the sensitivity of the apparatus to subtract the ambient xenon density accurately.

V. Conclusion

Measurements using TALIF spectroscopy have been presented in the plume of the ion thruster RIT-10. The feasibility of two TALIF schemes including excitation of the $6p'[1/2]_0$ and of the $6p'[3/2]_2$ levels has been shown. Relative particle density measurements have been obtained in the cold gas flow. During thruster operation, TALIF signals under variation of the beam current have been investigated. Critical issues such as saturation of the TALIF process have been described.

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