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Laser Induced Fluorescence in a collisional environment: a molecular probe for rapidly changing media

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Laser Induced Fluorescence (LIF) is nowadays a well established and widely used technique in gas discharges research, and one might wonder if there is really anything left to understand and investigate about it. The answer is yes, of course. The need for further deepening of the knowledge about LIF basically comes from its application to atmospheric pressure discharges, that is to highly collisional conditions. LIF, intended as a way to measure the concentration of transient species M , is in principle an absorption technique with a different observable, the fluorescence from an electronically excited state M prepared by absorption of resonant laser light. In a molecular case, M can be a single ro-vibronic state. Collision energy transfers (CET) processes involving M^* and the background molecules have an heavy influence on the fluorescence outcome. This is generally considered as a shortcoming, since the quantitative use of LIF for the characterization of the transient specie M requires a detailed knowledge of the collision frequencies. Reversing the point of view, this knowledge allows to use the laser prepared electronic state M as a transient quantum sensor for the medium composition, that can be used in hostile environments characterized by rapid changes (sub- μ s scale) of temperature, pressure and gas composition. This is possible since the CET processes with the background molecules redistribute the energy initially deposited in a single rovibronic state [1, 2], and thanks to the quantum nature of the molecular interaction that makes the CET processes depend strongly on the collisional partner. In this paper we shall present the general aspects of LIF in a collisional environment, introduce the idea of CET-LIF [3], and describe its application to the measurement of CO₂ dissociation in a nanosecond repetitively pulsed (NRP) spark discharge, using OH(A) state as the molecular sensor [4]. In parallel with the applications, we shall describe our work on the quantitative characterization of the CET processes in equilibrium [2] and non-equilibrium (work in progress) conditions.

References

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