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Aluminium laser-induced plasma: A collisional-radiative model

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The determination of the elementary composition of sample surfaces is a question widely open. Classical techniques (Castaing probe, atomic probe) involve experimental protocols which do not allow instantaneous and in-situ determinations The Laser Induced Breakdown Spectroscopy (LIBS) is a promising way to avoid these drawbacks [1, 2, 3] but it comes up against several problems to be solved in order to be generalised.

This technique is based on both analysis of the radiative emission of the plasma and comparison with reference spectra recorded from known samples. The wide range of possible spectra is the main limitation of the technique: instead of resorting to these databanks, we propose to model the complete behaviour of the plasma, mainly in early time corresponding to the breakdown. The present communication reports the elaboration of such a model for a test-case corresponding to an aluminium sample in vacuum interacting with a nanosecond laser pulse.

The early phase is very complex owing to the concomitance of very different phenomena and mutual influence. Simultaneous phenomena as laser light absorption, sample heating, melting, vaporisation, excitation and ionisation take place. Due to the characteristic time scales of these phenomena, the plasma is in chemical and thermal non equilibrium. We have therefore decided to pay our attention on these points by elaborating a Collisional-Radiative (CR) model accounting for 107 levels of Al, Al^+, Al^{++} and Al^{+++}. The procedure adopted to elaborate such a CR model has been already used successfully for other plasmas [4]. This type of tool is particularly suitable for describing non-equilibrium gaseous phases.

According to the laser fluence, the literature reports that different regimes can be observed. For aluminium, when the fluence is less than the threshold $F_0 = 8.5$ J cm^{-2}, the vaporisation is quiescent [5]. Conversely, when the fluence is higher than F_0, the vaporisation becomes strong and it induces ejection of matter by droplets: the related phase is called explosive. The development of CR models in multiphase flows has not been performed so far owing to the high degree of sophistication required. We have therefore arbitrarily reduced the range of application of our model to situations where $F < F_0$. This limitation leads also to moderate heating of the sample: the vaporisation of the metallic pool after melting can be therefore treated in a classical way, the temperature being less than the critical temperature $T_c = 6700$ K [6].

The coupling between the sample phenomena and the plasma can be simplified in a first approximation by assuming a time evolution of the interface temperature T_s similar to that of the laser pulse. The sensitivity of the results upon T_s can be therefore tested. The model developed with this simplification is called “model A”. The complete model solving the coupling between the sample and the laser pulse has also been elaborated and is called “model B”.

Figure 1 displays typical time evolutions resulting from the "model A" where the maximum value for T_s is 3000 K. It is interesting to note that the increase of the population densities (at the top of Fig.1) is very rapid when the decrease after the pulse is incomparably slower.

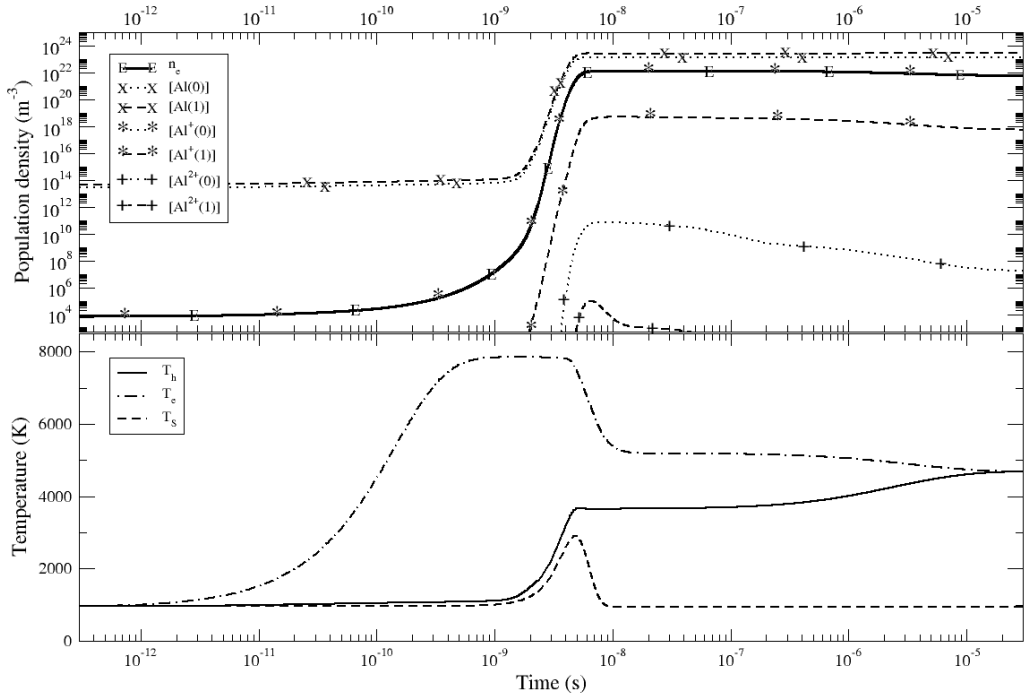

Fig. 1: Population density (up) and temperatures (down) of plasma components calculated by the CR model A.

The maximum value of electron density is found to be equal to some 10^{22} m^{-3} at the maximum of the laser pulse. This value is not so different from those reported in the literature [7, 8]. Besides, we can note the short duration needed to form the plasma (some nanoseconds). We can also note the persistence of the thermal non equilibrium long after the pulse ($T_e > T_h$ at the bottom of Fig.1) which indicates that for LIBS diagnostic in vacuum, the equilibrium is not necessarily fulfilled.

During the conference, we will present the details of the "model B" and some comparisons with the "model A". In particular, the evolution of excitation temperatures and the respective role of the elementary processes on the global behaviour of the plasma will be discussed.

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