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Simulation of phase explosion in the nanosecond laser ablation of aluminum

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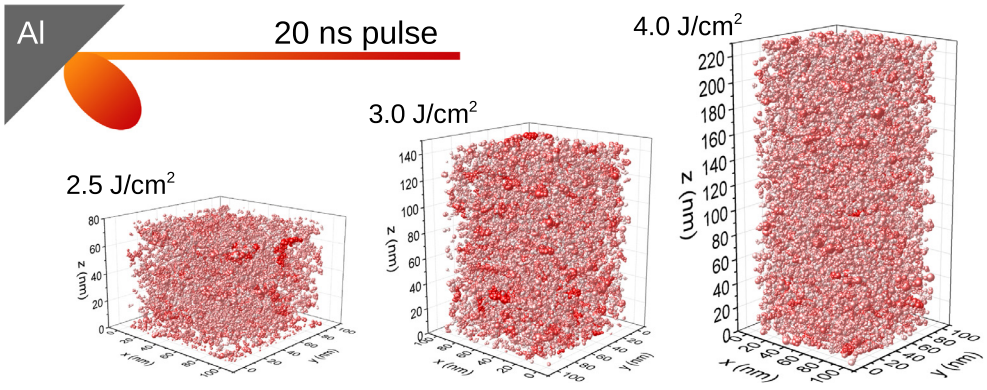
GRAPHICAL ABSTRACT



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ABSTRACT

Hypothesis: Vaporization, spallation and phase explosion are considered to be the main mechanisms contributing to the nanosecond laser ablation of metals. The theory of homogeneous nucleation, together with the dynamics of target heating, allows a space-time resolved simulation of the phase explosion mechanism.

Methods: The thermal phenomena occurring at the target surface are studied within the framework of a thermodynamic continuum approach. A 20 ns laser pulse of variable fluence and Gaussian time dependence was assumed. The temperature profile in the target external layers is studied through the heat diffusion equation. The vaporization from the surface is modeled assuming unsteady adiabatic expansion (UAE) of the vapor and a Monte Carlo (MC) method is used to describe the formation of liquid nanodroplets through phase explosion.

Results: Liquid nanodroplets in the ablated material are studied at different laser fluences. The size distribution of the nanodroplets formed in the phase explosion process is here reported and connections with experiments are discussed.

1. Introduction

The pulsed laser ablation of metals in vacuum allows the synthesis and deposition of small size nanoparticles (NPs) with peculiar positive features compared with conventional techniques, especially the good adhesion of highly nanostructured coatings on a large variety of substrates.

The material ejection in the nanosecond laser ablation of pure metals proceeds through thermal mechanisms, mainly vaporization, spallation and phase explosion [1,2], followed by some slower

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Table 1
List of symbols and abbreviations.

k_B	Boltzmann constant	FWHM	Full width half maximum
γ	Heat capacity ratio	KL	Knudsen layer
m	Atomic mass	UAE	Unsteady adiabatic expansion
α	Thermal diffusivity	FTCS	Forward time centered space
η	Relative volume fraction	MC	Monte Carlo
F	Laser fluence	NP	Nanoparticle

processes leading to the expulsion of large liquid clusters, usually observed in fast imaging experiments [3].

The complexity of the phenomena involved in the laser ablation of metallic targets reflects in some undesired effects in the deposited materials, such as a large dispersion in the size distribution of the deposited NPs.

In this context, the connection and comparison between experimental observations and theoretical models find both fundamental interest to clarify the physics of laser ablation and also deep implications in the new applications of high power laser pulses.

In the last years we studied the formation of liquid nanodroplets through the phase explosion mechanism, induced by the nanosecond laser irradiation of pure metals. In particular our work was based on the continuum description of the target material, following a thermodynamic study of the metastable liquid metals at temperature close to the thermodynamic critical point T_c.

First, we tried to directly connect the theory of homogeneous nucleation to the size distribution of the liquid nanodroplets formed in the phase explosion process [4], through a simplified computational method which assumed a constant temperature of the target. More recently we extended our simulations to include a linear time dependence of the target temperature [5].

In this paper we improve our computational scheme by considering a 20 ns full width half maximum (FWHM, see Table 1 for the list of symbols and abbreviations) laser pulse irradiating an aluminum bulk target and we numerically treat the heat diffusion problem in the target external layers. The target is spatially discretized so that ablation is modeled by the progressive removal of discrete layers.

The present improved computational approach allows us to describe the formation of liquid nanodroplets through the phase explosion mechanism, with temporal and spatial resolution. The phase composition of the sputtered material is studied as a function of time during a laser pulse and the size distribution of the produced aluminum nanodroplets is reported.

2. Theoretical aspects and computational methods

The main thermodynamic quantities of liquid aluminum in the deep metastable regime were estimated by using semi-empirical scaling laws consistent with the theory of critical exponents. In particular, to determine the thermodynamic critical point of aluminum, we adopted the method proposed by Blairs et al. [6] based on the experimental measurements of density and surface tension of the liquid metal at the melting point.

The temperature dependence of liquid and vapor density was derived from the coexistence curve analogously to our recent study [4]. Since both vaporization and phase explosion involve a liquid-vapor phase transition, a key quantity to be evaluated in our model is the enthalpy of vaporization, which was approximated by the Watson scaling law [7].

The saturated vapor pressure $p_s(T)$, describing the binodal curve of equilibrium phase change, was determined by integrating the Clausius-Clapeyron equation.

We adopted the empirical Guggenheim formula [8] for the temperature dependence of the surface tension, by using the experimental data provided by the comprehensive review of Keene [9].

2.1. Vaporization

The vaporization process was modeled according to the work of Kelly [10], by considering the evolution of the vaporized material from the Knudsen layer (KL), where the vapor atoms thermalize, to the UAE that leads the sputtered material away from the target. That method assumes the flow velocity at the boundary of the KL to be equal to the speed of sound:

$$u_K \approx \left(\frac{\gamma k_B T_K}{m}\right)^{1/2} = \left(\frac{5 k_B T_K}{3m}\right)^{1/2}, \tag{1}$$

where the subscript K refers to the KL outer boundary and the heat capacity ratio is assumed to be $\gamma = 5/3$. Moreover, the temperature and density jump across the KL is given by:

$$T_K = 0.669 T_s, \quad \rho_K = 0.308 \rho_s, \tag{2}$$

where T_s is the liquid surface temperature while ρ_s indicates the saturated vapor density at T_s. This model of vaporization is consistent with the numerical results of Knight [11] and allows an estimate of the surface recession velocity of the liquid surface u_l by simply applying the conservation relation $\rho_l u_l = \rho_K u_K$.

Finally, we assumed the recoil pressure on the liquid exerted by the vaporized material to be $p_l = 0.55 p_s(T_s)$ in the case of intense vaporization [11,12].

2.2. Phase explosion

When the target surface is irradiated by a high power nanosecond laser pulse, the external layers undergo an extremely rapid temperature increase, the nanosecond timescale allowing the lattice thermalization. In the fast heating of pure metals it has been proved that the normal boiling process due to the heterogeneous nucleation of vapor bubbles has a negligible kinetics [13]. Thus, the liquid phase becomes metastable, overcoming the boiling temperature, and it undergoes homogeneous vapor bubble nucleation when the temperature approaches the spinodal line, at approximately $0.9 T_c$.

The vapor pressure in the nucleated vapor bubbles p_v was calculated through the classical theory of homogeneous nucleation as reported by Debenedetti [14]:

$$p_v = p_s \exp\left[(p_l - p_s)\frac{m}{\rho_l k_B T}\right]. \tag{3}$$

In this work, we considered the classical nucleation theory as in our previous study [4], so that the size of spherical vapor bubbles which are in labile equilibrium with the metastable liquid is expressed by the critical radius:

$$r_c = \frac{\sigma}{p_v - p_l}, \tag{4}$$

where σ is the surface tension. Here we assumed the homogeneous nucleation process to be regulated by the steady-state nucleation rate, thus neglecting the time lag of homogeneous nucleation τ_{lag}. This simplification is consistent with our recent result, that showed that no significant nucleation time delay is expected when the heating rate is lower than 10^{12} K/s [5], as it is in our present simulations.

We adopted the same model reported in [5] to study the single vapor expansion dynamics in the metastable liquid, derived from the original method proposed by Lee et al. [15]. In brief, since the critical radius of vapor bubbles monotonically decreases with the temperature, during the heating process a nucleated vapor bubble spontaneously becomes unstable and starts growing.

As in our previous works, here we assumed as the phase explosion condition the reaching of a volume fraction occupied by vapor