Varied laser induced damage phenomena of gold coated gratings for pulse compression

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Abstract: In this paper, gold-coated gratings for pulse compression have been prepared and their laser damage experiments have been performed. Varied laser damage morphologies have been observed: when a 60fs-pulsed laser with energy density slightly higher than the damage threshold was used, damage morphology with a characteristic of discrete distribution of small pits was appeared. These damage pits are linearly distributed at the junction of ridges and grooves. If the laser energy density is much higher than the damage threshold, the gold films was overall ablated and the grating structure disappeared. Besides, if the gold film has poor adhesion, it was peeled off. When a 450ps-pulsed laser with energy density slightly higher than the damage threshold was used, part of grating ridges will be ablated and an obvious line exists between the ablated area and the unchanged area. In theory, the laser induced temperature field and stress field in gold-coated gratings were calculated based on the electromagnetic field using the finite element method. It is demonstrated that the temperature and thermal stress distribution characteristics are affected by the laser heating rate and the heat diffusion time (the calculated diffusion time ranges from 6fs to 450ps), which determines the laser damage characteristics. The possible damage drivers have electron hydrodynamic pressure, thermal ablation and thermal stress.

KeyWords: Diffraction gratings; Laser damage; Pulse compression.

1. Introduction

The chirped pulse amplification (CPA) technology is the main approach to achieve high-intensity short-pulse laser. Diffraction gratings are good candidates for stretching and compressing laser pulses in CPA [1-5]. To achieve high intensity laser pulse, the used pulse compression gratings (PCG) must meet the requirements of broadband high efficiency and high laser damage resistance. The final grating in the pulse compressor typically needs to endure the irradiation of highest power laser [6-8]. Metal-coated gratings used for pulse compression was first developed in 1980s [9]. But the rather low damage threshold limits the performances of the CPA laser system [10-13]. To improve the longevity of a short-pulse laser system, a better understanding of the damage mechanism and constantly improving the laser damage resistance of metal-coated gratings should be given more attention.

The damage threshold of gold-coated gratings is obviously less than that of gold films with even surface [12]. This is because that the grating structure will cause an uneven distribution of laser field and concentrated distributions of temperature and stress field. The damages of gold coated grating have different morphologies induced by laser irradiation with different pulse widths or different energy densities. The analyses of laser damage mechanisms of gold-coated gratings were usually performed based on the electromagnetic field distribution [5, 6]. While the electromagnetic field distribution is determined by grating structures, laser wavelength and laser incident angle. Varied laser induced damage phenomena cannot be interpreted only according to the electromagnetic field distribution. In this paper, the distributions of temperature field and stress field in gratings caused by laser pulse have been calculated, and the damage distribution, morphology and mechanism of gold coated gratings were analyzed accordingly.

2. Brief introduction of experimental process

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The fabrication processes of gold coated gratings can be described as follows. Firstly, the desired photoresist grating structures were manufactured by the holographic method. Then a gold layer was deposited by the method of magnetron sputtering or electron beam evaporation onto the initial grating structure to form gold-coated gratings. For the silica based gratings, the structure of photoresist gratings was transferred to the silica substrate by a reaction ion beam etching process before gold films deposition. The designed gold coated gratings have a spatial period of 574.7nm , a groove depth of 232nm and a duty cycle of 0.82. The -1st order diffraction efficiency was measured by P-polarized light from 700nm to 1000nm . The diffraction efficiency of the prepared grating is higher than 90% in the whole measuring wavelength range (from 700 to 1000nm).

The detailed laser damage experimental setup can be referred in [ref.14](#). The experiments were conducted using a CPA Ti: sapphire laser system. The system generated a p-polarized and near-Gaussian spatial profile pulse. The laser beam was focused onto the sample surface at an angle of 53° , and the focused beam diameters in normal direction were measured to be about 500 and $420\mu\text{m}$ for laser with pulse widths of 450ps and 60fs respectively. The laser damage test mode was “1-on-1”, in which a row of points on the sample are irradiated by laser pulses with different energy densities, and every point is irradiated once. A half-wave plate and a polarizer assembly were used to adjust the laser fluence. A 100X microscope was used to monitor the surface morphology of samples, and the image was displayed on a computer screen. Any surface morphology change which can be judged by human eyes will be taken as a permanent damage.

3. Theoretical basis and discussions

3.1 Electron and lattice temperature in gold

When a laser pulse irradiates on gold coated gratings, the light field is mainly distributed in the shallow surface of gold films. How much laser energy will be deposited in gratings depends on the absorption ability of the gold film, so the process of gold absorbing laser energy should be investigated firstly. The free electrons in gold can efficiently absorb laser energy and an electron can be heated at a rate of $dJ/dt = e^2 E^2 \tau_k / [m_e(1 + \omega^2 \tau_k^2)]$ [15], where J is the electron energy; ω is the laser frequency; E is the laser intensity; e is the elementary charge; m_e is the free electron mass; $\tau_k = 3\pi\hbar^3 / (4Ze^4 \ln\Lambda)$ for plasmon at low temperatures [16], where $\ln\Lambda$ is the Coulomb logarithm and a approximation of 2 was used in the calculation [17]; $\hbar$ is the Planck constant; Z is the mean ionization degree (here $Z=1$). Then the coulomb collision between the energetic electrons and the cold ions can transfer thermal energy from electrons to ions. This process can be described by a set of thermal equilibration equations: $dT_e/dt = (T_i - T_e)/\tau_{eq}$ and $dT_i/dt = C_e(T_e - T_i)/C_i + \tau_{eq}$ [18], where $C_i = 3N_i k_B$ and $C_e = 0.5\pi^2 k_B^2 T_e / E_f$ are lattice and electron specific heat; N_i and k_B are the ion number density and the Boltzmann constant respectively. E_f is the Fermi energy and the value of 5.53eV was used in the calculation [17]. T_i and T_e are ion temperature and electron temperature respectively. τ_{eq} is the relaxation time of the electron-phonon scattering and $\tau_{eq} = \tau_k m_i / m_e$ [16], where m_i is the ion mass.

In this section, the two-temperature model (TTM) was used to reveal two facts. 1) Both electrons and lattice in the gold can be effectively heated because of the high number density of free electrons. 2) The temperature difference between electrons and lattice is larger when the laser pulse width is shorter. The purpose of TTM calculation is to reveal the effect of electron-lattice interaction. The temperature distribution is less important in this case, and we focus more on the difference between electron and lattice temperatures as well as its evolution. For these purposes, the terms of thermal conduction in TTM were ignored. But in the next section, temperature distribution in gratings, the thermal conduction has been considered.

No considering the thermal conduction may cause an overestimate of temperatures. While, for a laser irradiation with a pulse width of 10ps , the thermal diffusion length at the end of laser irradiation is hundreds of nanometers for gold [12]. In this case, for a laser irradiation with a beam waist of hundreds of microns, the effect of thermal diffusion can be ignored. If the pulse width is longer than 10ps , the electron-lattice system will reach its equilibrium state before the end of laser irradiation [17]. Therefore, no considering the thermal diffusion has little effect on the analysis.

The basic energy absorption processes of materials are free electrons acceleration, followed by transferring energy to lattice by the electron-phonon scattering. For example, in the case of ps -pulsed width,

the time-dependent temperatures of electron and lattice under different electron number densities have been calculated without considering ionizations. The results are shown in Fig.1, in which the used calculation parameters were chosen according to those of gold. It can be seen that materials with a higher number density of free electrons have a stronger absorption capacity, and can absorb more laser energy. Besides, the efficiency of electrons heating lattice is also affected by the electron number density, and the lattice temperature rises faster for materials with a higher free electron density. Compared with dielectrics, metals have a higher efficiency to heat lattices.

The time-dependent temperatures of electron and lattice under laser irradiations with different pulse widths also have been calculated without changing the electron density and pulse energy density, and the results are shown in Fig.2. The thermal diffusion was ignored in calculation, so the absorbed energy can be evaluated directly according to the ultimate temperature rise. From figures 1(a) and 2(b)-(c), it can be seen that when the electron-lattice equilibrium state is arrived, the lattice temperatures reach the same value for laser irradiations with different pulse widths, so the absorbed laser energy can be considered as the same. For a certain pulse energy density, the laser pulse with a shorter pulse width has a higher power density, and it can heat electrons faster. So the difference between electrons temperature and lattice temperature is larger during the laser irradiation which has a shorter pulse width. In the case of laser irradiation with a pulse width of 10ps (Fig2(c)), the electron-lattice system has reached the equilibrium state when the laser irradiation is immediately vanished. While for the case of a 100fs-pulsed width (Fig2(a)), the electron-lattice system is far from the equilibrium state, and the electron temperature is much higher than the lattice temperature. Under this condition, the electron-lattice system eventually reach the equilibrium state 4ps after the laser irradiation (Fig2(b)).

3.2 Temperature distribution in gratings

The finite element method (FEM) in time domain was used in calculating the electric field, temperature and thermal stress fields in gratings, because it is an efficient and widely used method for calculating the electromagnetic field, temperature field and stress field separately [19]. In this paper, the FEM was used in these three fields coupled calculation. Besides, the main purpose of the calculation is to estimate the damage occurrence position. For this, the energy distribution in gratings can provide sufficient information, and it is concise and clear. So the electron and lattice temperature distributions were merged into energy distribution, and the TTM was not applied.

Firstly, Maxwell's equations were used to calculate the electromagnetic field distribution: $\nabla^2 E + k^2 \mu_r \epsilon_r E = 0$, where E is the electric field; μ_r is the relative permeability; ϵ_r is the relative permittivity; k is the wave number of free space and $k = \omega/c_0$, in which ω is laser frequency and c_0 is the speed of light in vacuum.

Based on the calculated electromagnetic field distribution, the thermal diffusion equation was used to calculate the temperature (T) distribution: $\rho C_p \partial T / \partial t = \nabla(k \nabla T) + Q$, where ρ is density; C_p is heat capacity; k is thermal conductivity; Q is the heat source, and $Q = \omega \epsilon'' |E|^2 / (8\pi)$, ϵ'' is the imaginary part of the permittivity [17].

Based on the calculated temperature distribution, Hooke's law was used to calculate the stress distribution. Hooke's law relates the stress tensor (s) to the strain tensor and temperature: $s = s_0 + C : (\epsilon - \epsilon_0 - \epsilon_T)$, where C is the 4th order elasticity tensor; ":" stands for the double-dot tensor product; s_0 and ϵ_0 are initial stresses and strains; $\epsilon_T = \alpha(T - T_0)$ is the thermal strain; α is the coefficient of thermal expansion; T_0 is the initial temperature. The strain tensor is written in terms of the displacement gradient (u): $\epsilon = 0.5 (\nabla u + \nabla u^T)$.

According to the measured -1st order diffraction efficiency of the prepared gratings, the gratings' structure parameters were fitted. The actual gold-coating of the prepared gratings has a spatial period of 585nm, a groove depth of 186nm and a duty cycle of 0.82. The measured and fitted diffraction efficiencies as well as the absorption rate were shown in Fig.3. At the wavelength of 800nm, the fitted 0 and -1st order diffraction efficiencies are 5.03% and 91.34% respectively. The absorption rate is about 3.63% because the efficiencies of the other order diffractions can be ignored.

Figure 4(a) shows the calculation model of gold coated gratings in this work. The grating structure is divided into three layers: the gold film layer with a thickness of 186nm, a spatial period of 585nm and a duty cycle of 0.82; the photoresist grating layer with a duty cycle of 0.79 and a groove depth of 217nm; and

the substrate layer with a thickness of 1200nm used in calculation. A P-polarized laser is incident as an incident angle of 53° from the left side in air, and it was excited on the upper boundary of the air layer. **The applied laser pulse has a top-hat time profile.** The diameter of the cross section of the used laser beam in experiments is about 500μm, so the laser irradiation area contains nearly a thousand grating periods. Periodic boundaries can be used to reduce the computational region. Therefore, in calculation, the left and right boundaries of the calculated domain were periodic boundaries. Besides, in temperature field calculation, the upper and lower boundaries of the calculated domain were set as thermal radiating boundaries (the net inward radiative heat flux is: $q = \xi \sigma (T_{\text{amb}}^4 - T^4)$, in which ξ is the surface emissivity and the value of 10 was used in the calculation; σ is the Stefan-Boltzmann constant; T_{amb} is the ambient temperature and $T_{\text{amb}} = T_0$ was used in calculation). The initial temperature was set as 273K. In thermal stress field calculation, the air layer was not included. The surface of the gold film was set as a free surface and the lower boundary of the substrate layer was set as a fixed boundary. No initial strain and stress exist in gratings.

The electromagnetic field distribution in gold coated gratings is shown in Fig.4(b) and the inset is a contour map. The used refractivity of air, photoresist, substrate, and gold are 1, 1.49, 1.52 and 0.18+5.00i respectively. It can be seen that a standing wave field was formed at the incident side in the air and the wave front is perpendicular to the laser incident direction. Due to the strong absorption and high reflection characteristics of gold films, the depth that the electromagnetic wave enters into the gold film is extremely limited, and the intensity of the electromagnetic field inside the grating is very low. Hence the laser cannot directly heat the photoresist and substrate, and these layers can only be heated by heat conduction.

The distribution of the deposited laser energy in gratings can be expressed using temperature distribution, and in this case the temperature includes electron temperature and lattice temperature. On the basis of the obtained electromagnetic field distribution, the typical temperature (energy) distribution in gratings under a laser irradiation was calculated. **In calculation, the mass densities of photoresist, substrate, and gold are 1180, 2202 and 19300 kg/m³ respectively, the heat conductivities of them are 0.18, 1.38 and 200 W/(m*K) respectively, and the heat capacities of them are 1470, 753 and 129 J/(kg*K) respectively[20-22]. Considering that the thermal conductivity of a metal will significantly decreased under high temperature, and the thermal conductivity has a great influence on the temperature distribution under high temperature during laser heating, the thermal conductivity of 200W/(m*K) has been taken in calculation instead of the normal temperature value of 318W/(m*K).**

It can be seen from Fig.4(c) that the temperature sharply decreases deep into the gold film, and the temperature rise of the substrate and the photoresist layer are also very small. Only the shallow surface of gold films can be heated directly by a laser irradiation, and the thermal ablation usually starts from the surface of gold films. The temperature distributions on gold films surface under laser irradiations with different pulse widths are show in Figs.5-6, and the sampling period of gold film surface was identified in the insert map in Fig.5 (a). The normalized laser field distribution will not change because of the invariant grating structures, laser wavelength and incident angle, so the normalized distribution of the laser energy deposition rate in gratings is unchanged. The change of temperature distributions under different pulse widths is caused by heat deposition rate and heat diffusion time. Heat deposition causes an uneven temperature rise and the formation of some hot spots on the gold film surface, while heat diffusion can eliminate these hot spots and decrease the temperature difference in different areas. The maximal temperature on the gold film surface decreases as the laser pulse width increases, and the laser damage threshold will increase accordingly when thermal ablations dominate the damage process.

As for the damage threshold prediction, if the damage process is dominated by thermal ablation (for instance, a ns-pulsed laser induced damage), the damage threshold can be directly evaluated based on the criterion of melting point or boiling point. But in the case of short pulsed laser induced damage (for instance, fs-pulses), the damage threshold cannot be evaluated based on the temperature field, not even the stress field. In this case, the damage process is considered to be dominated by electron hydrodynamic action, and the damage threshold is not linear with the electron temperature. But, the damage location in gratings must be the region with high electron temperature. Thereafter the damage location can be predicted according to the calculated deposited energy distribution.

Figure 5(a) shows the temperature distributions on gold films surface under a laser irradiation with a pulse width of 60fs. It can be seen that three areas have high temperature rise, including the top of the ridge

and two transition regions of grooves and ridges. In the groove-ridge transition region which is facing the incident light, the temperature is much greater than that of other regions, and hot spot is formed after the laser irradiation. In this case, according to the result reflected in Fig. 2(a), the deposited laser energy is mainly stored in the electron system, and hence the grating may be damaged by the electron hydrodynamic action. The hydrodynamic pressure of the electron system is $P_H = N_e k_b T_e$ [23], in which N_e , k_b and T_e are electron density, Boltzmann constant and electron temperature. The electron density is in the order of magnitude of $10^{28}/m^3$. If the electron temperature is in the order of magnitude of $10^4 K$, the hydrodynamic pressure is in the order of magnitude of GPa . This pressure is high enough to induce the damage of gold films. Besides, the distribution of the electron hydrodynamic pressure is determined by the electron temperature distribution. Therefore, at the junctions of grooves and ridges, the electron hydrodynamic pressure driven damages are likely to appear in the case of an *fs*-pulsed laser irradiation. This kind of damage usually involves materials eruption, and the deposited laser energy will be consumed. If the laser energy density is not high enough, the other types of damages will not happen after the initial damage. According to the result reflected in Fig.2(b), it can be deduced that the electron-lattice system will reach their equilibrium state after a few picoseconds by electron-phonon scattering, and the lattice temperature rises to its maximum. If the laser energy density is high enough, the initial electron hydrodynamic damages cannot prevent the lattice temperature from rising beyond the melting point, and the thermal ablation can happen. The characteristics of the thermal ablation are similar with those of the longer pulse induced damages, which will be discussed below.

Figure 5(b) shows the temperature distributions on gold films surface under a laser irradiation with a pulse width of $4.5ps$. In this case, the laser heating rate is slow, and hot spots at the junction of grooves and ridges are difficult to form because of the heat diffusion. Meanwhile, according to the result reflected in Fig.2, the difference between the electron temperature and the lattice temperature is smaller in this case compared to that in Fig.5(a), and the lattice-electron system reaches their equilibrium state when the laser irradiation disappears immediately. However, there are still two high-temperature areas, the ridge top and the groove-ridge junction region which is facing the incident light. In this case, the thermal ablation will start from these two positions. The occurrence probability of electron hydrodynamic damage decreases. Similarly, if the laser energy density is high enough, the other types of thermal ablation damages can happen. The characteristics of the thermal ablation are similar with those of the longer pulse induced damages.

According to the result reflected in Fig.2, for a laser irradiation with a pulse width of tens of picoseconds, the laser heating rate further decreases and the difference between the electron temperature and the lattice temperature can be ignored during laser irradiations. Electron-lattice system can be considered as being in an equilibrium state all the time. Figure 6(a) shows the temperature distributions on gold films surface under a laser irradiation with a pulse width of $45ps$. In this case, the temperature distribution on the ridge side wall which is facing the incident light is more uniform, while the temperature of the other side wall of the ridge is very low. If the laser irradiation has an energy density near the damage threshold, the thermal ablation will mainly occur on the top of ridges and the ridge side wall which is facing the incident light and keep the other side wall of ridges unchanged, especially for the section near grooves. Similarly, if the laser energy density is high enough, the other types of thermal ablation damages can happen. The characteristics of the thermal ablation are similar with those of the longer pulse induced damages.

Figure 6(b) shows the temperature distributions on gold films surface under a laser irradiation with a pulse width of $450ps$. The temperature distribution on gold films surface is more uniform because of the long time of heat diffusion. In this case, the main characteristic of the laser damage is an overall damage of the gold film.

3.3 Thermal stress distribution in gratings

If laser irradiations have pulse widths in the order of tens of picoseconds or longer, or have energy densities much higher than damage thresholds, the lattice thermal effect will become obvious. In these cases, the probability of thermal stress driving damages should be considered. Thermal stress distributions in gratings have been calculated based on the obtained temperature distribution. The calculating results are shown in Figs. 7-8. The used young modulus of photoresist, substrate, and gold are 2.33, 72 and 80 *GPa* respectively, the Poisson's ratios of them are 0.34, 0.17 and 0.42 respectively, and the coefficients of

thermal expansion of them are 7.0×10^{-5} , 0.055×10^{-5} and $1.443 \times 10^{-5} K^{-1}$ respectively. The sampling period of gold film surface was identified in the insert map in Fig.7 (a). The sampling period of gold-photoresist interface was identified in the insert map in Fig.8 (a).

Figure 7 shows the surface thermal stress distribution of gold films. It can be seen that the surface peak stress is smaller for the laser irradiation with a longer pulse width. The laser with a longer pulse width has a lower power density and heats gold films slower. Meanwhile, the heat diffusion time is longer and more energy will be conducted from the surface to the depth of gold films. Therefore, the surface temperature of gold films is smaller and the formed surface stress is also smaller for the laser irradiation with a longer pulse width. The surface stress peak of gold films is located in the junction region of grating ridges and grooves. Cracks or defects will appear in this place once the stress exceeds the gold strength, and possibly cause gratings failure.

Figure 8 shows the thermal stress distribution on the interface between gold films and photoresist gratings. It can be seen that the interface peak stress is larger for the laser irradiation with a longer pulse width, with which the heat diffusion time is longer and more energy will be conducted from the surface to the depth of gold films and the photoresist layer. As a result, the interface temperature is higher and the formed interface stress is also higher for the laser irradiation with a longer pulse width. The interface stress peak is located around the top of photoresist grating ridges. The development of this kind of stress concentration is likely to cause the crack formation at the interface when the interface stress exceeds the gold films adhesion strength before ablation damage happens. The growth of this kind of crack results in the shedding of gold films.

4. Experimental results discussions

Figure 9 shows a kind of laser induced damage morphology of gold coated gratings. The feature of this damage morphology is a discrete distribution of small pits, and these pits are linearly distributed at the junction of ridges and grooves. No clear damages appear in the other areas. According to the analysis of Fig.5 (a), these damage pits are probably caused by the electron hydrodynamic pressure. This is because that these damage pits occurred at the position of the calculated maximal field intensity in Fig. 5(a). Furthermore, these damage pits are concentrated distributed and their size is very small. These reflect that the energy diffusion time was very short before damage occurrence, and the damage probably happened during laser irradiation. During femtosecond pulsed laser irradiation, the electron-lattice system is far from equilibrium and the deposited energy is mainly stored in electron system. The damage can only be driven by hot electrons. Furthermore, the damage can be considered to be driven by electron hydrodynamic pressure by excluding the possibility of the Coulomb explosion for gold. The test damage threshold of this sample is $0.54 J/cm^2$ ("1-on-1" test mode, pulse width of 60fs.), and the energy density of the used test laser for Fig.9 is $0.63 J/cm^2$ (the energy densities of laser beams in this paper refer to the beam normal fluence). Although the test laser energy density is larger than the damage threshold, the material eruption consumed energy driven by electron hydrodynamic pressure can prevent the eventual lattice temperature from rising beyond the melting point. Eventually, no thermal ablation will occur.

Figure 10 shows another kind of laser induced damage morphology of gold coated gratings. The feature of this damage morphology is the thermal ablation of ridge tops and ridge side wall which is facing the incident light (as the thick line indicates in the insert map in Fig.10). The other side wall of ridges was kept unchanged, especially for the section near grooves. The ablation area of gold films surface is smooth, and the unchanged area keeps the original trace of films deposition. An obvious line exists between the ablation area and the unchanged area (as the dotted line indicates in Fig.10). In this case, the ablation damages have no discrete distribution characteristic reflected in Fig.9. This is because that the pulse width of the used laser for Fig. 9 is 60 fs and for Fig. 10 is 450 ps. Compared to the case of Fig. 9, the laser power density is much lower for Fig. 10, and the laser heating rate is much slower accordingly. Therefore in the case of Fig. 10, the thermal diffusion effect is obvious, and the local high temperature distribution in the sites of defects will be eliminated. The temperature distribution of gold films surface is uniform along the ridge extension direction. As a result, no discrete damage pits will appear. The test laser energy density for Fig.10 is $0.55 J/cm^2$, and it is near the test damage threshold of $0.51 J/cm^2$ ("1-on-1" test mode, pulse width of 450ps.). The used pulse width for Fig. 10 is 450ps, while the damage morphology is similar to the theoretical result of 45ps. This is probably because the calculation used thermal diffusion coefficient is too large. Actually, at high temperature, the electron heat conduction will be blocked in a certain degree by the lattice vibration.

Figure 11 shows another kind of laser induced overall ablation morphology of gold coated gratings. The periodic structure of gratings is disappeared. Some areas of gold films erupted out and the basic photoresist grating is bare with its periodic structure. The pulse width of the testing laser for Fig.11 is 60fs (“1-on-1” test mode), but the laser energy density of $0.90\text{J}/\text{cm}^2$ is much higher than the test damage threshold of $0.59\text{J}/\text{cm}^2$. The damage morphology in Fig. 9 usually occurs in the early stage of laser irradiation, and the materials eruption can consume some deposited energy. While for Fig. 11, the test laser energy density is much high than the gratings damage threshold, the remained energy after eruption and the deposited energy in the later stage of laser irradiation can heat the whole gold films to the melting point. When melting happened, under the action of surface tension, the gold films become flat.

The stress-induced damage morphologies are rarely observed in experiments. This is because that the metal elasticity modulus will decrease significantly as its temperature rises under actual circumstances [24, 25]. The thermal stress in gratings in actual cases will not be as high as theoretical values. Besides, the premise of the thermal stress formation is the formation of a temperature distribution, and higher peak stress usually needs higher temperature. Therefore, before stress-induced damage happening, thermal ablation may already happen. The damage morphology cannot be identified even if the stress-induced damages were occurred. However, these do not mean that stress driven damages of the gold coated gratings will not occur at all. If the quality of gold films is very poor (low strength) or gold films adhere badly to the photoresist and substrate, the stress-induced damage can be observed. Figure 12 shows a kind of damage morphology of films stripping. The test laser energy density is $0.95\text{J}/\text{cm}^2$ (“1-on-1” test mode), the wavelength is 800nm and the pulse width is 60fs. Unlike the other samples, the sample in Fig.12 was prepared by the method of electron beam evaporation. During films deposition, the kinetic energy of the evaporated atom cluster is not high enough, so the formed films have poor adhesion.

From Fig.12, it can be seen that the gold film already has a certain degree of ablation damage, and the grating structure has been smoothened in a certain degree. Compared with Fig.9, the laser energy density used for Fig.12 is a little bit higher, but the grating structure is not completely disappeared. This is probably because the shedding of gold films consumes part of the deposited energy. Meanwhile, because the gold film is very thin, its thermal expansion is free after films shedding, especially along the direction of films thickness. So the high thermal stress is difficult to form in gold films, and the grating structure is difficult to be smoothened.

From the above theoretical analysis and experimental results, it can be concluded that the stress induced damage can be suppressed by improving the quality and adhesion of gold films. As for the thermal ablations, because the high temperature region is distributed mainly on the surface of gold films, any method which can decrease the surface temperature may be helpful for improving the gratings damage threshold.

5. Conclusions

When a laser pulse irradiates on gold coated gratings, the light field is mainly distributed in the shallow surface of gold films. Only the shallow surface of gold films can be heated directly by a laser irradiation. For the case of fs-pulsed laser irradiation, linear distributed discrete damage pits induced by electron hydrodynamic pressure will appear at the junction of ridges and grooves. This is because that the deposited energy is mainly stored in the electron system, and electrons have no sufficient time to diffuse. The hot spots of heated electrons will form at the junction of ridges and grooves, and cause the damage. For laser irradiations with a pulse width of tens of picoseconds, there are two high-temperature areas on the gold film surface, and they are located on the top of ridges and at the groove-ridge junction region which is facing the incident light. The thermal ablation will start from these two positions. In this case, the lattice and electron systems can reach their equilibrium state when the laser irradiation disappears immediately. The damage will happen because of thermal ablation. Whereas, the thermal diffusion time is still short and the hot spots on grating ridges will not be eliminated. The damage still appears on part of the grating ridges. For laser irradiations with a pulse width of hundreds of picoseconds, electron-lattice system can be considered as being in an equilibrium state all the time. The thermal ablation will start from the ridge side wall which is facing the incident light. This because that the temperature distribution on the ridge side wall which is facing the incident light is more uniform, while the temperature of the other side wall is very small. For laser irradiations with a longer pulse width, the main characteristic of the laser damage is an overall damage of the gold film. In this case, the thermal diffusion time is long enough for the formation of

a relatively uniform temperature distribution on grating ridges. Besides, if the laser energy density is high enough, even for an fs-pulsed laser irradiation, different thermal ablations may happen. The characteristics of thermal ablations are similar with those of the longer pulse induced damages depending on the damage happen time. Because the test laser energy density is much high than the gratings damage threshold, the remained energy after initial damages and the deposited energy in the later stage of laser irradiation can heat the part or whole gold films to the melting point. When melting happened, under the action of surface tension, the melted part of gold films becomes flat. Finally, if the quality of gold films is very poor, or gold film adheres badly to the photoresist gratings, stress-induced damages can be observed. The main characteristics of stress-induced damages are peeling or cracking of gold films.

Acknowledgments

The work was supported by NSAF (No. U1430121). The authors gratefully acknowledge the financial support of the National Science Foundation of China (Grant 10704079), and the Open Research Fund of Key Laboratory of Material for High Power Lasers - Chinese Academy of Sciences.

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FIGURE CAPTIONS:

Figure 1 Electronic and lattice temperature of materials with different electronic densities under laser irradiation with a Gaussian time profile. In calculation, the used parameters for gold are: $N_i=5.87e28/m^3$, $\ln A=2$, $Z=1$ and $E_f=5.53eV^{[17]}$. The laser energy density is $1.0J/cm^2$, laser wavelength is $800nm$, the laser pulse width is $100ps$ and the initial temperature of materials is $300K$. (a) The electronic number density is equal to N_i (atomic number density). (b) The electronic number density is equal to $0.1 N_i$. (c) The electronic number density is equal to $0.01 N_i$. The dotted lines represent the electronic temperature and the solid lines represent the lattice temperature.

Figure 2 Electronic and lattice temperature of gold under laser irradiation with different laser pulse widths. The laser energy density is $1.0J/cm^2$, laser wavelength is $800nm$, the electronic number density is equal to N_i and the initial temperature of gold is $300K$. (a) The laser pulse width is $100fs$. (b) The laser pulse width is $100fs$. (c) The laser pulse width is $10ps$. The dotted lines represent the electronic temperature and the solid lines represent the lattice temperature.

Figure 3 The diffraction efficiency, absorption rate and cross section morphology (the inset) of gold coated gratings. The basic grating material is photoresist, and the surface gold film was deposited by the method of electron beam evaporation. The tested grating has a spatial period of $585nm$, a groove depth of $186nm$ and a duty cycle of 0.82 .

Figure 4 The calculation model (a), typical electromagnetic field (b) and temperature field (c) distribution in gold coated gratings. The used laser pulse width for (c) is $4.5ps$. In calculation, the thickness of gold film layer is $186nm$; the spatial period is $585nm$; the duty cycle is 0.82 ; the duty cycle of photoresist grating is 0.79 ; the groove depth is $217nm$. A P-polarized laser is incident as an incident angle of 53° .

Figure 5 The surface temperature distribution of gold coated gratings in different time during laser irradiation. In calculation, the mass densities of photoresist, substrate, and gold are 1180 , 2202 and $19300 kg/m^3$ respectively, the heat conductivities of them are 0.18 , 1.38 and $200 W/(m*K)$ respectively, and the heat capacities of them are 1470 , 753 and $129 J/(kg*K)$ respectively. The used calculating laser wavelength is $800nm$ and intensity is $0.5J/cm^2$. (a) The used laser pulse width is $60fs$. (b) The used laser pulse width is $4.5ps$.

Figure 6 The surface temperature distribution of gold coated gratings in different time during laser irradiation. The used calculating laser wavelength is $800nm$ and intensity is $0.5J/cm^2$. (a) The used laser pulse width is $45ps$. (b) The used laser pulse width is $450ps$.

Figure 7 The surface stress distribution of gold coated gratings in different time during laser irradiation. In calculation, the young modulus of photoresist, substrate, and gold are 2.33 , 72 and $80 GPa$ respectively, the Poisson's ratios of them are 0.34 , 0.17 and 0.42 respectively, and the coefficients of thermal expansion of them are $7.0*10^{-5}$, $0.055*10^{-5}$ and $1.443*10^{-5} K^{-1}$ respectively. The used calculating laser wavelength is $800nm$ and intensity is $0.5J/cm^2$. (a) the used laser pulse width is $45ps$, (b) the used laser pulse width is $450ps$.

Figure 8 The interface stress distribution of gold coated gratings in different time during laser irradiation. The used calculating laser wavelength is $800nm$ and intensity is $0.5J/cm^2$. (a) the used laser pulse width is $45ps$, (b) the used laser pulse width is $450ps$.

Figure 9 The laser induced damage morphology of gold coated gratings. The basic grating material is silica, and the surface gold film was deposited by the method of magnetron sputtering. The testing laser energy density is $0.63J/cm^2$, the wavelength is $800nm$ and the pulse width is $60fs$. The laser is incident from the top right side. This is different from the other figures in this paper just because of the different placement direction of samples in SEM test. A "1-on-1" test mode was used in experiment.

Figure 10 The laser induced damage morphology of gold coated gratings. The basic grating material is silica, and the surface gold film was deposited by the method of magnetron sputtering. The testing laser energy density is $0.55J/cm^2$, the wavelength is $800nm$ and the pulse width is $450ps$. The laser is incident from the top left side. A "1-on-1" test mode was used in experiment. The damaged zones are seen as the black ones in the SEM image.

Figure 11 The laser induced damage morphology of gold coated gratings. The basic grating material is photoresist, and the surface gold film was deposited by the method of magnetron sputtering. The testing laser energy density is $0.90J/cm^2$, the wavelength is $800nm$ and the pulse width is $60fs$. The laser is incident from the top left side. A “1-on-1” test mode was used in experiment.

Figure 12 The laser induced damage morphology of gold coated gratings. The basic grating material is photoresist, and the surface gold film was deposited by the method of electron beam evaporation. The testing laser energy density is $0.95J/cm^2$, the wavelength is $800nm$ and the pulse width is $60fs$. The laser is incident from the top left side. A “1-on-1” test mode was used in experiment.

FIGURES:

FIGURE1:

FIGURE2:

FIGURE3:

FIGURE4:

FIGURE5:

(a)

(b)

FIGURE6:

(a)

(b)

FIGURE7:

(a)

(b)

FIGURE8:

FIGURE9:

FIGURE10:

FIGURE11:

FIGURE12: