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Green Wavelength Femtosecond Laser Ablated Copper Surface

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Abstract

During green wavelength femtosecond laser ablation, d-band electrons are excited to become free and to participate in the absorption process. The increased electron temperature also induces the density of state shift and causes the gap between the d-band and the Fermi level to expand. The d-band electron transition effect during the laser ablation process causes nonlinear absorption, therefore, it should always be considered during simulations of laser-copper interaction.

This study used a single femtosecond laser pulse with a wavelength of 515 nm and a pulse duration of 300 fs to ablate copper with fluence 0.7 - 63 J/cm². The experimental results were compared with the theoretical results, where a modified Drude-critical point model was adopted to simulate the ablation depth. The modified model considered the electron transition effect and a two-temperature model that assumed both the linear and nonlinear absorption effect. Comparison of the experimental and simulated results revealed that the simulated ablation depth obtained using the nonlinear absorption model was consistent with the experimental results.

Keywords: non-linear absorption; femtosecond laser; two-temperature model

1. Introduction

In the past decades, there have been several experimental and theoretical studies on femtosecond laser irradiation on copper. For experimentation, laser parameters such as laser wavelength, fluence, and pulse duration have been used to investigate the surface morphology and ablation threshold [2-6]. On the other hand, the two-temperature model (TTM) with a dynamic optical model, linear absorption model, and phase explosion model have been used to investigate the mechanism of laser-material interaction [7-14]. Among the investigations on copper, the IR wavelengths regime [2, 3, 5-14] was the main focus, and the green wavelength regime has been rarely discussed.

Although some recent investigations were conducted on drilling copper with a femtosecond laser at two wavelengths, 1030 nm and 515 nm [4, 15], neither the ablation depth nor the theoretical investigation was presented. Thus, the laser-copper interaction for green wavelengths should be further investigated.

Nonlinear absorption effect has been proposed recently for femtosecond laser machining of transition metals (e.g., gold, silver, and copper) [15-20]. According to Bauer et al. [17], the electron has three different processes, intraband, interband excitation, and auger decay, during the femtosecond laser interaction with the transition metal. The interband excitation is where the d-band electron transitions above the Fermi level and causes nonlinear absorption. The phenomenon is similar to the laser interaction with semiconductors. Zayarny et al. [15] investigated the relationship between the ablation threshold at different wavelengths (1030 and 515 nm) for both copper and silver. The two-photon absorption effect has been proposed because the single-photon energy, for example, 1.20 eV for the wavelength of 1030 nm, is not high enough to force the electron transition from d-band above the Fermi level in the IR wavelength regime. Kudryashov et al. [18] used the pump-probe method to measure the transmission of gold films with different thicknesses and

proposed that the varied transmission coefficient is due to two-photon transition from d bands to s bands. Kudryashov et al. [19] also used single 515 nm femtosecond laser pulses to irradiate steel with different pulse duration. The multi-photon interband absorption was used to explain the relationship between ablation depth and fluence. Cheng et al. [20] predicted the structures generated on common and noble metals using different optical models. They proposed that, for noble metals, the interband absorption effect must be considered in the Drude model to enhance the accuracy.

For copper, the gap between the d-band and Fermi level is around 2.15 eV at room temperature [21]. Bevillon et al. [22, 23] proposed that the density of state (DOS) would vary depending on the electron temperature; the d-band would shift toward lower energy with increased electron temperature while the Fermi level would shift toward higher energy. This causes the gap between d-band and Fermi level to expand to around 7 - 8 eV when the electron temperature reaches 50,000 K. They also proposed that the free electron number would increase from 1.9 to 3.2 when the electron temperature is raised from 0 to 10^5 K because the d-band electrons would then reach the delocalized state and become free electrons.

In summary, the phenomena mentioned above are the reasons for considering the transition effect during the green wavelength femtosecond laser-copper interaction. Moreover, because the gap increases with electron temperature, the energy which forces the d-band electron to become a free electron is increased. Therefore, when the visible or near-infrared laser is used in the process, the non-linear absorption effect is induced at a high electron temperature. For copper, although the non-linear absorption phenomenon has been deduced by experimental methods, the non-linear absorption model has not been adopted to calculate the ablation depth and compare it with the experimental results.

In this study, a single femtosecond green wavelength laser pulse was used to ablate the copper. The ablation depth and threshold fluence were determined and compared with theoretical results. A TTM with a different absorption model and the dynamic optical model considering the transition

effect was used to calculate the electron and lattice temperature. The phase explosion model was adopted to calculate the ablation depth. Finally, the simulation results obtained by linear and non-linear absorption were compared with the experiment.

2. Experimental Setup

A polished copper (99.99%) sample was irradiated in the air with a femtosecond fiber laser machining system. The schematic of the experimental setup is shown in Figure 1. An infrared femtosecond fiber laser (KASMORO, mRadian Inc.) with a wavelength of 1030 nm, a pulse duration of 300 fs, a repetition rate of 3 kHz, and an average laser power of 1 W was used. The laser beam was passed through the barium borate (BBO) crystal to convert the laser beam with wavelength 515 nm, and a Gaussian profile with a diameter of 2.2 mm (at $1/e^2$). The laser beam was reflected by reflective mirrors, passed through an acousto-optic modulator (AOM), and subsequently entered an objective lens (numerical aperture NA=0.28). The laser could be controlled as a single pulse output through AOM. The focus laser beam diameter D was calculated around 2.2 μm by $D \sim 1.22 \lambda/\text{NA}$, where λ is laser wavelength. The laser powers used in this study were 0.04-3.6 mW at a repetition rate of 3 kHz. The laser fluences were calculated to be in the range of 0.7 - 63 J/cm². The morphology of the top view ablation profile was measured using scanning electron microscopy (Hitachi SU-8010). The crater depth and shapes were measured by atomic force microscopy (AFM, Bruker ICON).

Figure 1 A schematic of the experimental setup

3. Modeling

When femtosecond laser irradiates at the bulk metal surface, the free electron in the conduction band directly absorbs the laser energy. The increased electron temperature induces variations in the optical properties. These optical properties determine the laser energy absorption and affect the thermal ablation. Moreover, in transition metals (e.g., copper), the non-linear absorption effect must also be considered because the d-band electron transitions above the Fermi level. Thus, the dynamic optical model and the TTM with linear and non-linear absorption models are discussed in detail in this section.

3.1 Dynamic optical model

The Drude-critical point (DCP) model was used to simulate the dynamic optical properties when the femtosecond laser pulse irradiated the material surface. The DCP model included a classical Drude model and three Lorentz terms as shown below [24, 25]:

$$\begin{aligned}
\varepsilon(\omega) &= \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + \frac{i}{\tau_e})} \\
&\quad + \sum_{p=1}^3 B_p \Omega_p \left(\frac{e^{i\phi_p}}{\Omega_p - \omega - i\Gamma_p} + \frac{e^{-i\phi_p}}{\Omega_p + \omega + i\Gamma_p} \right) \\
&= \varepsilon_1 + i\varepsilon_2
\end{aligned} \tag{1}$$

where ε_∞ is the dielectric constant; ω_p is the plasma frequency; ω is the laser frequency; B_p is the weighting factor for three critical points; Γ_p is the broadening parameter; Ω_p and ϕ_p are the oscillator strength and phase, respectively, and τ_e is the electron relaxation time which can be described by Eq. (2):

$$\tau_e = \frac{1}{A_e T_e^2 + 1.4144 v_{eff}} \tag{2}$$

where T_e is the electron temperature; v_{eff} is the electron-lattice collision rate; and A_e is a constant. For copper, $A_e = 1.28 \times 10^7 \text{ s}^{-1} \text{ K}^{-2}$, v_{eff} can be calculated by the model proposed in [26], and the parameters in Eq. (1) can be found in reference [9].

Assuming that the incidence of the laser pulse is normal to the sample (along the z -axis), the surface reflectivity R and absorption coefficient α can be calculated by the Fresnel equations:

$$R(z, t) = \frac{(n(z, t) - 1)^2 + k^2(z, t)}{(n(z, t) + 1)^2 + k^2(z, t)} \tag{3}$$

$$\alpha(z, t) = \frac{2\omega k(z, t)}{c} \tag{4}$$

where c is the light speed in a vacuum; z is distance, t is time; and n and k are the normal refractive index and extinction coefficient, respectively. With calculated temperature-dependent optical properties from Eq. (1), the n and k can be calculated by Eqs. (5) and (6):

$$n(z, t) = \sqrt{\frac{\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2}}{2}} \tag{5}$$

$$k(z, t) = \sqrt{\frac{-\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2}}{2}} \tag{6}$$

In Eq. (4), the $\alpha(z, t)$ also equals the inverse of the optical penetration depth $\delta(z, t)$. In addition, the electron ballistic penetration depth δ_b was considered in the absorption coefficient in this study, as described by Eq. (7). The δ_b can be calculated by $\delta_b = V_e \times t_b$, where $t_b = 27$ fs is the Drude relaxation time [27]; and V_e is the electron velocity determined by Eq. (8). In Eq. (8), K_B and m_e are the Boltzmann constant and mass of an electron, respectively:

$$\alpha_{mod}(z, t) = \frac{1}{\delta(z, t) + \delta_b} \quad (7)$$

$$V_e = \sqrt{3K_B T_e / m_e} \quad (8)$$

Recently, J. Winter et al. used the pump-probe technique to measure the dynamic reflectivity of copper irradiated by a femtosecond laser with a wavelength of 1030 nm and 528 nm [28]. They proposed that there are two physical processes included during the laser process. First, the transition effect is associated with broadening parameters and is related to the collision frequency. Second, the transition effect increases the number of free electrons and induces the plasma frequency to vary with the increase in temperature. However, the broadening parameter plays an important role in the green wavelength regime because the photon energy is close to the gap between the d-band and Fermi level (2.15 eV). It would directly influence the d-band electrons and vary the collision frequency. Thus, based on [28], the broadening parameter Γ_1 was replaced by a temperature-dependent parameter, and the DCP model (Eq. (1)) was renamed DCP-mod. The temperature-dependent broadening parameter is defined as $\Gamma_1(T_e, T_l) = A_1 \tau_e$, where τ_e is the electron relaxation time and A_1 is a constant. For copper, $A_1=10.62$ which is derived by τ_e and Γ_1 at 300 K was used in this study.

Figure 2 shows the time history of dynamic reflectivity $R(0, t)$ calculated by the DCP-mod model on the surface of the Cu irradiated by a femtosecond laser (pulse duration $t_p = 680$ fs, wavelength 528 nm) of laser fluence 0.4 J/cm^2 . The red line is the experimental results from [28], and the blue line is obtained by Eq. (9). Note that the lasing starts from $t = -2t_p$, reaches its peak power at $t = 0$, and ends at $t = 2t_p$. The $R(0, t)$ increases, reaches a maximum at the time $t = -0.612$

ps, decreases quickly during the laser pulse, and then increases at a slower rate as the time is prolonged. The varying mechanism of reflectivity indicates that the broadening parameters varied with electron and lattice temperature, which induced the variation in dielectric function and was different from using near-infrared wavelength to ablate the copper [28]. The reason that the minimum value at $t = 0.544$ ps was lower than the experiment could be because the dynamic plasma wasn't considered in this study.

Figure 2. Comparison of dynamic reflectivity on the surface of the Cu calculated by DCP-mod and experimental data

3.2 Laser source term

To simulate the ultrashort laser-metal interaction, the TTM should always be adopted because the amount of time for laser interaction with the material is less than the electron-lattice coupling time. Also, in general, the one-dimensional (1D) heating problem can be used to analyze the ultrashort laser-metal interaction because the size of the heating spot is much larger than the heat-affected zone. The description of TTM is shown in the supplemental material.

The linear absorption laser heat source is often used to simulate the ultrashort pulse laser interaction with metal because the electron in a conduction band can absorb the laser energy directly. The linear absorption laser heat source with temperature-dependent optical properties can be described as follows:

$$S(z, t)_{linear} = \alpha_{mod} I(0, t) \exp^{-\alpha_{mod} z} \quad (10)$$

$$I(0, t) = \frac{0.94 \times F_0 [1 - R(0, t)]}{t_p} \exp^{-2.77 (\frac{t}{t_p})^2} \quad (11)$$

where α_{mod} is the linear-absorption coefficient (see Eq. (7)); $I(0, t)$ is the laser intensity at the front surface ($z = 0$); F_0 is the laser fluence; $R(0, t)$ is the reflectivity on the front surface that depends on the temperature (see Eq. (3)); and t_p is the pulse duration defined as the full width at the half maximum (FWHM) of the Gaussian pulse.

The non-linear absorption phenomenon may be considered when the ultrashort laser pulse is irradiated on transient metal [15-20, 22, 29, 30]. The laser heat source term can be modified from linear absorption to non-linear absorption, e.g., two-photon [18]. When the non-linear absorption effect is considered in the process, the process is similar to the laser interaction with semiconductors. To simplify, the carrier loss and auger recombination effect were not considered in this study. The non-linear laser heat source can be described as below:

$$S(z, t)_{non-linear} = \alpha_{mod} I(z, t) + \beta I(z, t)^2 \quad (12)$$

$$\frac{\partial I(z, t)}{\partial z} = -\alpha_{mod} I(z, t) - \beta I(z, t)^2 \quad (13)$$

where α_{mod} is the linear-absorption coefficient (see Eq. (7)); β is the two-photon absorption coefficient; and $I(z, t)$ is the non-linear absorption laser intensity. The temporal laser intensity distribution in the material at different depths can be obtained by Eq. (13) after obtaining the laser intensity on the front surface ($z = 0$) by Eq. (11). In this study, the Runge-Kutta fourth-order numerical method is adopted to solve Eq. (13). The calculated laser intensity $I(z, t)$ is substituted in Eq. (12) to solve the total laser heat source. An earlier study [19] proposed that the β can be

determined by the ablation threshold fluence and the experimental ablation depth, it can be described by Eq. (14):

$$\beta = \frac{t_p}{z_{abl}(1 - R_c)} \left(\frac{1}{F_{abl}} - \frac{1}{F_0} \right) \quad (14)$$

Where z_{abl} is the ablation depth; F_{abl} is the ablation threshold fluence; and R_c is the surface reflectivity at room temperature. The z_{abl} and F_{abl} are determined by the experimental results. In this study, it is given that z_{abl} and $F_0 = 0.7 - 63 \text{ J/cm}^2$ (see Figure 4), $t_p = 300 \text{ fs}$, $F_{abl} = 0.54 \text{ J/cm}^2$, $R_c = 0.58$ for copper with 515 nm, and the calculated β on the surface of the Cu irradiated by a femtosecond laser is shown in Figure 3. The result shows that the β was decreased on the input fluence regime.

Figure 3 Calculated two-photon absorption coefficient versus the fluence

3.3 Phase explosion model

In the process of femtosecond ablation of metal, the phase explosion is a dominant mechanism. When the femtosecond irradiates to the metal, the metal is superheated without boiling because the time is less than that required for normal boiling and normal vaporization so that heterogeneous nucleation is produced. When the superheated liquid temperature reaches $0.9 T_{tc}$ (thermodynamic

equilibrium critical temperature) [31], the tensile strength of the liquid material drops to zero and is ejected from the surface vapor and liquid mixture.

3.4 Numerical condition

In the numerical simulation, a single laser pulse of $t_p = 300$ fs and a wavelength of 515 nm were used to irradiate the Cu front surface, because considering the multi-photon absorption effect in the model, the carrier diffusion depth must be considered. Thus, the Cu thickness was set at 5 μm which was larger than the maximum carrier diffusion depth (3 μm) [32]. The initial electron and lattice temperature were set at 300 K. The finite difference method was used in this study, and the number of 2500 grids were divided. The grid sizes were 2 nm.

4. Results and discussion

4.1 Experiment results

The measured ablation depths were in the range of 209 - 1906 nm, respectively. Figure 4 presents the semi-logarithmic plot of the single ablation depth against the laser fluence. The straight line was obtained by curve-fitting from experimental data. The fitting equation could achieve the ablation threshold in Figure 4. The ablation threshold F_{abl} was around 0.54 J/cm² which is a little lower than the values found when using a femtosecond laser with a near-infrared wavelength, as previously reported [2, 10]. The AFM data of a copper surface irradiated by a single-shot femtosecond green wavelength laser with laser fluences of 0.7, 7, and 63 J/cm² are shown in the supplement material.

As shown in Figure 4, the ablation depth increased dramatically with the increase in fluence. It can be deduced by two reasons. First, in the femtosecond laser interaction with the metal process, the electron-phonon coupling factor (G) is the key parameter because it would influence the degree of electron coupling with the lattice and finally domain the ablation depth. According to an earlier

study [33], the G is dramatically increased when the electron temperature is more than 50,000 K. Thus, it would highly induce the deeper ablation depth at higher fluence input. Second, according to [20, 30], the interband absorption effect would induce more electron transition from d-band above Fermi level. These electrons contribute to the electron-electron scattering frequency and also induce the optical properties to vary. Therefore, it would increase the light absorption and cause deeper ablation depth.

Figure 4. The ablation depth (z_{abl}) against laser fluence (F_0)

4.2 Simulation results

In this study, the laser heat source $S(z, t)$ was investigated by linear and non-linear absorptions. In copper, at room temperature, the photon energy was higher than the gap between the d-band and Fermi level (2.15 eV) in the visible wavelength range (e.g., 515 nm, 2.4 eV). Linear absorption is the primary mechanism in low electron temperature. However, according to earlier studies [22], when the electron temperature increased above 10,000 K, the non-linear absorption effect influences and gradually dominates the absorption mechanism [15-17].

Figure 5 shows the comparison of the laser heat source calculated by linear absorption (see

Eq.(10)) and non-linear absorption (see Eq. (12)) on the front surface with different fluences. The laser pulse started at -0.6 ps, reached the peak value at 0 ps, and ended at 0.6 ps. The peak value calculated by the non-linear absorption model was always higher than the value calculated by the linear absorption model. As shown in Figure 5(a), the laser heat source calculated by the non-linear model was higher than the linear model in the low fluence regime. However, as shown in Figure 5(b), the difference in the value calculated by a different model gradually increased when the fluence was increased. The peak laser heat source by non-linear absorption was 20 times (0.7 J/cm^2) and 136 times (7 J/cm^2) that of linear absorption. The non-linear model enhanced the absorbed laser heat source and caused more ablation depth at high fluence.

Figure 5. Heat density temporal distributions calculated by different absorption models with fluences: (a) 0.7 J/cm^2 , (b) 7 J/cm^2

Figure 6 plots the laser heat source propagating into the material from $z = 0$ to $z = 800 \text{ nm}$ at a fluence of 0.7 J/cm^2 , $t = 0 \text{ ps}$, and different absorption models. The laser heat source, calculated by different models, showed exponential-like decay. Compared with these two models, the laser heat source calculated by the non-linear model was always higher than the linear model. It is evident that non-linear absorption enhances the volumetric laser heat source propagating into the material and increases the ablation depth.

Figure 6. Spatial profile of laser heat source propagating into the material at the fluence of 0.7 J/cm^2 and $t = 0 \text{ ps}$

Figure 7 illustrates the time evolution of electrons (T_e) and lattice (T_l) temperatures at the front side surface of the sample for different fluences and different absorption models. The results show that the electron was first thermalized when the laser irradiated on the sample, and the temperature increased immediately. After 0.5 ps, the electron is coupled with the lattice and transfers the temperature to the lattice. In this research, as the lattice temperature reached a critical point ($0.9 T_c = 6925 \text{ K}$), a phase explosion occurred, and the material experienced ablation, resulting in a constant lattice temperature.

The electron temperature calculated by the non-linear model was higher than that calculated by the linear model, especially 63 J/cm^2 . For 0.7 J/cm^2 , the T_e calculated by non-linear model ($2.8 \times 10^4 \text{ K}$) was slightly higher than the linear model ($2.6 \times 10^4 \text{ K}$). However, for 63 J/cm^2 , the T_e calculated by non-linear model ($2 \times 10^6 \text{ K}$) was much higher than linear model ($4 \times 10^5 \text{ K}$). It can be explained by the heat source term calculated by the non-linear model being higher than the linear model. Meanwhile, note that the G used in this paper was temperature-dependent. The higher T_e induces a stronger coupling effect. For example, for 0.7 J/cm^2 , the time of lattice to reach the critical point was around $t = 1.41 \text{ ps}$, which was lower than the linear model ($t = 1.83 \text{ ps}$). The

fluences used in this paper were all above the ablation threshold so that the lattice temperature reached the critical temperature and caused the material ablation. After a few ps or ns, depending on the fluence, the system reached an equilibrium state ($T_e = T_l$), and these two temperatures decrease together until the computing is over. This is not shown in this paper.

Figure 7. Time-dependence of the calculated electron and lattice temperature: (a) linear absorption model, (b) non-linear absorption model

Figure 8 presents the time history of the laser ablation depth with different fluences and different absorption models. In each model, the total ablation time in the highest fluence is much longer than lower fluence because the maximum electron temperature is higher so that the time of transfer of Te to Tl is longer.

The ablation depth by non-linear absorption was 16 times (0.7 J/cm^2) and three times (63 J/cm^2) that of linear absorption. It can be explained by the volumetric laser heat source, which, calculated by the non-linear model propagated into the material, was higher than the linear model and enhanced the ablation depth (Figure 6).

Figure 8. The ablation depth calculated by different laser fluences and different absorption models: (a) linear absorption model, (b) non-linear absorption model.

Figure 9 shows the comparison of experimental data and simulation results. The results calculated from the linear absorption model significantly underestimated the ablation depth. In contrast, in the non-linear absorption model, the results were in good agreement with experimental results. Note that the peak power density used in this was higher than the air breakdown threshold (e.g., 2.4×10^{14} W/cm²), and it reduced the laser fluence 10-25 % when irradiated on the front surface [34]. However, in this study, this effect is not considered so that the fluence was overestimated and caused deeper ablation depth. Interestingly, for the fluence 3.5-14 J/cm², the ablation depth was really close to the experimental results. This can be explained by the fact that β calculated by Eq. (14) was not very precise. In general, two different ablation thresholds can be determined when the input fluence increased from low to high [35]. However, in this study, for simplicity, the single ablation threshold was used. Nevertheless, the non-linear model was found to enhance the accuracy, and the results were in good agreement with the experimental results.

Figure 9. Comparison of the simulation and experimental data

5. Conclusion

The experimental and simulation results were reported for the single femtosecond green wavelength laser pulse ablation of copper with different fluences ($0.7 - 63 \text{ J/cm}^2$). The ablation threshold of copper in the green wavelength was around 0.54 J/cm^2 , obtained by the experimental procedure. A modified DCP model that considered the transition effect was used for the simulations to calculate the dynamic optical properties. The two-photon absorption coefficient was determined by considering the d-band shifting effect. The laser heat source term was modified to non-linear absorption in the TTM and compared with the linear absorption.

When the laser heat source was modified to non-linear absorption, the laser heat source calculated by the non-linear model could enhance the volumetric laser heat source propagating into the material, resulting in increased ablation depth. The ablation depth calculated from linear absorption underestimated the ablation depth by comparing the experimental and simulated results. However, the results calculated from the non-linear absorption model agreed with the experiments

over a wide range of laser fluences.

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Declaration

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Authors Contributions:

Yi-Hsien Liu: Investigation, Measurement, Data curation, Writing- Original draft preparation.

Chung-Wei Cheng: Conceptualization, Supervision, Writing- Original draft preparation.

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