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Modeling nanosecond pulsed laser ablation: A focus on temperature dependence of material properties

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Abstract

This paper presents a comprehensive model of nanosecond laser ablation of metals considering vaporization, phase explosion and plasma shielding. In addition, the effect of temperature dependent material properties is also considered. The results are in good agreement at low fluences, while the discrepancy marginally increases at higher fluences. The paper concludes by identifying some key challenges in the modeling of the process, which include, consideration to liquid and vapor ejection during phase explosion and the consequent shielding of the radiation due to the ejected liquid droplets.

Keywords: Laser ablation; Material properties; Vaporization; Phase explosion; Plasma shielding

Laser ablation using nanosecond (ns) pulses has several applications in micro-/nano-manufacturing. However, due to the complexities involved in the process, there is still a large scope in the modeling of the process. The process involves various phenomena such as target heating, material removal and plasma shielding. The absorption of laser radiation heats the target. Material removal in laser ablation using nanosecond pulses occurs mainly by vaporization and phase explosion. Phase explosion occurs only at fluences that can heat the target close to its critical temperature (T_c). Phase explosion occurs due to homogeneous nucleation of vapor bubbles that results in ejection of both liquid droplets and vapor. As the material is ejected, it interacts with the incoming laser radiation and absorbs a part of it, this is also called as plasma shielding.

The temperature of the target during laser ablation goes much beyond the normal boiling temperature. Consequently, the material properties also vary significantly. Therefore, the modeling of the process should necessarily involve consideration to temperature dependent material properties.

While there are a number of models that are focussed on vaporization and subsequent plasma formation, phase explosion has received very less attention [1,2]. This is mainly because of the unavailability of properties of metals above 4000 K due to the experimental challenges involved. Despite a wide variation in temperature during laser ablation, there exist a number of models that are based on constant values of material properties. Consideration to temperature dependence of optical properties has been attempted by a few researchers, notably, Gragossian *et al.* [3] using Fresnel equations along with Drude model. However, there exists a significant gap in the present knowledge. These include: unavailability of temperature dependent material properties at elevated temperatures and absence of a comprehensive model of laser ablation involving vaporization, phase explosion and plasma shielding. In this work, the material properties have been estimated using some general and empirical theories and Al has been used as the target material. In Section 1, the estimation of the temperature dependent material properties based on some general/empirical theories is discussed. This

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is followed by a description of the laser ablation model in Section 2. In Section 3, the results based on the model are presented. Finally, Section 4 summarizes the conclusions drawn based on the work.

1. Evaluation of material properties

The optical properties govern the amount of laser energy deposited and absorbed by the target, whereas, the thermophysical properties govern the temperature rise in the target. In addition, other properties such as latent heat of vaporization (ΔH_v), saturation vapor pressure (P_s) and surface tension (γ) govern vaporization and explosive boiling, which are the material removal mechanisms in laser ablation.

In this work, the critical temperature and density are taken as 8860 K and 280 kg/m^3 [4]. The solid density has been assumed to be constant ($\rho_m = 2700$ kg/m^3). In the liquid state, the density has been estimated by a modified Guggenheim formula [5] and the coefficients are obtained by fitting the experimental data of Gathers [6], available up to 4000 K. Specific heat is estimated using a scaling law [7], $c_P = c_{p0}[(T - T_m)/(T_c - T)]^c$ and the coefficients are obtained by fitting the curve to the experimental data [6]. The Watson equation is used to evaluate the latent heat of vaporization [8]. The saturated vapor pressure is calculated by solving the Clausius-Clapeyron equation, whereas the surface tension is evaluated using the modified Van der Waals equation [9]. The optical properties depend on target material, its temperature and the laser wavelength. They are estimated by accounting for both the interband and the intraband transition and the model developed by Vial et al. [10] is used. In estimating the optical properties, the electrical conductivity data reported by Korobenko et al. [11] is used. Using this electrical conductivity data, thermal conductivity is obtained using the Wiedemann-Frenz law.

2. Laser ablation model

2.1. Laser heating process

In this model, laser energy is absorbed in each layer leading to volumetric heat generation and a 1-D Fourier heat conduction equation is used to model the heat transfer process:

$$\rho c_p \left(\frac{\partial T}{\partial t}\right) = \nabla \cdot (K \nabla T) + \dot{q} \tag{1}$$

where, ρ is the density, c_p is the heat capacity, K is the thermal conductivity and $\dot{q}$ is the laser heat source term given by,

$$\dot{q}(x) = (1 - R)\alpha(x) I_L \exp\left(\int_0^x -\alpha(x) dx\right) \tag{2}$$

where, R is surface reflectivity, α is absorption coefficient and I_L is laser intensity. The temporal variation of the laser intensity profile (I_L) is described by a Gaussian profile with full width at half maximum (FWHM).

2.2. Material removal

Material removal is considered to occur by vaporization and phase explosion. Vaporization occurs at all temperatures and the surface recession due to vaporization is obtained using Knight's theory of vaporization [12]. Phase explosion is accounted by evaluating the homogeneous nucleation rate per unit volume using Carey's model [13], which is based on the classical nucleation theory In this work, as per the calculations for liquid Al, homogeneous nucleation rate is insignificant below a threshold temperature of $0.86T_c$ and reaches a very high value of 10^{38}m^{-3}s^{-1} at $0.9T_c$. Due to a very high nucleation rate at and above $0.9T_c$, it is assumed that the liquid layer instantaneously breaks down into vapor and liquid droplets and gets ejected out of the target. Therefore, in this work, phase explosion is assumed to occur as the target layer reaches $0.9T_c$. Ablation depth due to phase explosion is estimated using the width of the target layer reaching $0.9T_c$. The total ablation depth is estimated due to both vaporization and phase explosion.

2.3. Plasma shielding effect

Plasma shielding causes a drop in the laser intensity reaching the target surface. Radiation absorption by plasma occurs by Inverse Bremsstrahlung (IB) and photo-ionization(PI) processes [14]. The Plasma shielding effect is calculated in terms of the reduced laser intensity (I_{sh}) reaching the target surface, given by,

$$I_{sh} = I_L \exp\left(-\int_0^{h_p} (\alpha_{ib} + \alpha_{pi}) dx\right), \tag{3}$$

where, α_{ib} and α_{pi} are the absorption coefficients corresponding to IB and PI processes, respectively. The plasma height, h_{pl} is calculated by assuming that the plasma expands at a constant velocity equal to the acoustic velocity.

2.4. Numerical model

A numerical scheme based on finite volume method (FVM) is employed to solve the modeling equations as discussed in our earlier work [15]. The target is divided into a number of layers along the depth and each layer is considered as a finite volume with a unit crossectional area. The heat transfer equation (Eq. 1) is discretized using the Crank-Nicholson scheme and solved using Thomas algorithm.

3. Results and discussion

3.1. Variation in material properties

The results of the model presented in Section 2 are discussed in this section. The variation in material properties during the laser ablation process are shown in Fig. 1 and