



Ultrafast thermalization dynamics in two-layer metal films excited by temporally shaped femtosecond laser



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ABSTRACT

We theoretically investigated ultrafast thermalization dynamics in two-layer Au coated metal films excited by temporally shaped femtosecond laser. It is revealed that the thermalization processes at surface and interface of the two-layer metal films can be alternatively promoted via tailoring pulse separation and pulse energy ratio of temporally shaped double pulses. The results are explained as competitive electron–phonon coupling dynamics in the two-layer metal films, which can be well manipulated by temporally shaped femtosecond laser. This study provides basic understanding for optimizing laser thermal damage in layered metal films via tailoring femtosecond laser for a wide range of applications in the fields such as laser resonant cavity, photoelectric equipments, MEMS and biochips.

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1. Introduction

In recent years, the temporally shaped femtosecond laser, namely multi-pulse sequences with variable pulse separation have been attracting more and more interest in the community of laser-material processing. The high ablation efficiency, less thermal recast and high precision have been experimentally demonstrated via temporally shaped femtosecond laser processing of materials [1–4], which exhibits large advantages over single femtosecond laser pulse ablation. It is generally accepted that the potential advantages of temporally shaped femtosecond laser processing of materials are physically originated from the ultrafast thermalization dynamics, in which the laser energy deposition can be well controlled via tailoring temporal profiles of femtosecond laser pulses, leading to the potential merits of localized energy deposition and high processing precision.

Most of previous theoretical works focused on investigations of the ultrafast thermalization dynamics in single-layer medium via temporally shaped femtosecond laser [5–7]. Compared to single-layer medium, the multi-layer system can well serve as the ideal structure for satisfying the thermal, optical and electronic requirements in development of MEMS, photoelectric equipments and biochips [8–10]. The ultrafast thermalization dynamics in multi-layered metal mediums are significantly distinguished from that

of single-layer metal film due to the complex thermal dynamics at layer interface during femtosecond-to-picosecond thermal transfer periods [11–15]. For ultrafast laser excitation of two-layer metal films, the electrons system of surface layer is excited violently, which occurs on femtosecond timescale. Following that, the excited electrons dissipate its energy to local phonon and inner electrons system via two distinct ways of electron–phonon coupling and electron thermal diffusion processes. The previous important works are mainly related to single femtosecond pulse and the conventional femtosecond laser heating of layered metal films, or called repetitive-pulses heating. For example, Brian and Dai et.al. proposed a finite element-finite difference method for solving the heat transport equations in double-layered thin films via conventional femtosecond laser heating [16–18]. Those studies can well reveal the electron and phonon temperature distributions in two-layer metal films with a conventional femtosecond laser irradiation. The conventional femtosecond laser can usually output the repetitive-pulse with MHz or kHz repetition frequencies. It indicates that the pulse separation for a conventional femtosecond laser pulses train is on the timescale of μs and ms . Because the Fourier thermal diffusion can normally occur on the timescales of ns to ms. It potentially causes the Fourier thermal diffusion during the multi-pulses interval. As a result, the thermal effects can be usually presented during the conventional femtosecond laser heating process, leading to inaccurate ablation. As a temporally shaped femtosecond laser excitation of two-layer metal films, the thermalization dynamics across the layer interface trends to be even more complex compared to that of conventional ultrafast

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laser excitation. It is expected that the laser energy deposition and the ultrafast thermalization processes across the interface of two-layer metal films can be closely dependent on the parameters of temporally shaped femtosecond laser, such as pulse separation, pulse energy ratio, etc. It requires the further theoretical insight into the ultrafast laser heating process in order to avoid the Fourier thermal diffusion for high-quality micro and nanofabrication via a temporally shaped pulses ablation. However, to the best of our knowledge, the ultrafast thermalization dynamics across the layer interface of layered metal systems with respect to temporally shaped femtosecond laser excitation is still less investigated so far.

In this paper, we theoretically investigated the ultrafast thermalization processes across two-layer Au coated metal films excited by temporally shaped double femtosecond pulses. The non-equilibrium thermalization dynamics with respect to the relevance of pulse-to-pulse energy coupling into layered systems is proposed for exploring the ultrafast thermalization features in Au coated metal films. The effect of pulse separation and pulse energy ratio on the thermalization processes of Au coated two layer metal films are examined in details. The study provides strategy for optimizing thermal dynamics processes in layered metal film systems via tailoring femtosecond laser pulses.

2. Modeling and discussions

Due to the transient modifications of electron temperature during femtosecond laser excitation of metal film, the dielectric function of the metal target changes fast with time. Especially, as the temporally shaped femtosecond laser excitation of metal films, the dielectric function can be unprecedentedly modulated via the temporal parameters of laser pulses. As a result, the dynamic dielectric function with respect to temporally shaped laser excitation must be taken into account for well exploring the ultrafast thermalization processes in the two-layer Au coated metal films. The temperature-dependent complex dielectric function can be split into the real and imaginary parts as follows:

$$\varepsilon = 1 - \frac{\omega_p^2}{\omega^2 + \nu_m^2} + i \frac{\nu_m}{\omega} \frac{\omega_p^2}{\omega^2 + \nu_m^2} \quad (1)$$

where $\omega_p^2 = \frac{e^2 n_e}{\varepsilon_0 m_e}$ denotes the plasma frequency. According to Matthiessen's rule within the relaxation time approximation, the total scattering rate of electrons is the sum of the rates of the separate mechanisms: $\nu_m = 1/\tau_{ee} + 1/\tau_{ep}$, where τ_{ee} and τ_{ep} are the electron-electron and electron-phonon scattering times, depending on electron and phonon temperatures [19]. The electron and phonon temperature changes in metals abide by the two-temperature model, which is widely accepted for investigations of ultrashort laser interaction with metal films. For ultrafast laser excitation of the two-layer Au coated metal films, the electron system of surface Au film is excited violently, which occurs on femtosecond timescale. Following that, the excited electrons dissipate its energy to the phonon of bottom layer metal film via two distinct routes of the electron thermal diffusion across layer interface and the electron-phonon coupling in the bottom layer. Here, we introduce the two-temperature model, in which the dynamic Drude model with temperature-dependent thermal and optical parameters are taken into account for investigations of the thermal excitation processes in two-layer metal film with respect to relevance of double pulse energy coupling into targets. The two-temperature model with considering dynamic parameters can be presented as follows [20,21]:

$$C_e [T_e, T_p] \frac{\partial T_e}{\partial t} = \nabla \cdot (K_e [T_e, T_p] \nabla T_e) - G [T_e, T_p] (T_e - T_p) + Q \quad (2)$$

$$C_p \frac{\partial T_p}{\partial t} = G [T_e, T_p] (T_e - T_p) \quad (3)$$

Here, the coupled equations (2) and (3) describe femtosecond laser excitation and the following thermal relaxation processes.

For a wide range of electron temperature from 300 K to Femi temperature, the temperature-dependent electron heat conductivity during the two temperature relaxation processes is expressed as follows [22]:

$$K_e(T_e, T_p) = \chi \frac{(\theta_e^2 + 0.16)^{5/4} (\theta_e^2 + 0.44) \theta_e}{(\theta_e^2 + 0.092)^{1/2} (\theta_e^2 + \eta \theta_p)} \quad (4)$$

An analytical expression of the electron-phonon coupling strength was proposed by Zhang and Chen, which can be represented as follows [23]:

$$G[T_e, T_p] = G_0 [A_e/B_p (T_e + T_p) + 1] \quad (5)$$

where, G_0 is the electron-phonon coupling strength in room temperature, and the coefficients A_e and B_p are constants. The laser energy absorption rate Q is

$$Q(x, t) = \sqrt{\frac{4 \ln 2}{\pi}} \frac{1-R}{(\delta + \delta_b)} \sum_{i=1}^n \frac{F_i}{t_{pi}} \times \exp \left[\left(-\frac{x}{\delta + \delta_b} \right) - 4 \ln 2 \left(\frac{t - 2t_{pi} - (i-1)\Delta}{t_{pi}} \right)^2 \right] \quad (6)$$

Here, R is the Au film surface reflectivity, which is temperature and wavelength dependent parameter, t_{pi} is the FWHM (full width at half maximum) pulse duration of the respective pulses of temporally shaped pulse sequence. δ is the optical penetration depth, and δ_b is the electron ballistic transfer length for Au film, taken as 100 nm. F_i is the laser fluence of respective pulses of temporally shaped pulse sequence, which can be taken as the optional values in our simulations. n is the pulse number and Δ denotes the temporal separation between pulses. Considering the relationship between complex reflective index, n_c and complex dielectric function, $n_c = \sqrt{\varepsilon} = \sqrt{\varepsilon_1 + i\varepsilon_2}$, and applying the Fresnel law at the surface, we get the temperature-dependent reflectivity coefficient as follows:

$$R(T_e, T_p, \omega) = \frac{[\text{Re}(n_c) - 1]^2 + [\text{Im}(n_c)]^2}{[\text{Re}(n_c) + 1]^2 + [\text{Im}(n_c)]^2} \quad (7)$$

The temperature-dependent penetration depth of laser intensity into Au film by the metal plasma absorption through the free electron heating is calculated by

$$\delta(T_e, T_p, \omega) = \frac{2\omega \text{Im}(n_c)}{c} \quad (8)$$

During femtosecond-to-picosecond period, it is reasonable to assume that the heat energy loss from the film system to the surrounding as well as to the front surface can be neglected, and the perfect thermal insulation is assumed at the rear surface of two-layer films. Therefore, the exterior boundary conditions for the two-layer metal film can be written as

$$\left. \frac{\partial T_e^I}{\partial x} \right|_{\Omega} = \left. \frac{\partial T_e^{II}}{\partial x} \right|_{\Omega} = 0 \quad (9)$$

$$\left. \frac{\partial T_p^I}{\partial x} \right|_{\Omega} = \left. \frac{\partial T_p^{II}}{\partial x} \right|_{\Omega} = 0 \quad (10)$$

where, I denotes first layer film, and II denoting the second layer film. Ω indicates the exterior boundary of the two-layer film. For the interior interface of the two-layer film system, during femtosecond-to-picosecond period, the electron system exhibits perfect thermal conductance due to fast electrons coupling on sub-picosecond timescale. However, the phonon system presents incontinuous

thermal contact at interface due to quite large phonon thermal capacity compared to that of electron system during femtosecond-to-picosecond period. Therefore, the interior boundary conditions of the two-layer systems can be written as follows:

$$T_e^I|_{\Gamma} = T_e^{II}|_{\Gamma} \quad (11)$$

$$k_e^I \frac{\partial T_e^I}{\partial x}|_{\Gamma} = k_e^{II} \frac{\partial T_e^{II}}{\partial x}|_{\Gamma} = 0 \quad (12)$$

Because of the flexibility of Finite Element Method (FEM) in dealing with heat transfer equations, the coupling partial differential Eqs. (1) and (2) are simultaneously solved by the Finite Element Method. In the current FEM simulations, three main steps are employed for the calculation procedures. Firstly, we build the geometrical domain of a 2-D Au film with size of $5 \mu\text{m} \times 1 \mu\text{m}$; Secondly, the mesh generation procedure is carried out to divide the 2-D geometry into small patches. Following that, we introduce the mathematical expression into the meshed 2-D layered geometry, and solving the mathematical expression using FEM procedures package.

3. Results and discussion

The 2-D temperature fields evolutions for electrons and phonons of two-layer Au/Ni film triggered by temporally shaped double femtosecond pulses are shown in Fig. 1. The main material parameters used in the model are as follows: For Au film, $G_0 = 0.21 \times 10^{17} \text{ J m}^{-3} \text{ s}^{-1} \text{ K}^{-1}$, $C_p = 2.5 \times 10^6 \text{ J m}^{-3} \text{ K}^{-2}$, $A_e = 1.18 \times 10^7 \text{ s}^{-1} \text{ K}^{-2}$, $B_p = 1.25 \times 10^{11} \text{ s}^{-1} \text{ K}^{-1}$; For Ni film, $G_0 = 3.6 \times 10^{17} \text{ J m}^{-3} \text{ s}^{-1} \text{ K}^{-1}$, $C_p = 4.1 \times 10^6 \text{ J m}^{-3} \text{ K}^{-2}$, $A_e = 0.59 \times 10^7 \text{ s}^{-1} \text{ K}^{-2}$, $B_p = 1.4 \times 10^{11} \text{ s}^{-1} \text{ K}^{-1}$. It can be seen that the electron system of surface Au layer of Au/Ni film is heated at 100 fs, but the phonon system across the Au/Ni film keeps undisturbed at this time. At 1.5 ps, the electron system temperature of Au layer presents obvious drop, simultaneously, the phonon system temperature of Au layer gets slight rise due to participating of the electron–phonon coupling process during this period. More interestingly, the Ni layer phonon is slightly perturbed at this time. At 2 ps, a time of arrival of the second pulse, the electron system of Au layer is re-excited violently, leading to significant promotion of electron temperature of Au film. Because of persistent electron–phonon coupling process, the phonon system of Au/Ni film

is obviously heated at 2 ps. At 5 ps, the electron temperature of Au layer presents observable drop, however, the phonon temperature of Ni layer exhibits dramatic rise, which is attributed to the dominant electron–phonon coupling process in Ni layer compared to Au layer. The phonon temperature jump at layer interface originates from the dominant phonon heating at Ni layer during the electron–phonon coupling period. In fact, the Au/Ni film heating can be split into two steps. Firstly, the electron system of Au layer surface is excited on femtosecond timescale, simultaneously the electrons diffuse its energy to Ni layer via electron thermal diffusion across the layer interface. Following that, the phonon systems of Au/Ni film is heated due the electron–phonon coupling processes in respective layers. However, the phonon diffuses across the interface can be very tiny compared to the electron thermal diffusion due to quite weaker phonon thermal diffusion across the Au/Ni interface on the sub-picosecond to picosecond timescale. Considering the fact that the electron–phonon coupling strength for Ni ($3.6 \times 10^{17} \text{ J m}^{-3} \text{ s}^{-1} \text{ K}^{-1}$) is an order of magnitude larger than that of Au ($0.21 \times 10^{17} \text{ J m}^{-3} \text{ s}^{-1} \text{ K}^{-1}$). Therefore, the Ni layer phonon system can be dominantly heated due to the strong electron–phonon coupling in Ni layer, which potentially leads to the phonon temperature jump phenomenon. With time, the electron and phonon systems will ultimately reach thermal equilibrium across the two-layer film at tens of picoseconds. As the phonon temperature is high enough to reach the damage threshold for Au and Ni, the film system will be ablated as a result of the phase explosion mechanism.

The electron and phonon temperature field distributions at laser spot center along the depth of two-layer Au/Ni film at different delay time is shown in Fig. 2. We can see that the electron and phonon temperature distributions exhibit obvious dependences on the temporal separation of double femtosecond pulses. For the given laser fluence of 0.3 J/cm^2 , the temporal separation of 10 ps leads to the obvious increase of the electron temperature at surface layer (depth = 0) (Fig. 1(a)). However, the electron temperature at interface is slightly restrained for the pulse separation of 10 ps. Our previous investigations had showed that the electron–phonon coupling process can be manipulated by adjusting pulse separation of the temporally shaped femtosecond pulses [24]. The optimal pulse separation of 10 ps accounts for the electron–phonon coupling period with single pulse excitation for current laser parameters, beyond which the laser energy coupling into surface layer is weakened due to the crippling of double pulse incubation effect. On the contrary, the phonon system at interface of Au/Ni film is

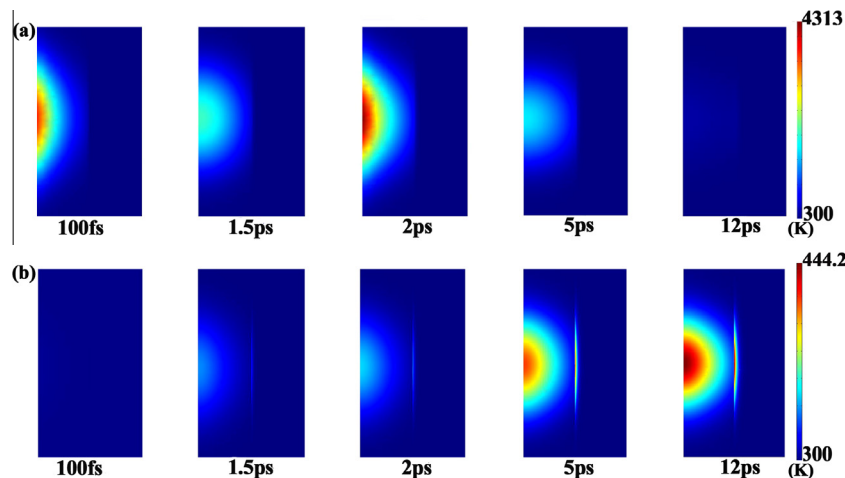


Fig. 1. The 2-D temperature fields evolutions for electrons and phonons of two-layer Au/Ni film triggered by temporally shaped double femtosecond pulses. $F_1 = F_2 = 0.1 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 30 \text{ fs}$, $\Delta = 2 \text{ ps}$. (a) Electron system and (b) Phonon system. The size of the 2-D two-layer film is $5 \mu\text{m} \times 1 \mu\text{m}$, and the thickness of surface Au layer and bottom Ni layer is $0.5 \mu\text{m}$, respectively.

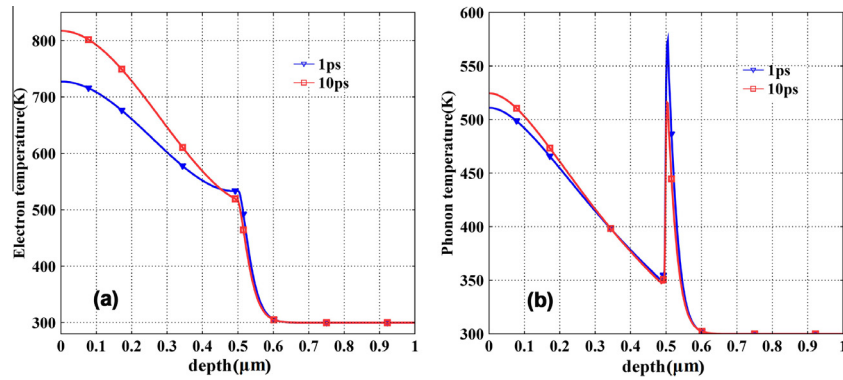


Fig. 2. The equilibrium electron–phonon temperature field distributions at laser spot center along the depth of two-layer Au/Ni film for temporally shaped double pulse excitation with different pulse separations. $F_1 = F_2 = 0.15 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 30 \text{ fs}$. (a) indicates the temperature field distribution for electron system at 14 ps; (b) denotes the temperature field distribution for electron system at 18 ps.

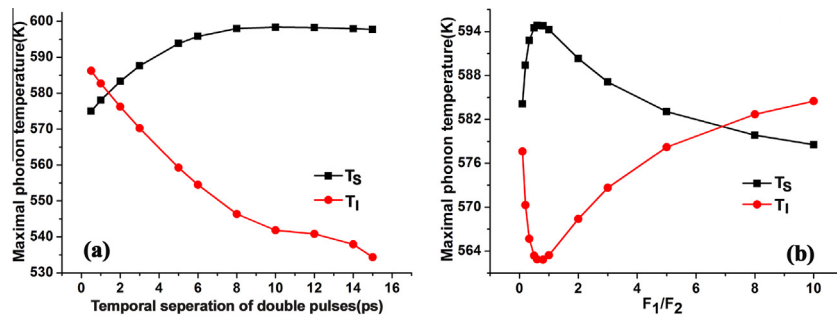


Fig. 3. The maximal surface and interface phonon temperatures for two-layer Au/Ni film as functions of temporal separation and pulse energy ratio of temporally shaped double femtosecond pulses. (a) $F_1 = F_2 = 0.2 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 30 \text{ fs}$; (b) $F_1 + F_2 = 0.4 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 30 \text{ fs}$.

preferentially heated at pulse separation of 1 ps. However, the surface phonon temperature is slightly lowered at the pulse separation of 1 ps. It indicates that the ultrafast thermalization dynamics in two-layer metal films can be well manipulated by adjusting pulse separation of temporally shaped femtosecond laser.

The maximal surface and interface phonon temperatures for two-layer Au/Ni film as functions of temporal separation and pulse energy ratio of temporally shaped double femtosecond pulses are shown in Fig. 3. It can be seen that the surface phonon temperature of Au/Ni film rises rapidly with increasing the pulse separation as it is less than 10 ps. Inversely, the interface phonon temperature of Au/Ni film exhibits obvious drop as the pulse separation is less than 10 ps. The phonon system can be disturbed via two energy transfer routes of the electron thermal diffusion across interface and the electron–phonon coupling at respective layers. The alternative thermalization at surface and interface of two layer Au/Ni film can be attributed to the competitive electron–phonon coupling dynamics at respective layers of Au/Ni film. As the electron–phonon coupling at the surface Au layer dominates the energy transfer, the electron thermal diffusion into the bottom layer film can be impaired due to energy conservation law, leading to weakened electron–phonon coupling process in Ni layer. As a result, the thermalization of localized phonon for Au/Ni film is restrained in Ni layer, which is simultaneously promoted at surface Au layer. As seen in Fig. 3(b), the preferential heating of surface layer Au film occurs at the pulse energy ratio of 1:1. As the pulse energy ratio departs from 1:1, the electron–phonon coupling dynamics at Au layer is weakened. As a result, the preferential phonon thermalization in Ni layer is promoted due to the large electron–phonon coupling strength for Ni compared to Au.

It should be emphasized that the phonon heating experiences a period of picoseconds, even at the zero ps delay of double pulses. It means that the phonon temperature is not promoted immediately, rather than rises on picosecond timescale, depending on the laser parameters such as the pulse separation and pulse energy ratio of temporally shaped double pulses. The results provide basic understanding for thermal loading processes during layered metal films heating for satisfaction of thermal, optical and electronic

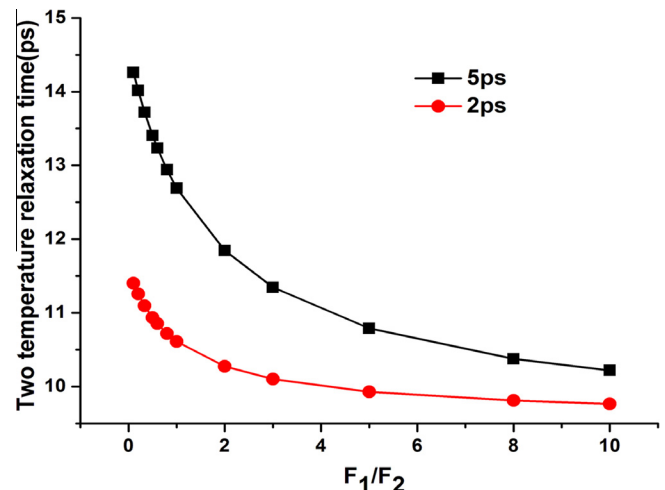


Fig. 4. The two-temperature relaxation time for two-layer Au/Ni film as functions of temporal separation and pulse energy ratio of temporally shaped double femtosecond pulses. (a) $F_1 = F_2 = 0.1 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 60 \text{ fs}$; (b) $F_1 + F_2 = 0.3 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 60 \text{ fs}$.

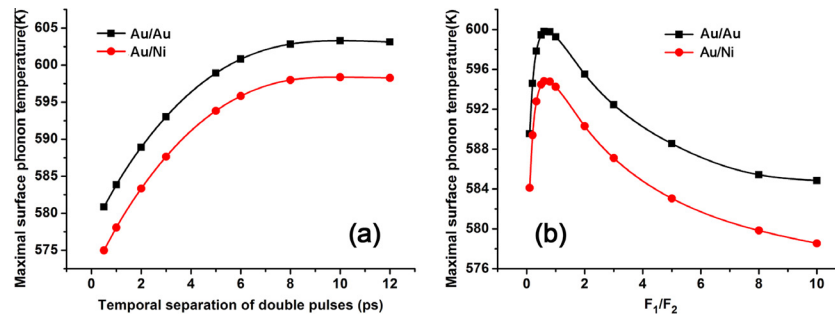


Fig. 5. The maximal phonon temperatures at surface of two-layer Au coated metal films as functions of temporal separation and pulse energy ratio of temporally shaped double femtosecond pulses. (a) $F_1 = F_2 = 0.2 \text{ J/cm}^2$, $\lambda = 800 \text{ nm}$, $t_{p1} = t_{p2} = 30 \text{ fs}$. (b) $F_1 + F_2 = 0.4 \text{ J/cm}^2$.

requirements in development of MEMs, photoelectric equipments and biochips.

The two-temperature relaxation time, which is defined as the non-equilibrium energy transfer period from excited electrons to lattices plays a crucial role in affecting thermalization dynamics in metals. It is generally accepted that a reduced two-temperature relaxation time, namely a large electron–phonon coupling strength can be extremely beneficial for enhancing the spacial localization of phonon thermalization in metals. Fig. 4 shows the two-temperature relaxation time for two-layer Au/Ni film as a function of the pulse energy ratio of temporally shaped double femtosecond pulses. We can see that the two-temperature relaxation time decreases rapidly with increasing the pulse energy ratio for the different pulse separations of 2 ps and 5 ps, respectively. Especially, as the pulse energy ratio is less than 1:1, the small pulse separation can lead to obvious drop of the two temperature relaxation time. However, as the pulse energy ratio is significantly increased (exceeding 5:1), the effect of pulse separation on the two temperature time becomes mild. The adjustment of the two temperature relaxation time is greatly beneficial for enhancing the localization of phonon thermalization. The results indicate that changing of pulse energy ratio and decrease of temporal separation can both cause modifications of the electron–phonon thermalization period, which is largely beneficial for regulating the spacial localization of phonon thermalization during the two-layer film ablation, leading to the potential merit of high fabrication precision.

The maximal phonon temperatures at surface of two-layer Au coated metal films as functions of temporal separation and pulse energy ratio of temporally shaped double femtosecond pulses are shown in Fig. 5. Here, the bottom layer metals are carefully selected as the typical transition Ni and noble metal Au. In fact, the Au/Au film can also be equally treated as a thick single layer medium. We can see that the phonon temperatures present similar tendency with varying temporal separation (Fig. 5(a)) and pulse energy ratio (Fig. 5(b)) for Au/Au and Au/Ni films. More interestingly, the maximal phonon temperature on surface of Au/Au film is always larger than that of Au/Ni film for different laser parameters. The different thermalization processes are physically originated from the distinct electron–phonon coupling strength for the bottom layer metal. In fact, the electron–phonon coupling strength for Ni is an order of magnitude larger than that of Au. The laser energy can be predominantly transferred to the bottom of Au/Ni film due to the strong electron–phonon coupling process in Ni layer compared to bottom Au layer for Au/Au film. As a result, the laser energy is dominantly transferred to phonon of Ni layer, leading to reduction of surface phonon temperature for Au/Ni film. It indicates that the different bottom layer configurations of two-layer Au coated film can change the absolute values of surface phonon temperature, but less influence general law of surface phonon temperature evolution. The results can be important for

understanding of the thermal loading process in two-layer metal films for well optimizing thermal deformation and thermal damage in layered metal films via irradiation of temporally shaped femtosecond laser.

4. Conclusion

In conclusion, we had theoretically investigated the ultrafast thermalization dynamics in two-layer Au coated metal films irradiated by temporally shaped double femtosecond laser pulses. It was revealed that surface phonon temperature of Au/Ni film rises rapidly with increasing the pulse separation as it is less than 10 ps. Inversely, the interface phonon temperature of Au/Ni film exhibits obvious drop as the pulse separation is less than 10 ps. Interestingly, the surface phonon temperature reaches peak for pulses energy ratio of 1:1. Simultaneously, the interface phonon temperature comes to minimum value at the optimal pulse energy ratio of 1:1. The results are explained as competitive electron–phonon coupling dynamics at respective layers of two-layer Au coated metal film, which can be manipulated by temporally shaped femtosecond laser. The main benefits of this work can be related to the controllable heating process at surface and interface layers of two-layer metallic film via temporally shaped femtosecond laser, which is beneficial for understanding of the thermalization dynamics for satisfying thermal requirements in development of MEMS, photoelectric equipments and biochips.

Conflict of interest

None declared.

Acknowledgments

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