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Modeling of femtosecond laser damage threshold on the two-layer metal films

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1. Introduction

The ultrashort pulsed lasers are widely used in a variety of fields including medical applications [1], material processing [2], pulsed laser deposition [3], molecular spectroscopy [4,5], ionization and dissociation of polyatomic molecules [6,7]. With the development of ultrashort laser based on chirped-pulse amplification [8], it is possible to carry out powerful femtosecond laser systems. For the sake of an increasing output power of such femtosecond laser systems, an important limiting factor in the high-power operation of lasers is the damage threshold of the optical components of the laser system. Hence, comparative damage threshold measurements on laser optical components are essential for the evaluation of different materials as well as different deposition techniques in respect to their applicability in high-power femtosecond laser systems. Due to the high reflectivity of gold surface in the infrared beyond 0.7 µm (an averaged reflectance is above 98%), gold coating optical components (mirrors and gratings, etc) are widely used in femtosecond pulsed laser systems (for example, Ti: Sapphire laser system) and infrared optical systems (for example, Terahertz system [9]).

The physics of femtosecond laser heating of gold films has been at the focus of scientific research for many years because of a large number of applications in comparison with other metals [10-13]. One particular research is the calculation of damage thresholds [14,15]. Laser heating of metal is a complex process [16], since

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ABSTRACT

The heating processes of the single-layer gold thin film and the two-layer film assembly of gold padded with other metal (silver, copper and nickel) irradiated by femtosecond laser pulse are studied by the two-temperature model. It is found that the substrate metal can change energy transport, which is corresponding to the temperature changing process, and the thermal equilibrium time. Compared with the single-layer gold film at the same laser fluence, the two-layer film structure can change the damage threshold of the gold surface. Our results indicate that we can maximize the damage threshold of the gold film surface by altering the thickness ratio of the gold layer and the substrate layer in the two-layer film assembly.

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the specific heat capacity of electron is low, irradiated by a laser beam, the electron temperature increases rapidly in an extremely short time, producing a great temperature difference between electron and lattice. The nonequilibrium energy transport, which is due to the electron-lattice coupling mechanism [17], will take place. Based on the heating process, many researchers worked on this problem to the heat transfer mechanism of multi-layer metal films by ultrashort pulsed laser [18,19]. In such processes, energy deposition in the surface layer results in a high electron temperature, which in turn leads to a buildup of the lattice temperature. Such a buildup increases the possibility of thermal damage of the material being processed. Introducing the padding layer under the surface layer has the potential of reducing laser damage. A widely accepted and developed model to predict this heat transfer process is the two-temperature model [20,21], which describes the nonequilibrium process of laser-metal interaction as three stages: depositing laser energy on the surface electrons, transport of energy by the electrons, and transfer of energy from hot electrons to lattice.

By modeling of the laser heating process the experimental cost can be minimized, optimization of affecting parameters can be provided, and the understanding of physical processes during the laser material interaction can be improved. In this paper, we presented a theoretical method with regard to the improvement of the damage threshold of the gold surface. Laser heating of the single-layer gold film or the two-layer gold film padded with other metal material (silver, copper, and nickel) was investigated, using the finite difference method. The distributions of electron temperature and lattice temperature are considered. The predicted results showed that the additional layer metal film material can influence the variation of film temperature. Furthermore, the single-layer gold film and the

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two-layer film consisting of gold and substrate materials as silver, copper and nickel were calculated with respect to their damage thresholds.

2. Mathematical model

The theoretical method to investigate the ultrashort lasermatter interaction is the well-known two-temperature model (TTM) [22]. Laser light is absorbed in metals by the conduction-band electrons within a few femtoseconds. After the fast thermalization of the laser energy in the conduction-band electrons may quickly diffuse and thereby transport their energy deep into the internal target (within a few femtoseconds). At the same time, the electrons transfer their energy to the lattice. The TTM describes the evolution of the temperature increase due to the absorption of a laser pulse within the solid and is applied to model physical phenomena like the energy transfer between electrons and lattice occurring during the target-laser interaction [23]. The one-dimension twotemperature equation is given below [11,24,25]:

$$C_{\rm e}\frac{\partial T_{\rm e}}{\partial t} = \frac{\partial}{\partial x}\left(k_{\rm e}\frac{\partial T_{\rm e}}{\partial x}\right) - G(T_{\rm e} - T_{\rm l}) + S \tag{1}$$

$$C_{\rm l}\frac{\partial T_{\rm l}}{\partial t} = \frac{\partial}{\partial x} \left(k_{\rm l} \frac{\partial T_{\rm e}}{\partial x} \right) + G(T_{\rm e} - T_{\rm l})$$
⁽²⁾

where t is the time, x is the depth, C_e is the electron heat capacity, C_l is the lattice heat capacity, k_e is the electron thermal conductivity, T_e is the electron temperature, T_l is the lattice temperature, G is the electron–lattice coupling factor [26], and S is the laser heat source. The heat source S can be modeled with a Gaussian temporal profile [27]:

$$S = \sqrt{\frac{\beta}{\pi}} \frac{(1-R)I}{t_{\rm p}\alpha} \exp\left[-\frac{x}{\alpha} - \beta\left(\frac{t-2t_{\rm p}}{t_{\rm p}}\right)^2\right]$$
(3)

where *R* is the target reflection coefficient, t_p is the full-width at the half maximum (FWHM) with the linear polarization, α is the penetration depth including the ballistic range, and *I* is the incident energy, $\beta = 4 \ln(2)$.

The electron heat capacity is proportional to the electron temperature when the electron temperature is less than the Fermi temperature as $C_e = \gamma T_e$ [28] and $\gamma = \pi^2 n_e k_B / 2T_F$, n_e is the density of the free electrons, and $k_{\rm B}$ is the Boltzmann's constant. The lattice heat capacity is set as a constant because of its relatively small variation as the temperature changes. The electron heat conductivity can be expressed as $k_e = k_{e0}BT_e/(AT_e^2 + BT_1)$ [29], where k_{e0} , A and B are the material constants. Many of the ultrafast laser heating analyses have been carried out with a constant electron-phonon coupling factor G. However, due to the significant changes in the electron and lattice temperatures caused by high-power laser heating, G should be temperature dependent $G = G_0(A(T_e + T_1)/B + 1)$, where G_0 is the coupling factor at room temperature [13,30]. The lattice thermal conductivity is therefore taken as 1% of the thermal conductivity of bulk metal since the mechanism of heat conduction in metal is mainly by electrons [12]. As the temperature changes, the variety of the lattice heat conductivity is relatively small, we think it is a constant.

Considering a one-dimensional two-layered thin film, Fig. 1 shows the schematic view of the laser heating, which indicates a two-layer metal film with an interface at x=l. For the two-layer thin film, the two-temperature equation (Eq. (1) and Eq. (2)) for studying thermal behavior in the thin film can be expressed as

$$C_{e}^{I}\frac{\partial T_{e}^{I}}{\partial t} = \frac{\partial}{\partial x}\left(k_{e}^{I}\frac{\partial T_{e}^{I}}{\partial x}\right) - G(T_{e}^{I} - T_{l}^{I}) + S^{I}$$
(4)



Fig. 1. Schematic of a two-layered film. The thickness of two-layered film is L = 200 nm. The thickness of top layer is *l*.

$$C_{l}^{1}\frac{\partial T_{l}^{1}}{\partial t} = \frac{\partial}{\partial x}\left(k_{l}^{1}\frac{\partial T_{l}^{1}}{\partial x}\right) + G(T_{e}^{1} - T_{l}^{1})$$
(5)

$$C_{\rm e}^{\rm II} \frac{\partial T_{\rm e}^{\rm II}}{\partial t} = \frac{\partial}{\partial x} \left(k_{\rm e}^{\rm II} \frac{\partial T_{\rm e}^{\rm II}}{\partial x} \right) - G(T_{\rm e}^{\rm II} - T_{\rm l}^{\rm II})$$
(6)

$$C_{l}^{II}\frac{\partial T_{l}^{II}}{\partial t} = \frac{\partial}{\partial x}\left(k_{l}^{II}\frac{\partial T_{l}^{II}}{\partial x}\right) + G(T_{e}^{II} - T_{l}^{II})$$
(7)

To solve Eqs. (4)–(7), the following initial and boundary conditions must be used. Before irradiated by the laser pulse, the electron and lattice sub-systems were considered to be at the same initial temperature ($T_0 = 300$ K)

$$T_{\rm e}^{\rm l}(x,0) = T_{\rm l}^{\rm l}(x,0) = T_{\rm 0}$$
(8)

$$T_{\rm e}^{\rm II}(x,0) = T_{\rm l}^{\rm II}(x,0) = T_{\rm 0}$$
(9)

The energy of the convective and radiative losses from the front and back surfaces of the two-layer film, in addition, is negligible during the femtosecond transient. The boundary conditions are formulated, as follows

$$\frac{\partial T_{e}^{I}}{\partial x}\Big|_{x=0} = \left.\frac{\partial T_{e}^{II}}{\partial x}\right|_{x=L} = 0$$
(10)

$$\frac{\partial T_1^{\mathrm{I}}}{\partial x}\Big|_{x=0} = \frac{\partial T_1^{\mathrm{II}}}{\partial x}\Big|_{x=L} = 0$$
(11)

At the interface of the film (x=l), the two-layer thin film which is in perfect thermal contact. Therefore we set the boundary conditions of the interface, as follows

$$T_{e}^{I}\Big|_{x=l} = T_{e}^{II}\Big|_{x=l}$$
(12)

$$T_{l}^{I}\Big|_{x=l} = T_{l}^{II}\Big|_{x=l}$$
(13)

$$k_{e}^{I} \frac{\partial T_{e}^{I}}{\partial x}\Big|_{x=l} = k_{e}^{II} \frac{\partial T_{e}^{II}}{\partial x}\Big|_{x=l}$$
(14)

$$k_{l}^{1} \frac{\partial T_{l}^{1}}{\partial x} \bigg|_{x=l} = k_{l}^{II} \frac{\partial T_{l}^{II}}{\partial x} \bigg|_{x=l}$$
(15)

3. Results and discussion

The numerical method are first performed to calculate a 200 nm thick single-layer gold film and three two-layer films, i.e., a 100 nm thick gold layer padding on a 100 nm thick silver layer, a 100 nm-thick copper layer or a 100 nm thick nickel layer. The laser light

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