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Thermal ablation of metal films by femtosecond laser bursts

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ABSTRACT

Thermal ablations of metal films by a single femtosecond laser pulse and by a laser burst were investigated using a comprehensive computational model, including a two-temperature model with dynamic optical properties, two phase change models for melting and evaporation under superheating, and a phase explosion criterion for ejection of the mixture of metastable liquid droplets and vapor. Numerical simulations were performed for copper films. The simulation of material ablation by the single pulse showed a good agreement with existing experimental data for a broad range of laser fluence 0.6–30 J/cm^2. In the study of material ablation by femtosecond laser bursts, it was found that under the same total fluence a laser burst with a pulse separation time of 50 ps or longer can significantly boost the ablation rate, compared to the single pulse. It was also demonstrated that for a given total fluence the ablation rate can be optimized by selecting a proper combination of pulse number and separation time in a laser burst.

1. Introduction

Due to the unique characteristics of material processing, interactions of ultrashort-pulsed laser with metals have received a great deal of attention in the past two decades [1–3]. It has been demonstrated that femtosecond (fs) laser material processing is a very effective means for precise micro/nano machining and surface structure modification of solid materials due to minimal heat affected zone and collateral damage, less debris contamination, and good reproducibility [4–6]. To date, it has become possible to generate any pulse shapes with recently developed optical devices. For example, a pulse train can consist of many bursts with a repetition rate from kHz to MHz, in which each burst consists of several pulses at a separation time of several tens of femtosecond to hundreds of picosecond (ps). The recent advancement of laser burst technology may provide another powerful means for ultrashort-pulsed laser applications.

In theoretical studies, two-temperature models (TTMs) [7–9] have been employed to simulate thermal problems for metals irradiated by ultrashort-pulsed lasers. For the fs-laser pulse train heating, Huang et al. [10] studied energy transport and melting process in a gold film. Temperature-dependent thermo-physical properties were employed; but, constant optical properties were assumed. No material ablation was studied. Jiang and Tsai [11] and later Sim et al. [12] employed the Drude model to calculate the dynamic changes of optical properties for predicting laser induced thermal response in metal films. Nonetheless, the calculated optical properties may not be accurate because the Drude model generally describes a higher reflectivity for metals. Chowdhury et al. [13] and Du et al. [14] respectively used constant optical properties and thermophysical properties which are valid for relatively low temperature to simulate thermal transport in gold films induced by multiple fs-laser pulses. None of the above works [11–14] investigated ultrafast melting/re-solidification and material ablation.

Besides the models themselves, thermophysical and optical properties of materials are a key factor that governs the solution accuracy in modeling ultrafast laser material interactions [15–17]. The former controls thermal transport and temperature distribution in a heated medium, while the latter dictates laser energy deposition which can affect the resulting thermal response. Of particular importance, the dynamic change in surface reflectivity (R) and absorption coefficient (α) inside a treated material due to elevated temperature could significantly alter the magnitude and distribution of laser energy deposition, leading to a different thermal response than that described with constant optical properties at room temperature. This may be the critical question which often does not allow directly comparing experimental and modeling data. To match experimental results, particular values for a constant surface reflectivity [18,19] or an absorbed laser fluence [20] together with a constant absorption coefficient at room

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Nomenclature

A_e	material constants for electron relaxation time ($K^{-2}\ s^{-1}$)
B_p	weighing factor of Lorentz oscillators
C	heat capacity ($J/m^3\ K$)
c	speed of light in vacuum (m/s)
f_1	normal refractive index
f_2	extinction coefficient
f_{rep}	repetition rate of pulse bursts (Hz)
G	electron–lattice coupling factor ($W/m^3\ K$)
$\hbar$	reduced Plank constant (J s)
$h_{\ell v}$	latent heat of evaporation (J/kg)
h_m	latent heat of fusion (J/kg)
J	laser fluence (J/m^2)
k	thermal conductivity (W/m K)
k_B	Boltzmann constant ($m^2\ kg/s^2\ K$)
L	film thickness (m)
m	electron mass (kg)
m_{opt}	effective electron mass (kg)
q	phonon wave vector (m^{-1})
R	reflectivity
R_g	specific gas constant (J/kg K)
s	longitudinal sound speed (m/s)
S	laser heat density (W/m^3)
t	time (s)
t_p	FWHM pulse width (s)
t_{sep}	separation time of pulses (s)
T	temperature (K)
T_m	normal melting point (K)
u	interfacial velocity (m/s)
u_0	maximum solid–liquid interfacial velocity (m/s)
v_F	Fermi velocity (m/s)
x	coordinate (m)

Greek symbols

α	absorption coefficient (m^{-1})
γ	total electron collision rate (1/s)
Γ_p	Lorentz oscillator spectral width (rad/s)
$\bar{\varepsilon}$	emissivity factor
ε	dielectric function
ε_1	real part of dielectric function
ε_2	imaginary part of dielectric function
ε_F	Fermi energy (eV)
$\upsilon_{e,p}$	collision rate between electron and phonon (s^{-1})
ρ	mass density (kg/m^3)
σ	Stefan–Boltzmann constant ($W/m^2\ K^4$)
τ_e	electron relaxation time (s)
ϕ_p	Lorentz oscillator phase
ω	laser frequency (Hz)
ω_D	plasma frequency (Hz)
Ω_p	Lorentz oscillator strength (rad/s)

Subscripts

e	electron
l	lattice
ℓ	liquid
ℓv	liquid–vapor interface
s	solid
sℓ	solid–liquid interface

temperature are frequently assumed in modeling ultrafast laser material ablation. Strictly speaking, they can only be considered as a trial-and-error approach. The dynamic changes in optical properties indeed could play an important role in the laser energy absorption, particularly for metal films heated by fs-laser bursts due to the incubation effect from the repeated laser pulse heating.

In this paper, thermal ablations of metal films by a single fs-laser pulse and by a laser burst are investigated using a comprehensive computational model, including a TTM with dynamic optical properties, two phase change models for melting and evaporation under superheating, and a phase explosion criterion for ejection of the mixture of metastable liquid droplets and vapor. A critical point model with three Lorentzian terms demonstrated by Ren et al. [17] is incorporated into the TTM to characterize temperature-dependent optical properties. All the thermophysical properties employed are valid for a wide range of temperature. Numerical analysis is performed for copper films. The material ablation is first studied for a single fs-laser pulse, and the simulated ablation depths are compared with existing experimental data. Then, the burst modes are investigated for their performance of material ablation. The numerical results of thermal response and ablation depth generated by the fs-laser bursts having different pulse number and separation time are presented and discussed.

2. Mathematical models

Consider that a free standing metal film of thickness L and initial temperature T_i is irradiated at its front surface ($x = 0$) by a single laser pulse or by a pulse train. Fig. 1 depicts the structure of a laser pulse train consisting of two bursts with a repetition rate f_{rep}, in which each burst consists of three pulses with a separation time t_{sep}. All the laser pulses are Gaussian in time with a full width at half maximum (FWHM) t_p. For simplicity, the problem is approximated to be one dimensional since laser spot sizes are much larger than the thermally affected depth.

2.1. Two-temperature model

In the TTM for pure metals, heat conduction in lattice is often neglected due to the fact that the thermal conductivity of lattice is much smaller than that of electrons. However, heat conduction in the lattice could also be of importance for precisely describing lattice thermal response, particularly when phase change(s) are considered. In that case, a TTM that includes heat conduction in the lattice [9] should be employed:

$$C_e\frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x}\left(k_e\frac{\partial T_e}{\partial x}\right) - G(T_e - T_l) + S \tag{1}$$

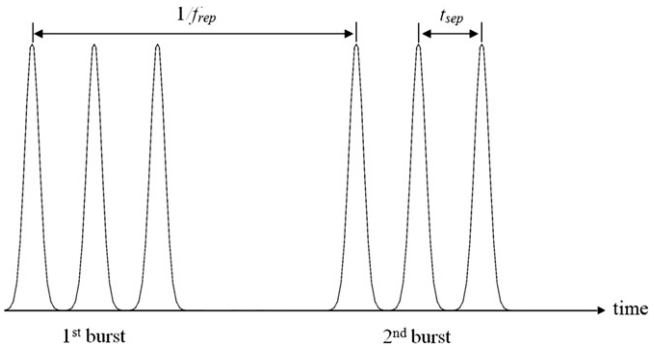

Fig. 1. Illustration of a laser pulse train.