ADVANCEMENTS IN PLASMA CHARACTERIZATION USING TWO-PHOTON POLARIZATION SPECTROSCOPY

Arne Meindl[1], Stefan Loehle[1], Irina Kistner[2], Andreas Schulz[2], and Stefanos Fasoulas[3]

[1]*High Enthalpy Flow Diagnostics Group HEFDiG, Institute of Space Systems IRS, University of Stuttgart, Germany*
[2]*Institute of Interfacial Process Engineering and Plasma Technology IGVP, University of Stuttgart, Germany*
[3]*Institute of Space Systems IRS, University of Stuttgart, Germany*

ABSTRACT

O_2 plasma created in a microwave powered plasma torch at atmospheric pressure is investigated. Measurements of electronic excitation temperature and relative ground-state density of atomic oxygen are carried out using optical emission spectroscopy and two-photon induced polarization spectroscopy respectively. The results presented in this paper indicate that the gas temperature is largely independent of the microwave power. Changes to the microwave power manifest as variations of the plasma volume which is observed as a drop in relative atomic oxygen number density. These findings are in agreement with previous investigations with other plasma species at similar conditions.

Key words: laser spectroscopy; atomic oxygen; plasma.

1. INTRODUCTION

Two-photon induced polarization spectroscopy (TIPS) has been used for detection of various molecules [11, 4] as well as of atomic hydrogen and xenon in the past [3]. The TIPS technique has been in further development at the High Enthalpy Flow Diagnostics Group (HEFDiG) of the Institute of Space Systems (IRS) in Stuttgart since 2016. Its scope of application has been extended to include atomic oxygen ground-state detection. The study presented in this paper features some of the very first measurements of atomic oxygen ever performed using TIPS. The setup as it is in use today has been established in 2018 [9]. This includes a detailed approach for absolute number density calibration using xenon as reference species. A first attempt at absolute number density calibration was presented at the AIAA Science and Technology Forum earlier this year [10]. This paper presents results obtained in an atmospheric pressure O_2 plasma during the ongoing development of the TIPS technique and characterization of the plasma torch.

The plasma torch used in this study is being developed at the Institute of Interfacial Process Engineering and Plasma Technology (IGVP) [6, 12]. It is depicted in Fig. 1 during operation. The microwave excitation primarily affects free electrons that transfer the energy to the oxygen molecules in the gas through collisions thus creating the plasma.

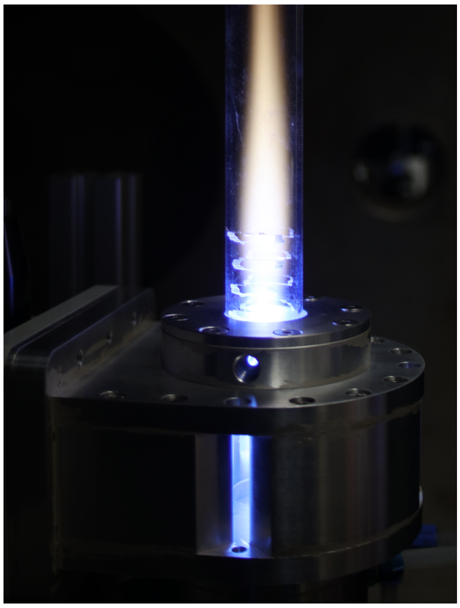

Figure 1. Microwave plasma torch operating with O_2.

2. EXPERIMENT

TIPS is a laser diagnostic technique that relies on the simultaneous absorption of two photons, one being contributed by a strong, circularly polarized pump beam and the other by a weaker, linearly polarized probe beam [3]. The two beams are crossed at a slight angle thus defining the measurement volume. Through consideration of the two-photon selection rules while choosing a suitable two-photon absorption transition to probe, it can be assured that the absorption of one photon from each beam induces a polarization rotation within the probe beam. The induced polarization rotation can be detected behind an analyzer which is a linear polarizer in crossed alignment

with the linear polarization of the probe beam.
The TIPS signal is created as the photons are absorbed thus rendering the technique immune to quenching effects. Therefore, the TIPS technique is suited for local detection of atomic oxygen at the challenging conditions of atmospheric pressure O_2-plasma.
In addition to the TIPS measurements, optical emission spectroscopy (OES) is used to expand the knowledge of the plasma conditions, since the O_2 plasma created with this plasma torch is not well understood yet.

2.1. Laser Diagnostic Setup

Figure 2 shows a schematic of the laser diagnostic setup used in this study. It has been described in detail in the past [9]. Both laser beams are created using the same source, a pulsed Nd:YAG-pumped tunable dye laser system operating at 20Hz. The laser beam is split in 9:1

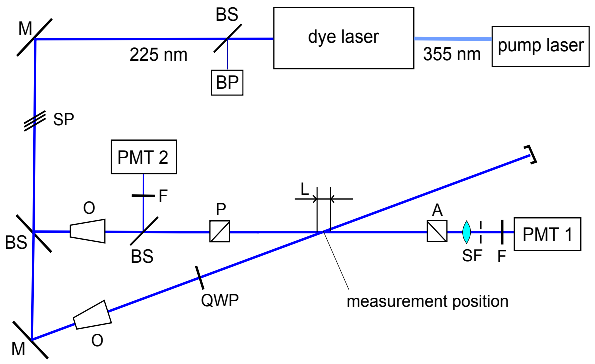

Figure 2. TIPS experimental setup. A - analyzer, BP - beam profiler, BS - beam splitter, F - filters, L - overlap length, M - mirror, O - focusing optics, P - linear polarizer, PMT - photomultiplier tube, QWP - quarter wave plate, SF - spatial filter, SP - stack polarizer.

ratio into a strong, circularly polarized pump beam and a weak, linearly polarized probe beam. When polarization rotation of the probe beam occurs, the irradiance E_1 detected by PMT 1 increases while the irradiance E_2 detected by PMT 2 remains unchanged. The measurement signal of TIPS is the ratio $\frac{E_1}{E_2}$. Through scanning of the laser wavelength over the two-photon absorption line of atomic oxygen, a polarization profile is obtained.

2.2. Measurements in O_2 Plasma

For the investigation with oxygen as operating gas, the plasma torch is equipped with a quartz tube that extends out vertically in order to stabilize the gas flow above the resonator exit and minimize entrainment of ambient air. Slits in the quartz tube allow the laser beams to reach the intended overlap position while avoiding any influence of the birefringent quartz on their polarizations. Figure 3 details the measurement positions of TIPS and OES above the resonator exit of the plasma torch.

Figure 3. Measurement positions of TIPS and OES above the plasma torch resonator exit.

The TIPS measurement position is located about 3 mm above the resonator exit in the center of the plasma flow. The overlap has a length of 9 mm. At the measurement position, the focused pump and probe beams are 300 μm and 200 μm in diameter respectively.
The OES spectra are collected using an Echelle spectrometer (LTB, *Aryelle 150*) and an optical fiber with one end fixed next to the plasma torch in the same vertical position above the resonator exit as the laser beams. This results in a line-of-sight measurement through the entire plasma flow with a diameter of 5 mm.
TIPS and OES measurements are performed for two different microwave power settings, 1 kW and 2 kW, as well as multiple O_2 volumetric flow rates between 10 l/min and 25 l/min. TIPS and OES are performed consecutively.

3. DATA ANALYSIS

The OES spectra are calibrated using an Ulbricht sphere placed at the measurement position. Dark frames are recorded separately and subtracted during the calibration. Figure 4 shows a resulting calibrated spectrum.
The atomic oxygen emission lines around 777 nm and 844 nm are prominent features of the spectrum. They are used in the further data analysis. Voigt convolved spectra simulated using PARADE [7, 13] are fitted to the atomic lines in order to determine electronic excitation temperatures for the different plasma conditions.

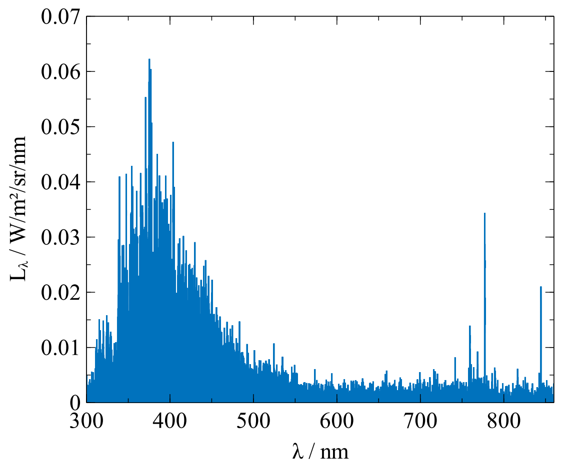

Figure 4. Optical emission spectrum calibrated for radiance L_λ of plasma created with 1 kW microwave power and 10 l/min volumetric O_2 flow rate.

The intense broad feature between 300 nm and 500 nm is not investigated further, because the radiance of the Ulbricht sphere used for calibration drops to near zero towards 400 nm. Hence, a different calibration source would be necessary for a meaningful investigation of this wavelength range. The emission from near UV into the blue spectral range might be due to O_2^+ though a possible contribution by second order emission from O_2 Schumann-Runge cannot be ruled out since no low-cut filter is used during the measurement.

In order to analyze the polarization profiles recorded with TIPS, the lineshape has to be modelled with several different contributions in mind. Absorption, dispersion, and inaccuracies in the alignment of the analyzing polarizer all influence to the overall shape of the polarization profile. The absorption and dispersion are linked via the Kramers-Kronig relations [1, 2]. They are subject to Gaussian as well as Lorentzian shaped broadening contributions. A complex error function $W(x + \mathrm{i}y)$ can be used to describe both absorption and dispersion lineshapes, χ_{abs} and χ_{disp}, as functions of the detuning from the absorption central wavelength $\Delta\lambda$ and the Gaussian and Lorentzian half widths at half maximum (HWHM), Γ_D and Γ_L [8, 14]:

$$\chi_{abs}(\Delta\lambda) = \frac{1}{\sqrt{\pi}\Gamma'_D}\mathrm{Re}[W(x(\Delta\lambda) + \mathrm{i}y)] \quad (1)$$

$$\chi_{disp}(\Delta\lambda) = \frac{1}{\sqrt{\pi}\Gamma'_D}\mathrm{Im}[W(x(\Delta\lambda) + \mathrm{i}y)] \quad (2)$$

$$\Gamma'_D = \frac{\Gamma_D}{\sqrt{\ln 2}} \qquad x(\Delta\lambda) = \frac{\Delta\lambda}{\Gamma'_D} \qquad y = \frac{\Gamma_L}{\Gamma_D}. \quad (3)$$

The absorption and dispersion profiles are combined with an asymmetry $\pm\alpha$ caused by imperfect crossing of the linear polarizers in order to model the polarization profile $P(\Delta\lambda)$ as measured with TIPS:

$$P(\Delta\lambda) = \chi_{abs}(\Delta\lambda)^2 + (\chi_{disp}(\Delta\lambda) \pm \alpha)^2. \quad (4)$$

The lineshape model is implemented into a nonlinear fitting algorithm with fitting parameters Γ_D, Γ_L, $\pm\alpha$, absorption central wavelength λ_0, and a scaling factor S. An example of a polarization profile fit to measured data is shown in Fig. 5.

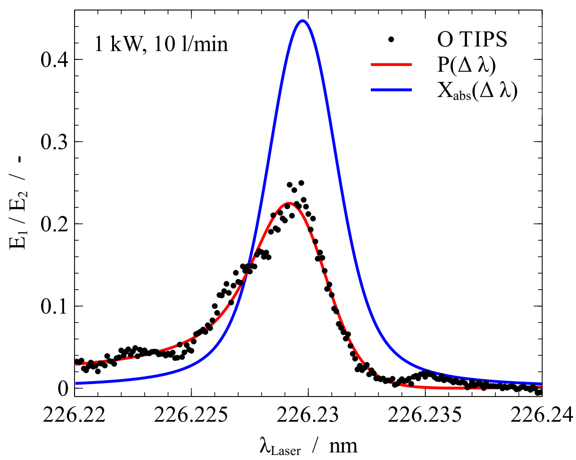

Figure 5. TIPS scan and profile fit of O $2p^4\,{}^3P_0 \rightarrow 3p\,{}^3P_0$ in plasma created with 1 kW microwave power and 10 l/min volumetric O_2 flow rate.

During the fitting process, the profiles are constructed using area normalized Voigt absorption profiles that are subsequently scaled to the data via parameter S. Thus, both the parameter S as well as the integrated absorption profile of the probe beam can be used directly for measurements of relative ground-state number densities of atomic oxygen for the different plasma conditions.
Γ_D potentially influences the magnitude of the scaling parameter S as can be seen in equations (1) and (2), which could impact the relative number density measurements. Therefore, Γ_D is fixed to 1.281 pm during fitting which corresponds to a translational temperature of 4000 K. Investigations with humid air plasma performed by Leins et al. [6] during the development of the plasma torch have shown that the maximum gas temperature is determined by the microwave excitation mechanism and is therefore not expected to exceed this value at this study's TIPS measurement position.

4. RESULTS AND DISCUSSION

Figure 6 shows the electronic excitation temperature derived from the PARADE fits to the OES spectra for the different plasma conditions. At 1 kW microwave power, the 20 l/min flow rate condition has not been investigated.

A systematic uncertainty analysis has not been performed

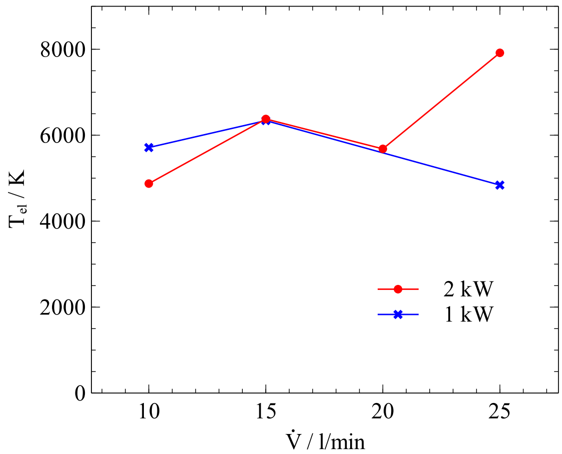

Figure 6. Electronic excitation temperature T_{el} for different microwave powers and volumetric O_2 flow rates.

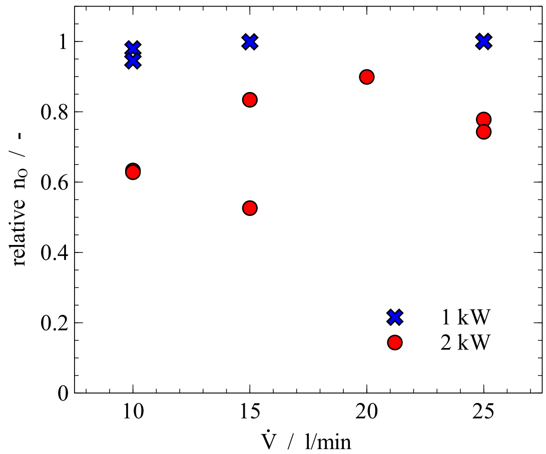

Figure 7. Relative atomic oxygen ground-state number densities normalized to peak number density for different microwave powers and volumetric O_2 flow rates.

for the results of this study. However, due to the fact that the electronic excitation temperature is measured using only the 777 nm and 844 nm emission lines, an uncertainty of ± 1000 K should be expected here as a conservative estimate. Therefore, the measurements indicate that the electronic excitation temperature is unaffected by microwave power and flow rate across most of the investigated conditions. This is consistent with the previous investigations of air plasma that showed a mostly condition-independent temperature behaviour [5, 6]. This indicates that any error introduced through the locking of Γ_D during the TIPS lineshape fitting would be the same across these conditions and therefore not affect the evaluation of relative number densities.

The only exception is the electronic excitation temperature measured for the 2 kW, 25 l/min condition. It is significantly higher than the rest. A likely explanation for this behaviour is the high temperature core of the plasma, which is normally confined to the resonator and thus below the measurement position, extending further downstream, and therefore out of the resonator, with increasing power and flow rate [5]. If it reaches the OES measurement volume, it would result in a higher measured electronic excitation temperature. The OES measurement volume extends further upstream than the TIPS measurement volume as can be seen in Fig. 3, this could be the reason why this effect is not present in the TIPS data. For future investigations, it would be advisable to limit the upstream extent of the OES measurement volume with respect to the TIPS position for measurements this close to the resonator exit.

The relative ground-state number densities of atomic oxygen measured with TIPS are presented in Fig. 7. The plot contains at least two measurements per condition with the exceptions being conditions 1 kW, 15 l/min and 2 kW, 20 l/min which consist of a single measurement each. Again, the 20 l/min condition was not investigated for 1 kW microwave power and no systematic uncertainty analysis was performed. However, a good repeatability of the results can be attested to most conditions. There is only one condition (2 kW, 15 l/min) where the results of multiple measurements are not very close or even in exact agreement.

For the 1 kW conditions, the number density is consistently high and stable. However, increasing the microwave power results in a lower number density. The fact that the temperature does not change with increasing power means that the additional power results in expansion of the plasma volume instead and consequently lower local number densities at the measurement position.

5. CONCLUSION AND OUTLOOK

The O_2 plasma torch has been investigated with regards to electronic excitation temperature and relative ground-state density of atomic oxygen using OES and TIPS respectively. The results indicate that the gas temperature is largely unaffected by variations to microwave power and gas flow. Different conditions result in variations of the plasma volume. For plasma created by microwave excitation, this behaviour can be observed when the frequency of the exciting collisions of free electrons and heavy particles approaches an optimal value equal to the microwave frequency. The improved understanding of the plasma torch behaviour with regards to microwave power and flow rate achieved by this study will be key in optimizing the experimental setup and methodology for future investigations of this type.

With regards to the development of the TIPS technique, the results are promising due to the achieved repeatability. The development of the technique will continue and measurements of absolute ground-state number densities of atomic oxygen through calibration with xenon as reference species will be pursued in future investigations.

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