Analysis of 46.9-nm Pulsed Laser Radiation Aftereffects in Sc/Si Multilayer X-Ray Mirrors

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Summary. Specific structural changes in Sc/Si multilayers (MLs) irradiated by nanosecond 46.9-nm single laser pulses with fluences of 0.04-5.00 J/cm^2 were studied by methods of SEM and cross-sectional TEM. The threshold damage was found to be 0.08 J/cm^2. The ML melts down under the fluence F >0.08 J/cm^2, and the exothermic reaction of silicide formation starts. Main degradation mechanisms of MLs are discussed. The results of this study can be used for development of advanced multilayer mirrors capable handling the intense radiation conditions of new generation coherent X-ray sources.

1 Introduction

Multilayer (ML) X-ray mirrors are widely used as optical components in the regions of UV and soft X-rays (1-50 nm) due to their high efficiency and flexibility in the parameters and forms. Under aggressive environment (heating, ion irradiation and exposure to high-power laser radiation) they can degrade as a result of the structural nonequilibrium conditions [1-4]. Growing power of new generation coherent X-ray sources (tabletop laser [5], FEL [6] and others) makes the stability of ML optical properties even more critical.

The Sc/Si MLs developed for the wavelength range 35-50 nm have been successfully used in many applications with the capillary-discharge laser generating at the wavelength λ=46.9 nm [7]. Knowledge of radiation-induced degradation mechanisms in the Sc/Si structures will help finding high-end edges of the application and the ways to enhance their stability.

On the other hand, the very laser radiation represents a specific interest for the surface processing [8]. Diffusion mixing of the layers and formation of the chemical compounds at moderate temperatures indicate the excessive free energy of the MLs. Moreover, the energy released in the mixing process can trigger self-sustained reaction observed in multilayer nanofoils [9]. In this work we discuss features of a single-shot laser influence onto Sc/Si ML degradation process related to the nonequilibrium of the layered structure.

2 Experimental

Sc/Si MLs with the period of ~27 nm were deposited onto silicon and float glass substrates by the method of DC magnetron sputtering. Part of Sc-containing layer in the period was ~0.5. Number of the periods was 10 (Sc/Si/10) on glass substrate and 33 (Sc/Si/33) on Si. MLs deposited onto both type of substrates were exposed to the focused laser radiation operating at λ=46.9 nm. The energy of the pulsed (1.2 ns) laser beam was ~ 0.13 mJ. To vary the fluence value (F) we translated ML sample with respect to the beam focus. Each laser shot stroke the sample surface in a new ML region at normal incidence.

Scanning electron microscope JSM-820 was used to get the information on surface morphology and chemical composition of Sc/Si MLs. Cross-sectional images were produced with the help of transmission electron microscope PEM-U (SELMI, Ukraine) at accelerating voltage of 100 kV.

3 Results and discussions

3.1 Scanning electron microscopy study

We investigated different laser imprints (LIs) obtained with the change of irradiated area on the Sc/Si ML surface, which corresponded to a set of the fluences in the range of 0.04-5.00 J/cm^2. We observed traces of a ML melting at the fluence values starting from F~0.08 J/cm^2. This fluence corresponds to the melting threshold predicted using the thermal diffusion model [10] for the MLs with average thermal characteristics being between Sc and Si.

An incidental change in the morphology of the molten surface was observed at the fluences up to F~0.3 J/cm^2 (fig. 1*a*) in the irradiated areas of Sc/Si/33 ML deposited on Si substrate. Note that the rectangular undamaged area in the center of LI is a shadow of the ML sample inherited from the irradiation scheme [11]. Further growth of the fluence resulted in the melt cracking structures. Cracks advent is an evidence of deep ML melting; better contrast SEM images obtained with the reflected electrons (penetration depth up to ~1 μm) have denoted this fact. First signs of evaporation were revealed at F~0.9 J/cm^2 (fig. 1*b*) in the form of small micron-sized pits (visible in fig. 1*b* as black dots inside the LI center) indicating a melt boiling. Simultaneously a “crown” was formed as a result of pressing out the melt to the LI rim by excessive vapor pressure. The thermal diffusion model gives the lower threshold fluence for evaporation to be F~0.2 J/cm^2. Perhaps, the evaporation process would start before the cracks appearing at the surface, but to define the threshold value more precisely one need to use more accurate and sensitive instruments (for example, probe beam deflection technique [12]). At F~2.2 J/cm^2 the active evaporation or ablation of Sc/Si/33 ML becomes visible. The specific features of this stage are a crater formation in the LI center (fig. 1*c*) and appearing solidified drops around the LI. According to electron microanalysis there is no Sc within the crater of evaporated region, i.e. ML is completely absent in this region. The crater as a rule occupies less than a half the LI area. Presence of the drops means the ML being removed from the center in liquid phase as well. So, the mechanism of a combined ablation is characteristic for the Sc/Si ML: an ejection of the melt and vaporization; it is similar to that for metals [13].

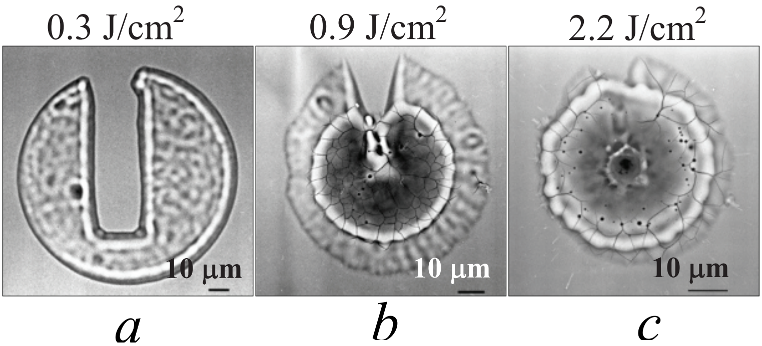

Fig. 1. SEM images of Sc/Si/33 ML on Si substrate irradiated with 46.9-nm laser pulses of different fluences.

Despite the fact that ~97% of absorbed energy is concentrated in top 10 periods, melting and ablation in Sc/Si/10 ML deposited on the glass goes in completely different way as compared to Sc/Si/33 ML. Melting as a rule is accompanied by cracking and flaking the ML practically up to F~0.5 J/cm^2 (fig. 2*a*). An ablation processes in some LI areas begins at F~0.6 J/cm^2 (fig. 2*b*) that is considerably less than that for Sc/Si/33 (2.2 J/cm^2). Fig. 2*c* shows SEM picture for LI at 1.4 J/cm^2 with ML removed from the most LIs. Judging from the presence of the drops the ablation mechanism here didn't change. The rest of laser energy (~3%) is concentrated within the substrate surface because of high absorption [14] and low heat conductivity of the glass [15] compared to Si and Sc [16]. However this energy is insufficient to reduce the observed threshold of the ablation for Sc/Si/10 ML at least to one-third. Taking into account the strong absorption of practically all incident energy in the ML material we expected the similar behavior of MLs with 10 and 33 periods under the irradiation. We believe the difference of heat conductivity in glass and the ML material is the reason of observed distinction.

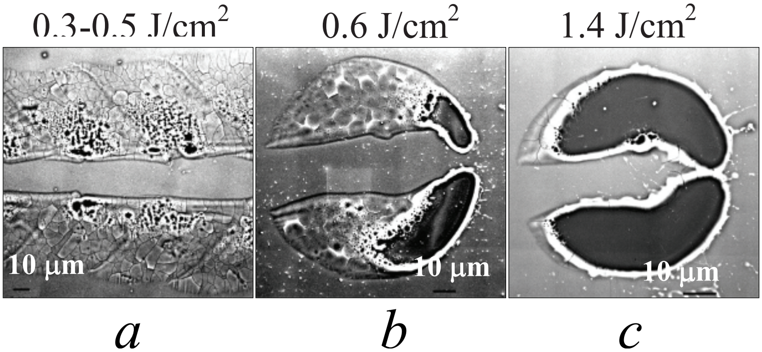

Fig. 2. SEM images of Sc/Si/10 ML on float glass substrate irradiated with 46.9-nm laser pulses of different fluences.

According to our calculation, at F~0.4 J/cm^2 the laser radiation can ablate the Sc/Si/10 ML material completely. However, it looks like it doesn't take place (see fig. 2a). Our estimations show that even at F~0.6 J/cm^2 (fig. 2b) the bottom two periods cannot be melted by the absorbed laser energy directly. Complete ablation can occur at F≥1.4 J/cm^2, with all the layers being melted under the laser irradiation.

3.2 Transmission electron microscopy study

Fig. 2 shows cross-sectional image of the Sc/Si/33 ML after irradiation by the laser beam at F~0.13 J/cm^2 (laser source is on the left). The most of

ML has been molten, and according to electron diffraction analysis the alloy composition corresponds to Sc_3Si_5 silicide. Only 7 periods of 33 survived at the substrate (on the right). This ratio of molten and survived volumes indicates the fluence approaching the full structure melting threshold for the Sc/Si/33 ML. The estimations, however, show that only 4 periods can be molten at that fluency, i.e. only one-sixth of the value observed in the experiment. Such a discrepancy can be explained by the suggestion that the formation of the Sc_3Si_5 silicide results in a release of up to 570 kJ/mol [17]. This energy is enough to heat and melt the material. For the short laser pulse duration (~1.2 ns) no efficient diffusion process can occur, therefore that melting is believed to be the main mechanism of ML degradation.

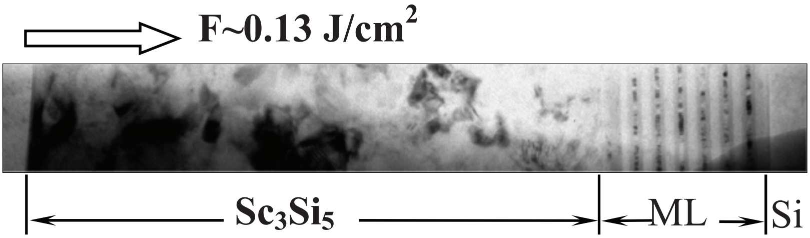

Fig. 3. Cross-sectional TEM image of Sc/Si/33 ML after irradiation at fluence of 0.13 J/cm^2. Laser beam falls from the left. Substrate (S) is on the right.

So, we see that melting of the Sc/Si ML by the laser beam initiates the exothermic reaction, which, from one hand, facilitates the ML ablation (for $F \geq 1.4$ J/cm^2) and, from the other hand, enlarges the molten region at low fluences ($F<0.6$ J/cm^2). The smallest fluence capable melting the surface layers and activating the reaction of silicide formation is calculated to be about 0.08-0.10 J/cm^2 that is rather close to the observed experimental value of 0.08 J/cm^2.

Note that the presented experimental data are a solid ground for the use of the special barrier layers in Sc-Si and Si-Sc interfaces for increasing radiation resistivity of Sc/Si MLs near the laser-radiation induced melting thresholds.

4 Conclusions

We have studied the specific features of Sc/Si ML degradation under the influence of nanosecond 46.9-nm laser pulses. The layer melting at the minimum fluence of 0.08 J/cm^2 was shown to be the main mechanism of ML degradation. The melting initiates the silicide formation and the liberation of considerable amount of heat. In the case of high fluences ($F \geq 1.4$ J/cm^2) this heat facilitates early ML ablation and at low fluences ($F < 0.6$ J/cm^2) it increases the material's molten volume in several times.

References

1. Stearns, D.G.: 'Thermally induced structural modification of Mo-Si multilayers', *J. Appl. Phys.*, **67,** 2415-2427, 1990.
2. Penkov, A.V.: 'Thermal dependence of ion-beam mixing in Mo/Si multilayer periodic coatings', *Metallofiz. Noveysh. Tekhnol.*, **28**, #2, 183-192, 2006 (in Russian).
3. MacGowan, B.J.: 'Investigation of damage to multilayer optics in X-ray laser cavity: W/C, Wre/C, WC/C, stainless-steel/C, and Cr_3C_2/C mirrors', *J. X-ray Sci. Technol.*, **3,** 231-282, 1993.
4. Leguern, F.: 'Experimental study of simulation of the damage induce to various multilayer interferential mirrors by soft X-ray plasma-laser sources', *J. X-ray Sci. Technol.*, **7,** 271-283, 1997.
5. Grisham, M.: 'Damage to extreme-ultraviolet Sc/Si multilayer mirrors exposed to intense 46.9-nm laser pulses', *Opt. Lett.*, **29**, 620-622, 2004.
6. Yu, L.H.: 'First Ultraviolet High-Gain Harmonic-Generation Free-Electron Laser', *Phys. Rev. Lett.*, **91,** 074801, 2003.
7. Benware, B.R.: 'Generation and application of a high-average-power polarized soft-x-ray laser beam', *J. Opt. Soc. Am.* B, **18,** 1041-1045, 2001.
8. Rosenfeld, A.: 'Picosecond UV-ablation of Au and Ni films', *Appl. Surf. Sci.*, **96-98,** 339-442, 1996.
9. Mann, A.B.: 'Predicting the characteristics of self-propagating exothermic reactions in multilayer foils', *J. Appl. Phys.*, **82,** 1178-1188, 1997.
10. Siegel, J.: 'UV-laser ablation of ductile and brittle metal films', *Appl. Phys.* A, **64,** 213-218, 1997.
11. Benware B.R.: 'Focusing of a tabletop soft x-ray laser beam and laser ablation', *Opt. Lett.*, **24,** 1714-1716, 1999.
12. Matthias, E.: 'In-situ investigation of laser ablation of thin films', *Thin Solid Films*, **254,** 139-146, 1995.
13. Veiko, V.P.: 'Two-phase mechanism of laser-induced removal of thin absorbing films. II. Experiment', *J. Phys. D: Appl. Phys.*, **13,** 1571-1575, 1980.
14. Henke, B.L.: 'At. Data Nuclear Data Tables', **54,** 181-342, 1993.

15. Samsonov G.V. (ed.): *Handbook on physical and chemical properties of oxides*, Metallurgiya, 1978 (in Russian).
16. Samsonov G.V. (ed.): *Handbook on properties of elements. Ch. 1. Physical properties*, Metallurgiya, 1976 (in Russian).
17. Golutvin, Yu.M.: 'Enthalpy of silicide formation in Sc-Si system', *Izv. AN SSSR. Metals*, #**6,** 47-54, 1984 (in Russian).