Usefulness of ps-TALIF to measure gas temperature and collisional cross-sections

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In this work, we will discuss about the application of ps-TALIF to H_2 plasmas at low to moderate pressures to obtain more for meaurement of gas temperatures as well as collisional cross-sections, in addition to the H-atom densities. TALIF has become the standard approach to measure atom densities in reactive environments [1]. The principle of TALIF is to excite ground-state atoms to a fluorescing state by absorption of two photons of a pulsed laser and subsequently recording the resulting fluorescence signal. Combinination of ps-laser with a streak camera having ps-time resolution allows for application of TALIF for diagnosing reactive moderate to high pressure plasmas where the effective lifetimes τ_X of the fluorescence are of sub-nanosecond or ps-time scales[1].

In addition to the measurement of atomic densities, measurement of τ_H serves as a probe of local plasma conditions as it encapsulates all the radiative and collisional quenching processes

$$\frac{1}{\tau_i} = A_H + Q_H = A_H + \frac{P}{k_B T_g} \sum_{j}^{quenchants} k_{Q_{i/j}} x_j \tag{1}$$

Where Q_H and A_H are the quenching rate and the total Einstein coefficient of radiative decay of the excited state. P is the local pressure, T_g is the gas temperature, k_B is the Boltzmann constant, and x_j is the molar fraction of the quenchant j. One can identify, 3 distinct regimes based on the radiative and collisional cross-sections

- Radiation dominated regime at extremely low pressures i.e. $A_H >> Q_H$
- Collision dominated regime at high pressures where $A_H << Q_H$
- Competition between radiative and collisional processes at intermediate pressures i.e. $A_H \sim Q_H$

Therefore, the measurement of τ_H and exploitation of equation 1 allows one to gain more insight about the reactive environment.

At low pressure conditions, the depopulation mechanism of the laser excited state H(n=3) is dominated through radiative processes. In fact, the normally optically thick Lyman_β becomes optically thin in these limits and its contribution to the overall radiative lifetimes A_H can no more be ignored. With respect to collisional quenching of the laser excited state, molecular H_2 is the most dominant. Previous experimental studies have reported a wide range of collisional quenching cross-section $\sigma_{Q_{H/H_2}}$ between 65 to 156 A^2. Measurement of τ_H over a range of low pressure, where the radiative processes are dominant, allows for the measurement of the radiative decay rate $1/A_H$ and collisional quenching cross-section $\sigma_{Q_{H/H_2}}$ using the Stern-Volmer plot. ps-TALIF measurements was performed on a Sairem Hi-wave source at pressures between 0.01 to 3 mbar. Lyman_β line is only partially trapped in these conditions and $1/A_H$ is no more constant. This implies that Stern-Volmer method cannot be used to determine the quenching cross-sections. In such a scenario, it is necessary to use a collisional-radiative model in order to obtain the collisional cross-sections. Using such an approach we deduced a value of 98±10 A^2 for $\sigma_{Q_{H/H_2}}$ [2].

Experiments were also performed on a moderate pressure microwave plasma torch to investigate the validity of $\sigma_{Q_{H/H_2}}$ estimated from low pressure conditions. In fact at moderate to high pressure conditions dominated by the highly collisional regime, one can estimate the gas temperature directly from the measured fluorescence lifetimes τ_H following Similarly, with the knowledge of radiative and collisional decay constants, it would be possible to deduce the gas temperature T_g by rewriting equation 1 as

$$T_g = \frac{8}{\pi k_B}\left(\frac{P\tau_i}{1-\tau_i A_i}\sum_j \frac{x_j \sigma_{Q_{i/j}}}{\sqrt{\mu_{i/j}}}\right)^2 \tag{2}$$

The peak of the gas temperatures obtained from τ_H that represent the emissive region of the plasma has been compared with rotational temperature determined from the emission spectra of H_2 rotational bands of R branch of the transition $G^1\sum_g^+$, $\nu' = 0 \rightarrow B^1\sum_u^+$, $\nu'' = 0$, and lower and upper limits of the Q-Branch $d^3\prod_u$, $\nu' = 0 \rightarrow a^3\sum_g^+$, ν''= 0 of the Fulcher-α band. With regard to the σ_{H/H_2} of H_2 molecule, values ranging between 65 [3] to 156 [4] A^2 have been reported in the literature and the gas temperatures estimated using the respective σ_{H/H_2} are either too low when compared to the rotational temperature of Fulcher or far too high to be realistic (c.f. Figure 1(a)). This validates the value of σ_{H/H_2} (98 A^2) measured for the experiments at lower pressure.

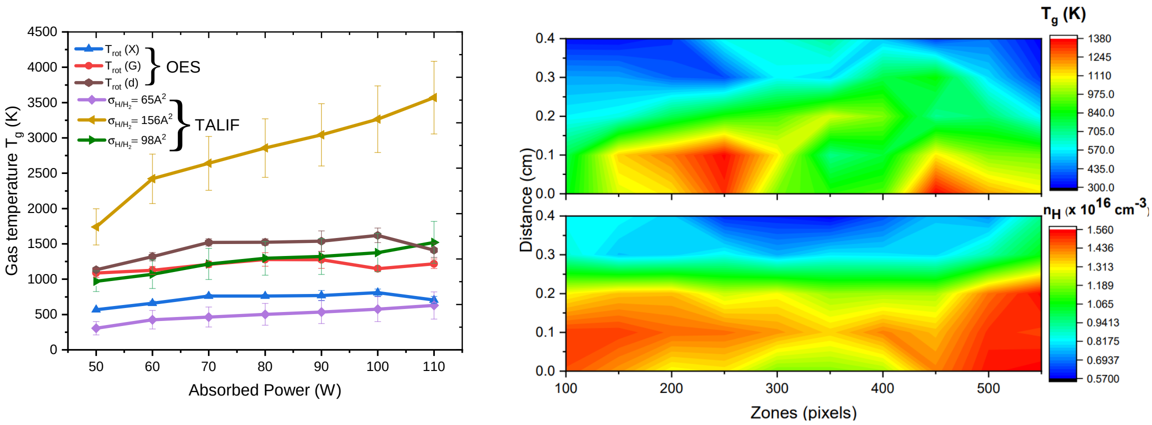

Fig. 1: (a) Comparison of gas temperatures measured using OES and ps-TALIF and (b) Constructed 2D contour plots of H-atom density and T_g at 100 mbar.

Furtheremore, the present methodology of gas temperature measurement allows us to make simultaneous measurement of H-atom density as well as gas temperature. Figure 1 (b) shows the contour plot of the H-atom density and gas temperatures measured from the ps-TALIF procedure.

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[1] Gazeli, K et al., *plasma* **1(4)** (2018) 145–171.
[2] Duluard, CY et al., *PSST* **33(1)** (2024) 015003.
[3] Bitttner, J et al., *Chemical Physics Letters* **143(6)** (1988) 571–576.
[4] Catherinot, A et al., *Chemical Physics Letters* **18(3)** (1978) 1097.