

Femtosecond TALIF at high pressure and cell calibration developments

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1. Context

Conventional ns TALIF operates in the quadratic regime, for which the integrated fluorescence signal is proportional to the square of the laser intensity. However, several limitations exist. The accuracy of TALIF measurements depend on the uncertainty of the two-photon absorption cross sections, which is on the order of 50% for the used kinetic schemes. Another drawback is the need for the accurate measurement of the fluorescence decay time, which is highly dependent on the quenching rates. As quenching acts as a competitive channel, the fluorescence decay can become as short as a few tens of ps in atmospheric pressure plasmas. Their measurement is therefore very challenging and may not be possible even with the use of streak cameras. Short laser pulses (ps, fs) have been used to both enable the measurement of the fluorescence decay rates at high pressure and to prevent photolytic effects. However, high laser intensities of the order of hundreds of GW/cm² to PW/cm² with very large spectral width can easily be reached with ultra-short lasers and consequently other phenomena are expected. In this work, we follow the path set in [1,2] and present the results of VUV absolute calibration of the fs-TALIF diagnostic. By working in the very high laser intensity regime, quenching rates are lower than photoionization and stimulated emission and therefore the technique becomes quench-free. To analyze the fluorescence signal, the calibration was performed with a known source of the same atom as the probed atom, calibrated by VUV-absorption spectroscopy (at about 120 nm) at the Synchrotron SOLEIL facility in Saclay, France.

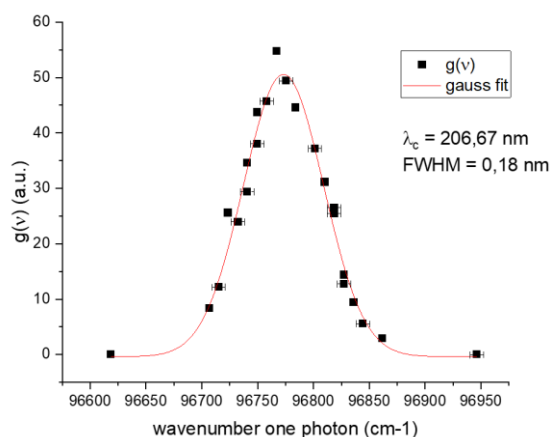


Fig. 1: Laser two-photon absorption profile $g(\nu)$.

References

[1] Stancu G.-D., Plasma Sources Sci. Technol. **29** (2020) 054001

[2] Dumitrache et al. Plasma Sources Sci. Technol. **31** (2022) 015004

2. Fs-laser system

A Ti:sapphire laser system (Spectra Physics Solstice ACE) produced femtosecond pulses and operated in the wavelength range $\lambda = 780\text{--}830 \text{ nm}$. The energy per pulse of the fourth harmonic was $\sim 20 \text{ mJ}$ at a repetition frequency of 1 kHz and tuned for the wavelength range 204-207 nm. In figure 1, a typical two-photon absorption profile measured for N atoms is shown. Note that this appears very large (about 90 cm^{-1} for FWHM), because of the double convolution broadening mechanism that is dominated by the fs laser ($g(\nu) = f_{\text{abs}} \otimes f_{\text{laser}} \otimes f_{\text{laser}}$).

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