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## **ACCEPTED MANUSCRIPT**

## Modeling Plasma Heating by ns Laser Pulse

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#### Abstract

The transition to breakdown of a weakly ionized gas, considering inverse bremsstrahlung, has been investigated using a state-to-state self-consistent model for gas discharges, mimicking a ns laser pulse. The paper is focused on the role of the initial ionization on the plasma formation. The results give the hint that some anomalous behaviors, such as signal enhancement by metal nanoparticles, can be attributed to this feature. This approach has been applied to hydrogen gas regarded as a simplified model for LIBS plasmas, as a full kinetic scheme is available, including the collisional–radiative model for atoms and molecules. The model allows the influence of different parameters to be investigated, such as the initial electron molar fraction, on the ionization growth.

Keywords: LIBS, ns Laser Pulse, Plasma Modeling, Hydrogen state-to-state chemical kinetics

#### 1. Introduction

Nowadays laser-induced plasmas (LIP) find a large number of applications, from pulsed laser deposition (PLD) [1–3] to adrotherapy [4]. Among them, laser induced breakdown spectroscopy (LIBS) [5, 6] is the most popular technology, due to its simplicity, the wide set of samples that can be analyzed (liquid [7], solid [8], gas [9]) and the possibility to operate in different conditions (e.g. underwater [10], on martian surface [11–13]). In investigating the emission spectra local thermodynamic equilibrium (LTE) is usually considered [14], even if the possibility of non-equilibrium conditions has been also investigated [15–18].

The spectral signature of LIBS is the consequence of the amount of ablated material (number of emitters) and of the plasma temperature (excited states), which are influenced by different portions of the laser pulse [19]: the first photons initiate the plasma, by evaporating the surface and by multi-photon ionization. These mechanisms produce a weakly ionized gas with low electron temperature which is heated by the upcoming radiation. The early stage evolution of the plasma temperature, characterized by strong nonequilibrium, is determined by the interaction of the electrons emitted by the surface with the electric field of the laser pulse (inverse bremsstrahlung) [20, 21] that leads to a fully ionized gas (breakdown) observed as a laser spark, characterized by strong continuous emission. The heating rate strongly depends on the initial number of electrons in the plasma, which can be influenced by the surface topography and composition. The interest in this aspect araises from the need of explaining the dependence of the LIBS signal on the surface roughness [22, 23] or the enhancement due to the presence of metal nanoparticles on the surface of the target (NELIBS [24, 25]). The metal nanoparticles deposited on a surface increase the initial number of electrons because the photoemission current can be two orders of magnitude higher than the bulk metal [26]. Moreover, the nanoparticles evaporated by the laser pulse release in the gas metallic atoms that have an ionization potential much lower than the target particles, and therefore can be multiply ionized also at lower plasma temperature and with low applied field, presenting a higher ionization efficiency with respect to the target particles. The evolution of the plasma in its first stage determines temperature, pressure

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