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Detection of surface changes of materials caused by intense irradiation with laser-plasma EUV source utilizing scattered or luminescent radiation excited with the EUV pulses

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ABSTRACT Extreme ultraviolet (EUV) radiation is absorbed in a thin surface layer of any material. Irradiation of material samples with intense EUV pulses may cause different surface changes. Some of them, especially connected with material desorption, can be clearly visible using an optical or electron microscope. Other changes concerning crystal structure or chemical composition may not be visible under the microscope. They can however be detected using the EUV radiation itself. In this paper a new method of measurement of surface changes by irradiation with a laser-plasma EUV source is presented. The radiation was collected and focused on a material surface using a specially designed multifoil collector. Radiation scattered or excited in the material was detected with the use of a Wolter-type mirror coupled to a back-illuminated CCD camera. Depending on material samples, images with different intensity distributions were registered. For some samples, the intensity distributions of the images obtained before and after irradiation were slightly different. The intensity differences in such cases allowed us to obtain differential images. The appearance of such images was assumed to be evidence of surface changes.

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1 Introduction

Interaction of soft X-rays or extreme ultraviolet (EUV) radiation with materials may cause many different effects. As a result of photon absorption a hole in the valence or core band can be created and an electron is released from the surface or appears in the conduction band. The hole in the core band can decay by the Auger mechanism with creation of electronic excitations of different types. The excitations can decay in either radiative or non-radiative recombination processes. The radiationless recombination may cause defects and chemical changes in irradiated material. An absorption length of EUV radiation in any material is very short. It means that the changes in the irradiated material can occur only in a surface layer with a thickness of tens of nanometres. If the EUV fluence is sufficiently high, material desorption from the surface can occur. If the fluence is relatively low only some physical or chemical structure of the surface can be modified.

Many experiments concerning interaction of the radiation with different materials were performed using synchrotrons or free electron lasers (FELs). It seems however that similar investigations could be carried out using laser-plasma EUV sources. There exist excellent multilayer selective optics enabling us to collect EUV radiation in different wavelength ranges. The most common are the optics for $\lambda = 13.5$ nm. This wavelength is connected with the possibility of production of high-quality Mo/Si multilayer optics having the reflectivity close to a theoretical value of about 70%. The radiation may also be collected using wide band grazing incidence optics. In this case the whole radiation from the soft X-ray range to EUV can be utilized. In both cases a wide collection angle enables us to obtain 10^{15}–10^{17} photons/cm^2 in a single pulse with time duration of the order of 1 ns. It means that the radiation fluence of the focused radiation can be comparable with the fluence of focused radiation of a vacuum ultraviolet (VUV)-FEL and much higher compared to synchrotron radiation. Of course, the pulse duration of a FEL is 4–5 orders of magnitude shorter compared to laser plasma. It gives then respectively higher power density in a single pulse. The average power, however, can be comparable. It means that laser-plasma sources can be applied instead of synchrotrons or FELs for some applications requiring EUV radiation with a fluence of 0.01–1 J/cm^2 and not very high power density. It can also be used in some investigations offering other ranges of radiation parameters in comparison with synchrotrons or FELs.

The usefulness of laser-plasma sources was demonstrated in some micromachining experiments. Such investigations were performed initially using synchrotron radiation [1]. Experiments with laser-plasma sources concerned mainly polymers [2–5]; however, micromachining of inorganic materials was also performed [6]. Even if the EUV fluence is too low for material desorption [7], the radiation causes some changes that can modify radiation scat-

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tered at the surface or influence luminescent radiation excited in the material. Spectral investigation of the luminescent radiation can give information about the electronic structure of crystals [8]. Depending on the experiment, the radiation used for excitation can be mono- or polychromatic. Depending on the material, the excited radiation may be emitted in the visible [9, 10], the ultraviolet [11, 12] or even in the EUV wavelength range [13, 14]. The investigations concerning such phenomena were performed using synchrotron, FEL and X-ray laser radiation for different materials. In some of the experiments the spectra were modified after irradiation with a certain dose, due to modification of the chemical structure [14, 15].

In this paper selected results of interaction of EUV radiation with different inorganic materials using a 10-Hz laser-plasma EUV source based on a laser-irradiated gas puff target [16] are presented. We propose a simple method for detection of surface changes under intense EUV irradiation by utilizing the EUV radiation itself. Luminescent and scattered radiation emitted from irradiated samples were measured in a wide spectral range from EUV to infrared (IR). Some evidence of material changes was revealed.

2 Experiment

The investigations were performed using an experimental setup presented in Fig. 1. In the experiments, a 10-Hz laser-plasma EUV source was used. The source was based on a double-stream gas puff target irradiated with 4-ns, 0.8-J Nd:YAG laser pulses. The EUV radiation was collected and focused using a multifoil optic. Detailed description of the source and the optic can be found elsewhere [16, 17]. In this experiment plasma was created in a Xe target. Radiation in this case was emitted in a wide spectral range from IR to EUV. The EUV part of the radiation was selected using a 140-nm Zr filter. The spectrum of the focused EUV radiation was also modified in relation to the direct plasma radiation because of the relatively low reflection coefficient of gold-plated collector mirrors below $\lambda \sim 8$ nm. To measure the spectrum an additional 200-nm Zr foil was mounted at a focal plane of the collector. The spectra were taken using a 5000 l/mm transmission grating coupled to a back-illuminated CCD camera. An optical axis of the spectrograph was located coaxially with the axis of the collector. There was no direct radiation along the axis because of mechanical construction of the collector, so that only the radiation scattered on the Zr foil could be registered. The spectrum contains a large number of unresolved lines with the dominating feature in the wavelength range around 11 nm and does not differ much from the spectrum of direct Xe plasma radiation in the wavelength range $\lambda \sim 6$–18 nm. A typical Xe spectrum of the focused EUV radiation is presented in Fig. 2. EUV radiation from the source was used both for creation of surface changes and for detection of the changes. Samples for irradiation were mounted in the EUV radiation collector focal plane. The samples were irradiated during 2–4 min with 10-Hz repetition rate and maximum fluence $\sim 30\ \mathrm{mJ/cm^2}$ [17]. No filter was applied for irradiation. It means that surface modifications were created using unlimited radiation from the source. For detection of surface changes the 140-nm Zr filter was mounted between the EUV collector and a sample. The wavelength range in this case was limited to $\lambda \sim$ 6–18 nm. Transmission of the filter in this range was about 50%. The radiation intensity was then much lower compared to irradiation without any filter. It means that in this case material changes caused by incident radiation were less significant than for unlimited irradiation. It was however possible to obtain scattering or luminescence with an intensity sufficient for registration. Radiation registered within the wavelength range $\lambda \sim 6$–18 nm could be either luminescent or scattered. For longer wavelengths radiation was emitted from the sample as a result of luminescence. The registrations were performed using an imaging system containing a Wolter-type optic and a back-illuminated CCD camera. The images were taken for radiation emitted under an angle of 60° with respect to the collector axis.

The magnification of the measuring system was 10 and the resolution, mainly limited by the resolution of the CCD detector, was 5 μm. Without any additional filter the system allowed us to register images in a whole wavelength range from IR to EUV. Measurements in selected wavelength ranges were performed using different filters mounted between the irradiated samples and the Wolter optic. The radiation was registered in a wavelength range $\lambda \sim 6$–18 nm using a 200-nm Zr foil, $\lambda \sim 17$–70 nm using a 750-nm Al foil, and $\lambda > 130$ nm using a CaF_2 filter. There was no filter for the wavelength range $\lambda \sim 70$–130 nm. The in-

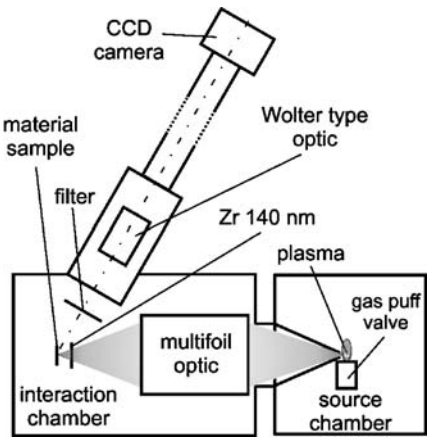

FIGURE 1 Schematic view of the experimental setup for irradiation of material samples and registration of scattered and luminescent radiation

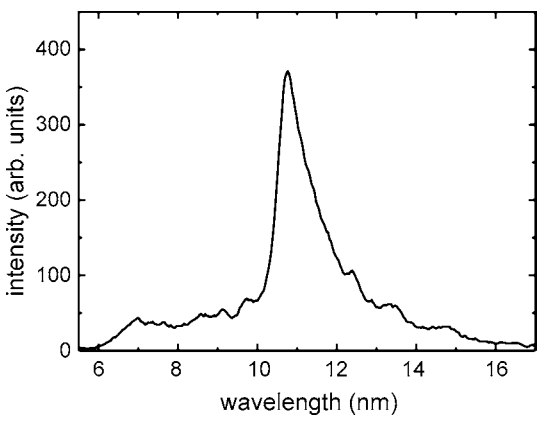

FIGURE 2 A typical spectrum of xenon plasma EUV radiation focused with the multifoil collector and filtered using a 140-nm Zr foil

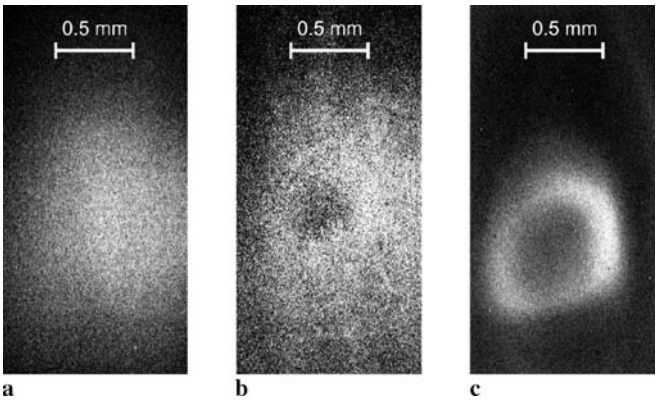

FIGURE 3 Differential images indicating surface changes arisen during intense EUV irradiation of the following material samples: (**a**) 30-μm Al foil, (**b**) CaF_2, (**c**) mica crystal

tensity in this range could be estimated as a difference between radiation registered without any filter and with the use of the filters mentioned above. Using such an experimental setup, measurements concerning intensity changes of luminescent or scattered radiation after EUV irradiation were carried out. The images were registered before and after irradiation. For some materials the irradiation resulted in changes sufficiently strong to be revealed from differential images. Such an image could be obtained as a difference of intensity distributions between images registered before and after irradiation.

3 Results

The measurements were performed for different materials, either elements or chemical compounds. Intense irradiation of some material samples with maximum fluence caused different effects. For very thin (750-nm) Al foil, for example, there could be noticed deformations connected with melting of the metal. The deformations could be easily observed in visible light. Similar evidence of melting was revealed for 10-μm Sn foil. Such deformations did not occur for thick Al foil and also for other metallic foils with relatively high melting temperatures (Mo, Ta, Fe). There were also no visible deformations of surfaces of non-metallic inorganic materials (Si, mica, CaF_2) after intense irradiation with EUV pulses even using a scanning electron microscope. For some material samples, however, surface changes could be revealed by utilizing scattered EUV or luminescent radiation excited with EUV. The measurements were performed with the experimental procedure described above. The most interesting results with clearly visible differential images were obtained for three material samples: 30-μm Al foil, mica crystal and CaF_2. The images are presented in Fig. 3. There can be noticed a significant difference between images obtained for aluminum and for mica. For aluminum the differential images were obtained by subtraction of intensity distributions registered before irradiation from the distributions registered afterwards. For mica there was no image without earlier irradiation. It means that the differential image is the same as the image obtained after irradiation. There can be noticed a characteristic annular structure with lower intensity at the centre. A similar structure can be noticed also for CaF_2. Intensity profiles taken across the images are shown in Fig. 4. For aluminum the intensity distribution corresponds moreover to the intensity distribution of the focused EUV radiation with a maximum at the centre. It means that the surface changes are of the same kind independent of the EUV intensity. There was probably melting of a thin surface layer under EUV irradiation followed by its fast freezing. The intensity profiles for CaF_2 and especially for mica are quite different. It can be noticed that the profiles for both materials have a minimum at the centre. It means that the material surface changes depend on the EUV fluence. For CaF_2 the images could be obtained in a wavelength range $\lambda > 70$ nm. For shorter wavelengths with the use of the 200-nm Zr or 750-nm Al there was no image. Radiation registered in this case was then shifted towards much longer wavelengths in relation to the incident radiation. For mica the images could be registered without any filter or using the 200-nm Zr filter. There was no image using other filters. It means that the radiation was emitted in the same wavelength range as the incident radiation $\lambda \sim 6$–18 nm and could also be emitted in the wavelength range not covered by any filter $\lambda \sim 70$–130 nm. For CaF_2 the images came from luminescent radiation only while for mica it could be either luminescent or scattered radiation. To adjudicate what was the origin of the radiation, spectral measurements are necessary. Nevertheless, it is clear that at least for CaF_2 for relatively low EUV fluence some processes took place, changing luminescent properties of the material. For the highest

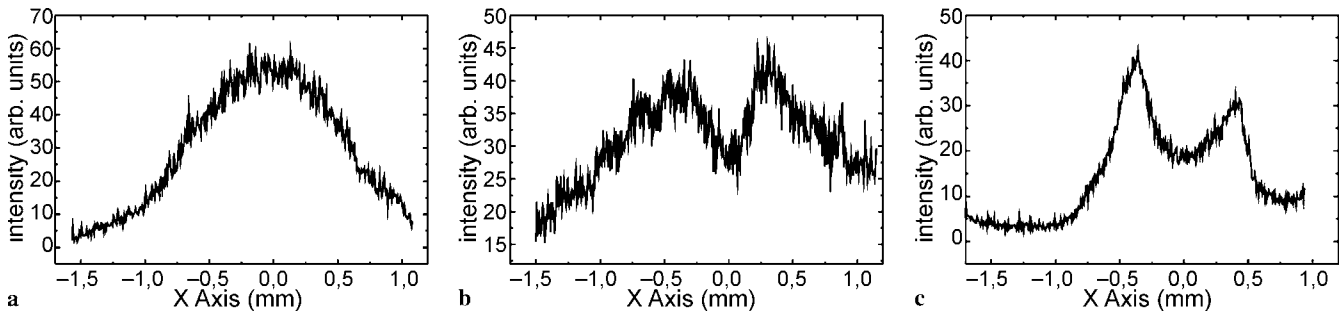

FIGURE 4 Intensity profiles taken across the differential images shown in Fig. 3 respectively for the samples: (**a**) 30-μm Al foil, (**b**) CaF_2, (**c**) mica crystal

fluence close to the focus centre, chemical decomposition of the material or its evaporation can occur. In this area luminescent properties can be similar to the unchanged material.

In summary, first results of irradiation of inorganic materials using a laboratory EUV source based on a laser-irradiated gas puff target were presented. A simple method of detection of surface changes utilizing the EUV radiation itself was proposed. This method enables us to control surface changes of materials during irradiation. By replacing the imaging system with a spectrograph, more precise investigations of physical and chemical surface changes could be performed.

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