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Temperature field analysis of optical coatings induced by millisecond and nanosecond lasers

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To study the differences between long-pulse and short-pulse laser-induced damage in optical dielectric thin films, temperature distributions in single-layer HfO_2 films, multi-layer HfO_2/SiO_2 films, and HfO_2 films with high-absorptive inclusions induced by 1 ms and 10 ns lasers are analyzed based on the temperature field theory. Through our calculations, the damage property differences between millisecond and nanosecond lasers are summarized. The results for single-layer films show that 1 ms laser is easier to damage the substrate than 10 ns laser. For multi-layer films, the laser field effect is weaker when irradiating by 1 ms laser. Furthermore, when inclusions are introduced, the film is easier to be damaged by 10 ns laser, which means that 10 ns laser is more sensitive to the inclusions.

Keywords: laser damage, thin films, thermal effects.

1. Introduction

Optical coatings are important components in a laser system but easy to be damaged. Damages of these components induced by the laser beam limit the output power and energy of the lasers. There has been much attention paid, during nearly 40 years of studies, to high-power short-pulse (nanoseconds or below) laser-induced damage in optical film components. Film damage induced by high-energy long-pulse laser with pulse width of milliseconds was little mentioned. Nowadays, for the advantage of smaller loss during transmission, no plasma shielding, no self-focus, larger pulse energy and high efficiency of energy coupling, long-pulse laser has got more and more applications in the field of laser processing and laser damage. And, as a result of longer acting time and larger affected area of the long-pulse laser, the thermal damage of films induced by long-pulse lasers is predicted to be distinguished from short-pulse. Our preliminary studies have shown in the experiment that there are indeed some differences between long-pulse laser damage of optical materials and short-pulse laser damage in damage thresholds and damage morphologies but there is no further analysis [1–3]. So it is important and significant to further study the damage property differences between long-pulse and short-pulse lasers and summarize the characters in theory.

Two major effects can be involved in the interaction between the laser and the thin film. One is the thermal effect of thin film absorption, the other is the laser field effect [4]. In general, these two effects exist together but one of them usually dominates, especially for multi-layers [5–8]. Besides, sometimes during the film deposition process, nanoparticle inclusions may be introduced into the film body [9]. These high-absorptive inclusions will absorb more laser energy and reduce the damage threshold. So the inclusion effect on the damage performance between long-pulse and short-pulse lasers will also be discussed in this paper. In order to study the differences between long-pulse and short-pulse laser-induced damage in optical dielectric thin films, a simple model of single-layer HfO_2 films is analyzed at first. Then we go further for the models of multi-layer films and films with high-absorptive inclusions. The temperature distributions in single-layer HfO_2 films, multi-layer HfO_2/SiO_2 films, and HfO_2 films with high-absorptive inclusions induced by 1 ms and 10 ns lasers are analyzed based on the temperature field theory. The damage property differences between millisecond and nanosecond lasers are summarized from the temperature field theory.

2. Model and theory

A scheme of the film model used for our calculations is illustrated in Fig. 1. Because of the axisymmetric property of the laser irradiating film, an axisymmetric model is established. Laser-induced damage to optical thin films is believed to be a thermally dominant process when the irradiating laser is infrared and its pulse length is a nanosecond or longer [10]. And the process is a consequence of the temperature rise induced by thermal absorption and the laser field effect [6, 8]. The temperature field in films can be calculated from the thermal diffusion equation, considering the thermal source item introduced by absorptions of both intrinsic coating materials and defects [11], which has been indicated reasonable by many researchers [12–15]. The detailed

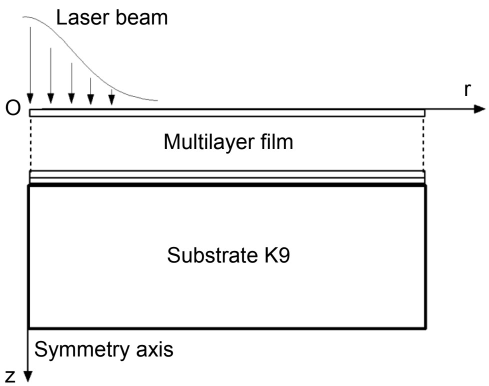

Fig. 1. Scheme of the film model irradiated by a laser.

computational theory and method can be referred to our previous work in Refs. [3] and [16].

3. Calculational results and analysis

The temperature distributions in single-layer HfO_2 films, multi-layer HfO_2/SiO_2 films, and HfO_2 films with a high-absorptive platinum (Pt) inclusion induced by 1 ms and 10 ns lasers are analyzed based on the temperature field theory. The substrates for the three films are K9 glass. The material parameters used for our calculations are summarized in Table 1 [3, 16]. The wavelength of two pulsed lasers is 1064 nm.

3.1. Single-layer films

In the past studies on single-layer optical thin films, the thickness can be ranged from 200 nm to 2000 nm [17–19]. In the present study, the single-layer HfO_2 films of 500 nm, 1000 nm, 1500 nm, and 2000 nm are taken into consideration. For the convenience of analysis, laser energy was chosen based on a criterion that causes the maximum temperature of film components reach melting point of the least material (the minimum laser energy that causes damage to film components) [20]. The $1/e^2$ spot

T a b l e 1. Material parameters used for our calculations.

Materials	K9	HfO_2	SiO_2	Pt
Refractive index	1.52	1.985	1.465	3.7
Absorption coefficient [1/m]	1.181	354.315	141.726	7×10^7
Specific heat [J/(kg·K)]	858	480	841	132.6
Thermal conductivity [W/(m·K)]	1.5	2.0	1.19	80
Density [kg/m^3]	2510	9680	2500	21450

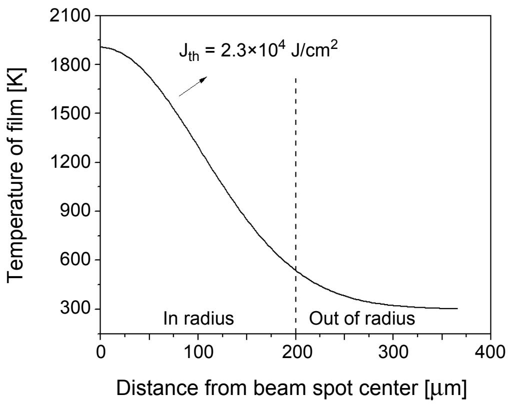

Fig. 2. The 1-ms laser-induced surface temperature distribution of 500 nm single-layer HfO_2 films along the radial direction.

radius is 200 μm. Then the laser fluency correspondingly is called melting damage threshold J_{th}. In our calculations, J_{th} used for 1 ms laser is 2.3×10^4 J/cm^2, and for 10 ns laser is 159 J/cm^2. Figure 2 shows the temperature distribution on the surface of 500 nm single-layer HfO_2 films along the radial direction irradiated by 1 ms laser. The results show that the radial temperature distribution agrees with the Gaussian beam. This distribution form is similar for the 10 ns case and does not cause significant differences between 10 ns and 1 ms laser-induced damages. So the radial temperature distribution will not be discussed any more in the following text.

Figure 3 shows the temperature distribution along the axial direction of 500 nm HfO_2 films at the laser center spot. It can be found from Fig. 3 that for 10 ns laser film interaction case, the maximum temperature is 4196 K, and it decreases to 1758 K at the interface of film and substrate, and then quickly drops to initial temperature at a shallow depth of 750 nm which affects the substrate very little. In comparison,

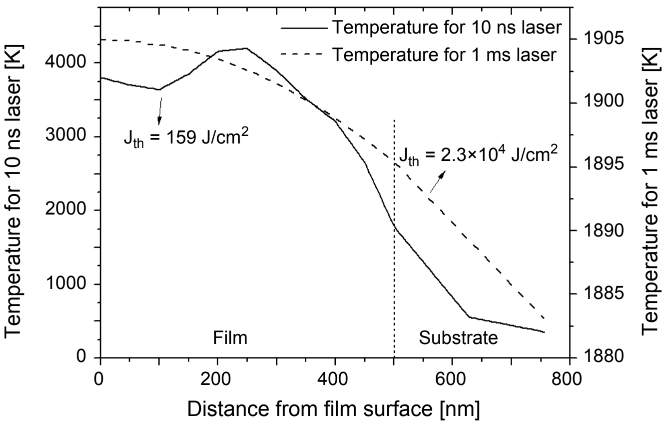

Fig. 3. Temperature distribution along the axial direction of 500 nm HfO_2 films at the laser spot center.

the heat affected depth of 1 ms laser is much bigger than that of 10 ns. The temperature from film surface to interface decreases slightly, only 10 K (1905–1895 K). According to our calculations, the temperature drops to the initial temperature at a depth more than 80 μm, which is 10^7 times deeper than that of 10 ns. By reason that the film temperature and the substrate temperature are so close to each other, the film damage and substrate damage induced by 1 ms laser are difficult to be separated. The two damages are usually observed happening together, which is proved by our previous work (Ref. [3]). In addition, it can be found from Fig. 3 that the axial temperature distribution forms for the two pulsed lasers are also different. Fluctuations appear for 10 ns laser case while monotone decreasing for 1 ms laser case. We think that the fluctuations for 10 ns laser are the result of the laser field effect during irradiation. The discussions about the laser field effect will be described in detail in Section 3.2.

Figure 4 shows the temperature rise processes of the 500 nm HfO_2 film and K9 substrate for 10 ns and 1 ms laser cases. For 10 ns laser film interaction, the temper-

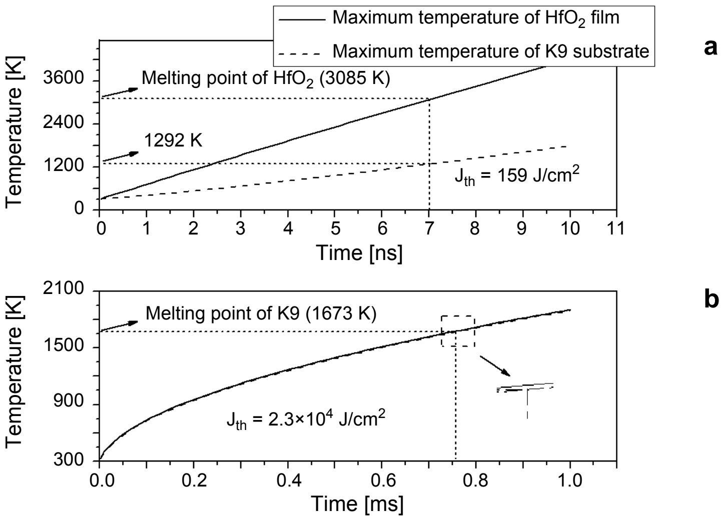

Fig. 4. Temperature rise processes of the 500 nm HfO_2 film and K9 substrate irradiated by 10 ns laser (**a**) and 1 ms laser (**b**).

atures of the film and the substrate are significantly different. At the time $t = 7$ ns, the temperature of HfO_2 film has reached the melting point 3085 K, while the temperature of K9 is 1292 K, below the melting point 1673 K. It means that for 10 ns pulse irradiation, the temperature rise causes melting damage first in the film host, and originates the damage from the film to the substrate as laser energy increases. For 1 ms laser film interaction, the temperature rise of substrate almost overlaps that of the film. At the time $t = 0.76$ ms, the maximum temperature of the film and substrate are 1680 K and 1673 K, respectively. That means that when K9 substrate reaches the melting point (1673 K), the temperature of the film host is still far below the melting threshold (3085 K). So the damage induced by 1 ms laser will originate at the substrate, which is significantly distinguished from 10 ns laser-induced damage. Furthermore, it should be noticed that in our calculations for single-layer HfO_2 films of other thickness 1000 nm, 1500 nm and 2000 nm, there are similar differences between 1 ms long-pulse and 10 ns short-pulse lasers, which is shown in Figs. 2–4. The conclusions of their distinction are the same.

3.2. Multi-layer films

In Section 3.1 we have mentioned the laser field effect during laser film interaction. In order to further study the differences between long-pulse and short-pulse laser film interactions affected by the laser field effect, an HfO_2/SiO_2 multi-layer high-reflection (HR) coating with the obvious standing-wave effect was used for temperature field analysis. The film structure of HfO_2/SiO_2 HR coating was G|(HL)^{12}H|A, where H denoted the high index material HfO_2 with one QWOT (quarter wavelength optical thickness) and L denoted the low index material SiO_2 with one QWOT. Figures 5

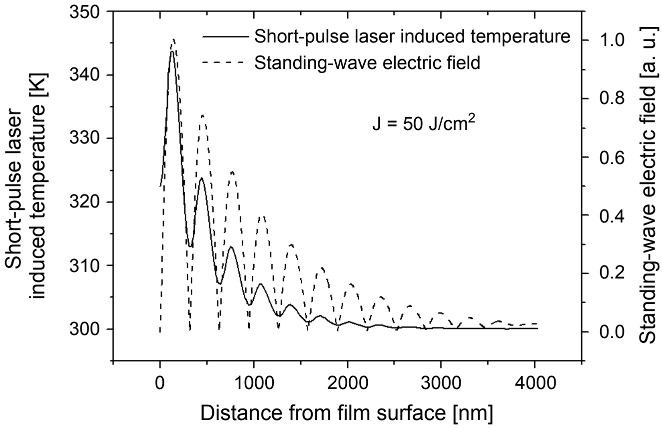

Fig. 5. Axial temperature and laser field distributions in HfO_2/SiO_2 HR coatings irradiated by 10 ns laser.

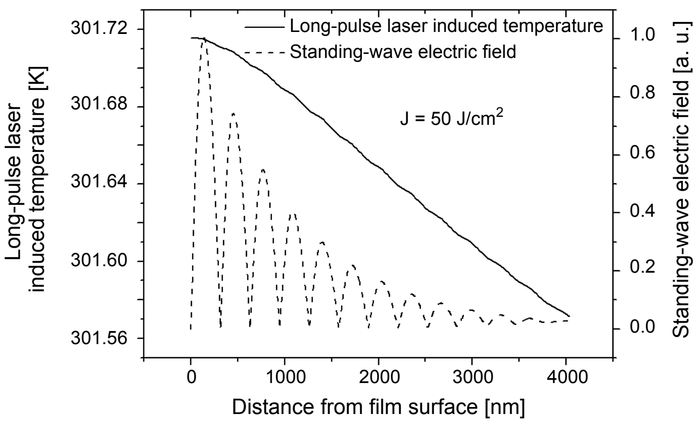

Fig. 6. Axial temperature and laser field distributions in HfO_2/SiO_2 HR coatings irradiated by 1 ms laser.

and 6 show the axial temperature distribution and laser field distribution in the films for 10 ns and 1 ms laser, respectively. The laser energy density used for our calculation is 50 J/cm^2.

It can be found from Fig. 5 that for 10 ns laser, the trend of temperature distribution in film is in general in accord with that of the laser field. However, Fig. 6 for 1 ms laser shows a different point. This difference is caused by the thermal diffusion length μ_{th} which can be expressed as [21]:

$$\mu_{\text{th}} = \sqrt{\frac{\kappa\tau}{\rho c}}$$

where κ, ρ, c are the thermal conductivity, the density and the specific heat of material, respectively, τ is the pulse duration of the laser. Calculated from the formula above, when the pulse duration is 10 ns, the thermal diffusion length of HfO_2 is approximately

Table 2. List of thermal diffusion lengths and thickness of materials.

Pulse duration	10 ns	1 ms
Thermal diffusion length of HfO_2	65 nm	21 μm
Thermal diffusion length of SiO_2	74 nm	23 μm
Thickness of a HfO_2 layer	134.01 nm	134.01 nm
Thickness of a SiO_2 layer	181.57 nm	181.57 nm
The total thickness of HfO_2/SiO_2 films	3920.97 nm	3920.97 nm

65 nm and that of SiO_2 is 74 nm (as shown in Tab. 2). When the pulse duration is 1 ms, the thermal diffusion lengths of HfO_2 and SiO_2 are 21 μm and 23 μm, respectively. It is worth noticing that for 10 ns laser the thermal diffusion lengths of HfO_2 and SiO_2 are both smaller than their respective thickness (HfO_2 134.01 nm, SiO_2 181.57 nm), and for 1 ms laser the thermal diffusion lengths of both materials are so big that they are even bigger than the total thickness of films (3920.97 nm). This is significant. It means that for 10 ns laser film interaction, the absorptive heat could not even transfer out of each film layer efficiently in such a short time. The temperature rises in the film host and cannot distribute uniformly, then appears a form in accordance with the laser field. On the contrary, for 1 ms laser film interaction, the heat transfers out efficiently to the substrate and the temperature in film appears relatively uniform. The laser field effect for 1 ms laser is much weaker than that of 10 ns.

It can be known from the analysis above that for 10 ns laser film interaction, the temperature rises sharply at the peak of the electric field distribution which usually locates at the high–low reflective index interfaces. The high temperatures make the interface area change (physically or chemically) so that the nonlinear absorption increases and produces still higher temperatures. Such a process makes these high–low interfaces more prone to laser-induced damage than other areas within multilayer structure. If the first high–low interface is destroyed and splashed by laser, the second high–low interface exposed to the laser will be the next. The same situation exists for the other high–low interfaces. Early in the year 1997, QIAN ZHAO *et al.* [13] had seen in real time the high–low interfaces destroyed one by one during their experiments. However, for 1 ms laser film interaction, since the temperature field in the film is not affected by the laser field and distributes relatively uniformly, as well as its bigger heat affects deeply, the 1 ms laser-induced damage to film coatings is always observed as a whole in the film and the substrate (see in Ref. [3]).

3.3. Films with high-absorptive inclusion

As we know, during thin film deposition, metal inclusions such as platinum or gold can usually be introduced into film coatings due to impurity of raw material and sputtering of metal electric gun and crucible. For example, in HfO_2 films deposited by jet vapor deposition (JVD), Pt was usually used as the top electrode which will introduce the Pt inclusions [22]. High absorption of laser energy by these inclusions should be a weak link during laser film interaction. There have been many researches

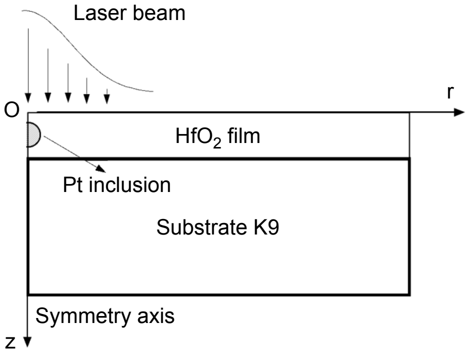

Fig. 7. Model of a film with high-absorptive inclusion irradiated by a laser.

on the inclusion effect of laser damage in nanosecond regime, but little reported in millisecond regime. For the purpose of studying the damage differences between long-pulse and short-pulse lasers when inclusion introduced, the temperature field analysis of 500 nm HfO_2 film with a high-absorptive Pt inclusion (30 nm radius and 250 nm depth) for both 10 ns and 1ms lasers was carried out. The film model with Pt inclusion is shown in Fig. 7, which is similar to Fig. 1 where just a spherical high absorption region was introduced into the film. From Ref. [21], the source term of Pt inclusion is related to the absorption cross-section Q and the incident intensity I:

$$\int A(\mathbf{r}, t)\,\mathrm{d}_{\mathrm{r}}^{3} = QI$$

The absorption cross-section is determined by the Mie theory. So that

$$Q = Q\left(\frac{2\pi r_0}{\lambda}, n'\right)$$

where r_0 is the radius of the inclusion, λ is the wavelength of the incident radiation in the material and n' is the imaginary part of the index of refraction of the inclusion,

$$n' = \frac{4\pi\alpha}{\lambda}$$

where α is the absorption coefficient of the inclusion.

In our calculations, to make results comparable, we set up the certain laser fluencies for 1 ms and 10 ns laser, respectively, which can cause the same temperature rise ($\Delta T_{1\,\mathrm{ms}} = \Delta T_{10\,\mathrm{ns}} = 1$ K) under the condition that there is no Pt inclusion (inclusion radius $r_0 = 0$). That is to say, there is the same effect for certain laser fluencies when $r_0 = 0$. As a result of that, when Pt inclusion is taken into consideration ($r_0 > 0$), the inclusion effects on film temperature for 1 ms and 10 ns lasers are then obvious and comparable. The calculational results of the film temperature rise are listed in Tab. 3.

It can be noticed from Tab. 3 that the introduction of high-absorptive inclusion has a greater influence on 10 ns laser induced temperature. After Pt inclusion is incorporated into HfO_2 films, the maximum temperature rise for 10 ns laser goes up

T a b l e 3. Changes of laser-induced film temperature rises after the introduction of Pt inclusion.

Films	Without Pt inclusion	With Pt inclusion
10 ns laser-induced temperature rise [K]	1	880
1 ms laser-induced temperature rise [K]	1	245

T a b l e 4. Changes of laser-induced film temperature rises after the introduction of Pt inclusion.

Inclusion sizes and depths [nm]	30 nm radius, 150 nm depth	30 nm radius, 350 nm depth	15 nm radius, 250 nm depth	45 nm radius, 250 nm depth
10 ns laser-induced temperature rise [K]	1050	1480	700	680
1 ms laser-induced temperature rise [K]	320	400	200	190

to 880 K, which is nearly 4 times more than that of 1 ms laser (245 K). It is because the thermal diffusion length under 1 ms laser is large enough for the heat to transfer out and distribute uniform so that the temperature will not rise sharply by the inclusion effect. However, the case of 10 ns laser is different because the film temperature rises sharply to be melted and damaged more easily. As it is known, a coating's laser-induced damage threshold (LIDT) can be deduced by applying a temperature criterion chosen as the exceeding of the melting temperature [12, 20]. By this criterion, it can be drawn that the 10 ns LIDT of films will be reduced much more than 1 ms LIDT after inclusion introduced. In other words, it can be said that 10 ns laser is more sensitive to high-absorptive inclusions than the 1 ms one.

The above results are based on a certain inclusion radius and depth. In fact, for our calculations, the radius and depth of inclusion will affect the specific values of temperature rise, but the property differences between 1 ms and 10 ns lasers are not affected. Just as Table 4 shows, we have calculated the case of four other inclusions with different sizes and depths in films. It has been found that 10 ns laser will cause more temperature rises than the 1 ms one for all the cases, which means that 10 ns laser is more sensitive to high-absorptive inclusions than 1 ms laser. The more detailed information about the influence of inclusion size and location can be found in our another work (Ref. [23]).

4. Conclusions

An axisymmetric model of optical dielectric thin films irradiated by laser is established. The temperature distributions in single-layer HfO_2 films, multi-layer HfO_2/SiO_2 films, and HfO_2 films with a Pt high-absorptive inclusion induced by 1 ms and 10 ns lasers are calculated. The damage property differences between millisecond and nanosecond lasers are summarized. The results show that for 1 ms laser, heat affected depth is larger. The substrate is easier to be melt damaged and even can occur prior to film layers. 1 ms laser film interaction is much less affected by the laser field effect than 10 ns case so that the damage by 1 ms laser is always observed as a whole in film and substrate while the damage by 10 ns laser is observed as the high–low interfaces destroyed one by one. If the film contains inclusions, it will be easier to be damaged by 10 ns laser, which means that 10 ns laser is more sensitive to the inclusions than 1 ms laser.

This article presents the information about the damage differences between 1 ms and 10 ns laser-induced damage in optical dielectric thin films. The results of our study can provide evaluation of the damage performances and theoretic foundation for the investigation into the differences of laser film interaction mechanisms between long-pulse and short-pulse lasers.

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