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Ultrafast investigation of electron dynamics in the gold-coated two-layer metal films

Anmin Chen [a], Laizhi Sui [a], Ying Shi [a], Yuanfei Jiang [a], Dapeng Yang [b,*], Hang Liu [a], Mingxing Jin [a,*], Dajun Ding [a]

[a] Institute of Atomic and Molecular Physics, Jilin University, Changchun 130012, People's Republic of China
[b] Key Laboratory of Geo-exploration Instrumentation Ministry of Education, College of Instrumentation & Electrical Engineering, Jilin University, Changchun 130012, People's Republic of China

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ABSTRACT

The gold-coated metal thin films are widely used in modern engineering applications. In this paper, the ultrafast electron dynamics of gold-coated two-layer thin films has been investigated by ultrafast time-resolved pump–probe experiment. The dependence of the surface electron temperature on the film structure was considered based on the two-temperature model at the different two-layer film structure. The effect of laser fluence (3, 6 and 17 mJ/cm^2), and two-layer film thickness (the thickness of 50 nm and 100 nm gold layer) is considered. The theoretical predictions are compared with experimental data, which agree well with both thermal model and transient reflectivity.

1. Introduction

With the development of ultrashort laser based on chirped-pulse amplification [1], it is possible to carry out powerful femtosecond laser systems. The femtosecond pulsed lasers are widely used in a variety of fields including material processing [2], pulsed laser deposition [3], molecular spectroscopy [4,5], ionization and dissociation of polyatomic molecules [6,7], and so on. For the sake of increasing the 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 [8]). The interaction of femtosecond laser and gold film has been a challenging research topic. During pulsed laser irradiation of gold film, the electron–electron interaction time is very short, on the order of femtoseconds, compared with electron–lattice interaction time, which is on the order of picoseconds. It has been assumed that the incident photon energy of the laser beam is absorbed instantaneously by the free electrons of the metal and is confined close to the surface. Hence, a strong nonequilibrium is created between the electrons and lattices. The thermal energy possessed by these "hot" electrons diffuses deeper into the film.

Optical pump–probe measurement using femtosecond laser has been proven to be a very sensitive tool for the investigation of electron dynamics in metals. They have been used to study many phenomena of fundamental and applied interests such as optical orientation of spin, ultrafast demagnetization, and ultrafast excitation of coherent lattices. Their potential for material characterization is illustrated by measurements of the electron–lattice coupling constant in metals [9], hot electron linear and angular momentum relaxation times and nonlinear susceptibility tensor components in metals, and the spin wave mode spectrum of nanomagnets. For the interaction of the laser and metal, the investigation of ultrafast electron dynamics investigation has been reported using the pump–probe measurement by many researchers [10–13]. In these electron dynamics studies, the single-layer metals have been used. The electron dynamics of the multi-layer metal has been investigated by Ibrahim et al. [14]. The Au/Cr two-layer film had been experimentally studied by Qiu and Tien [15]. And the Au/glass two-layer film had been experimentally studied by Wang and Ma et al. [16]. However, the researches of the ultrafast electron dynamics are still lacking for the multi-layer metal films.

In this paper, we report the experimental results of the transient reflectivity of the gold-coated two-layer film using the femtosecond pump–probe technique for three different pump powers. Experimental results show that the reflectivity change increases with the power of the pump laser. Numerical solutions of the two-temperature model (TTM) are compared with experimental results. The distributions of electron temperature and lattice temperature are considered. The results show that the substrate layer chrome film can influence the variation of gold film temperature.

* Corresponding authors.
E-mail addresses: ydp@jlu.edu.cn (D. Yang), mxjin@jlu.edu.cn (M. Jin).

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2. Mathematical model

The theoretical method to investigate the ultrashort laser–matter interaction is the well-known two-temperature model [17]. 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 [18]. The one-dimension two-temperature equation is given below [19,20]:

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

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

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 [21], and S is the laser heat source. The heat source S can be modeled with a Gaussian temporal profile [22]:

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

Where $R=0.369$ (the wavelength of the pump beam is 400 nm) is the target reflection coefficient, t_p is the full-width at the half maximum (FWHM) with the linear polarization, α is the absorption coefficient and I is the incident energy, $\beta=4\ln(2)$.

The reflectivity (R) of metal is mainly due to the Drude free electron model. The electrical permittivity ε (dielectric function) of metals modeled as a plasma, is expressed as [23]

$$\varepsilon=\varepsilon_1+i\varepsilon_2 \tag{4}$$

$$\varepsilon_1=1-\frac{\omega_p^2\tau^2}{1+\omega^2\tau^2} \tag{5}$$

$$\varepsilon_2=\frac{\omega_p^2\tau^2}{\omega(1+\omega^2\tau^2)} \tag{6}$$

ω is laser frequency, $\omega_p=\sqrt{n_e e^2/(m_e\varepsilon_0)}$ is the plasma frequency, n_e is the density of the free electrons, m_e is the mass of electron and ε_0 is the electrical permittivity of free space. τ is the electron relaxation time. In general, for good conductors, the e–e collision rate may be determined by $\upsilon_{e-e}=AT_e^2$ whereas the e–ph collision rate is independent of T_e, but proportional to T_l, namely, $\upsilon_{e-ph}=BT_l$. Here A and B are constants, and both contribute to the electron collision frequency υ. A relationship between the electron relaxation time τ and the e–e and $e-ph$ collision rates for electron temperatures below the Fermi temperature is given by

$$\frac{1}{\tau}=\upsilon=\upsilon_{e-e}+\upsilon_{e-ph}=AT_e^2+BT_l. \tag{7}$$

We then work out n and κ, the real and imaginary parts of the complex refractive index. These are:

$$\varepsilon_1=n^2-\kappa^2 \tag{8}$$

$$\varepsilon_2=2n\kappa \tag{9}$$

and

$$n=\sqrt{\frac{\varepsilon_1+\sqrt{\varepsilon_1^2+\varepsilon_2^2}}{2}} \tag{10}$$

$$\kappa=\sqrt{\frac{-\varepsilon_1+\sqrt{\varepsilon_1^2+\varepsilon_2^2}}{2}}. \tag{11}$$

The reflectivity depends on both n and κ and is given by

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

The absorption coefficient is determined by κ, and is given by

$$\alpha=\frac{2\kappa\omega}{c}. \tag{13}$$

The electron heat capacity is proportional to the electron temperature when the electron temperature is less than the Fermi temperature as $C_e=\gamma T_e$ [24] and $\gamma=\pi^2 n_e k_B/2T_F$. n_e is the density of the free electrons, k_B is the Boltzmann's constant and T_F is Fermi temperature. 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 $k_e=k_{e0}BT_e/(AT_e^2+BT_l)$ [25], where k_{e0}, A and B are the constants. Many of the ultrafast laser heating analyses have been carried out with a constant electron–lattice 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 ($G=G_0(A(T_e+T_l)/B+1)$, where G_0 is the coupling factor at room temperature) [26]. 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 due to electrons [27]. As the temperature changes, the variety of the lattice heat conductivity is relatively small and it is assumed 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 (Eqs. (1) and (2)) for studying thermal behavior in the thin film can be expressed as

$$C_e^{I}\frac{\partial T_e^{I}}{\partial t}=\frac{\partial}{\partial x}\left(k_e^{I}\frac{\partial T_e^{I}}{\partial x}\right)-G\left(T_e^{I}-T_l^{I}\right)+S^{I} \tag{14}$$

$$C_l^{I}\frac{\partial T_l^{I}}{\partial t}=\frac{\partial}{\partial x}\left(k_l^{I}\frac{\partial T_l^{I}}{\partial x}\right)+G\left(T_e^{I}-T_l^{I}\right) \tag{15}$$

$$C_e^{II}\frac{\partial T_e^{II}}{\partial t}=\frac{\partial}{\partial x}\left(k_e^{II}\frac{\partial T_e^{II}}{\partial x}\right)-G\left(T_e^{II}-T_l^{II}\right) \tag{16}$$

$$C_l^{II}\frac{\partial T_l^{II}}{\partial t}=\frac{\partial}{\partial x}\left(k_l^{II}\frac{\partial T_l^{II}}{\partial x}\right)+G\left(T_e^{II}-T_l^{II}\right). \tag{17}$$

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 are considered to be at the same initial temperature ($T_0=300$ K)

$$T_e^{I}(x,0)=T_l^{I}(x,0)=T_0 \tag{18}$$