Low electron temperature in ablating materials formed by picosecond soft x-ray laser pulses

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ABSTRACT

To study the ablation process induced by the soft x-ray laser pulse, we investigated the electron temperature of the ablating material. Focused soft x-ray laser pulses having a wavelength of 13.9 nm and duration of 7 ps were irradiated onto the LiF, Al, and Cu surfaces, and we observed the optical emission from the surfaces by use of an optical camera. On sample surfaces, we could confirm damage structures, but no emission signal in the visible spectral range during ablation could be observed. Then, we estimated the electron temperature in the ablating matter. To consider the radiation from a heated layer, we supposed a black-body radiator as an object. The calculation result was that the electron temperature was estimated to be lower than 1 eV and the process duration was shorter than 1000 ps. The theoretical model calculation suggests the spallative ablation for the interaction between the soft x-ray laser and materials. The driving force for the spallation is an increasing pressure appearing in the heated layer, and the change of the surface is considered to be due to a splash of a molten layer. The model calculation predicts that the soft x-ray laser with the fluence around the ablation threshold can create an electron temperature around 1 eV in a material. The experimental result is in good accordance with the theoretical prediction. Our investigation implies that the spallative ablation occurs in the low electron temperature region of a non-equilibrium state of warm dense matter.

Keywords: soft x-ray laser, laser ablation, laser produced plasma, electron temperature

1. INTRODUCTION

The interactions of short pulse lasers with matter are interesting in both of technological applications for manufactures and physical phenomena. Lasers with short durations abilities to make high temperature and high pressure in materials. Until now, we have reported on the ablation and/or modification structures on the material surfaces induced by the soft x-ray laser (SXRL) pulses. On aluminum (Al) surface, we found that unique nanometer sized conical structures were created[1,2]. On gold (Au) and copper (Cu) surfaces, nanometer scale ripple-like structures were formed[3]. On silicon (Si) surface, deep holes having several hundred nanometers in depth appeared after the multiple SXRL pulse irradiation[3]. The wavelength of the SXRL beam is shorter than those of visible or infrared lasers, which means that an SXRL beam potentially has the ability to draw small patterns on material surfaces. Hence, the SXRL is a candidate a tool to fabricate nanometer scale structures.

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We have also studied the surface ablation/modification mechanism theoretically. In the low fluence region, especially around the ablation threshold, spallation phenomena are predicted[4–7]. The driving force for the spallation is an increasing pressure appearing inside the heating layer. Around the ablation threshold, it is considered that the first change of the surface structure is due to a splash of a molten layer, because the dependence of the molten depth on the fluence agrees with experimental results[6,7]. The theoretical approach for an interaction model of a SXRL pulse with material surfaces plays a crucial role in the explanation of the nanometer scale surface modification.

The penetration of SXRL pulse into matter during surface ablation/modification process accompanies heating of the electrons in the matter. During the heating, the density is of the order of solid state density, so that such a process occurs in a non-equilibrium state with relatively high electron temperature, so called warm dense matter (WDM). However, the details of the surface ablation/modification process have yet to be clearly resolved. One of the difficulties in the theoretical description is the characteristic time of equilibrium states between electrons and ions, which is comparable to the ablation time including heat transfer, phase transitions, and appearance of shock waves. Therefore, experimental results are very important to describe the fundamental process of the surface ablation/modification mechanism.

To study the ablation/modification mechanism experimentally, especially in the soft x-ray range, spatial, temporal, and spectral analysis experiments are necessary. Then, we have initiated an experimental study for investigation of the surface ablation/modification mechanism by SXRL pulses. In this paper, we discuss the electron temperature in ablating matters formed by picosecond SXRL pulses.

2. EXPERIMENT

The SXRL irradiation experiment was carried out at the SXRL facility at Japan Atomic Energy Agency (JAEA). The experimental setup is shown in Fig. 1 schematically. The SXRL pulse, which was generated from silver (Ag) plasma mediums used in an oscillator-amplifier configuration, was simply focused on a sample surface by using a Mo/Si multilayer coated spherical mirror. The characteristics of the SXRL pulse were wavelength of 13.9 nm (photon energy of 89 eV), bandwidth of narrower than 10^{-4}, and duration of 7 ps[8]. The Mo/Si multilayer coating was optimized for soft x-rays with a wavelength of 13.9 nm at normal incidence. A 0.2 μm thick Zr filters were placed in front of the spherical mirror in order to reduce the scattered optical radiations from the Ag plasma. At this wavelength with normal incident irradiation, almost the entire incident energy of x-ray photons was absorbed by the samples. The typical energy of the SXRL pulse reaching on the sample surface was approximately 48 nJ, and fluence was estimated to be around 20 mJ/cm^2, which was calculated as an average value in the damaged area[1,3].

Emission light in the visible spectral range from a sample surface induced by the SXRL irradiation was observed by an optical camera. A complementary metal oxide semiconductor (CMOS) device with approximately 24 mm x 16 mm in

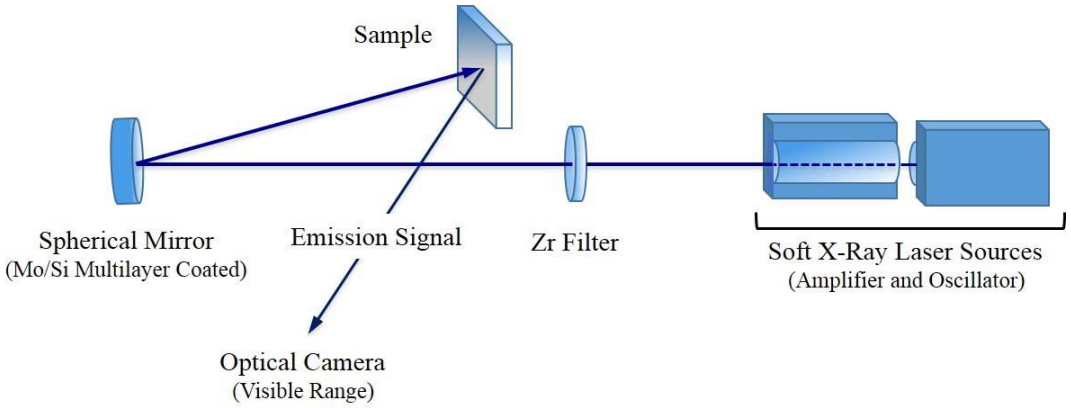

Figure 1. Schematic diagram of experimental setup. Soft x-ray laser pulse generated from Ag plasma mediums is focused on the sample surface. Emission light in the visible range from the sample surface is observed by an optical camera.

size, which had the pixel size of 4.8 μm x 4.8 μm, was used as an image sensor of the camera. The optical system observed radiation in the spectral region from 400 nm to 800 nm (in the photon energy region of 1.54–3.08 eV). The detectable energy threshold was estimated to be approximately 0.016 nJ/pixel (1.6 x 10^{-11} J/pixel). The optical image of the emitted light was transferred from vacuum to air through a quartz viewport. The SXRL irradiation was performed in a vacuum, and the camera with the CMOS detector was in air.

Lithium fluoride (LiF), Al, and Cu plates were used as sample targets. LiF is a typical dielectric with well-known characteristics. Al is a metal with a simple zone structure and Cu has a complicated electron spectra having *d*-electrons. We expected that the surface ablation/modification mechanism for such different solid materials would be similar, because the x-rays are absorbed mainly by internal shells in the material. We used rather thick samples: the thickness of the LiF plate was 2 mm, and those of the Al and Cu plates were 1 mm. All sample plates were mounted on a movable sample holder in order to ensure equal focusing conditions. The fresh surfaces were placed at the best focal position of the SXRL beam.

3. RESULTS AND DISCUSSION

Emission light in the visible range from every sample surfaces during the irradiations of SXRL pulses were captured by an optical camera. Figure 2 shows observed results for sample surfaces. Luminescent from Cerium doped Lutetium Yttrium Orthosilicate (Ce: $Lu_{2-x}Y_xSiO_5$; Ce: LYSO) scintillator was also shown for a reference. Ce: LYSO scintillator has luminescence with wavelengths of 420–480 nm in visible range, which can be generated by SXRL beam. As shown in Fig. 2, we could confirm the arrival of SXRL pulse (impossible to see by eyes) on the sample place by observing luminescence from the scintillator. On the samples, we could not observe the any optical signal from irradiated LiF surface even by adjusting the brightness and contrast of the acquired images. However, a damage structure was created on the LiF surface by a SXRL pulse irradiation. Figure 3(a) shows the beam pattern of the SXRL pulse recorded on the LiF sample surface obtained with a fluorescent microscope. On a LiF crystal, stable color centers (CCs) are formed by the SXRL pulse irradiation[9]. By observation of the photoluminescence pattern from the CCs, we could confirm the irradiated position, easily. At the center of the fluorescent image on LiF surface, as shown in Fig. 3(b), we could find the ablation structure induced by the SXRL pulse irradiation. By using the same setup, we also irradiated Al and Cu sample surfaces. As shown in Fig. 2, we could not observe any emission signals, despite the fact that clear damage on the target surfaces occurred. The results obtained with the dielectric and metal samples were the same, even though material properties, such as electric resistivity, melting point, x-ray attenuation length, and so on, were different. It can be concluded that the ablation and/or surface modification process induced by the SXRL pulse will occur without a plasma production.

The experimental results obtained above mean that the photon intensity reaching the CMOS pixels, i.e., integrated energy of visible light during the exposure time of the camera, emitted from the ablation plasma produced by only the SXRL pulse, did not exceed the detectable energy threshold of the CMOS detector. The production of WDM having a low electron temperature is a possible plasma-less ablation process in materials whether they are dielectric or metals. We could consider the plasma-less ablation process should be an important process to consider as a SXRL interaction mechanism with all matter.

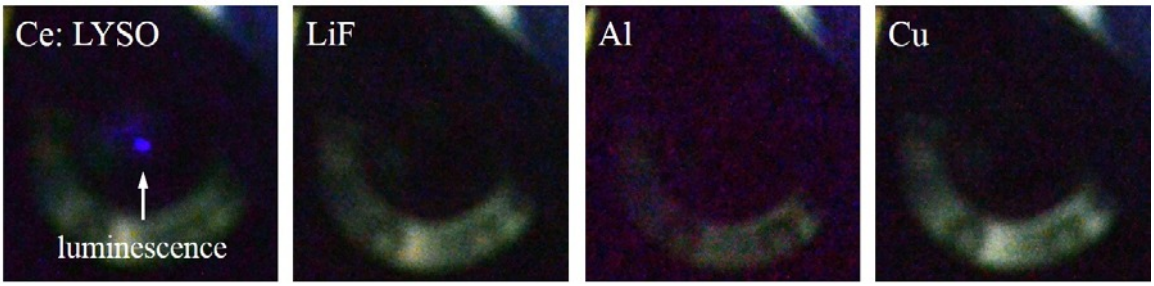

Figure 2. Acquired mages of the SXRL pulse irradiated LiF, Al, and Cu sample surfaces by the CMOS detector. Luminescence from the Ce: LYSO scintillation generated by SXRL beam are shown as reference.

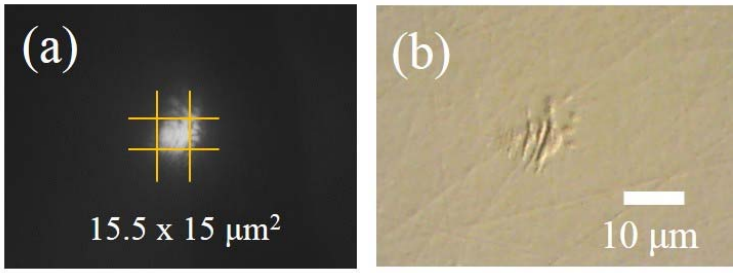

Figure 3. (a) Photoluminescence image from color centers on LiF surface observed by a fluorescence microscope. (b) Ablation structure created on the LiF surface, which was formed in the center of the photoluminescence image.

The atomistic model calculation predicts that the absorbed energy of SXRL pulse having a fluence of 65 mJ/cm^2 for a surface modification creates an electron temperature of around 1 eV in a material[7]. In this experiment, the fluences of the focused SXRL pulses were lower than 30 mJ/cm^2, so the electron temperature would be smaller than 1 eV. If the electron temperature is high and/or duration time of plasma emission induced by the SXRL pulse irradiation is relatively long, the optical emission signal will be observed by the CMOS detector. However, in this experiment, no emission signal could be observed, hence, we will be able to estimate an upper limit of the electron temperature and duration of plasma[10].

The penetration of the SXRL pulse into the target in an ablation or a modification process is accompanied by heating of the electrons, so such a process occurs in an extremely non-equilibrium state in the system. To consider the radiation from a heated or molten layer, we suppose a black-body radiator as an object. Suppose that the temporal behavior of the target electron temperature, T_e, can be described by a Gaussian;

$$T_e = T_m \exp\left(-\frac{t^2}{{t_{em}}^2}\right) \tag{1}$$

where T_m is the maximum electron temperature and t_{em} characterizes the emission duration. It should be noted that dependence shown in Eq. (1) is an approximation, but we believe that the Gaussian function can be used to approximate the dependence of target temperature in time. Taking into account the imaging properties of the CMOS camera system of the observable spectral region, we can calculate the total energy of the optical photons reaching a camera cell. The details have been discussed in Ref. 10. Calculated results for various durations are shown in Fig. 4. The experimentally

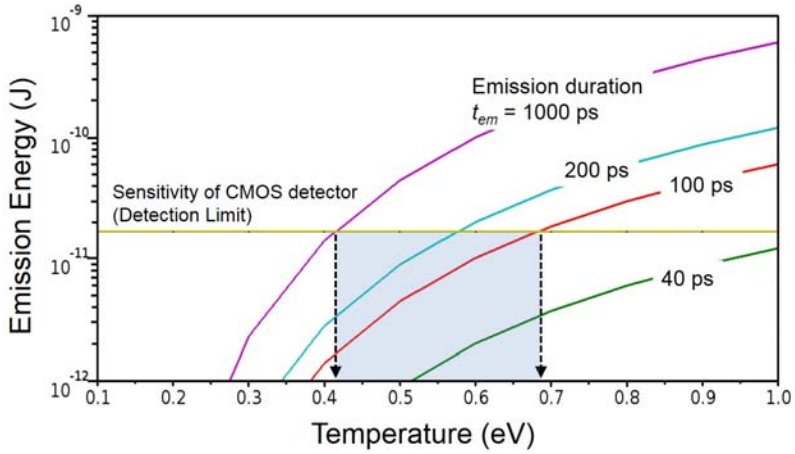

Figure 4. Dependencies of emission energy on the electron temperature for various emission duration. Figure is reprinted from Fig. 5 in Ref. 10 with permission from American Institute of Physics. Copyright 2014, AIP Publishing LLC.

determined detectable energy threshold is also shown. From the consideration of the calculated curves, if T_m is higher than 0.7 eV and t_{em} is longer than 100 ps, we should observe an emission signal from the irradiation surface by the system. On the other hand, T_m is lower than 0.4 eV and t_{em} is shorter than 1,000 ps, no image should be observed. In this case, we can determine an upper limit of the target temperature. If we assume t_{em} ~ 100–1,000 ps, the temperature is estimated to be T_m ~ 0.4–0.7 eV.

If the SXRL induced electron temperature is higher and/or duration of emission is longer than the estimated values mentioned above, the optical emission signal will be observed by the CMOS detector. Based on the calculation results shown in Fig. 4, we can estimate the duration of emission, which is necessary for reaching the detectable energy of the CMOS detector. Figure 5 shows the estimated duration, which will satisfy a detectable intensity of emission signal from sample surface. Needed emission duration for detectable intensity of the CMOS detector changes abruptly with exponential function. If the electron temperature of ablating matter is very low, for example less than 0.3 eV, it may be impossible to detect the emission signal, because extremely long duration longer than 2,000 ps is needed. An ablation process, especially SXRL induced surface modification process, such long duration of emission is difficult to assume. Actual process appearing a black-body condition would continue as long as several hundred ps[1,6]. However, if the electron temperature becomes higher, emission light will be detected quite easily. At the electron temperature higher than 1 eV, the needed emission duration for detection is shorter than 10 ps, which is compatible or shorter than the SXRL pulse width.

In this experiment, no emission signal could be observed, hence, we will be able to estimate an upper limit of the electron temperature and duration of heated matters. For a duration of 100–1,000 ps, the temperature will be estimated to be 0.4–0.7 eV. Our experimental results are in good accordance with the theoretical predictions. Under a low fluence region for SXRL pulse, the electron temperature less than 1 eV is considered reasonably, which is, however, enough for the nanometer scale surface modification on surface.

4. SUMMARY

In this study, we demonstrated that the focused picosecond soft x-ray laser pulses could induce ablation of a dielectric and metals at very low electron temperature. In spite of the use of sensitive detector, the any emission light signal accompanying the surface ablation/modification caused by SXRL pulse irradiation was not observed. These results allowed us to estimate the maximum electron temperature of the ablating matter to be 0.4–0.7 eV for the duration of 100–1,000 ps, which is a quite low electron temperature region of WDM. We can conclude that our experimental study confirms the theoretical model of an atomistic simulation, which predicts the splash of molten layers occurring under low electron temperature less than 1 eV and which creates the nanometer scale surface modifications due to spallative ablation.

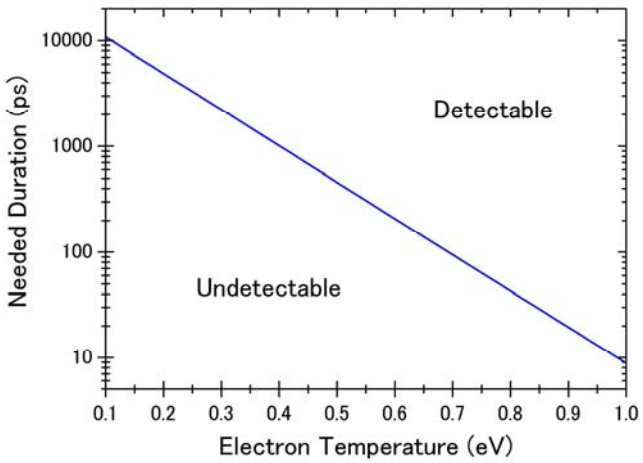

Figure 5. Estimated duration of emission from sample surface as a function of electron temperature.

It is important to consider the plasma-less ablation and/or surface modification process for a possibility of SXRL interaction mechanism with matter.

Now, we are carrying out the experiments to understand the ablation mechanism on more complex subjects, such as multilayered films. We will report the results elsewhere[11].

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