Conventional and ultrashort laser diagnostics for fundamental studies of atmospheric pressure plasmas

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Based on the principle of stimulated emission introduced by Albert Einstein in 1917, laser sources have been incessantly developed since their discovery in the 1960s. Generated in continuous or pulsed mode, laser diagnostic techniques are today essential tools for the fundamental understanding of energy distribution, kinetic and dynamic processes in reactive and non-reactive plasma environments, as well as for plasma engineering. With photons of wavelength ranging primarily from UV to Mid-IR, they enable probing with high sensitivity, selectivity, spatial and temporal resolutions key plasma parameters such as species densities, pressures, temperatures, velocities, flux distributions and E&M fields.

Due to their strong potential of applications in the domains of materials, energy, environment, transport, health or agriculture, atmospheric pressure plasmas have attracted great scientific interest in recent years. In particular, non-equilibrium reactive plasmas generated under ambient conditions are complex environments governed by multi-physics interactions, are usually confined and present transient behaviors. Owing to much larger collision rates than at low pressures, they exhibit fast kinetic and dynamic processes with characteristic times often less than one ns and with reduced plasma volumes (e.g. sub-mm) dominated by large species, temperature and field gradients.

These peculiarities of atmospheric plasmas call into question the implementation of conventional laser diagnostic methods. For example, the sensitivity of classical absorption spectroscopy (AS) is lower because of the small absorption length, whereas the selectivity is poorer due to the large collisional broadening that enhances the spectral overlap. Furthermore, evaluating the density or temperature of plasma species becomes intricate because the line-of-sight AS signals represent integrated values over a highly non-uniform plasma for which even the absorption length is an unknow [1,2]. Significant challenges are also present in the case of the laser induced fluorescence (LIF) diagnostics. Although the sensitivity, selectivity, spatial and temporal resolution are exceptional, the uncertainties using LIF with single-photon or multi-photon absorption methods will increase, due to the quenching phenomena that are orders of magnitude larger than for low pressure plasmas. Additionally, photolytic processes in reactive plasmas will perturb measurements by intrusively increasing the densities of probed species [3,4,5]. Laser scattering techniques, which include elastic, inelastic, resonant or non-resonant, coherent or non-coherent scattering processes, are confronted either with a lack of sensitivity and selectivity, or with the sharp temperature and species gradients over reduced and transient plasma volumes [6,7].

The employment of ultrashort lasers such as mode-locked ps and fs for plasma and combustion diagnostics has increased over the past two decades. Large improvements in diagnostics have been reported particularly for multi-photon techniques, where the very high instantaneous intensity and photon statics favor the probability of laser-plasma interactions. For example, fluorescence techniques for species detection demonstrate higher sensitivities and photolytic-free capabilities [8,9,10], second harmonic generation methods for electric field measurements exhibit higher sensitivities [11], whereas coherent anti-stokes Raman spectroscopy methods allow for single-shot species and temperature measurements [12].

It should be noted that these developments pose new challenges for accurate description of the laser-plasma interactions. For instance, experiments employing ps and fs lasers for two-photon absorption laser induced fluorescence (TALIF) are reported for photon intensities on the order of TWcm^{-2} or even higher, while conventional ns lasers have intensities usually below GWcm^{-2} [10]. Their photon statistics are determined by the mode-locked characteristic which is very different from that of ns lasers (e.g. multimode with stochastic phase fluctuations). As the second-order correlation factor increases by several orders of magnitude, the two-photon transition probability increases and hence, the sensitivity of the method. New phenomena, such as Stark detuning and coherent excitation are expected. At the Heisenberg limit, a 100-fs laser will have a spectral width of ~146 cm^{-1}, which is a few orders of magnitude larger than for a conventional ns laser (e.g. ~ 0.1 cm^{-1}). Therefore, excitation of multiple transitions and species is more likely. Furthermore, employing fs lasers, coherent processes are expected for two-photon excitations. Indeed, the decoherence time scale for laboratory plasmas are orders of magnitude greater than the excitation time. For example, the characteristic Doppler decoherence time at room temperature for hydrogen, nitrogen and oxygen atoms, for two-photon transitions at 97492 cm^{-1}, 96750 cm^{-1} and 88631 cm^{-1}, respectively, is on the order of ps and tens of ps, while the collisional decoherence time at atmospheric pressure plasmas is hundreds of ps or longer. This implies that appropriate models must consider coherence terms as described by the density matrix equations [3].

In Fig. 1 and 2, examples of two-photon excitation probability for the O atom with laser intensities of 1 $GWcm^{-2}$ and 100 $GWcm^{-2}$, and pulse widths of 6 ns and 200 ps, respectively, are computed using a density matrix model for negligible quenching (Fig. 1) and for typical quenching of atmospheric pressure plasmas (Fig. 2). We notice that probability amplitudes for 1 $GWcm^{-2}$ are significantly different, whereas at 100 $GWcm^{-2}$ they exhibit Rabi oscillations, and they are almost identical. Consequently, in high intensity regime the fluorescence probability, which is proportional to excitation probability, can become independent on quenching phenomena. This is a great advantage for accurate measurements of species using LIF techniques, particularly for non-uniform transient plasmas at atmospheric or high pressures, where quenching can vary considerably in time and space due to changes of the temperature and nature of colliders. Note that a classical ns LIF technique would require a tremendous number of experiments for quenching characterization.

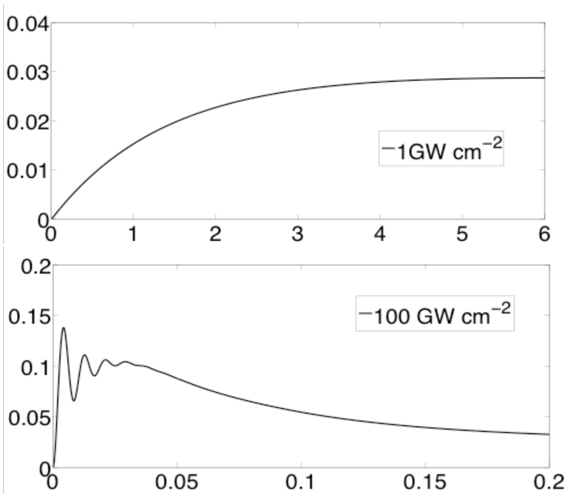

Fig. 1: Excitation probability for O atoms for laser intensities of 1 $GWcm^{-2}$ & 100 $GWcm^{-2}$, and pulse width of 6 ns & 200 ps, respectively, for negligible quenching.

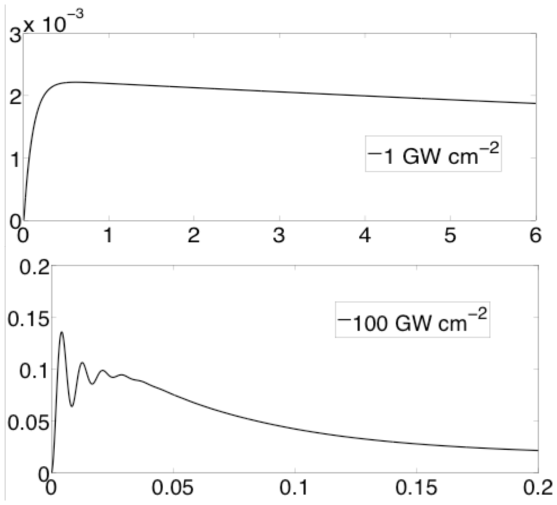

Fig. 2: Excitation probability for O atoms for laser intensities of 1 $GWcm^{-2}$ & 100 $GWcm^{-2}$, and pulse widths of 6 ns & 200 ps, respectively, for quencing rate of $9x10^9$ Hz.

In this contribution, insights and perspectives will be presented for conventional and ultrafast laser plasma diagnostics, such as time-resolved absorption enhanced by cavities or calibration techniques employed for femtosecond laser induced fluorescence. Examples of fundamental investigations of atmospheric pressure plasmas generated by nanosecond and microwave discharges using laser techniques will be shown along with plasma energy branching, kinetic and dynamic studies.

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