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RESEARCH ARTICLE

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Metal absorptivity in femtosecond pulsed laser ablation

Abstract In this paper, the effect of the absorptivity of metal on femtosecond pulsed laser ablation is investigated. The formulas for the absorptivity depending on target temperature are derived from Maxwell Equations and the Lambert-Beer's law. Based on this, a new two-temperature model is proposed to describe the femtosecond pulsed laser ablation with metal. Then, using Au as an example, a finite difference method is employed to simulate the space-dependent and time-dependent absorptivity and the target temperature. The temperature evolution of our model is compared with the result obtained form the heat conduction model taking the absorptivity as constant. It is shown that the absorptivity plays an important role in the femtosecond pulsed laser ablation. The results of this paper are helpful in choosing the best technical parameters in femtosecond pulsed laser ablation.

Keywords absorptivity, femtosecond pulsed laser ablation, two-temperature model, target temperature

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1 Introduction

Significant advances in tunable solid-state lasers and the chirped pulse amplification technique by using Ti: sapphire, Cr: LiSAF, and Cr: LiCAF laser media have led to a challenging phase of application research [1]. The characteristics of the interaction of femtosecond (fs) laser and materials can be comprehended by the study of target surface temperature. However, the understanding on the femtosecond pulsed laser and the materials mechanism lag behind experimental studies. Although some researchers employ the investigation of fs laser ablation, the researches are limited to transparent materials [2].

Absorptivity is an important parameter in the interaction of a laser and the target. It refers to the magnitude of the ratio of absorbed energy to incident energy and describes the absorbing ability of the target to laser energy [3]. In the initial stage of femtosecond laser ablation with materials, the thermal mechanism still dominates almost all the physical effects. So far, there are some experimental and theoretical studies about absorptivity. However, few literatures investigate the absorptivity theoretically in detail in femtosecond laser ablation using a metal material [4].

In our previous work, we investigated the effect of absorptivity on nanosecond laser ablation. Recently, we found that there are some new interesting characteristics in femtosecond laser ablation. In this paper, the absorption mechanism in the interaction process of pulsed laser with the material is investigated in detail. According to the Maxwell Equation and the Lambert-Beer's law, as an example of metal, we deduce the formulas for the temperature-dependence of absorptivity. The formula is introduced to the two-temperature model to describe femtosecond laser ablation. The effects of the absorptivity on metal temperature are investigated principally.

2 Metal target absorptivity

The optical characteristics of a material can be described by the complex refractive index:

$$n_c = n + i\kappa \tag{1}$$

where n is the refractive index, κ is extinction coefficient.

It is assumed that a pulsed laser beam irradiates on a target surface vertically as shown in Fig.1. x refers to the spa-

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tial coordinate in the direction normal to the target surface with the origin located at the surface. 0^+ refers to the position that is illimitably close to the interface inside the target, and 0^- is the position that is illimitably close to the interface outside the target.

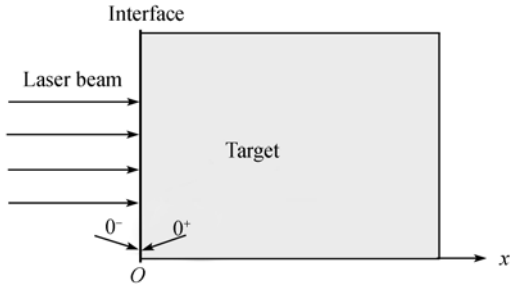

Fig.1 The geometry of laser irradiation.

The electric field intensity in the target can be described as follows:

$$E^+ = E_0^+ \, \mathrm{e}^{\mathrm{i}\omega(\frac{x}{\upsilon}-t)} \tag{2}$$

where E^+ is the electric field intensity in the target, E_0^+ is the electric field intensity at $x = 0^+$, ω is the incident laser angle frequency, υ is the laser propagation velocity in the target satisfying the following relation [6]:

$$\upsilon = c/n_{\mathrm{c}} \tag{3}$$

where c is the laser propagation velocity in vacuum.

Substituting Eqs. (1) and (3) into Eq. (2) yields

$$E^+ = E_0^+ \, \mathrm{e}^{-\mathrm{i}\omega t} \, \mathrm{e}^{\mathrm{i}\omega\frac{xn_{\mathrm{c}}}{\upsilon}} \, \mathrm{e}^{-\omega\frac{xk}{c}} \tag{4}$$

By the operation of the module to Eq. (4), we have

$$\left|E^+\right|^2 = E^+E^{+*} = \left|E_0^+\right|^2 \mathrm{e}^{-2\omega\frac{xk}{c}} \tag{5}$$

According to the Lambert-Beer-Bouguer Law, when a laser irradiates a target surface, the laser intensity attenuates in exponent in the target, which can be expressed by [7]

$$I^+(x,t) = b\beta I^-(0,t)\mathrm{e}^{-bx} \tag{6}$$

where $I^+(x,t)\,[W/m^2]$ is the intensity at a given time t and depth x in the target, $I^-(0, t)$ is the intensity of the incident laser. β is the absorptivity of the target, b is the absorption coefficient of the target, $1/b$ has the dimension of length and is termed as the penetration depth where the laser intensity is $1/e$ time of the irradiated laser intensity.

In the target, the intensity is equal to the square of the module of the electric filed intensity, namely $\left|E^+\right|^2 = I^+(x,t)$. Based on this relation, comparison of Eq.(5) with Eq.(6) gives the following relation:

$$b = \frac{2\omega k}{c} = \frac{4\pi\upsilon k}{c} = \frac{4\pi k}{\lambda_0} \tag{7}$$

where λ_0 is the laser wavelength in vacuum.

Substituting Eq. (4) into Maxwell equations in an isotropical medium, we obtain the refractive index n and the extinction coefficient k as the functions of the conductance σ and the permittivity ε as follows [5]:

$$\begin{aligned} n^2 &= \frac{1}{2}\varepsilon\left\{\left[1+\left(\frac{\sigma}{\omega\varepsilon\varepsilon_0}\right)^2\right]^{0.5}+1\right\} \\ k^2 &= \frac{1}{2}\varepsilon\left\{\left[1+\left(\frac{\sigma}{\omega\varepsilon\varepsilon_0}\right)^2\right]^{0.5}-1\right\} \end{aligned} \tag{8}$$

where ε_0 is the permittivity of vacuum, which is a constant $\varepsilon_0 = 8.85\times10^{-12}\,\mathrm{C}^2\cdot\mathrm{N}^{-1}\cdot\mathrm{m}^{-2}$.

For general metal, the conductance is high. Usually, we can regard that

$$\left(\frac{\sigma}{\varepsilon\varepsilon_0\omega}\right)^2 \gg 1 \tag{9}$$

So, Eq. (9) can be simplified by

$$n \approx k \approx \sqrt{\frac{\sigma}{2\omega\varepsilon_0}} \tag{10}$$

In the condition of laser irradiation, σ can be presented as the linear increasing function of temperature T in the form [8]:

$$\sigma(T) = \frac{\sigma_0}{1+\alpha(T-T_1)} \tag{11}$$

where T represents the target temperature at a given t and x, σ_0 is the target conductance at initial temperature T_0, α is the target temperature coefficient of resistance. For general metal, α is in the range $\sim(400\text{–}700)\times10^{-5}\,\mathrm{K}^{-1}$ [9].

When a laser irradiates a target surface vertically, the target absorptivity is generally described as follows [8]:

$$\beta = 1-R = 1-\frac{(n-1)^2-\kappa^2}{(n-1)+\kappa^2} = \frac{2\kappa^2}{(n-1)^2+\kappa^2} \tag{12}$$

Substituting Eqs.(10) and (11) into Eq.(12) gives

$$\beta(T) = 2\sqrt{\frac{4\pi\upsilon\varepsilon_0}{\sigma}} = \sqrt{\frac{4\pi c\varepsilon_0\left[1+\alpha\left(T-T_0\right)\right]}{\lambda_0\sigma_0}} \tag{13}$$

This is the most important relationship that we derived in this paper indicating the relation between the resistance temperature coefficient, the laser wavelength, the target temperature and the absorptivity.

3 Two-temperature model

In general, three energy transfer stages during femtosecond laser irradiation of metals have been identified [10]. Initially, the free electrons absorb the energy from the laser. This stage is characterized by a lack of thermal equilibrium among the electrons. In the second state, the electrons reach thermal equilibrium and the density of states can now be represented by the Fermi distribution. However, the electrons and the lattice are still at two different temperatures. In the final stage, the electrons and the lattice reach thermal equilibrium and thermal diffusion carries the energy into the bulk. A two-temperature model to predict the non-equilibrium temperature distribution between electrons and the lattice during femtosecond laser irradiation of metals was first described by Anisimov *et al*. [11].

It treats electrons and the lattice as two separate subsystems with different temperatures governed by respective equations.

In the two-temperature model, the electron temperature T_e, and the lattice temperature T_i are subject to two coupled one-dimensional governing equations:

$$\rho C_e \frac{\partial}{\partial t} T_e = \frac{\partial}{\partial x} K_e \frac{\partial}{\partial x} T_e - g\left(T_e - T_i\right) + S\left(x,t\right)$$
$$\rho C_i \frac{\partial}{\partial t} T_i = g\left(T_e - T_i\right) \tag{14}$$

where C_e=0.71×10$^{-4}T_e(x,y)$, K_e are specific heat and thermal conductivity of the electron. C_i is specific heat of the lattice, ρ is target density, g (Te-Ti) is the electron-lattice coupling term, which shows that the energy transfer from electrons to the lattice is proportional to their temperature difference. $S(x,t)$ is the laser heating source term expressed as [12]

$$S(x,t) = bI^{+}(x,t)\mathrm{e}^{-bx} = b\beta I^{-}(0,t)\mathrm{e}^{-bx} \tag{15}$$

The laser incident intensity can be expressed by a Gauss function [13–14]:

$$I^{-}(0^{-},t) = I_0 \mathrm{e}^{\frac{-(t-\tau)^2}{2\zeta^2}} \tag{16}$$

where I_0 is the maximal intensity of the incident laser at the outside surface, τ is the half-width of

the pulse. The temporal shape of the laser pulse may be changed with the parameter ζ [13].

Substitution of Eqs. (13) and (16) into Eq. (15), gives

$$S(x,t) = b\beta I^{-1}(0,t)\mathrm{e}^{-bx}$$
$$= bI_0\sqrt{\frac{4\pi c\varepsilon_0[1+\alpha(T_e-T_0)]}{\lambda_0\sigma_0}}\,\mathrm{e}^{\frac{-(t-\tau)^2}{2\zeta^2}}\,\mathrm{e}^{-bx} \tag{17}$$

What needs to be emphasized is that, we introduce the lattice temperature in the formula (17). The reason is that the resistivity of metal, which is the reciprocal of conductance, causes the dispersion of electrons in the lattice by libration. The lattice acoustics wave forms a potential energy fluctuation described by the average extraction value of ion displacement. According to the energy average law, the average extraction value of displacement has a direct ratio to temperature. At high temperature, the phonon number of the lattice acoustics wave has a direct ratio to temperature. The more the phonon number, the more the electron is dispersed. Therefore, the resistivity of metal has a direct ratio to temperature. Then, we adopt the lattice temperature here [14].

During the pulse of a femtosecond laser, there is no vaporization because the pulse width (*fs*) is much shorter than the coupling time of the electron and lattice (*ps*). The time for the surface to start to vaporize is [8]

$$\tau' = \frac{\pi\rho C}{4k}\left(\frac{kT_v}{\beta I_0}\right)^2 \tag{18}$$

where T_v is the vaporization temperature.

With the equation of energy conservation and adiabatic hypothesis, the boundary conditions as the irradiation of laser can be depicted by

$$-K\left.\frac{\partial T(x,t)}{\partial x}\right|_{x=0} = \beta I_0 \mathrm{e}^{\frac{-(t-\tau)^2}{2\sigma^2}}, \quad 0<t<\tau \tag{19}$$

$$\left.\frac{\partial T(x,t)}{\partial x}\right|_{x=\delta} = 0, \quad 0<t<\tau \tag{20}$$

where β_0 is the absorptivity of the target surface, $\delta = 3.46\sqrt{\alpha_s \tau}$ [15].

After the end of the pulse, the boundary conditions are

$$-K\left.\frac{\partial T(x,t)}{\partial x}\right|_{x=0} = 0, \quad \tau<t<\tau' \tag{21}$$

$$\left.\frac{\partial T(x,t)}{\partial x}\right|_{x=\delta} = 0, \quad \tau<t<\tau' \tag{22}$$

The initial condition is

$$T\big|_{t=0} = T_0 \tag{23}$$

4 Results and discussion

Using Au as an example, the two-temperature Eq. (14) is solved by a finite difference method. It is assumed that the target is irradiated by an amplified Ti:sapphire laser system generating 60 fs pulse of about 0.078 mJ/cm^2 with central wavelength of 800 nm. During the simulation, a time step of 1×10^{-15} s and depth step of 1×10^{-8} m were used. The initial temperature is 273 K. Table 1 provides the parameter values of Au from Ref. [15].

Table 1 Thermal and optical property of Au.

K_e /(W·cm^{-1}·K^{-1})	ρ /(g·cm^{-3})	C_i /(J·g^{-1}·K^{-1})	β_0	g /(W·cm^{-3}·k^{-1})	α / K^{-1}	σ_0 /(Ω^{-1}·m^{-1})
3.11	19.3	0.131	0.051	2.6×10^{11}	3.24×10^{-3}	4.16×10^{7}

First, the ablation process is analyzed by studying the evolution of temperature. The electron and the lattice temperature on the surface are shown in Fig.2. It is seen that the electron temperature on the surface is increased quickly to the peak value of 10 000 K, while the lattice temperature does not increase as fast. Due to the electron-lattice coupling, the electron temperature starts to decrease and the lattice temperature increase while they reach approximately the same value after 15 ps. Because the heat capacity of electron is 1—2 order smaller than that of the lattice, and the pulse width of the femtosecond laser is much shorter than the electron relaxation time, energy is not lost by the electron-photon coupling. Therefore, the electron temperature increases quickly, while the lattice temperature does not increase as fast.

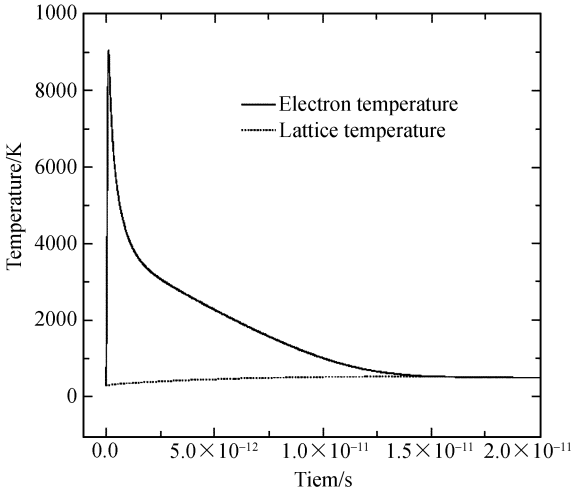

Fig.2 The temperature evolution with time.

When the laser irradiates the metal target, the target will absorb the laser energy and be heated due to the interaction of the laser with the electrons and the ion in the target. The increase of the target temperature results in the enhancing of the target resistivity. Generally, the resistivity can be regarded as a linear increasing function of target lattice temperature [8]. Moreover, the absorptivity is an increasing function of the target resistivity according to Ref. [16]. Therefore, the temporal evolution of target absorptivity and the target lattice temperature have a similar trend. Thus, at a given depth, the evolution of the target absorptivity increases with time, as shown in Fig.3 (a). Under the condition of this work, the lattice temperature decreases with the position, so the absorptivity also decreases with the position, as shown in Fig.3 (b).

In order to investigate the effect of the absorptivity on the femtosecond laser ablation, we plot the target surface temperature evolutions obtained from the two ablation models in Fig.4. Curve 1 is obtained from our model, considering the variation of the absorptivity, while the other is obtained from the model without considering this variation, which can be described by Eqs. (16)—(18), regarding the absorptivity as constant. We can see that the effect of the absorptivity mainly behaves in that the curve considering the variation is obviously higher than the other one during the period of $1.0\times10^{-13}\text{s}\leqslant t\leqslant15.0\times10^{-13}\text{s}$. During $0\leqslant t\leqslant1.0\times10^{-13}\text{s}$, the two curves nearly overlap each other.

During the laser irradiation, the temperature evolution in the target is formed as a result of the superposition of (1) heating due to laser-energy absorption, (2) cooling of the target surface due to vaporization and (3) cooling due to thermal conductivity [17]. In this paper, the thermal conduc-

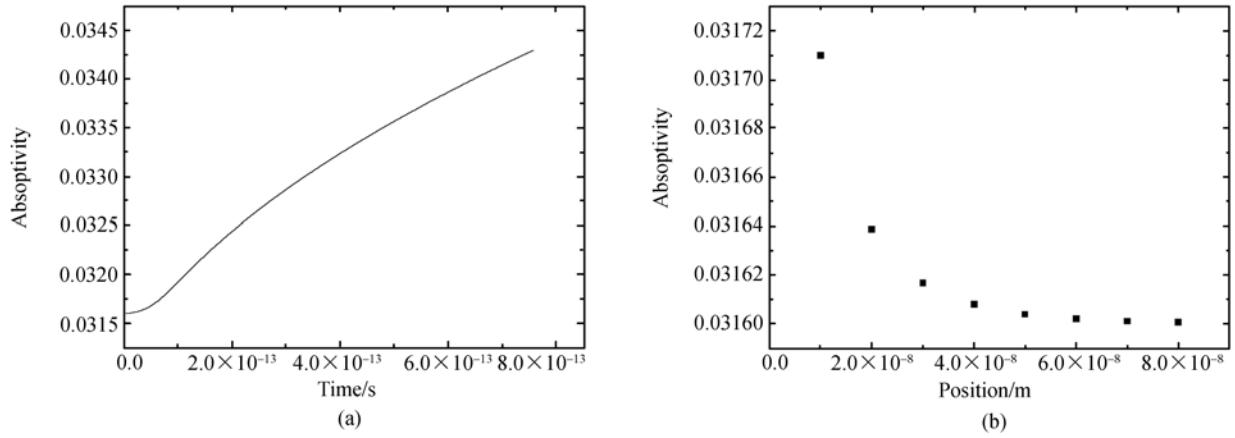

Fig.3 The time and position dependence of the absorptivity.

tivity is assumed as a constant. The vaporization effect has not been analyzed to stand the optical chrematistic of metal in femtosecond laser ablation. We emphatically investigate the effect of the heating due to laser-energy absorption. Absorption of the laser energy is a volume process governed by the Beer's Law, that is, $I(x,t)=\beta I(0,t)\exp(-bx)$. For the evolutions the absorptivity are slow, the difference of the absorbed energy is quite small for both cases considering and not considering the variation, during the period of $0\leqslant t\leqslant 1.0\times10^{-13}$ s. Thus, the two curves nearly overlap each other. However, in the course of $1.0\times10^{-13}\text{ s}\leqslant t\leqslant 15.0\times10^{-13}$ s, as the variation is considered, the target absorptivity changes sharply with the increasing time, that is, it increases quickly. According to the Beer law, the energy absorbed in the case where variation of absorptivity is considered is much little than the case without considering the variation. Therefore, the difference of the two curves is bigger. Then, we can deduce that once we neglect the variation of the absorptivity, the calculated temperature will be less than the actual case.

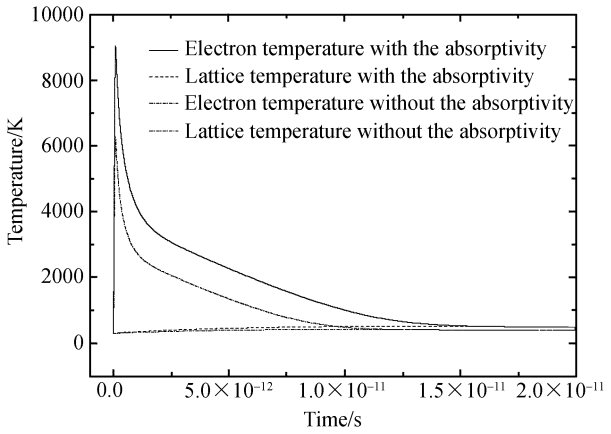

Fig.4 Comparison of target surface temperature.

5 Conclusions

In this paper, the formula for the target absorptivity is obtained based on the Maxwell equation. Then, the two-temperature ablation model is presented with variation of absorptivity. Next, using Au as an example, the two-temperature ablation model is solved by a finite difference method. The time-dependent target temperature and the absorptivity are obtained. The calculated temperature is compared to the results of the model without considering the variation of the target absorptivity. It shows that the two curves nearly overlap each other at the $0\leqslant t\leqslant 1.0\times10^{-13}$ s, while the temperature considering the variation is obviously higher than the case not considering the variation during $1.0\times10^{-13}\text{ s}\leqslant t\leqslant 15.0\times10^{-13}$ s. In this paper, all the physical results are explained in detail from the viewpoint of the law of conservation of energy.

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