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Ultrafast dynamics of laser thermal excitation in gold film triggered by temporally shaped double pulses

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ABSTRACT

We present the theoretical studies on ultrafast thermal excitation dynamics of gold film irradiated by temporally shaped femtosecond laser double pulses. It is revealed that the gold film thermalization efficiency can be greatly promoted by optimizing the pulse energy ratio, pulse separation and pulse exchange of temporally shaped femtosecond double pulses. The phenomena are explained as the enhanced laser energy coupling into the electron subsystem due to the interplay of pulse-to-pulse energy coupling. This study should be helpful for basic understanding of the ultrafast thermal dynamics for optimizing the gold film excitation process via shaping femtosecond laser for the potential applications such as thermal plastic deformation effects, laser ablation and laser excitation spectrum.

1. Introduction

In recent years, laser thermal excitations via temporally shaped laser pulses has been attracting more and more attention due to the interesting synergetic effects arising from the relevance of the pulse-to-pulse energy coupling into target material [1–4]. Experiments with several delayed femtosecond laser pulses were demonstrated to be advantageous in laser material processing [5–8]. For example, the ablation morphology can be optimized and ablation efficiency is promoted by controlling the temporal separation of shaped femtosecond laser pulses. Further, Donnelly et al. experimentally explained the nickel ablation features with double pulse irradiation, demonstrating that the maximum ablated volume of metallic nickel occurs at the optimized pulse separation from 10 ps to 100 ps [9].

Although the progressive works observed in experiment for demonstrating the advantages of double pulse ablation using temporally shaped laser, the potential mechanisms for the temporally shaped laser excitation of metal films in the early stage of the ablation are still puzzling for well optimizing the dynamics processes. In the early stage of the temporally shaped femtosecond laser interaction of metals, the metastable electron subsystem between the delayed pulses can play an important role in the thermal excitation dynamics. The metastable electron subsystem in gold film produced by pre-pulse of the delayed double pulses will definitively affect the total laser energy coupling into metals and finally the target ablation characteristics. For well describing the metal thermal dynamics upon temporally shaped femtosecond laser excitation, the two-temperature model (TTM) must be properly modified to contain the non-equilibrium thermal relaxation dynamics of the metastable electron subsystem between the pulse-to-pulse separation for accommodating the actual thermal excitation processes. Most of previous works considering the effects of the pulse delay (or pulse separation) on material processing are mainly based on experimental explorations [10–13]. Although Jiang et al. proposed a dynamics model for investigations of the pulse train excitation of metals [14], the model mostly consider the effect of pulse separation on material heating process. In fact, the gold film thermal excitation process can be not only affected by pulse separation, but also by the even more configurations such as pulse energy ratio, pulse exchange, etc. Therefore, it becomes very urgent to understand the fundamental dynamics of temporally femtosecond laser interactions with gold film for well optimizing the ablation process. However, to the best of our knowledge, the laser thermal excitation in gold film with respect to the temporally shaped pulse configurations of pulse energy ratio, pulse exchange are less investigated so far.

In this paper, we theoretically investigated the ultrafast thermal excitation process in gold film triggered by temporally shaped femtosecond laser double pulses. The transient modifications of

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gold film excitation arising from pulse-to-pulse relevance are presented to reveal the potential energy coupling process. The effects of temporally shaped double pulses excitation on gold film thermalization processes are examined in details. This study will be beneficial for optimizing the thermalization process of temporally shaped femtosecond laser interaction of metals, providing for basis for advancing a wide range of applications such as ablation, surface detections and laser excitation spectrum.

2. Modeling and methods

Due to the transient modifications of the temperature of the electron subsystem during the ultrafast laser excitation of gold film, the gold dielectric function can be changed by the electron temperature. As a result, the dielectric function of gold film can be significantly modulated via manipulating the temporally shaped femtosecond laser. Therefore, the temperature-dependent dielectric function must be taken into account for exploring gold ablation dynamics via temporally shaped laser. The temperature-dependent complex dielectric function can be split into the real and imaginary parts as follows [15]:

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

where $\omega_p^2 = e^2 n_e/\varepsilon_0 m_e$ 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_{e-e} and τ_{e-p} are the electron–electron and electron–phonon scattering times, depending on electron and phonon temperatures [16]. The temperature-dependent Drude model can be physically important for investigations of the transient optical reflectivity and absorption. The dynamics process of laser energy coupling into gold film under the excitation of double femtosecond pulses can therefore be well predicted with respect to the temperature-dependent Drude model. The electron and phonon temperature changes in gold film comply with the two temperature model, which is most widely accepted for investigations of ultrashort laser interaction with all kinds of materials, especially the metal targets. As double femtosecond laser pulses interactions with gold target, the intermediate electron system excited by the pre-pulse can essentially affect the laser energy coupling with the excited high-temperature electrons system of gold film. Here, we introduce the extended Two Temperature Model (TTM), in which the temperature dependent thermal and optical parameters are fully taken into account for investigations of the thermal excitation dynamics of gold film. For investigations of femtosecond laser thermal excitation in gold film, it is reasonable to assume that the hydrodynamics process can be ignored because of the quite longer hydrodynamics period compared to the electron–phonon coupling period (ps timescale). As a result, the validity of the TTM can be assured for the chosen picosecond time domain, which is appropriate for description of the nonequilibrium thermal excitation process in gold film. Here, the density variation effects due to the hydrodynamic expansion is also ignored on picosecond timescale for investigations of isochoric heating of gold film excited by temporally shaped double femtosecond pulses. Before material eruption, the energy release in gold film is mainly dominated by the electron–phonon relaxation mechanism. In such situation, the maximum heating efficiency with respect to double pulse separation can exhibit dissimilar behavior from the eruption process. Different from the ablation process that considers the hydrodynamics dynamics with respect to pulse-to-pulse coupling into erupted plume, the current model concentrates on the heating efficiency with respect to the synthetic effect of laser energy coupling into non-expansion gold film. The current works can well predict the thermal dynamics process in the early stage in non-swelling gold film with respect to double pulse separation, which can serve for optimizing the nondestructive material thermal excitation applications. The thermal excitation processes of femtosecond laser interactions with metals are described as follows:

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

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

Here, the coupled Eqs. (2) and (3) describe the ultrashort laser pulses excitation and the following thermal relaxation process. In fact, the excited electron system can uncouple with phonon system on femtosecond timescale, but relax with lattice on the timescale of picoseconds. The thermal properties of excited electron system such as the thermal capacity and thermal conductivity can be well defined at the non-equilibrium electron–phonon coupling state. In the current model, the piece function of electron thermal capacity C_e is applied, which considers the *s*- and *d*-band contributions in different temperature regimes as also found in our previous publication [16]. As the electron temperature is less than T_F/π^2, the electron thermal capacity is directly proportional to the electron temperature. In such situation, the *s*-band contribution plays a dominant role, and the *d*-band excitation can be completely ignored; For the electron temperature increasing from T_F/π^2 to $3T_F/\pi^2$, the *s*-band and *d*-band excitations can both contribute to the thermal capacity for gold film target. As the electron temperature increases from $3T_F/\pi^2$ to T_F, the *d*-band electron excitation dominates over the *s*-band contribution. In indicates that the thermal excitation of *d*-band can play a significant role as the electron temperature is high enough to approach the Fermi level. As a result, in the current model, the piece function of electron thermal capacity can describe the thermal dynamics in gold film for ablation with optimized variations in the 5% range (300 K–900 K).

For a wide range of electron temperature ranging from 300 K to Fermi temperature, the electron heat conductivity during the two temperature relaxation processes is written as follows [17]:

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

An analytical expression of the electron–phonon coupling strength was proposed by Zhang and Chen, which can be represented as follows [16]:

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

where, G_0 is the electron–phonon coupling strength in room temperature, and the coefficients A_e and B_p are constants.

In fact, the *d*-band excitation will lead to the increase of the electron–phonon coupling strength. Therefore, laser energy can be locally absorbed due to participating of d-band excitation. On the contrary, the d-band excitation may lead to a higher electron thermal conductivity than that without d-band electron, which in turn may result in strong diffusion of hot electrons.

The laser energy absorption rate Q is

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

Here, R is the gold 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 sequences. δ is the optical penetration depth, and δ_b is the electron ballistic transfer length for gold film, taken as 100 nm. F_i is the laser fluence of respective pulses of the temporally shaped pulse sequence, which can be taken as 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:

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

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

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

In the current model, the optical parameters of optical absorption, reflectivity and the thermodynamic parameters such as electron–phonon coupling strength and electron thermal conductivity can be transiently modified, which provides the possibility for enhancing laser energy coupling into gold film. Because of the flexibility of finite element method in dealing with the heat transfer equations, the partial differential Eqs. (1) and (2) are simultaneously solved by the Finite Element Method (FEM). In the current FEM simulations, three main steps are employed for the calculation procedures. Firstly, we build the geometrical domain of a 2D gold film with size of 8 μm × 200 nm; Secondly, the mesh generation procedure is carried out to divide the 2D gold geometry into patches. Following that, we introduce the mathematical expression into the meshed 2D gold film geometry, and solving the mathematical expression using FEM procedures package. The initial conditions for electrons and phonons are assumed to be room temperature. During the femtosecond-to-picosecond time period, it is reasonable to assume that heat losses from the metal film to the surrounding as well as to the front surface are neglected, and the perfect thermal insulation between gold film with substrate is assumed at the rear surface. In the current FEM simulations procedures, the gold film geometry is nonlinearly meshed across the laser spot space and the time step size is sett as 1 fs which is smaller than the laser pulse duration for precisely calculating the temporal evolution behavior of the ultrafast thermal excitation in gold film.

3. Result and discussions

The temporal evolution of phonon thermalization on gold film surface excited by the temporally shaped double femtosecond pulses with different pulse energy ratios are shown in Fig. 1. We can see that the phonon temperature exhibits piecewise increase with increasing delay time and appears turning point at delay time of

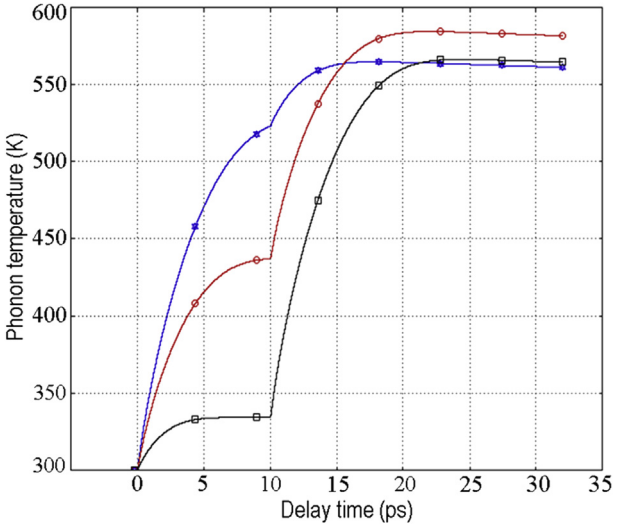

Fig. 1. The temporal evolution of phonon thermalization processes on gold film surface under temporally shaped double femtosecond pulses excitation with different pulse energy ratios. The circle-decorated line represents pulse energy ratio of 1:1, the square-decorated line represents 1:4, the star-decorated line represents 4:1. (The total laser fluence is 0.4 J/cm^2, the thickness of gold film is 0.2 um, $t_{p1} = t_{p2} = 30$ fs, $\Delta = 10$ ps).

10 ps. It originates from the two-step thermal excitation processes with the shaped double femtosecond pulses irradiation. More interestingly, we can see that the phonons subsystem exhibits discrepant thermalization processes for the different pulse energy ratios. The maximal phonon thermalization efficiency occurs at the optimal energy ratio of 1:1. When the pulse energy ratio departures from 1:1, the phonon thermalization efficiency is lowered sharply. In fact, the surface phonon temperature can exhibit slight drop on picosecond timescale after the electron–phonon relaxation termination due to the weak role of the Fourier thermal diffusion mechanism on picosecond time domain. Therefore, we have estimated the phonon temperature as the constants on tens of ps after the electron–phonon relaxation termination. The approximation can be reasonable for the thermal expansion in non-swelling gold film up to tens of ps.

Fig. 2 shows the maximal temperature of electrons and phonons on gold film surface as a function of energy ratio of shaped double pulses. We can see that the electron temperature reaches minimum at the energy ratio of 1:1 for the different laser fluences of 0.3 J/cm^2, 0.4 J/cm^2 and 0.5 J/cm^2, respectively [Fig. 2(a)]. However, as the pulse energy ratio is less than 1:1, the increase of pulse energy ratio will cause drop of electron temperature. Once the energy ratio exceeds 1:1, the electron temperature takes on a rapid increase with increasing pulse energy ratio. The phonon temperature achieves its maximum at the preferred energy ratio of 1:1 [Fig. 2(b)]. The phonon temperature variations with pulse energy ratio can be attributed to the alternative laser-electron subsystem coupling with respect to the pulse-to-pulse energy allocations. As the pulse energy ratio is less than 1:1, the pre-pulse is not competent for warm-up of gold film, leading to less formation of high-temperature electron system before the arrival of main pulse. As a result, the main pulse less exhibits enhanced coupling with gold film. However, as the pulse energy ratio is continuously increased and approaches 1:1, the high-temperature electrons can be affluently generated, the main pulse energy coupling with gold film is significantly enhanced due to the high-absorptive characteristic of high-temperature electron, leading to the promotion of the phonon temperature. Once the pulse energy ratio exceeds 1:1, although the

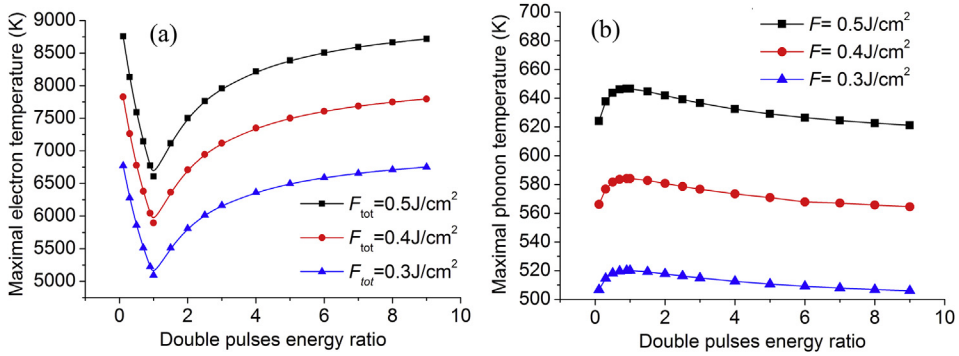

Fig. 2. The maximal temperatures of electron and phonon in gold film as a function of energy ratio of shaped double pulses. (The thickness of gold film is 0.2 um, $t_{p1} = t_{p2} = 30$ fs, $\Delta = 10$ ps).

pre-pulse energy is large enough for producing the high-temperature electrons, the small energy allocation of main pulse is not qualified for effective heating of gold film. Therefore, the phonon temperature presents slow drop.

In fact, the phonon heating is caused the electron–phonon coupling mechanism, meaning that the increase of the phonon energy is in the cost of losing the electron energy. It indicates that the lowering of the electron temperature will lead to rise of phonon temperature. The enhanced dynamics of the electron–phonon coupling can contribute to drop of electron temperature and synchronously promotion of phonon temperature. As a result, the counterintuitive situation of minimal electron temperature leading to maximal phonon temperature can be understood based on the fact that the enhanced electron–phonon coupling dynamics via manipulating double pulse parameters. It indicates that the optimal pulse energy ratio of 1:1 will be both beneficial for reducing the electron temperature and enhancing subsequent phonon excitation process, which potentially results in the promotion of the laser thermalization process in the gold film target.

The maximal phonon temperature on gold film surface as a function of pulse separation of the temporally shaped femtosecond double pulses is shown in Fig. 3. We can see that the phonon temperature incipiently presents rapid increase and eventually exhibits saturated rise. The maximal phonon temperature occurs at the optimal pulse energy ratio of 1:1. The dependences of phonon temperature on double pulse separation can be explained as follows: as the pulse separation is less than the electron–phonon relaxation time of gold film with single pulse irradiation, the

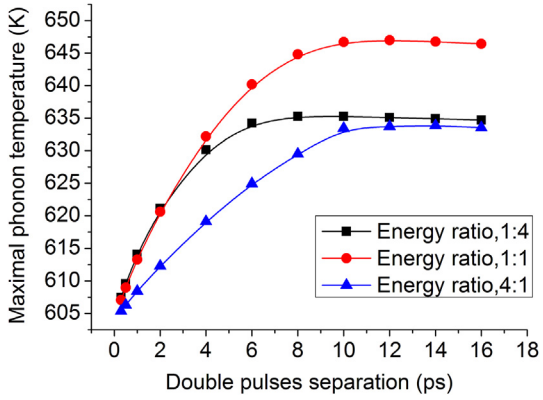

Fig. 3. The maximal phonon temperature as a function of pulse separation of shaped femtosecond laser double pulses (the total laser fluence is 0.5 J/cm^2, $t_{p1} = t_{p2} = 30$ fs, the thickness of gold film is 0.2 um).

excited high-temperature electrons state can less decay on gold film surface before the arrival of the following main pulse. The laser energy coupling with the excited electron subsystem can be enhanced significantly. As a result, the phonon temperature presents rapid increase with increasing the pulse separation of the temporally shaped double pulses. However, once the pulse separation exceeds the electron–phonon relaxation time of first pulse excitation of gold film (6 ps, 8 ps and 10 ps for energy ratios of 1:4, 1:1 and 4:1, respectively), due to the vanishing of high-temperature electron subsystem on gold film surface before the arrival of the main pulse, the pulse coupling enhancement does not occur during the second pulse excitation of gold film surface. As a result, as the pulse separation exceeds ~10 ps, the phonon temperature rise becomes tardy with increasing pulse separation and finally gets saturation. In addition, as the double pulses overlap, the double pulses perform like a whole big pulse, the heating efficiency gets the minimum compared to double pulse excitation of gold film.

Fig. 4 shows the maximal temperature of electron and phonon on gold film surface excited by double pulse configurations with exchanged position in time domain. We can see that the increases of electron and phonon temperature presents linear tendency with increasing fluence. More interestingly, the different pulse configurations can lead to completely opposite results of the electron and phonon temperature. For fs + ps double pulses irradiation, the electron temperature in gold film is significantly suppressed compared to that with ps + fs double pulses excitation [Fig. 4(a)]. On the contrary, the fs + ps double pulses excitation will lead to the promotion of phonon temperature [Fig. 4(b)]. The observable promotion of phonon temperature via optimizing double pulse duration configurations can be attributed to the enhanced laser energy inpouring into gold film arising from the relevance of pulse-to-pulse coupling with high-temperature electron subsystem on gold film surface. For fs + ps double pulses excitation, the femtosecond pulse pre-heats the gold film target, producing high-temperature electron subsystem on gold film surface, the second picosecond pulse then interacts with the excited high temperature electron system. In this way, the coupling of the second pulse with the high temperature electron can be enhanced as a result of the reduced reflectivity according the Drude model [17]. For ps + fs double pulses excitation, however, the relative low peak power of the pre-picosecond pulse is not qualified for production of high-temperature electron subsystem for enhancing the laser-gold film coupling. As a result, the following femtosecond pulse dose not exhibit enhanced energy coupling into gold film. In short, the studies fundamentally clarify the ultrafast thermal excitation dynamics for enhancing gold film thermalization efficiency with fs + ps double pulse excitation, which is proved to be superior over the ps + fs double pulse excitation.

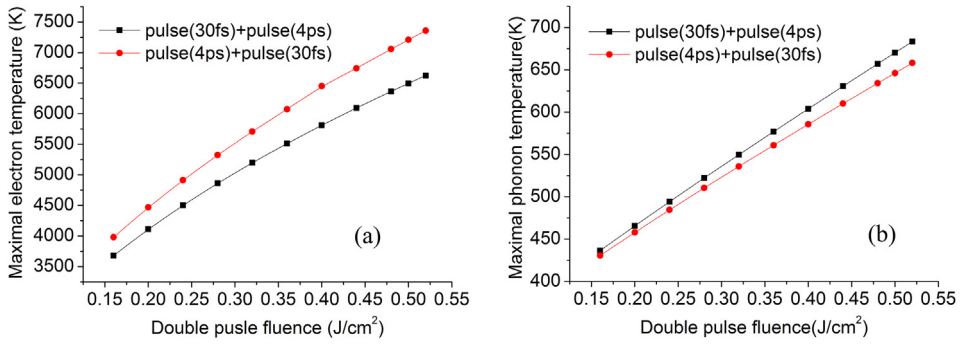

Fig. 4. The maximal temperatures of electron and phonon of gold film excited by double fs–ps and ps–fs pulses. (a) Electron temperature, (b) phonon temperature ($\Delta = 10$ ps, the thickness of gold film is 0.2 um).

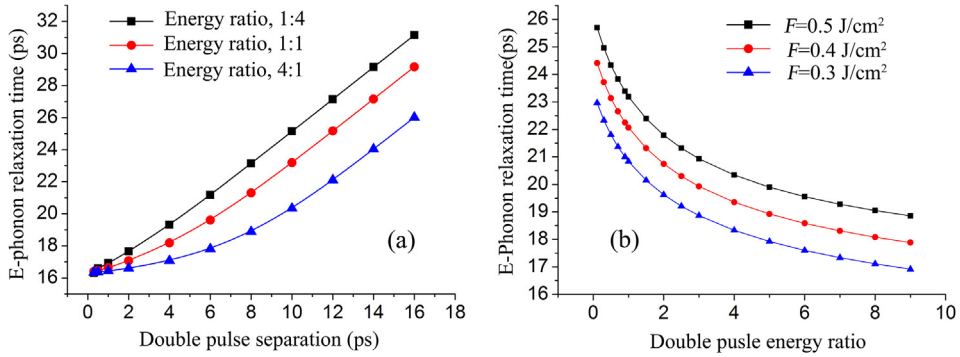

Fig. 5. The dependences of electron–phonon relaxation time (period) on the laser parameters (pulse separation and pulse energy ratio) of temporally shaped femtosecond laser double pulses. (a) The total double pulses fluence is 0.5 J/cm^2, $t_{p1} = t_{p2} = 30$ fs, the thickness of gold film is 0.2 um; (b) $t_{p1} = t_{p2} = 30$ fs, $\Delta = 10$ ps, the thickness of gold film is 0.2 um.

The electron–phonon coupling processes can play an important role in processing of metallic targets under ultrashort laser pulses irradiation in ablation regime. Our previous studies revealed that the electron–phonon relaxation processes can be significantly affected by laser fluence, pulse duration and film thickness [18]. Herein, we will present the investigations on the dependences of the electron–phonon relaxation time on the pulse energy ratio and pulse separation of the temporally shaped double femtosecond pulses. The electron–phonon relaxation time is defined as the energy exchange period between electrons and phonons after the temporally shaped double pulses irradiation. We can see that increase of the pulse separation will give rise to the increase of the electron–phonon relaxation time [Fig. 5(a)]. The difference of the electron–phonon relaxation time for different pulse energy ratios becomes more sharp as the pulse separation persistently increases. However, the increasing the pulse energy ratio will cause obvious drops of the electron–phonon relaxation time as shown in Fig. 5(b). The electron–phonon relaxation time presents increase tendency with increasing total fluence of the temporally shaped double femtosecond pulses. The results provide for the flexible route for controlling of the electron–phonon relaxation processes for optimal processing of gold film via adjusting the temporally shaped femtosecond laser.

4. Conclusion

We theoretically investigated the ultrafast thermal excitation processes in gold film irradiated by temporally shaped femtosecond laser double pulses. It is revealed that the maximal thermalization efficiency of gold film occurs at the optimal pulse energy ratio of 1:1 and the preferred pulse separation of 10 ps. The electron–phonon relaxation time decreases with increasing the pulse energy ratio, but presents increase tendency with increasing pulse separation. In addition, it is revealed that the fs + ps double pulses can bring in potential benefit for enhancing the phonon thermalization efficiency of gold film. The study will be helpful for promoting gold film thermal excitation efficiency via optimizing the temporally shaped femtosecond laser parameters.

Acknowledgments

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