Applied Surface Science 361 (2016) 41–48

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Femtosecond laser ablation of copper at high laser fluence: Modeling and experimental comparison

C.W. Cheng[a,*], S.Y. Wang[b], K.P. Chang[b], J.K. Chen[c]

[a] Department of Mechanical Engineering, National Chiao Tung University, No. 1001, Ta Hsueh Road, Hsinchu 30010, Taiwan
[b] ITRI Southern Campus, Industrial Technology Research Institute, No. 8, Gongyan Rd., Liujia District, Tainan City 734, Taiwan
[c] Department of Mechanical and Aerospace Engineering, University of Missouri, Columbia, MO 65211, USA

ARTICLE INFO

Article history:
Received 13 July 2015
Received in revised form
29 September 2015
Accepted 6 November 2015
Available online 19 November 2015

Keywords:
Femtosecond laser
Two-temperature model
Phase explosion model

ABSTRACT

Thermal ablation of a copper foil surface by a single femtosecond laser pulse of duration 120 fs and wavelength 800 nm was investigated herein both theoretically and experimentally. A 1D two-temperature model with temperature-dependent material properties was considered, including the extended Drude model for dynamic optical properties. The rapid phase change and phase explosion models were incorporated to simulate the material ablation process. The simulated ablation depths agree well with the experimental measurements for the high laser fluence regime ranging from 6.1 to 63.4 J/cm^2.

1. Introduction

Femtosecond lasers have been successfully employed in surface structural modification, drilling and cutting because of the minimal heat affected zone [1] and the unique capability to create quasi-periodic nanostructures by single laser beam irradiation [2–4]; thus, they are considered as a promising tool in precise micro/nano material processing.

Many theoretical and experimental works on ultrashort (pico- or femto-second) laser-material interactions have been reported since the early 1990s. In the theoretical studies on femtosecond laser ablation of copper, most of the works focus on the use of a two-temperature (TTM) model [1,5–7], or a hydrodynamic model [8]. On the other hand, experiments with single- or multi-shot ablation of copper under different laser parameters have also been presented, e.g., laser fluence [1,9–11], incident angle and polarization [12], and pulse duration [13]. Two different ablation regimes are found, and the ablation rates are shown to be dependent on either optical penetration depth or electron heat penetration depth [1].

As most previously-published experimental results for the ablation rate of copper are limited to low laser fluences, i.e. <10 J/cm^2, most theoretical studies have only compared simulation results within this regime of fluence [7,5]. In our previous study, numerical simulation and experimental comparison were investigated for femtosecond laser ablation of copper by multi-pulses (i.e. grooving) with laser fluence above 10 J/cm^2 [14]. However, due to the change in laser intensity distribution over the ablated crater surface of the target as a result of the multi-pulse effect, the discrepancy between simulation and actual experiment was revealed.

In this work, a 1D two-temperature model with temperature-dependent material properties, i.e. dynamic optical properties and thermophysical properties was developed. Together with the rapid phase changes, a phase explosion model for ejecting metastable liquid and vapor was considered. A comparison between the simulation and experimental results for ablation of a copper foil surface by a single pulse femtosecond laser with high laser fluence of 6.1–63.4 J/cm^2 was carried out.

2. Modeling

Considering that a copper foil is normally irradiated by a femtosecond laser pulse on the front surface ($z = 0$), a 1D two-temperature model can be used in this simulation since the laser spot size is often much larger than the thermally-affected depth. The 1D TTM is given as a coupled set of nonlinear differential equations [15,16]:

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

---

[*] Corresponding author. Tel.: +886 3 5712121 55126; fax: +886 3 5720634.
E-mail address: weicheng@nctu.edu.tw (C.W. Cheng).

http://dx.doi.org/10.1016/j.apsusc.2015.11.055

$$C_l\frac{\partial T_l}{\partial t}=\frac{\partial}{\partial z}\left(k_l\frac{\partial T_l}{\partial z}\right)+G(T_e-T_l) \tag{2}$$

where the subscript e and l denote electron and lattice, respectively, C is heat capacity, k is thermal conductivity, G is the electron–phonon coupling factor, S is laser heat density, t is time, and z is distance; the laser beam is propagated along the z-axis. For temperature-dependent optical properties, the heat density S can be expressed as [14]:

$$S(z,t)=0.94\frac{[1-R(0,t)]F_o}{t_p}\frac{1}{\delta(z,t)+\delta_b}\exp\left[-\int_0^z\frac{1}{\delta(z,t)+\delta_b}\mathrm{d}z-2.77\left(\frac{t}{t_p}\right)^2\right] \tag{3}$$

where F_o is the laser peak fluence, $R(0, t)$ is the temperature-dependent surface reflectivity of the material at $z = 0$, $\delta(z, t) = 1/\alpha(z, t)$ is the temperature-dependent optical penetration depth, δ_b is the ballistic electron penetration depth, and t_p is the full width half maximum (FWHM) of the Gaussian temporal pulse. The lasing starts from $t = -2t_p$, reaches its peak power at $t = 0$, and ends at $t = 2t_p$. The laser energy outside this time period is ignored because it is too small to significantly alter the results.

The δ_b is added to take into account the effects of the ballistic motion of photon-excited hot electrons in contributing to penetration depth [6,17]. A constant value of δ_b is often computed based on the assumption that the excited, non-equilibrium electrons could penetrate into the non-excited region at Fermi velocity [18]. In this work, it is determined by $\delta_b = \nu_e \times t_b$, where $t_b = 27$ fs is the Drude relaxation time [19], and ν_e is the electron velocity calculated by:

$$\nu_e=\sqrt{\frac{2k_BT_e}{m_e}} \tag{4}$$

where k_B is the Boltzmann constant, and m_e is the mass of an electron.

The dynamic optical properties of surface reflectivity and absorption coefficient during laser irradiation could significantly alter the irradiated laser energy absorption and influence the distribution of laser heat density, respectively. Recently, an extended Drude model which accurately characterizes the reflectivity and absorption coefficient for gold film at different temperatures was presented [20]. That model is modified here by taking the interband transition effect into account, as shown below:

$$\varepsilon(\omega)=\varepsilon_\infty-\frac{\omega_p^2}{\omega(\omega+i\gamma_D)}-\frac{f\Omega_L^2}{(\omega^2-\Omega_L^2)+iA\gamma_D\omega}=\varepsilon_1+i\varepsilon_2 \tag{5}$$

where ε_∞ is the dielectric constant, ω_p is plasma frequency, ω is laser frequency, γ_D is the damping coefficient which equals the inverse of electron relaxation time (τ_e), Ω_L represents the oscillator strength of the Lorentz oscillators, f is a weighting factor and A is a constant. The following values are optimized for copper: $\varepsilon_\infty = 9.4286$, $\omega_p = 1.3593 \times 10^{16}$ Hz, $\Omega_L = 1.7668 \times 10^{15}$ Hz, $f = 3.6355$ and $A = 44.5275$. The τ_e is expressed in the form:

$$\tau_e=\frac{1}{A_eT_e^2+1.4144\upsilon_{e,p}} \tag{6}$$

where A_e represents the material constants for electron relaxation time, and $\upsilon_{e,p}$ is the electron–phonon collision rate which depends on both electron and lattice temperature. For copper, A_e is 1.75×10^7 K^{-2}/s and $\upsilon_{e,p}$ is calculated by the model proposed in [21]. The details of the above parameters in the modified Drude model for copper can be found in reference [14].

With calculated temperature-dependent ε_1 and ε_2 values from Eq. (5), the normal refractive index (f_1) and extinction coefficient (f_2) can be calculated by:

$$f_1(z,t)=\frac{1}{\sqrt{2}}\left[\left(\varepsilon_1^2+\varepsilon_2^2\right)^{1/2}+\varepsilon_1\right]^{1/2} \tag{7}$$

$$f_2(z,t)=\frac{1}{\sqrt{2}}\left[\left(\varepsilon_1^2+\varepsilon_2^2\right)^{1/2}-\varepsilon_1\right]^{1/2} \tag{8}$$

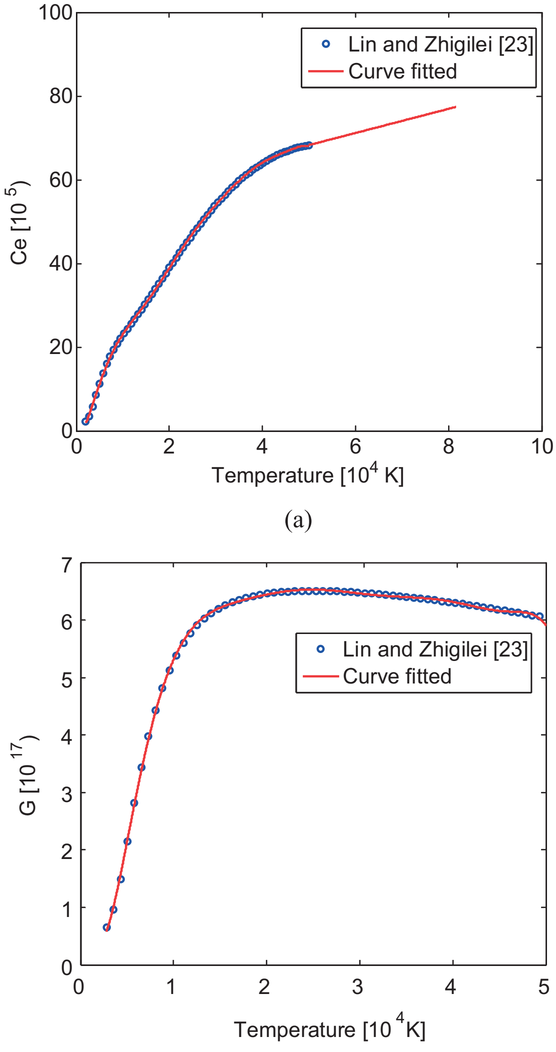
Assume the incidence of the laser beam is normal to the material surface; the surface reflectivity R and absorption coefficient α can be determined by Fresnel functions:

$$R(z,t)=\frac{(f_1-1)^2+f_2^2}{(f_1+1)^2+f_2^2} \tag{9}$$

$$\alpha(z,t)=\frac{2\omega f_2}{c} \tag{10}$$

where c indicates light speed in a vacuum.

The thermophysical properties of Eqs. (1) and (2), C, k, G, control thermal transport and temperature distributions in the laser-irradiated material. In this study, polynomial functions adapted from Lin and Zhigilei [22] are used to describe C_e and G for copper over a range of electron temperature ($\leq 5 \times 10^4$ K). Fig. 1 shows



(a)

(b)

Fig. 1. Temperature dependent (a) C_e [J/m^3/K] and (b) G [W/m^3/K] of copper.