Study of Ablation Threshold of Ionic Crystals for Capillary-discharge XUV Laser Radiation

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Abstract. Ionic crystals are promising materials for testing XUV source in PLD (pulsed laser deposition) arrangement. In this investigation of LiF we use an incidence angle of 20° to study characteristics of ablation and laser-matter interaction under conditions similar as in the PLD arrangement. Single-shot ablation threshold was determined. Also CaF2 was irradiated by XUV radiation and ablation threshold relative to LiF was determined. Irradiated samples have been investigated by Nomarski (DIC—Differential Interference Contrast) microscopy and optical profiler (WLI—white light interferometry).

Introduction

Growing of a thin film by PLD method [Chrisey et al., 1994] has some conveniences as growing thin films at lower temperature (in comparison with thermal evaporation) since the simplicity and stoichiometry is maintained. Nanosecond pulses of 46.9-nm radiation provided by 10-Hz capillary-discharge Ne-like Ar laser—CDL [Heinbuch et al., 2005] can be used for deposition of UV-Vis-NIR transparent dielectric materials with a very weak linear absorption at wavelengths of lasers commonly used for PLD. This CDL system was recently installed in Prague at the Institute of Physics AS CR. Deposition techniques employing the synchrotron radiation are rather close to quasi-continuous sputtering or electron evaporation of thin films than to PLD. The ablation of transparent dielectrics has become more successful by using the ultra-short (picoseconds and femtosecond) pulses when nonlinear absorption of laser radiation occurs at high intensities. However, the ultra-short pulses of UV-Vis-NIR laser radiation are not very suitable to be used in PLD, because the ablation rate is often quite low. Interaction of extreme ultraviolet radiation (XUV) with matter differs from that of the long-wavelength (i.e., UV-Vis-NIR) radiation. The interaction of the short-wavelength radiation occurs mostly due to the photo-effect in atoms of the irradiated material. Thus the absorption coefficient depends mainly on the elemental composition and density of the irradiated material. Contrary to the long-wavelength radiation, there is a little influence on the fine chemical and electronic structure of the particular material. This is the reason why both organic and inorganic dielectrics, which are transparent in UV-Vis-NIR spectral regions, strongly absorb the extreme ultraviolet radiation.

In this article some initial experiments are presented. Experimental parameters varied during these experiments were number of pulses, focus-surface distance, and angle of incidence.

Experimental

The principle of the desk-top capillary-discharge laser system (in Fig. 1 denoted as CDL; [Heinbuch et al., 2005]) is based on lasing of neon-like argon (Ar8+) ions. The laser radiation wavelength is given by transition between states 1S0(2p1/23p1/2)J=0 and 1P1(2p1/23s1/2)J=1 of Ne-like Ar8+.
ions. The capillary is 21-cm long with an inside diameter of 3.2 mm. Capillary is made of Al$_2$O$_3$. An 80–90 kV voltage pulse is applied on the electrodes of the capillary and is followed by a production of capillary plasma in pre-ionized Ar gas at a pressure of 380 mTorr. Under such conditions a z-pinch effect occurs in the capillary plasma. The system emits laser radiation at 46.9 nm wavelength. The laser delivers a beam of 26.44-eV photons with an average energy of several μJ per pulse at the 3-Hz repetition rate; the pulse duration is 1.5 ns. This laser source produces a beam with a radial intensity distribution having a minimum in the centre. The beam profile has an annular shape resulting from a refraction of the amplified rays by radial electron density gradients in the plasma column.

Focusing is provided by a spherical multilayer Sc/Si mirror with focal length of 25 cm and reflectivity of 30 % (1 in Fig. 1). The mirror was designed and manufactured to have the maximum reflectivity at 46.9 nm and the angle between the incident and reflected beam around 6°. More details can be found in [Vyšín et al., 2009]. The beam diameter is about 1 cm at the mirror position.

This simple experimental setup of the laser radiation source and focusing brings several limitations to our investigation, i.e., fixed laser wavelength, shot-to-shot laser output fluctuations, unchangeable, relatively low energy in a single shot (varying with the repetition rate of laser pulses and the capillary aging), finite spot size limiting the maximum fluence (pulse energy over beam area). Here we are following two ways how to change the fluence: (1) choosing different sample-mirror distance and (2) changing the angle between the surface normal and the direction of the focused laser beam. Both ways result to a change in the spot size; in the first way, due to the divergence of the focused beam. Later, the normal-incidence beam spot area should be divided by a cosine of the incidence angle. An advantage of the second way is likely a reduction of the interaction between the incident laser beam and expanding plasma plume owing to their different directions of propagation. This finds a potential use in the PLD arrangement with this XUV source.

The sample manipulator (4 in Fig. 1) enables motion in three axes. The position change along the z axis was used for changing the focus-surface distance. A CCD camera (3 in Fig. 1) served as a visual inspection of the sample positions with respect to the beam. Single crystals of LiF, CaF$_2$ with a thickness of 1 mm and 2 mm, respectively, were used as the samples. Data analysis was done using the open source software Gwyddion [Nečas, Klapetek].

Results and discussion

In order to estimate the single-shot ablation threshold, we have exposed the LiF surface to series of 10, 50, and 100 CDL shots under an incidence angle of 20°. In Fig. 2 we can see the LiF surface after 10 shots exposure at the focus position. In the analysis of these craters we have taken into account that the attenuation length (depth where 1/e of energy is absorbed) is lower than the attenuation length for normal incidence. The total irradiated area of the tilted surface becomes wider in the direction of propagation under these conditions. Thus, the pulse energy (which is kept constant)
Figure 2. A WLI image of LiF surface with a multi-shot damage pattern after 10-shot exposure at the focus position. The angle between the surface normal and the beam direction (arrow) was 20°. The total irradiated area is 3700 μm² and the area of ablated crater projected to the surface plane is 900 μm². Depth of the crater is 0.23 μm.

is distributed over a larger area than the focal spot area for the normal incidence. This decrease of fluence leads to a reduction of crater diameters because the local fluence at the projected beam tail drops below the ablation threshold. This threshold can be computed from a simple equation using the Lambert-Beer law \[ \text{Liu}, 1982 \] \[ d = a \cdot \ln\left(\frac{F}{F_{th}}\right) \] where \( d \) is the maximum crater depth, \( a \) is the attenuation length of the material, \( F \) is the peak fluence and \( F_{th} \) is the ablation threshold fluence. When fluence does not step over the threshold then only desorption process occurs.

Fig. 3 shows craters’ profiles created by the CDL beam under similar irradiation conditions measured by the optical (WLI) profiler. Craters were produced by 10 shots, every crater at the different z-position (mirror-surface distance). In the LiF crater profile we can clearly distinguish between the material eroded in desorption (i.e., low efficient) and ablation (i.e., high efficient) regimes. See refs [Chalupský et al., 2009; Juha et al., 2009] for more details about these erosion regimes under extreme ultraviolet exposure. The maximum fluence (\( F \); related to maximum depth) at different z-positions is computed from equation \( F = \frac{E}{S_{\text{foc}}} \), where \( E \) is the pulse energy and \( S_{\text{foc}} \) is the beam area at 1/e of maximum estimated in a Gaussian beam approach as \( S_{\text{foc}} = (\text{crater surface area} * \)

Figure 3. Profiles of craters created in LiF in tight focus (black solid line), \( z = +10 \mu m \) (gray line), \( z = 21 \mu m \) (black dashed line), and \( z = 31 \mu m \) (gray dashed line) by 10 CDL shots. The angle between the surface normal and the beam direction (arrow in Fig. 2) was 20°. The profiles were measured along the beam propagation direction, i.e., dashed line in Fig. 2.
1 10 100
0.01
0.1
1
Ablation depth [µm]
Number of pulses
CaF2
LiF
Figure 4. Maximum depth of craters ablated by various numbers of CDL pulses (i.e., 1, 5, 10, 20, 50, 100, 200 and 400) accumulated in LiF (circles) and CaF2 (squares) for normal incidence.

attenuation length / maximum crater depth). For different z-positions 0, 10, 21, and 31 µm we compute the maximum fluence as 0.24, 0.02, 0.01, and 0.007 J/cm², respectively. The threshold fluence (pulse energy 2 µJ, single shot) computed from dependence “maximum depth vs. fluence” with known (attenuation length at fixed incidence angle 20°) \( a = 12.8 \text{ nm} \) is \( F_{th} = 0.1 \text{ J/cm}^2 \).

Fig. 4 shows the dependence of the maximum crater depth on number of pulses. We can clearly see a linear dependence. From the ablation rate of different materials we can compute a relative ablation threshold by dividing ablation depth per pulse by attenuation depth of the irradiated material. For all compared craters in CaF2 and LiF the ablation rates are nearly the same. Therefore, we have to consider that from this experiment the ablation threshold of CaF2 is greater than the ablation threshold of LiF in our conditions. This assertion follows from values of attenuation depth 16.8 nm (CaF2) > 13.6 nm (LiF) [Henke et al., 1993]. Ratio of the ablation rate \( d \) (the same for both materials) and attenuation depth \( a \) is inversely proportional with the ablation threshold \( F_{th} \) (\( d/a = \ln(F/F_{th}) \)).

Conclusion
Ablation rate of ionic crystals LiF and CaF2 is governed by attenuation length of XUV radiation in a particular material. Ablation threshold for LiF was experimentally determined as 0.1 J/cm² for incidence angle 20°. This value corresponds to normal incidence value [Ritucci., 2006]. The ablation process is, however, very similar for both materials. Area around the damage craters is clean (free of cracks, particles, and melts). The eroded volume is large enough to enable testing deposition of thin layers. Maximum depths of craters ablated by multiple CDL pulses depend almost linearly on number of shots for calcium fluoride. A saturation behavior was registered only for LiF for hundreds of pulses. This result is going to be verified by next investigation.

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