JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 15, No. 9 - 10, September – October 2013, p. 943 - 947

The size effects of inclusions on laser induced film damage

JINGHUA HAN[a], YAGUO LI [b], WEIXING FAN[a], CHANGTAO HE[c], PINGQIU WANG[d], RUIHUA NIU[d] ,GUOYING FENG[a*], QUANXI LIU[d]

[a] *School of Electronics and Information Engineering, Sichuan University, Chengdu, 610064, China*

[b] *Fine Optical Engineering Research Center, Chengdu 610041, China*

[c] *XinJiang Astronomical Observatory, Chinese Academy of Sciences, Urumqi 830011 , China*

[d] *Southwest Institute of Technical Physics, Chengdu 610041,China*

The damage of thin films due to inclusions can be caused by thermal accumulation, interference of scattered light with the incident laser beam and laser induced plasma. The synergic effects of these three mechanisms determine the damage characteristics of optical thin films, each of which is related to the size of inclusions in thin films. Our research results show that the films are damaged owing to the localized interference of light and the melting of thin films when the radii of inclusion particles lie within 24 nm and 40 nm. The laser-induced plasma resulting from the evaporation and ionization of thin film material will be generated when the radii of inclusion particles are larger than 40 nm. Those smaller than 24 nm, can be regarded as safe and free from inducing damage.

(Received August 8, 2012; accepted September 18, 2013)

Keywords: Laser-induced damage, Inclusion, Thermal effect, Scattering, Laser plasma

1. Introduction

Laser-induced damage in optical materials remains a great challenge in high power laser systems [1,2]; in particular, thin films are more susceptible to damage [3,4]. Thereby, the studies on the damage mechanism are badly needed. The researches have been extensively carried out with the advent of lasers [5]. Many physical models have been put forward in an attempt to explain the mechanisms of laser-induced damage in thin films, among which the inclusion induced damage has been accepted widely due to successful explanation of the variation in laser induced damage thresholds (LIDT) with laser pulse duration [6], damage microstructure and morphology [7,8], the influence of inclusion density on the LIDT [9], and so forth.

The film damage by nanosecond laser pulses is associated with the second-phase inclusion particles, which absorb the incident laser light strongly. The absorption from inclusions in thin films to laser light will trigger thermal breakdown in materials and ionization of optical materials [10,11]. In addition, inclusions can reflect and/or scatter the incident laser beam and the scattered/reflected light may interfere with the incident laser beam [12]. Therefore the inclusions affect the incident laser by means of absorbing, scattering /reflecting and interfering the incident light which determinate the damage morphologies of thin films together [13]. We in this article have analyzed each damage mechanics first, then investigated the damage morphologies resulting from different mechanisms, and finally, come to the conclusion that the damage mechanism of thin films is determined by inclusion size.

2. The inclusion-induced thermal breakdown in thin films

The inclusion particles can absorb the incident laser strongly, which will rapidly raise the temperature of particle as well as the surroundings because of the heat diffusion. Supposing the particle shape is spherical, the temperature on the particle surface can be described by the equation (1) [14].

$$T(L)=\frac{3\varepsilon_\lambda JR^2}{2C_{vi}(R+x)D}[(q-m)^{-1} erfc\frac{L}{2(Dt)^{1/2}}-(q-m)^{-1}\exp\left(\frac{(q-m)L}{2R}+\frac{(q-m)^2 D_g t}{4R^2}\right)$$
$$\times erfc\left(\frac{L}{2(Dt)^{1/2}}+\frac{(q-m)(D_g t)^{1/2}}{2R}\right)-(q+m)^{-1} erfc\frac{x}{2(Dt)^{1/2}}+(q+m)^{-1}\times\exp\left(\frac{(q+m)L}{2R}+\frac{(q+m)^2 Dt}{4R^2}\right)$$
$$\times erfc\left(\frac{L}{2(Dt)^{1/2}}+\frac{(q+m)(Dt)^{1/2}}{2R}\right)] \qquad (1)$$

The parameters used in this paper are as follows [15]:

Where $\varepsilon_\lambda\left(spectral\ emissivity\ of\ inclusion\right)=0.3$, $D\left(thermal\ diffivity\ of\ film\right)=3\times10^{-3}cm^2s^{-1}$, $C_v\left(heat\ capacity\ per\ unit\ mass\ of\ film\right)=0.9cal\ cm^{-3}K^{-1}$

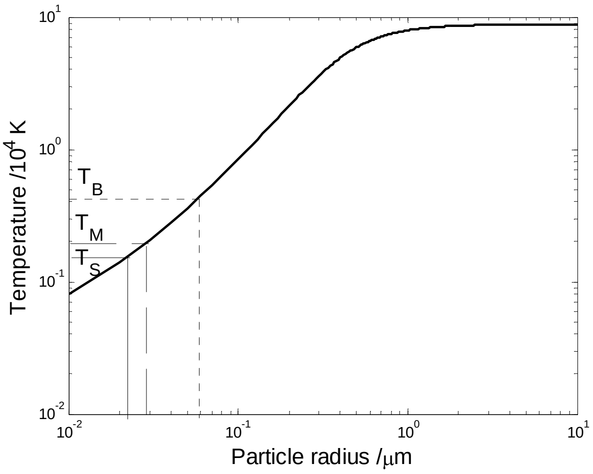
$C_{vi}\left(heat\ capacity\ per\ unit\ mass\ of\ inclusion\right) = 0.66cal\ cm^{-3}K^{-1}$

, $q = {3C_v}/{C_{vi}}$, $m = \left[q\left(q-4\right)\right]^{1/2}$,

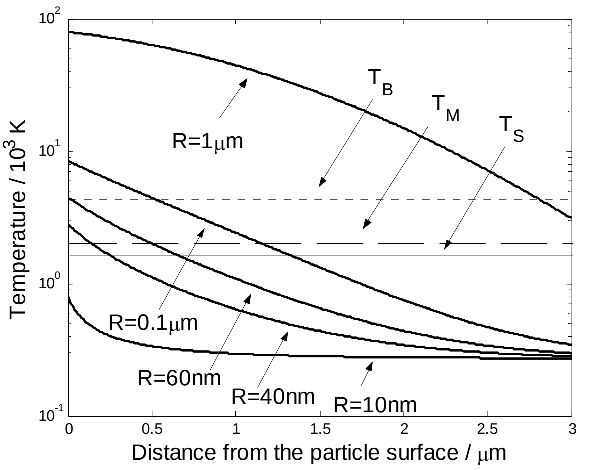
$\mathrm{Ts}\left(softing\ point\ of\ film\right) = 1900K$, ,

$\mathrm{T_B}\left(boiling\ point\ of\ film\right)$=2500K。

The temperature in surrounding medium and on particle surface for a typical laser pulse (Energy: 500 mJ, pulse width (FWHM): 20 ns, beam radius of Gaussian beam 7 mm) can be shown in Figure 1 and 2.



a



b

Fig. 1 The temperature variation on the particle surface (a) and in surrounding medium (b) with particle size

It can be noted from our simulation results that the temperature rises sharply for small inclusions; however, when the size of inclusions is further increased, both the absorption and diffusion will be enhanced and in the end the temperature will level off because of the balance between the deposited energy and the diffused heat. The temperature on the particle surface increases successively to softening and boiling points of thin film materials with increasing inclusion sizes and tends to be steady when the inclusion exceeds 1 µm (Fig. 1(a)).

The temperature of thin film adjacent to inclusions will rise correspondingly with the heat diffusion. The temperature rise in films increases with the particle size and gradually decreases with the distance away from the particle surface. As shown in Fig. 1(b), when the particle radius is 60 nm, the film temperature decreases to boiling and softening points at the distance of about 0.4 um and 0.6 um from particle surface, respectively, much larger than the particle size, which alludes that the thermal damage range can be as large as the order of microns for nanometer inclusion particles. Hamza and his colleagues have investigated the laser induced SiO_2 thin films damage by imbedded gold particles and found that the damage range was of the order of microns[16], consistent to some extent with our simulation above.

3. The inclusion-induced scattering/reflecting and interfering

The inclusions in the thin films can not only absorb the laser pulse energy but scatter it. The scattered or reflected light by metal inclusions may interfere with the incident laser, resulting in interference fringes.

Presuming that the laser of plane electromagnetic wave is normal incident on the films, the scattered and reflected lights satisfy Helmhotz formula. Ignoring reflectivity variation with angle, the definite solution to amplitude distribution of the reflected light (scattered light) can be written as:

in the intermediate region:

$$\Delta r + k^2 r = 0 \tag{2.a}$$

on the internal boundary:

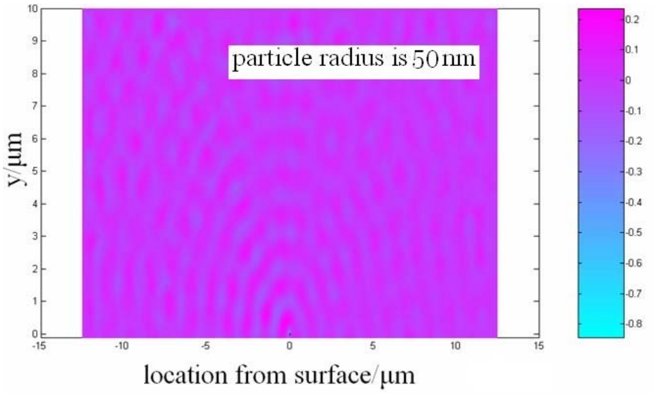
$$r = -e^{-ikx} \tag{2.b}$$

on the outer boundary:

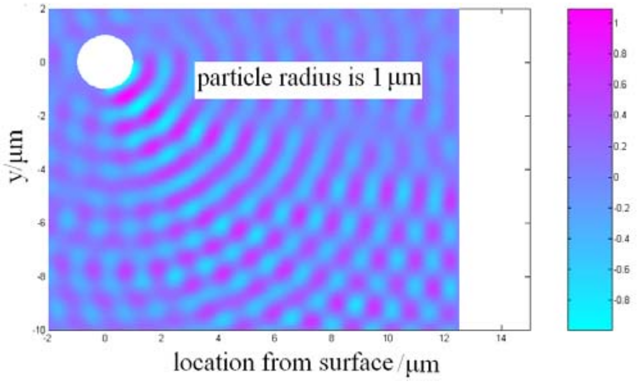
$$\partial r/\partial n = -ik \tag{2.c}$$

where $k = 2\pi/\lambda$ is wave number, (2.a) is the Helmhotz equation, (2.b) is the first boundary condition which regards the scattered light as wave source, (2.c) is a second type boundary condition, derived from absorbing boundary conditions that the scattered light is absorbed by the surrounding border without being reflected.

Here the incident laser wave is 1.064 µm, the wave number k equals to 6 µm^{-1}, and the depth of inclusion is equal to 1µm. Using the PDE toolbox in MATLAB and finite element method, the amplitude distribution of reflected light (scattered light) can be illustrated:



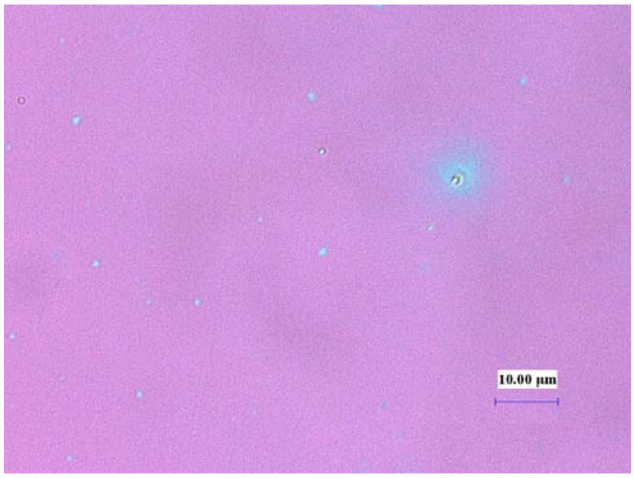
a



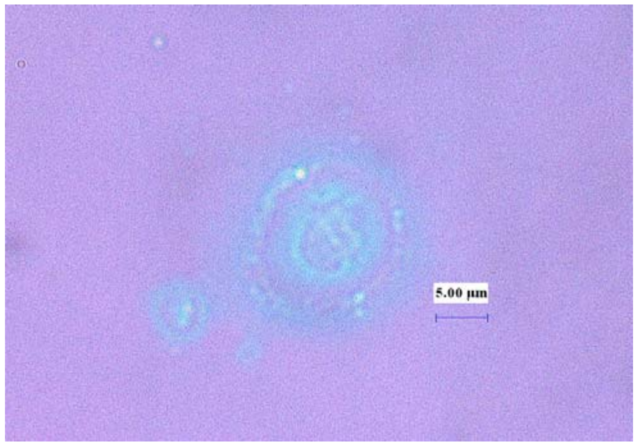
b

Fig.2 The amplitude distribution of reflected light (scattered light) owing to the particle radius of 50 nm (a) and 1μm(b).

Fig. 2 presents that the difference in the ripple intervals scattered by a large (1000 nm) and small (50 nm) particles is minute and the intervals is ~1 μm for both conditions. Since the liquid surface tension decreases with increasing temperature, the molten film has a tendency to expand from hotter to colder regions [17,18]. The coherence of the scattered light and the incident laser contributes to circle-like distribution of temperature. In this way , the ripple damage morphologies appear after the solidification of the molten film material. The representative damage morphologies were imaged (Fig. 3).



a



b

Fig. 3 The ripple damage of SiO_2 antireflection coating at 1.064 μm(examined with an optical microscopy (VHX-600, Keyence, Japan))

Comparing the two photos, it can be found that Fig. 3(b) has several ripples while there are only the damage points in Fig. 3(a). The different ripple numbers may be the consequence of the different damage area of the melted film by various particle sizes. From the results shown in Fig. 1, the highest temperatures of particles with radii <24 nm cannot reach the softening point of thin films and will not cause damage. On the contrary, for the particles with radii between ~ 24 nm and ~40 nm, the melted range will be enlarged with the particle size, producing ripple-shaped damage with various sizes in thin films. If the particles radii are larger than ~40 nm, the highest temperature will be beyond the boiling point of thin films as well as the evaporation is ionized readily upon the laser irradiation due to which the laser plasma will be produced. The laser plasma can cause severer damage to the thin films. We will analyze the effect of laser-induced plasma in the following section.

4. The inclusion-induced laser plasma

Once the laser plasma forms, most of subsequent laser pulse energy will be deposited due to the inverse bremsstrahlung effect, leading to dramatic rise in temperature and enhancing the radiation effects of plasma [19, 20]. The laser plasma possesses more effective ionization effects on thin films than the incident laser, because the radiation spectrum of laser plasma has a wide distribution from deep ultraviolet to soft X-ray [21], which is much shorter than incident laser in wavelength [22]. Under the synergic effects of thermal and ionization, the laser-irradiated zone can be divided into ionization, gas, liquid and solid layer from outside to inside. Therefore, the physical state composition would be rather complex: ionization state of ions, vaporization of the molecules or atoms, molecular clusters, as well as liquid and solid particles [23,24].

With the rapid expansion of laser plasma, the shock wave can be formed [25]. Assuming that the shock wave is released instantaneously at a point, the blast-wave theory is applicable to predict the variation of hemispherical shock wave radius with the laser-matter interaction time (Equation (4) in reference [26]). Using the same parameters in section 2, the variation can be plotted in Fig. 4.

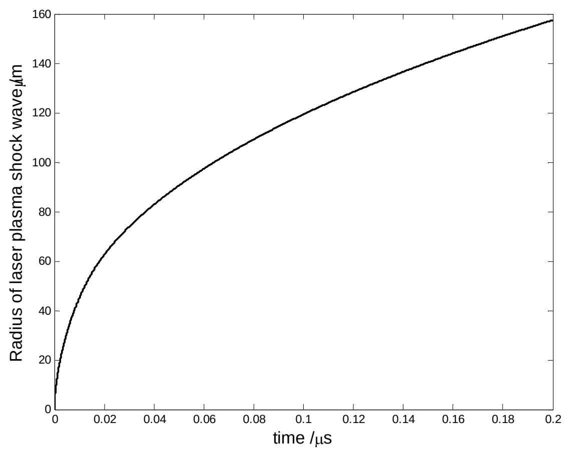
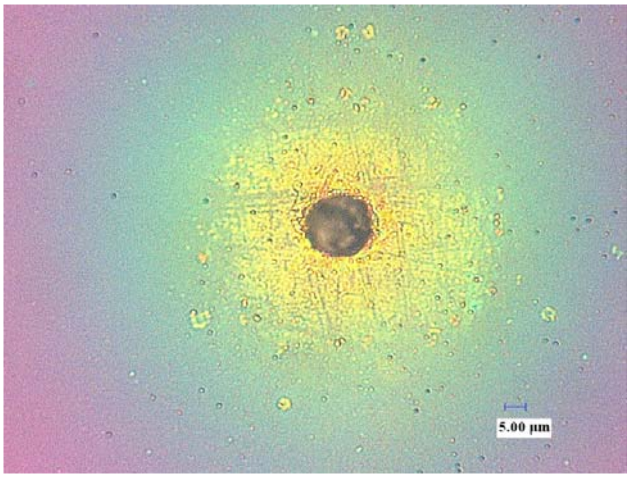
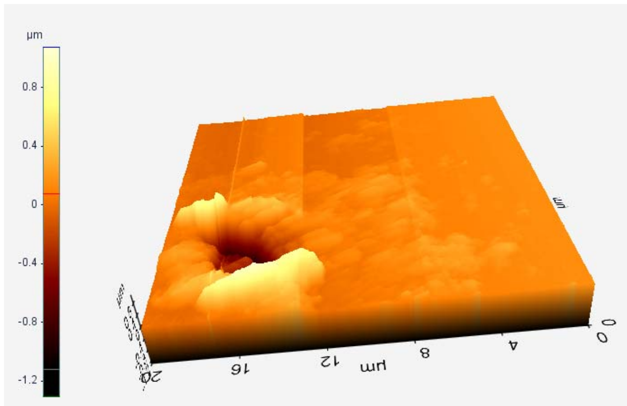


Fig. 4. The diffusion of laser plasma shock wave with acting time of laser pulse.

As shown in Fig. 4, the diffusion velocity of shock wave is fast at early stage and slows down gradually. On account of the repulsive force of laser plasma shock wave, the complex mixture of vaporization and ionization will spread out rapidly, cool down and deposit around the inclusion particles and therefore the pits are formed. The closer to the core of the pit, the more the mixture and the thicker the deposition layer [23]. The typical damage morphologies by laser plasma are presented in Fig. 5.



a



b

Fig. 5 The pit-shaped damage observed by a microscope (a) and Atomic Force Microscopy (AFM) (b)

The pit-shaped damage can be divided into pit and color-changing region, with the radii of about 7 μm and 100 μm, respectively. The depth of pit is several microns. It is clear that the color transits from yellow to green and then to purple that is the color of thin film. The AFM three-dimensional morphology shows that this region is covered with deposition layer that gradually thinned outwards.

5. Conclusion

There are three kinds of damage effects of inclusion on the thin films: thermal effects, scattering /interfering effects and laser plasma effects, which are all influenced by particle size. When the particle radius is <24nm, the absorbed laser pulse energy will diffuse to surrounding medium and the temperature cannot reach the softening points of thin films, so the damage would not occur. For the particle radii between ~24 nm and 40nm, the film material near the inclusions will be melted and the scattering /interfering ripples can be formed. If the temperature is higher than boiling point for the particle radius >40nm, the laser plasma will be produced and the ionization effect and shock effect of laser plasma on the thin films will initiate pit-shaped damage.

Acknowledgements

This research was in part financed by the National Natural Science Foundation of China (Grant Nos. 60890200), and the National Science and Technology Major Project (GFZX0205010803.22).

References

[1] W. Huang, W. Han, F. Wang,et al., Chinese Phys. Lett. **26,** 017901(2009).
[2] J Honig., Opt. Eng. **43.**2904(2004).
[3] M. Reichling, A. Bodemann, N. Kaiser., Thin Solid Films **320,** 264 (1998).
[4] Z. L. Wu, C. J. Stolz, S. C. Weakley, et al., Appl. Opt. **40**, 1897 (2001).
[5] Y Xu, L Zhang, D Wu, et al., JOSA B **22**, 1899 (2005) .
[6] M.F. Koldunov, A.A. Manenkov., SPIE.**3578**, 212 (1998).
[7] J. Dijon, T. Poiroux, C. Desrumaux., SPIE **2966**, 315 (1997).
[8] T. A. Wiggins, R. S. Reid., Appl. Opt. **21** 1675 (1982).
[9] JY Natoli, L. Gallais, H.Akhouayri , et al., Appl. Opt. **41** 3156 (2002).
[10] Michael D. Feit, Alexander M. Rubenchik, et al., SPIE.**3047** 480 (1997).
[11] MD Feit, AM Rubenchik, DR Faux, et al., SPIE **2966** 417 (1996).
[12] FY Genin, K Michlitsch, J Furr, et al., SPIE. **2966** 126 (1997).
[13] FY Génin, MD Feit, MR Kozlowski, et al., Appl. Opt. **39** 3654(2000).
[14] RW Hopper, DR Uhlmann., J.Appl.Phys. **41** 4023(1970).
[15] RN Raman, MJ Matthews, JJ Adams, et al., Opt. Express **18** 1094(2010).
[16] Hamza AV,Siekhaus W J,Rubenchik A M,et al., SPIE.**4679** 96(2002).
[17] Gorodetesky G, Kozyaka T G, Melcher R L et al., Appl . Phys. Lett . **46** 828(1985).
[18] Lemons R A , Bocsh M A., Appl .Phys. Lett **.39**,343(1981).
[19] F. Docchio, P. Regondi, M. R. C. Capon et al., Appl. Opt. **27,**3661(1988).
[20] C. H. Fan, J. Sun, J.P .Longtin., J. Heat trans. **124**, 275(2002).
[21] B. A. M. Hansson, L. Rymell, M. Berglund,et al., Microeng. Eng. **53** 667(2000).
[22] L. V. Keldysh., Sov. Phys. JETP **20** ,1307(1964).
[23] JH Yoo, SH Jeong, R Greif, et al., J.Appl.Phys. **88** 1638(2000).
[24] AV Pakhomov, MS Thompson, DA Gregory., J. Phys. D: Appl Phys. **36** 2067(2003).
[25] RK Singh, J Narayan., Phys. Rev. B**4113** 8843(1990).
[26] Alberto Salleo., Dissertation for the Ph. D. of University of California,Berkeley p:105 (2001).

*Corresponding author: hjhscu@gmail.com,
guoying_feng@yahoo.com.cn