X-ray laser-induced ablation of lead compounds

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ABSTRACT

The recent commissioning of a X-ray free-electron laser triggered an extensive research in the area of X-ray ablation of high-Z, high-density materials. Such compounds should be used to shorten an effective attenuation length for obtaining clean ablation imprints required for the focused beam analysis. Compounds of lead (Z=82) represent the materials of first choice. In this contribution, single-shot ablation thresholds are reported for $PbWO_4$ and PbI_2 exposed to ultra-short pulses of extreme ultraviolet radiation and X-rays at FLASH and LCLS facilities, respectively. Interestingly, the threshold reaches only 0.11 J/cm^2 at 1.55 nm in lead tungstate although a value of 0.4 J/cm^2 is expected according to the wavelength dependence of an attenuation length and the threshold value determined in the XUV spectral region, i.e., 79 mJ/cm^2 at a FEL wavelength of 13.5 nm. Mechanisms of ablation processes are discussed to explain this discrepancy. Lead iodide shows at 1.55 nm significantly lower ablation threshold than tungstate although an attenuation length of the radiation is in both materials quite the same. Lower thermal and radiation stability of PbI_2 is responsible for this finding.

Keywords: extreme ultraviolet laser, x-ray laser, free-electron laser, radiation damage, laser ablation, damage thresholds, single-shot damage, focused beam characterization

1. INTRODUCTION

In the last decade, new interaction and imaging experiments (see for example [1-3]) were made possible by bringing short-wavelength (λ < 100 nm) free-electron lasers (FEL; see for example [4]) into operation and making them ready for users from scientific community. Another milestone was achieved by obtaining the first lasing in the X-ray spectral

Damage to VUV, EUV, and X-ray Optics III, edited by Libor Juha, Saša Bajt, Richard A. London, Proc. of SPIE Vol. 8077, 807718 · © 2011 SPIE · CCC code: 0277-786X/11/$18 · doi: 10.1117/12.890134

region at the LCLS facility [5,6] (Linac Coherent Light Source; Menlo Park, CA) in April 2009. Two more X-ray free-electron lasers are currently either in commissioning (Spring-8 Angstrom Compact free electron LAser - SACLA in Japan [7]) or under construction (European XFEL in Germany [8]). So, there is an urgent need to modify techniques and instrumentation developed for FELs working in the extreme ultraviolet and the soft X-ray spectral regions for the purpose of their use at X-ray FEL facilities.

At extreme ultraviolet and soft X-ray laser facilities, an ablation of suitable materials has been utilized for visualization and characterization of both lateral and longitudinal distribution of radiation intensity in the focused laser beam [9-11]. Choosing the material most suitable for this purpose, we should take into account an assumption of a local action of deposited energy on ablating material. Materials ablated by short-wavelength radiation in a non-thermal manner, without massive melting and influence of other unwanted phase transitions, represent the best choice. For the wavelengths longer than 10 nm, aliphatic organic polymers, e.g., poly(methyl methacrylate) – PMMA, have been proven as material serving to this purpose very well. Moreover, PMMA is also widely used in electron-beam, EUV, and x-ray lithography as a resist so that its radiation-chemical and radiation-physical properties are very well understood.

However, detailed investigation of the wavelength dependence of PMMA ablation characteristics at the FLASH facility (for more details about the facility, its beamlines and focusing systems see [12,13]) revealed an increase of surface roughness with decreasing wavelength (Fig. 1). Using X-rays to ablate the polymer, the roughness, resulting from increasing attenuation length of radiation and kinetic energy of photoelectrons, of crater surface make determination of the beam characteristics from crater contours, areas and shapes very difficult and unreliable. AFM image of such a rough surface obtained at LCLS can be seen in Fig. 2. Therefore we should abandon materials like PMMA when we are working at X-ray FEL facilities, turning to heavy solids securing short attenuation length of X-rays.

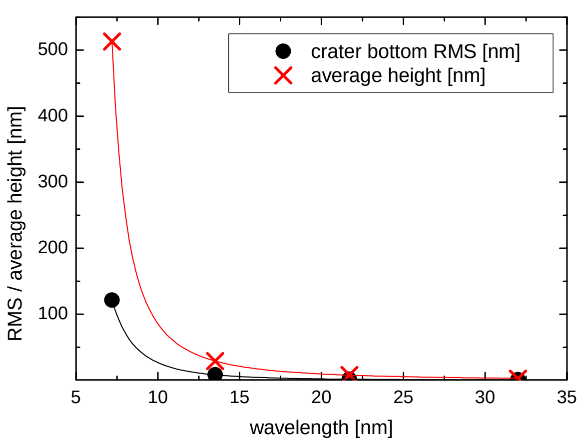

Figure 1: Wavelength dependence of a roughness of PMMA ablated at FLASH tuned from 32 nm to 7.1 nm.

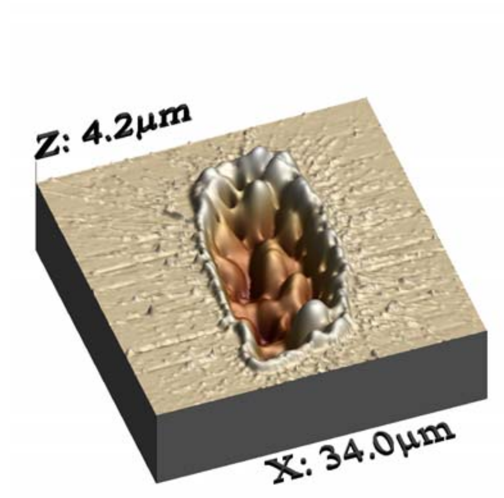

Figure 2: PMMA ablated by LCLS tuned at 1.5 nm.

Attenuation length of X-rays becomes much shorter in materials like lead tungstate ($PbWO_4$; frequently used as a scintillator at HEP facilities [14]) because of its high mean atomic number (Pb,W) and high density (= 8.3 g/cm^3) resulting in a stronger interaction with short-wavelength radiation. Comparable attenuation length we may get in lead iodide (PbI_2) which is often utilized as a semiconductor detector of ionizing radiation [15]. In this contribution, we summarize initial results of experiments carried out at the LCLS facility with the focused X-ray laser beam to determine the single-shot damage thresholds in two lead-containing monocrystalline materials, i.e., lead tungstate and lead iodide. For $PbWO_4$, the results will be compared to the ablation data obtained in the XUV spectral region at the FLASH facility in Hamburg.

2. EXPERIMENTAL

The experiment was carried out using the SXR experimental station [16] of the LCLS facility [5,6]. The SASE (Self Amplified Spontaneous Emission) FEL X-ray source was operated with the electron bunch charge 250 pC and 13 undulator segments tunable to deliver X-ray photons with energies between 800 eV and 2000 eV (λ from 1.55 nm to 0.62 nm). Under these conditions, LCLS is expected to be working in its saturation regime. The duration of a single pulse was about 100 fs. An average pulse energy was of about 1 mJ in an unattenuated beam. Gas attenuator was used to adjust a fluence level at the sample surface. Pulse energy was determined for each shot by nitrogen-fluorescence detector. The transmission of the beamline was measured by using a photo-ionization Gas Monitor Detector (GMD) filled with a suitable rare gas. The LCLS beam was focused on the sample by Kirkpatrick-Baez mirrors with a focal length of 1.2 m [17]. The focusing system guaranties beam diameter at the sample surface of several micrometers in the tight focus. Ablation phenomena were studied changing sample position along the focused beam and varying pulse energy doing z-scans and F-scans, respectively. Proper sample positioning and focused beam characterization were secured with help of ablation imprint techniques [18]. After each shot fired on the sample surface, the target was moved to an unexposed position. Each sample was positioned perpendicularly to the incident LCLS beam in its tight focus.

Single crystals of lead tungstate have been grown by Czochralski technique at the Institute of Physics in Prague. PbI_2 was prepared by direct synthesis from lead and iodine at the Prague Institute of Chemical Technology. The material was further purified by zone melting and grown by Bridgman-Stockbarger method [19]. Irradiated surfaces of both materials were investigated using Nomarski (DIC - differential interference contrast) optical microscope (BX51M DIC microscope, Olympus; Japan) and by an AFM microscope working in tapping mode (D3100 NanoScope Dimension controlled by NanoScope IV Control Station, Veeco; USA).

3. RESULTS AND DISCUSSION

DIC image of a typical crater created by the focused LCLS beam in $PbWO_4$ above the ablation threshold can be seen in Fig. 3. Athough in the center of the damage pattern a circular region can be indicated with traces of surface melting, outer ablation contour is clean and sharp. Both the focal spot area and single-shot ablation threshold were determined from the plot (Fig. 4) of damaged surface areas as a function of a pulse energy logarithm, i.e, by Liu's technique. Details of the method may be found in refs. [9,10,20]. The focal spot area and the ablation threshold inferred from the data in Fig. 4 are A_{eff} = (229±6) μm^2 and F_{th} = (0.11±0.03) J/cm^2, respectively.

Figure 3: The DIC image of a crater formed in $PbWO_4$ by the focused LCLS beam near the ablation threshold.

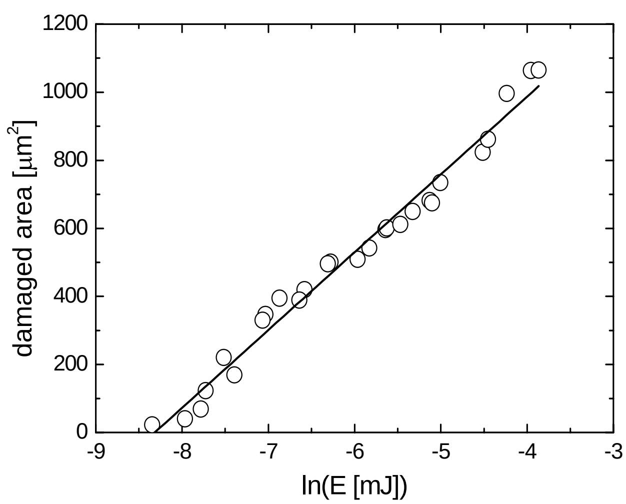

Figure 4: The dependence of $PbWO_4$ surface area damaged by 800-eV LCLS radiation on the pulse energy.

The ablation threshold values obtained for $PbWO_4$ irradiated by a single ultra-short pulse of XUV (FLASH) and X-ray (LCLS) laser radiation at different wavelengths are summarized in Tab. 1. For X-rays, the ablation threshold does not seem to be strongly dependent on a photon energy although an attenuation length listed from Henke's tables [21] changes a lot with changing wavelength. All the thresholds are slightly above 0.1 J/cm^2 from 800 eV to 2 keV. For photons with an energy of 800 eV, the first ablative damage appears around 110 mJ/cm^2. This is a much lower value than expected taking into account the threshold obtained in the extreme ultraviolet spectral region at 13.5 nm, i.e., 79 mJ/cm^2, and the ratio of attenuation lenths

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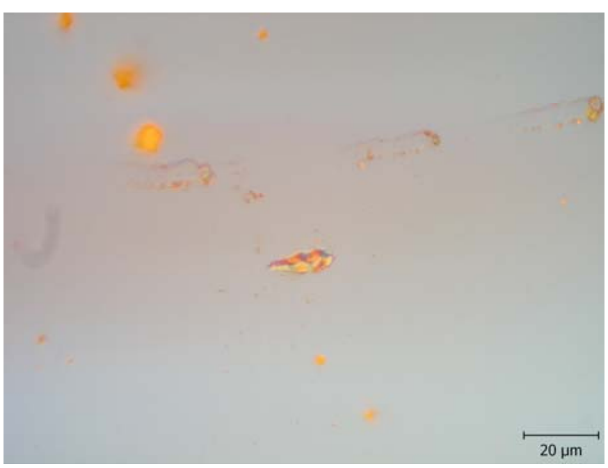
Table 1: Single-shot damage thresholds in lead tungstate irradiated by free-electron lasers tuned at various wavelengths in XUV and X-ray spectral regions.

wavelength [nm]	photon energy [eV]	atten. length Henke [nm]	atten. length [nm]	threshold fluence [mJ/cm^2]	evaluation method
13.5	91.7	33.4	-	79	measured: F-scan
1.55	800	170	-	400	extrapolated from 13.5 nm
1.55	800	170	380	110	measured: F-scan
0.77	1600	675	-	104	measured: z-scan
0.62	2000	509	-	~1200	extrapolated from 13.5 nm
0.62	2000	509	-	~150	measured: F-scan

at these two wavelenths, i.e., 1.55 nm and 13.5 nm. According to Henke's tables [21], the expected value should be around 400 mJ/cm^2 to reach at the wavelength of 1.55 nm a surface energy density corresponding to the threshold obtained at 13.5 nm. Since the threshold is in principle given by surface energy density rate, the expected value should be at LCLS even higher because a LCLS pulse duration ranges around 100 fs while FLASH provides pulses of 30 fs. Moreover, the effective attenuation length determined from LCLS ablation data seems to be longer than a value listed in Henke's tables. An effective attenuation length of 380 nm was found for 800-eV photons, while Henke [21] gives 170 nm. This is another factor that could cause further decrease of estimated threshold value.

The effect is even more remarkable at a wavelength of 0.62 nm, which is the shortest wavelength achievable at the SXR experimental station. The wavelength scaling provides a threshold estimate around 1.2 J/cm^2 while actually obtained value is by an order of magnitude lower, i.e., 0.15 J/cm^2. This finding can be considered as an evidence of the non-thermal character of X-ray ablation in this case.



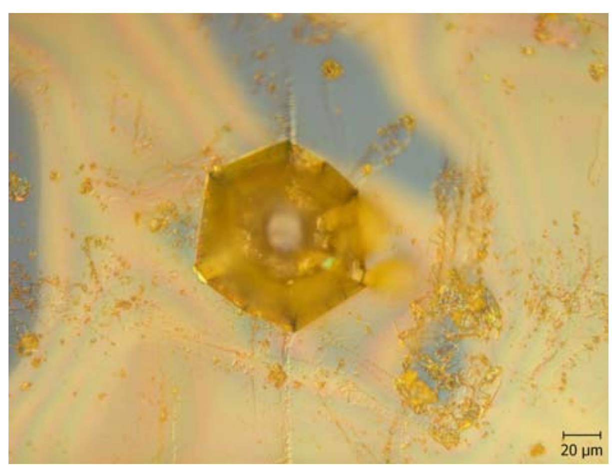


Figure 5: DIC images of PbI_2 irradiated (left) near and (right) above the damage threshold by 800-eV LCLS radiation.

Monocrystalline PbI_2 exhibits a single-shot ablation threshold around 4 mJ/cm^2 when irradiated by 1.5-nm LCLS radiation (Fig. 5). Although there are wide error bars, i.e., (-2;+32) mJ/cm^2, the threshold of PbI_2 is clearly much lower than that obtained in $PbWO_4$ at the same LCLS wavelength. An attenuation length of 1.55-nm radiation is according to Henke [21] quite the same in both materials, i.e., 170 nm and 166 nm for $PbWO_4$ and PbI_2, respectively. The reason is that the mean atomic number of lead iodide is greater than of tungstate but iodide density is slightly lower (= 6.1 g/cm^3). PbI_2 has lower thermal and radiation resistance than $PbWO_4$. Melting points of $PbWO_4$ and PbI_2 are 1398 K [14] and 685 K [22], respectively. PbI_2 [23-25] seems to be less radiation stable than $PbWO_4$ [26,27] under comparable irradiation conditions. Experiments with conventional sources of ionizing radiation are in progress to compare PbI_2 and $PbWO_4$ radiation resistance at the same irradiation conditions. Mechanical damage to lead iodide single crystal appears at high fluences. In Fig. 5 on the right, a hexagonal pattern is broken out the layered structure of PbI_2 because of a high strain rate in the near surface layer. The shape of the pattern originates from the hexagonal symmetry of PbI_2 crystal lattice (i.e., layered structure of *C* 6 type - CdI_2). The nature of mechanical-like damage to solids irradiated by a single LCLS pulse was recently discussed in ref. [28] for low-Z materials.

4. CONCLUSIONS

The dependence of an area of $PbWO_4$ surface damaged by a single shot of 1.55-nm (800 eV) laser radiation on pulse energy shows clearly that ablation of this material by a single laser shot begins at F_{th} = 110 mJ/cm^2. However, wavelength scaling of FLASH results gives an expected value of F_{th} = 400 mJ/cm^2. At 0.62 nm, the difference between the value experimentally determined at the LCLS facility and the value estimated by scaling attenuation lengths is even greater. It indicates a key role of non-thermal, photon energy dependent processes in the X-ray ablation. Observed behavior cannot be explained only by the wavelength dependence of energy density rate on the sample surface and in the near-surface region. LCLS-irradiated PbI_2 exhibits much lower single-shot ablation threshold than $PbWO_4$. This is not surprising because PbI_2 is thermally and radiation less resistant than $PbWO_4$.

ACKNOWLEDGEMENTS

This work was partially funded by the Czech Ministry of Education (Projects LC510, LC528, LA08024, ME10046), Czech Science Foundation (Grants P108/11/1312 and P205/11/0571) and by Academy of Sciences of the Czech Republic (Grants Z10100523, IAA400100701, and KAN300100702). The research leading to the results presented here has received funding from EC's 6th FP under grant agreement No. RII3-CT-2003-506350. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Portions of this research were carried out on the SXR Instrument at the Linac Coherent Light Source (LCLS), a division of SLAC National Accelerator Laboratory and an Office of Science user facility operated by Stanford University for the U.S. Department of Energy. The SXR Instrument is funded by a consortium whose membership includes the LCLS, Stanford University through the Stanford Institute for Materials Energy Sciences (SIMES), Lawrence Berkeley National Laboratory (LBNL), University of Hamburg through the BMBF priority program FSP 301, and the Center for Free Electron Laser Science (CFEL).

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