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Ultrafast thermal dynamics of nano-ripples formation via laser double pulses excitation

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ABSTRACT

The ultrafast thermal dynamics of nano-ripples formation on gold film via ultrafast laser double pulses excitation is theoretically investigated by numerical simulations. The non-equilibrium thermal modulations with respect to the electron and phonon energy transfers within gold film is proposed for predicting the nano-ripples formation. It is revealed that the nano-ripples contrast on gold film surface can be well controlled via tuning the pulse energy ratio, pulse separation and pulse exchange of ultrafast laser double-pulse. It is attributed to the tunable energy transfer routes between the electron thermal diffusion and the electron–phonon coupling via tuning double pulses parameters. The study provides theoretical basis for producing high-contrast ripples for a wide range application in the fields such as high-absorptive solar cells, surface friction devices and super-hydrophobic surface.

1. Introduction

Laser-induced periodic surface structures (LIPSSs), also called ripples, have attracted much interest for preparing submicron-scale-structures for both scientific and practical purposes [1–6]. The ripples formation is widely accepted as the interference between the incident laser and surface scattering wave, which may originate from the surface roughness, surface instability and surface plasmon excitation [7–11]. Based on the surface wave interference theory, the ripples period can be predicted with respect to the laser parameters such as laser polarization, incident angle and laser wavelength.

Recently, femtosecond laser double pulses with fs–ps temporal separations have found great potential in material patterning [12–14]. It is experimentally observed that the patterned nano-ripples features can be well controlled via the femtosecond laser double-pulse train processing [15]. It rises a challenge for the conventional surface wave interference theory, which fails to predict the thermal modification of nano-ripples period because of the ignorance of material excitation. Bonse et.al. investigated the electron excitation role in affecting the ripples formation, revealing that the electron density can be an important role in regulating the ripples period [16]. Generally, it is accepted that the ultrafast non-equilibrium thermal dynamics can be mainly responsible for regulating the thermal ablation features [17–21]. For a metallic target, typical noble metal of gold, it is believed that the non-equilibrium thermal dynamics in the target material can be significantly modified by femtosecond laser double pulses excitation [22]. The thermal dynamics for regulating nano-ripples features can be expected to play an important role in affecting the performances of the rippled surface devices. It raises great interest for exploring the thermal dynamics of the nano-ripples formation on a metal surface with respect to femtosecond laser double pulses excitation. Especially, it is important to understand the fundamental dynamics of nano-ripples formation with respect to the non-equilibrium thermal excitations by the complex parameters of femtosecond laser double pulses. However, the nano-ripples contrast modulation with respect to the non-equilibrium thermal excitation via tailoring femtosecond laser double pulses is less investigated so far.

In this paper, the ultrafast thermal dynamics for patterning nano-ripples on gold film via femtosecond laser double pulses excitation is theoretically investigated. The periodical phonon temperature modulation for regulating the ripples profile was explored in details based on Finite Element simulations. The interplay of the electron thermal diffusion and the electron–phonon coupling are proposed for revealing the modulated ripples profile in the early stage of ripples formation. The theoretical study is helpful for understanding the basic thermal dynamics of nano-ripples formation thus providing the strategy for producing controllable nano-ripples structures via tuning the complex parameters of femtosecond laser double pulses.

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2. Theory and methods

As femtosecond laser irradiation of metallic film surface, the interference between surface scattering wave and the incident femtosecond laser is taken as the nano-ripples formation mechanism. The generation of surface scattering wave like the surface plasmon polaritons is physically originated from surface roughness of target [23,24]. In current investigation, the laser interference patterns for patterning nano-ripples are equalized as the periodically modulated laser source. The assumption can be tenable at the early stage of the thermal dynamics of the ripples formation on metallic on ps timescale. Here, we have not taken into the ripples formation mechanism as a main concern for simplifying the problem for investigations of the ripples contrast modulation. As femtosecond laser double pulses irradiation of gold film, the laser spot is modulated on gold film surface due to the interference between surface plasmon polaritons and the incident initial phase of the femtosecond laser double pulses. Then the second pulse will interact with the temperature-modulated surface. The energy diffusion can be described as follows: firstly the excited surface electron diffuses it's energy into the inner region of gold film on the timescale of several femtoseconds. Then the electron energy transfers into the lattice through the electron–phonon coupling process. Finally the electron and phonon systems will reach the thermal equilibrium, typically on the order of picoseconds. During the ultrafast non-equilibrium thermal relaxation process, the nano-ripples most likely formed due to the periodic temperature modulation on gold film and phase explosion mechanism. The ultrafast non-equilibrium thermal relaxation manipulated by femtosecond laser double pulses plays an important role in regulating the spatially modulated phonon temperature. The thermal excitations of gold film described by the two-temperature relaxation mechanism are written as follows [25,26]:

$$C_e\frac{\partial T_e}{\partial t} = \nabla(K_e\nabla T_e) - g(T_e - T_p) + Q \tag{1}$$

$$C_p\frac{\partial T_p}{\partial t} = g(T_e - T_p) \tag{2}$$

The electron thermal conductivity depending on the electron temperature and the phonon temperatures is expressed as follows:

$$K_e(T_e, T_p) = \chi\frac{\left(\theta_e^2 + 0.16\right)^{5/4}\left(\theta_e^2 + 0.44\right)\theta_e}{\left(\theta_e^2 + 0.092\right)^{1/2}\left(\theta_e^2 + \eta\theta_p\right)} \tag{3}$$

where $\theta_e=T_e/T_F$ and $\theta_p=T_p/T_F$ are the normalized electron and phonon temperatures with T_F denoting the Fermi temperature; χ and η are material constants. The analytical expression of the electron–phonon energy coupling strength can be represented as follows:

$$g = g_0\left[A_e/B_p\left(T_e + T_p\right) + 1\right] \tag{4}$$

where g_0 is the electron–phonon energy coupling strength in room temperature, the coefficients A_e and B_p are constants.

3. Results and discussions

The normalized phonon temperature modulations on gold film surface with respect to the electron thermal conductivity and the electron–phonon coupling strength in electron–phonon non-equilibrium state is shown in Fig. 1. We can see that for a given electron–phonon coupling, a large electron thermal conductivity of 1.5 Ke definitely causes the decline of ripple contrasts, which is defined as $R=(T_{max}-T_{min})/(T_{max}+T_{min})\times 100\%$. Here, T_{max} and T_{min} is the maximal and minimal absolute phonon temperatures at the central modulation region. It means that the electron thermal conductivity is playing an important role in dominating the laser energy deposition, which is unfavorable for generating the high contrast nano-ripples on gold film surface. On the contrary, as seen in Fig. 1(b), for a given electron thermal conductivity, a large electron–phonon coupling strength of 1.5 g can obviously lead to the rise of the normalized phonon temperature on gold film surface. As the phonon temperature is high enough to cause the phase explosion, the ripples is substantially patterned on gold film surface. It indicates that the electron thermal diffusion and electron–phonon coupling processes are both important in regulating the phonon temperature profile contrast, and finally the ripples contrast. Lin et al. has predicted that the electron–phonon energy coupling strength and the electron thermal conductivity can be affected by electron and phonon temperatures in the non-

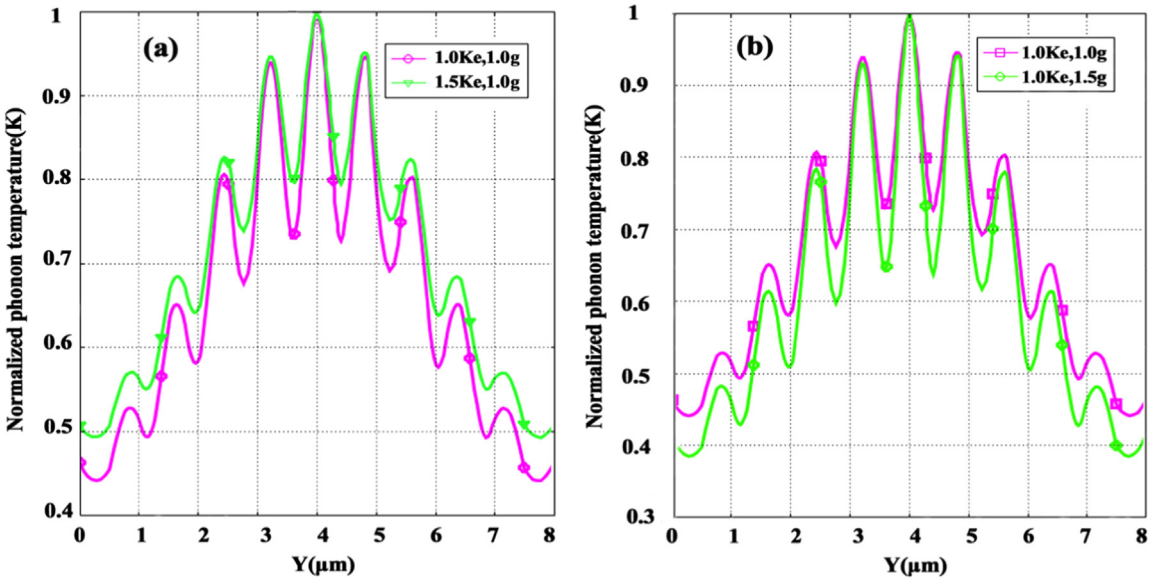

Fig. 1. The normalized phonon temperature field distribution on gold film surface with respect to the thermal parameters of the electron thermal conductivity and the electron-phonon coupling strength. (a) Depending on electron thermal conductivity and (b) depending on electron-phonon coupling strength.

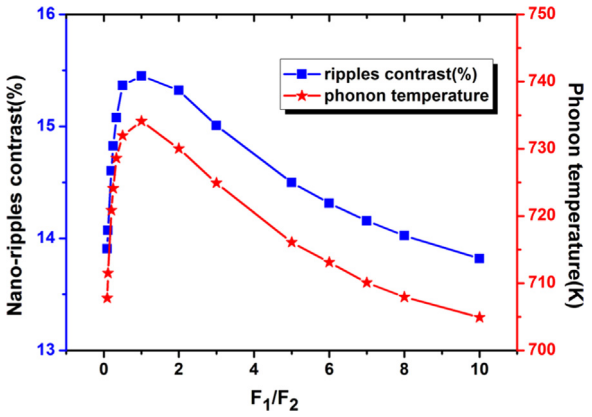

Fig. 2. The nano-ripples contrast and the localized surface phonon temperature on gold film at the termination of electron–phonon energy coupling period as function of the energy ratio of femtosecond laser double pulses. Total pulses fluence is 0.45 J/cm^2, pulses duration 60 fs, laser wavelength 800 nm, and double pulses separation 5 ps.

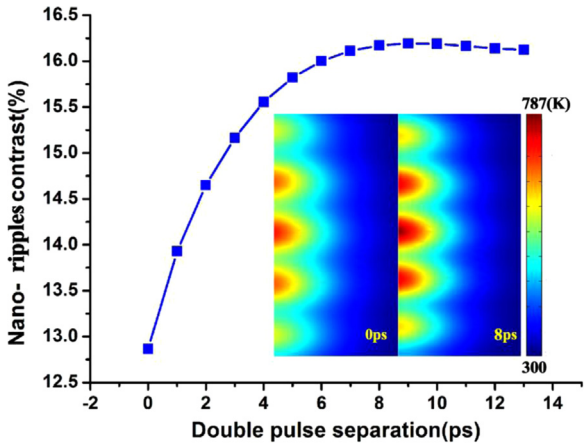

Fig. 3. The nano-ripples contrast modulation as a function of the temporal separation of femtosecond laser double pulses. The inset color drawing shows the 2-D phonon temperature field distribution when the pulses separation is 0 ps and 8 ps, respectively. Total pulses fluence is 0.5 J/cm^2, pulses duration 100 fs, laser wavelength 800 nm. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

equilibrium electron–phonon coupling state [26]. Our previous study also found that the material temperature can be well regulated by manipulating the laser parameters [22]. As a result, it is expected that the nano-ripples contrast can be well controlled via optimizing the complex laser parameters of femtosecond laser double pulses.

The nano-ripples contrast as a function of the pulses energy ratio of double femtosecond laser pulses are shown in Fig. 2. We can see that the nano-ripples contrast increases rapidly with increasing the pulses energy ratio of femtosecond laser double pulses as the energy ratio is less than 1:1. However, the nano-ripples contrast decreases quickly as the energy ratio exceeds 1:1. In fact, as the pulses energy ratio is less than 1:1, the localized surface phonon temperature on gold film surface increases quickly with the increasing pulses energy ratio as seen in Fig. 2. It indicates that the electron–phonon coupling is dominating the energy transfer to form the localized phonon temperature modulation, which potentially causes the promoted ripples contrast. On the contrary, as the pulses energy ratio exceeds 1:1, the increase of the pulse energy ratio causes the drop of the localized surface phonon temperature, indicating that the electron thermal diffusion plays an important role in the laser energy diffusion into the inner bulk of gold film. As a result, the nano-ripples contrast is lowered due to weakened phonon energy deposition on gold film surface. The results are basically helpful for optimizing the nano-ripples contrast via controlling the pulse energy ratio of the temporally shaped double femtosecond pulses.

The nano-ripples contrast as a function of the temporal separation of femtosecond laser double pulses is shown in Fig. 3. The inset with color drawing shows the modulated phonon temperature profiles across the gold film at the given double pulse separations of 0 ps and 8 ps, respectively. It can be seen that the nano-ripples contrast increases quickly with increasing the pulses separation as it is less than 8 ps. It is calculated that the nano-ripples contrast increases by 27% with the optimal pulses separation of 8 ps compared to that of single pulse with the identical fluence. Once the pulses separation exceeds 8 ps, the nano-ripples contrast exhibits a slow drop. It is attributed to the competitive energy transfer routes between the electron–phonon coupling and the electron thermal diffusion during nano-ripples formation. As the pulses separation is less than 8 ps, the electron–phonon coupling strength is strengthened with increasing of double pulses separation, leading to the rise of nano-ripples contrast. However, as the pulses separation exceeds 8 ps, the nano-ripples contrast is lowered due to concurrent processes of the electron thermal diffusion strengthening and electron–phonon energy coupling weakening. The localized phonon temperature on gold film surface with the pulses separation of 8 ps is apparently higher than that of double pulses separation 0 ps. It shows the electron thermal diffusion is slowed, and the electron–phonon coupling is promoted as double pulses separation is at 8 ps compared to that at 0 ps, which enables the formation of high-contrast nano-ripples on gold film surface.

The nano-ripples contrast as a function of laser fluence with respect to the pulse exchange of the femtosecond and picoseconds double pulses are shown in Fig. 4. We can see that the nano-ripples contrast increases rapidly with increasing the total fluence of both the fs–ps and ps–fs double pulses. More interestingly, the ps–fs double pulses excitation definitely causes the promotion of the nano-ripples contrast. However, the nano-ripples contrast of gold film is lowered for fs–ps double pulses excitation. It indicates that the ps–fs double pulses excitation is helpful for promotion of the contrast of patterned nano-ripples. However, the electron thermal diffusion dominates the energy transfer during the ripples formation in case of fs–ps double pulses excitation. It potentially causes the laser energy penetration into the electron system of inner bulk of the gold film, but local phonon at surface is less heated. As a result, the contrast of nano-ripples on gold film

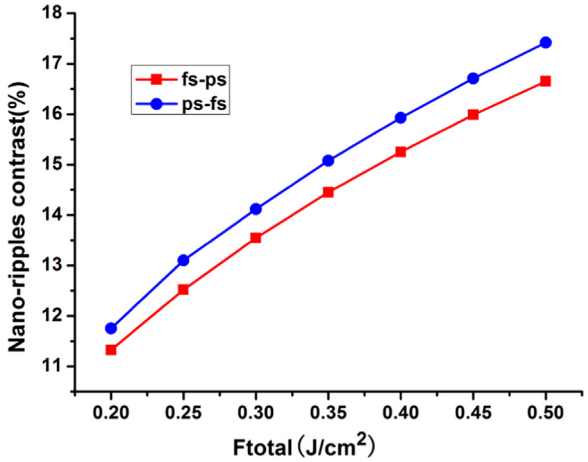

Fig. 4. The nano-ripples contrast as a function of laser fluence with respect to fs–ps and ps–fs double pulses. Fs pulse duration is 60 fs, ps pulse duration 2 ps, laser wavelength 800 nm, and pulses separation 5 ps.

surface is lowered via fs–ps double pulses excitation. The results are helpful for generation of high-contrast nano-ripples on metallic film surface via tuning the temporal positions of double fs–ps laser pulses.

4. Conclusions

We have theoretically investigated the ultrafast thermal modulation dynamics for patterning nano-ripples on gold film via femtosecond laser double pulses excitation. It is revealed that the nano-ripples contrast increases rapidly with increasing double pulses energy ratio as the pulses energy ratio is less than 1:1. However, the nano-ripples contrast drops quickly as the energy ratio exceeds 1:1. The nano-ripples contrast can be obviously promoted with increasing the pulses separation as it is less than 8 ps, once the pulses separation exceeds 8 ps, the nano-ripples contrast presents slow drop. In addition, the ps–fs double-pulse excitation can definitely improves the contrast of nano-ripples.

Acknowledgments

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