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Effect of laser parameters on laser ablation and laser-induced plasma formation: A numerical modeling investigation

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Abstract

A comprehensive numerical model has recently been developed for nanosecond (ns) laser ablation of metallic targets, describing the processes of target heating, melting and vaporization, the resulting plume expansion in 1 atm helium gas, as well as plasma formation in the plume. In the present paper, we investigate the influence of laser parameters, i.e., laser irradiance, pulse duration and wavelength, on typical calculation results, such as the target temperature, melt and evaporation characteristics, the plume expansion velocity, plume (plasma) temperature and ionization degree, densities of neutrals, ions and electrons in the plume, as well as the laser absorption characteristics in the plume (plasma shielding). Comparison is made with experimental data from literature, whenever available, and in general, good agreement is reached between our model predictions and experimental results. Therefore, the model can be useful to predict trends in target and plume (plasma) characteristics, which are difficult to obtain experimentally.

Keywords: Laser ablation; Laser-induced plasma; LIBS; Modeling; Plume expansion; Laser parameters; Laser absorption

1. Introduction

Lasers are widely applied in analytical spectrometry, for the analysis of solid materials. Several analytical techniques make use of the effects of laser–solid interaction, in different regimes of laser irradiance, including matrix assisted laser desorption ionization (MALDI) (e.g., [1]), laser microprobe mass spectrometry (e.g., [2]), laser-induced breakdown spectrometry (LIBS) (e.g., [3–5]), as well as laser ablation (LA) used as sample introduction method for inductively coupled plasma mass spectrometry (LA–ICP-MS) or atomic emission spectrometry (LA–ICP-AES) (e.g, [6–8]).

LIBS and LA for sample introduction in the ICP make use of similar laser operating conditions. Typically, Nd:YAG lasers (with wavelengths ranging from 1064 nm down to the UV range) or excimer lasers (in the UV range) are applied. Typical pulse durations are in the order of several nanoseconds (ns), although there is a trend to scale down to fs-pulse duration, and the laser irradiance varies from 10^8 till above 10^{10} W/cm^2. Generally, the evaporated plume expansion takes place in 1 atm background gas, for instance in He, Ar or air.

Consequently, although LIBS and LA are based on different mechanisms of detection, the processes occurring during and after laser–solid interaction are quite similar. In the case of metallic targets, the laser causes heating of the solid target, followed by melting and evaporation of some of the target material. The evaporated material expands, and because of the high temperature, a plasma is formed in the material plume. This plasma contains electrons, ions, neutral species, as well as excited species. The emission of light originating from the excited species in the plasma constitutes the analytical signal measured by LIBS. During the plume expansion, the temperature drops down, and eventually, the evaporated atoms will undergo condensation, resulting in the formation of nm-sized particles. Besides, also larger (μm-sized) particles can be formed, e.g., by

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splashing of the molten target, due to the recoil pressure of the material plume. In the case of LA as sample introduction method for the ICP, these particulates are transported, in the form of aerosols, from the laser plume into the ICP, where they will be vaporized (atomized) again, as well as ionized and/or excited, and subsequently measured with mass spectrometry or atomic emission spectrometry.

Recently, we have developed a comprehensive numerical model, which describes the processes occurring during the laser ablation of metallic targets, and the subsequent expansion of the evaporated material plume, as well as plasma formation. The original model was developed for expansion in vacuum [9,10], and was subsequently extended for expansion in 1 atm He gas, which appeared to be a much more complicated task [11,12]. The model does not yet include mechanisms for particle formation, but this task is planned for the next stage.

It is worth to mention that there exist a variety of models in literature for laser ablation under vacuum (or low pressure, i.e., up to 100 Pa) conditions (for a comprehensive literature overview, see Ref. [10]). However, the number of models that have been developed for laser ablation under 1 atm background gas pressure, is very limited, and these model investigations [13–17] often apply the hydrodynamic equations only for the vapor species, like in vacuum conditions, whereas it is obvious that a binary gas mixture (i.e., metal vapor and background gas), as well as the interactions between vapor and gas, need to be considered. A hydrodynamic model, which describes the behavior of both vapor and background gas, has been developed by Gnedovets and Gusarov, and it is applied to expansion in a background gas at 1 atm, but for a long laser pulse (ms-range) at very low laser irradiance (i.e., 10^4–10^5 W/cm^2), so that no plasma is formed [18–20]. Recently, Gusarov and Smurov have applied this model to shorter laser pulses (ns-range) at a laser irradiance in the order of 10^9 W/cm^2, hence conditions typical for LA and LIBS, but without taking into account the formation of a plasma [21]. It is, however, clear that plasma formation is important at these conditions. To our knowledge, there exist no models yet that describe the laser ablation with expansion in 1 atm background gas, including the plasma formation, except for the model developed in our group [11,12].

In the present paper, we have applied our model to a wide range of laser operating conditions, i.e., laser irradiance, pulse duration and wavelength, which are typical for LIBS and LA as sample introduction method for the ICP.

There are many experimental studies reported in the literature, about the effect of laser parameters on the evaporation process, plume and plasma characteristics, and analytical performance of LIBS, LA–ICP-MS and LA–ICP-AES. Russo and co-workers [22–24] have investigated the effect of laser irradiance on electron density and plume temperature, for a Nd:YAG laser at 266 nm and glass or Si samples, in the irradiance range of 10^9–8×10^{10} W/cm^2, and they have derived a simple scaling law, with a different behavior below or above 2×10^{10} W/cm^2. In Ref. [25], the effect of laser irradiance, in the range of 1.2–3×10^8 W/cm^2, on the electron density is reported, but for plume expansion in vacuum. Russo's group has also studied the effect of laser irradiance on the mass ablation rate of Cu [26], and they found a different power law, for a laser irradiance below or above 3×10^8 W/cm^2, attributed to plasma shielding. Moreover, still a different irradiance behavior was observed at a laser irradiance above 10^{10} W/cm^2, probably due to self-focusing or phase explosion [24,27]. Another scaling law, for the relation between mass ablation rate on one hand, and laser irradiance and wavelength on the other hand, was presented in Ref. [28]. The effect of laser fluence on the ablation rate was also reported by Horn et al. [29], up to a fluence of about 45 J/cm^2, for a Nd:YAG laser at 266 nm and an excimer laser at 193 nm, in He or Ar background gas, and a range of different sample materials, and it was found that the ablation rate increases linearly with fluence [29]. The influence of laser irradiance on the temporal evolution of the plasma is studied by Laserna et al. with a dynamic microphone, for a Nd:YAG laser at 1064 nm, in the irradiance range of 10^9–10^{10} W/cm^2, and it is observed that a higher laser irradiance yields larger plasma volumes [30]. Shannon et al. and Chan and Russo [26,31] have described the effect of laser irradiance on the ICP emission intensity, with an excimer laser at 248 nm and a Nd:YAG laser at 266 nm, for several different sample types, and laser irradiance values ranging from 10^7 W/cm^2 till above 10^{10} W/cm^2. It is found that the signal intensity as a function of laser irradiance exhibits two different slopes, with a steeper slope below a certain irradiance value, and a smaller slope above this value [26,31]. The value of this irradiance depends on the laser pulse duration, i.e., it is higher for ps than for ns pulses. This roll-off behavior is again attributed to plasma shielding (see above) [26,31]. The same authors have also reported the effect of laser irradiance on the measured particle size distributions [32] and on elemental fractionation [33]. The fraction of larger particles seems to decrease as a function of laser irradiance, up to a value of 4–5×10^8 W/cm^2, which is found to be similar to the threshold value for plasma shielding [32]. Above this value, the particle size distribution remains more or less unchanged, but the total number of particles keeps increasing [32]. Furthermore, it is observed that fractionation occurs for a laser irradiance below 6×10^8 W/cm^2, and above 2×10^{10} W/cm^2, but it is found to be unimportant in the region in between [33]. In Ref. [34] it is shown that the lifetime of clusters in the plasma is reduced at higher laser irradiance. Finally, Sdorra and Niemax have studied the effect of laser energy, in the range of 2–20 mJ/pulse, for a Nd:YAG laser of 1064 nm and five different background gases, and it was reported that the measured plasma temperature and the emission intensity in the laser induced plasma generally increase with laser energy [35].