Combined continuous–microscopic modeling of laser plume expansion

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Abstract

A hybrid model is developed to study the dynamics of laser plume expansion in vacuum or into a background gas. The method takes advantages of both continuous and microscopic simulation approaches. As a result of the combination of two different numerical methods, such as, large particles and direct Monte Carlo simulation, the model describes high-rate laser ablation for a wide range of background pressures (from zero to hundreds Pa). The model is used to investigate laser plume interaction with background gases. Particularly, the plume–gas mixing and energy exchange are taken into account. The dynamics of the laser plume expansion is investigated. Snowplow effect is observed at sufficiently high pressures. At smaller pressures, strong plume–gas mixing takes place near the contact surface. The simulation results explain experimentally obtained spatial maps of the reaction products formed during the plume expansion into a reactive background gas.

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1. Introduction

The process of laser ablation is widely used for surface treatment, laser cleaning and thin film deposition [1]. For further development of these fields, one needs a better understanding of basic ablation mechanisms. A large variety of both experimental and theoretical studies already clarified many aspects of the ablation process. The complexity of the involved physical phenomena, however, still does not allow its complete theoretical description.

To model laser plume expansion, two basic approaches have been used. In the first or macroscopic approach, the plume is described by a system of gas–dynamic equations that may be solved by using different analytical and numerical methods [2,3]. In the second, or microscopic approach, the plume and background gas are represented by a number of atoms or molecules, and the time evolution of the flow is calculated by using a Direct Simulation Monte Carlo (DSMC) method [4,5]. Both of these approaches have their advantages and limitations. For example, gas–dynamic description, which is based on the assumption of the Maxwellian velocity distribution and frequent collisions, is well suitable for rather dense plumes. On the contrary, Monte Carlo approach is preferable for the transitional and rarefied flows and...
for regions with non-Maxwellian velocity distributions. In addition, mass diffusion and heat exchange, which make gas–dynamic approaches more complicated, are naturally treated by DSMC.

Herein, we present a new multi-scale model based on a combination of the macroscopic and the microscopic approaches. The former approach is used for the expansion of a dense newly formed plume, whereas the latter is applied in the subsequent calculations. As a result of the combination, the model describes high-rate laser ablation for a sufficiently long time and a wide range of the background pressure. Furthermore, the developed model provides an adequate description of the plume–background gas interaction and mixing. The calculated plume evolution is first compared with the experimental results obtained in the presence of inert gases. The calculation results are furthermore used for the interpretation of the experiments performed in the presence of a reactive gas.

2. Model

At the very beginning of the plume development and expansion, the plume–background gas diffusion is negligible, and energy exchange can be disregarded [6]. At this initial stage, numerical gas dynamics is well suitable for the plume expansion. Later, the mass diffusion and the energy exchange are taken into account by using the direct Monte Carlo simulation. The developed model is three-dimensional and uses axial symmetry. We assume that a laser beam with radius $R_0$ and pulse duration $\tau$ interacts with a plane target ($z \leq 0$). The half-space $z > 0$ is filled by a background gas (pressure $P_b$, density $\rho_b$).

2.1. Gas–dynamic part

At the beginning, we consider the plasma plume as a non-viscose and non-heat-conductive vapor containing atoms, ions and electrons. We use a one-fluid two-temperature approximation [7–9], which is based on the solution of the following system

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \vec{V}) = 0, \quad (1)$$

$$\frac{\partial \rho U}{\partial t} + \text{div}(\rho U \vec{W}) + \frac{\partial P}{\partial z} = 0, \quad (2)$$

$$\frac{\partial \rho V}{\partial t} + \text{div}(\rho V \vec{W}) + \frac{\partial P}{\partial r} = 0, \quad (3)$$

$$\frac{\partial \rho E_a}{\partial t} + \text{div}(\rho E_a \vec{W}) + \text{div}(P_a \vec{W}) = Q_{ei} + Q_{ea}, \quad (4)$$

$$\frac{\partial \rho E}{\partial t} + \text{div}(\rho E \vec{W}) + \text{div}(P \vec{W}) = -F, \quad (5)$$

$$E = e_a + e_e + \frac{\|\vec{W}\|^2}{2}, \quad E_a = e_a + \frac{\|\vec{W}\|^2}{2}, \quad (6)$$

$$P = P_a + P_e, \quad P_a = (\gamma - 1)\rho e_a, \quad P_e = (\gamma - 1)\rho e_e, \quad (7)$$

where $\rho$ is the density, $U$ and $V$ are $Z$ and $R$ components of the velocity vector $\vec{W}$; $\gamma$ the specific heat ratio; $e_a$ is the thermal energy of ions and neutrals; $e_e$ the thermal energy of electron gas; $P_a$ and $P_e$ the partial pressures of the heavy (ions and neutrals) and light (electrons) gases. In Eqs. (4) and (5), $Q_{ei}$ and $Q_{ea}$ describe the energy exchange between the electron gas and the gas composed of ions and neutrals, and $F$ is the energy lost by electrons for ionization. The system is solved numerically by using the method of large particles (LP) [8]. For simplicity, we use the equilibrium ionization–recombination kinetics for only singly charged ions and neglect photo-processes. Initial boundary conditions at the surface $z = 0$ are calculated as a heat source resulting from the absorption of the laser radiation corresponding to a laser pulse with a Gaussian temporal shape and pulse duration $\tau$. The temperature distribution along the laser-irradiated area is assumed to be uniform. The initial flow parameters at $z = 0$ and $r < R_0$ are calculated using the jump conditions at the Knudsen layer boundary [10]. The contact boundary is determined using the mass percentage procedure described in [7]. The large particle calculations are performed until the time $t_0 \sim 10\tau$. The switch time $t_0$ is chosen so that it is long enough for the plume density to diminish by several orders of magnitude and to account for the ionization–recombination processes in the plasma plume. This time is, however, short enough so that the mass diffusion and the heat exchange between the plume and the background gas are insignificant at $t < t_0$.

2.2. Monte Carlo part

At $t > t_0$, the laser plume and the background gas are modeled by an ensemble of simulated particles
(about $10^6$). These simulated particles are introduced based on the LP gas–dynamic parameters obtained at $t = t_0$. The physical space is divided into a network of cells with dimensions $L_c$ smaller than the local mean free path. Time is incremented by a time step, which is about $L_c/v$, where $v$ is the mean velocity of the particles in the cell. The calculations are continued by using a typical DSMC procedure, which consists in the repetition of two steps: (i) the calculation of a representative set of collisions in each cell with a real frequency; and (ii) the movement of the simulated particles [4,5]. In these calculations, the LP solution of (1)–(7) at $t = t_0$ serves as initial state for the Monte Carlo stage, in which both the cell size and the time step are small enough, so that no discontinuity takes place during the switch from the LP part to the DSMC part.

### 3. Experiments

The experimental results presented in this paper have been obtained by using two different techniques, time-of-flight (TOF) and laser-induced fluorescence (LIF). Two different laser set-ups have been used for material ablation in these techniques. Thus, the TOF results for Ge target are obtained using an ArF laser with 193 nm wavelength and 15 ns pulse duration (FWHM). The LIF results for AlO are obtained using Nd:YAG laser (266 nm) with 5 ns pulse. More details about experimental procedure can be found elsewhere [11,12].

### 4. Results and discussion

The developed model has been used for a series of calculations of laser plume expansion in the presence of a background gas. Hereafter, we present the simulation results obtained for laser ablation of solid targets (Al and Ge) in the presence of several background gases.

First, let us consider the calculated positions of the plume front and of the plume central density maximum as a function of time (Fig. 1). The simulation dependencies obtained for Ge in the presence of Ar agree with the experimental results. Both the simulation and the experimental results show that the increase in the background pressure leads to the deceleration of the plume.

If the background pressure is high enough, a snowplow effect can be observed in the two-dimensional contour maps of the plume and gas densities (Fig. 2). This effect implies the compression of the background gas in front of the expanding plume (Fig. 3a). In addition, the plume front also becomes compressed (Fig. 3b). The calculation results also clearly demonstrate an intense mixing of the plume and gas species.

Collective effects, such as plume and gas compression, take place if the frequency of collisions in the plume is still high when the plume dimensions become comparable to the mean free path in the background gas. Assuming radial plume expansion, the required number density of the background gas $n_b \geq \Sigma_{b}^{-1}(n^*/N)^{1/3}$, where $N$ is a total number of the ablated species; $\Sigma_{b}$ is the collision cross-section in the background gas; and $n^*$ is the plume number density. Taking $n^* \sim 10^{17}$ cm$^{-3}$, $N \sim 10^{16}$, and $\Sigma_{b} \sim 10^{-15}$ cm$^2$ yields $n_b \sim 2 \times 10^{15}$ cm$^{-3}$. If, furthermore, the background gas is at room temperature, the required pressure $P_b^* = n_b k T_b$ is higher than about 10 Pa, where $k$ is the Boltzmann’s constant. More precisely, conditions that are needed for the plume snowplowing can be obtained from our calculations by gradually increasing the background pressure [13].

At small laser fluences, plume expansion is under-sonic, so that it is accompanied by shock-wave
formation. Monte Carlo simulation, which easily treats diffusion, is well suitable for this case. If, furthermore, laser fluence several times exceeds the ablation threshold, the laser plume expansion is super-sonic, and two shock waves are formed. One of them (external) propagates in the background gas, whereas the other (internal) moves backward inside the plume [14–18]. The proposed hybrid model is still valid in this case. Alternative gas–dynamic description including a turbulent stage of Rayleigh Taylor instability [18,19] is also relevant if the sizes of all shock waves are much larger than corresponding local mean free paths.

At small pressures, the plume–gas mixing is stronger than in the snowplow mode. This plume expansion mode with an enhanced plume–gas mixing is preferable for the reactive laser ablation. Typical background pressures used to produce molecules by laser ablation depend on the molecular weigh of the background gas. For aluminum ablation in oxygen, these pressures range from 10 to 30 Pa [12]. This regime, which was particularly difficult for previous gas–dynamic simulations, is considered below.

The spatial maps of the ground-state AlO molecules formed as a result of the reactive laser ablation are shown in Fig. 4. Interestingly, the distributions obtained at oxygen pressure of 13 Pa show two maxima at the plume periphery. The observation of these maxima cannot be explained by the plume snowplow. Vortices, as a viscous effect, would hardly appear at such small background pressures. In fact, the mean free path of the gas molecules is on the order of plume dimensions at the pressures used in these experiments.

To explain the experimental distributions, we focus at the regions with stronger reactivity. Intuitively, without considering details of the reaction kinetics and their temperature dependencies, the reactive rates are higher in the regions with stronger mixing of the reaction components. Thus, the formation of the AlO molecules more probably occurs in the regions where both aluminum and oxygen are present. To obtain an approximate picture of these regions, we plot the
product of the number densities of aluminum and oxygen (Fig. 4a). The plots obtained at the same pressures as the experimental results show a surprising similarity with the experiments (Fig. 4b), with the two maxima at the plume periphery. Based on the comparison of these distributions with the velocity flow-fields, we can attribute the maximum, which is closer to the target, to the back-scattering of the ablated and background species. The second maximum can be explained by the radial expansion of the plume that starts after the plume experiences the presence of the gas.

5. Summary

To summarize, we have presented a model that allows the calculation of laser plume expansion for a wide range of laser fluences and background pressures. This model treats the plume interaction with the background gas, taking into account the mixing and energy exchange of the plume and gas species. Particularly, calculation results have explained the experimental results obtained during reactive laser ablation. The regions with stronger mixing of the reaction components have been shown to correspond to the regions where more molecules are observed in the experiments.

Laser plume expansion into several background gases has been modeled. Calculation dependencies agree with the experimental results. The performed simulation has visualized plume snowplow at larger pressures. This effect implies the formation of two compressed layers, one being at the plume edge and the other one appearing in the ambient gas in front of the plume. At smaller pressures, strong plume–gas mixing takes place particularly at the plume periphery. The pressure value corresponding to the transition between these regimes depends on the experimental conditions, such as laser parameters, ablated material, and ambient gas. In particular, the simulation shows that, at the same laser fluence, the compression of the plume is more pronounced if laser spot is larger. The calculated pressure values agree with the experimental ones.

The effects of the background gas on the ablated particles are extremely significant during the PLD of thin films. In fact, thin film qualities, such as crystalline or amorphous structure, are strongly affected by the energy of the species arriving at the substrate. The developed approach can be also used to study the formation of nano-particles by laser ablation.

References