Short pulse soft x-ray laser ablation research

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ABSTRACT

The short pulse laser ablation have been extensively studied for confirmation and discussion of damage formation including estimations of damage thresholds and probabilities of surface machining. Irradiation examines by the femtosecond soft x-ray laser reveals formations of smooth craters on silica glass surfaces, and the appropriate selection of the laser wavelength can make the nanometer size modification on silicon surface in the vicinity of the damage threshold. On the other hand, we also revealed that damage thresholds and modification structures obtained by the picosecond soft x-ray laser irradiation provide the same results as the femtosecond soft x-ray laser. This means that not only femtosecond but also picosecond soft x-ray laser irradiation experiments can be contribute to deep understandings of the ablation phenomena.

Keywords: short pulse soft x-ray laser, ablation, damage threshold, damage structure

1. INTRODUCTION

The developments of short pulse lasers having picosecond or femtosecond duration have advanced dramatically. Not only ordinary optical lasers but also high harmonic generation lasers, plasma based x-ray lasers, and free electron lasers have been developed, and the oscillation wavelengths emitted from these lasers vary from the optical to x-ray region. The direct interaction of a laser pulse with matter is ablation. As a result of the interaction, laser pulses produce high temperature and pressure in matter, light emission, and damage structures on surface. One of the attractive and important subjects in the laser–matter interaction is soft x-ray laser ablation, because the ablation threshold induced by the short pulse laser is lower than that of optical- or long-pulse lasers [1, 2] and the modification structures depend on target materials [3]. Low ablation threshold and formation of characteristic modification structure may lead an ability to draw characteristic nanometer scale patterns on the material surface directly. To apply soft x-ray laser ablation to new fields, ablation process and mechanism have to be investigated experimentally and theoretically. Experimental results can give important information for the construction of ablation models. The calculation results can then be compared with the experimental results to explain ablation dynamics.

Model calculations of ablation dynamics have been also studied. The theoretical model for lithium fluoride (LiF) crystal predicted spallation phenomenon for the picosecond soft x-ray laser ablation [4]. Model calculation showed that the soft x-ray laser of 10 mJ/cm^2 fluence removed 40 nm thick surface layer. This value was coincident with experimental results. The driving force of damage formation was an increasing inertial pressure appearing inside of the irradiated heating layer. In aluminum (Al) case, the model calculation also predicted the appearance of inertial stress [5]. However, the damage threshold of the Al surface was approximately 15 mJ/cm^2 [3]. This value was smaller than the calculated ablation threshold of Al (75 mJ/cm^2), but was near the melting threshold of 6.5 mJ/cm^2 [6]. For the gold (Au) case, the ablation threshold predicted by the model calculation was about 90 mJ/cm^2, which was larger than the experimental result of about 20 mJ/cm^2 [3, 6]. This value was also near the melting threshold of Au (39 mJ/cm^2), and the modification depths agreed with melting depths [7]. The theoretical approach for an interaction model plays a crucial role in the explanation of the surface modification. However, it seems that there is still large gap between the experimental results and the theoretical predictions. To understand the ablation process more deeply and accurately, systematic studies of soft x-ray laser ablation are necessary. In this article, we report on experimental results of short pulse soft x-ray laser ablation of semiconductor silicon, insulator silica glass, and metal aluminum around damage thresholds.

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2. EXPERIMENTS

2.1 Femtosecond soft x-ray laser irradiation experiment

The femtosecond soft x-ray laser irradiation experiment was configured on the soft x-ray beamline of BL1 at the SPring-8 Angstrom Compact Free Electron Laser (SACLA) facility [8]. The soft x-ray free electron laser pulse converted from the electron beam had a wavelength of 13.5 nm (photon energy of 92 eV) or 10.3 nm (120 eV). A pulse width of soft x-ray free electron laser was 70 fs. A Kirkpatrick–Baez (K–B) mirror was used to focus the soft x-ray free electron laser beam onto the sample surface. The focal spot size defined by the full width at half maximum was evaluated by the knife-edge scanning method to be 10.5 µm x 8.6 µm. Zirconium (Zr) and silicon (Si) thin film filters with various thicknesses were adapted to attenuate the pulse energy. The transmittance of each thin-film filter was measured at the Photon Factory of the High Energy Accelerator Research Organization (KEK-PF). The details of femtosecond soft x-ray laser irradiation experiment was described in Ref. 9.

2.2 Picosecond soft x-ray laser experiment

The picosecond soft x-ray laser pulse irradiation experiment was carried out by use of a plasma-based soft x-ray laser system at the x-ray laser facility of the National Institutes for Quantum and Radiological Science and Technology. The soft x-ray laser pulse was generated from silver (Ag) plasma media using an oscillator–amplifier configuration with double Ag tape targets. The soft x-ray laser had a wavelength (photon energy) and pulse width of 13.9 nm (89 eV) and 7 ps, respectively. The intensity of each pulse was reduced by Zr thin film filters. The soft x-ray laser pulse was focused onto the sample surface by the multilayer coated spherical mirror. The details of the picosecond soft x-ray laser irradiation experiment was described in Ref. 3.

3. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show atomic force microscope (AFM) images of damage structures on the Si surface formed by femtosecond soft x-ray laser pulse irradiations [9]. In the case of the 13.5 nm wavelength irradiation, the first changes of silicon surface appeared around the irradiation fluence of 150–250 mJ/cm^2. In this fluence region, only an increase of the roughness of the surface could be observed (not shown). When the laser fluence became large and reached around 300 mJ/cm^2, dome structures as shown in Fig. 1(a) was formed. The hole structures appeared at the irradiation fluence above 400 mJ/cm^2. On the other hand, the first change at the 10.3 nm irradiation was the formation of small crater as shown in Fig. 1(b). The crater structure appeared at a fluence of 107 mJ/cm^2, and the crater structure grew with an increase of laser fluence. The damage thresholds, which formed hole (crater) structure, at 13.5 nm and 10.3 nm irradiations were estimated to be 418 mJ/cm^2 and 106 mJ/cm^2, respectively. The results of femtosecond x-ray laser ablation experiments showed that damage structures and damage fluences were different, when irradiation wavelength were different. Next, experimental damage fluences were compared with the theoretical values [10]. The theoretical calculation predicted that the ablation threshold at 13.5 nm and 10.3 nm irradiations were about 1200 mJ/cm^2 and 90 mJ/cm^2, respectively. The experimental damage threshold at 10.3 nm irradiation was coincident with the experimental value. However, the damage threshold at 13.5 nm was quite smaller than the calculated value, but was near the calculated melting threshold. This means that there

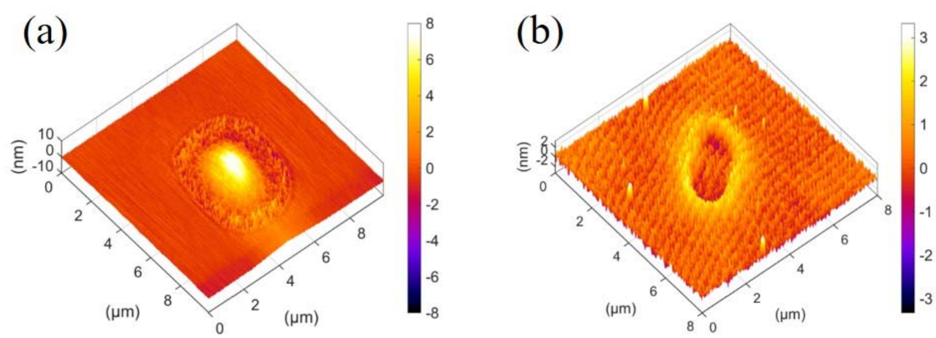

Figure 1. AFM images of damage structures on silicon surface by the irradiations of wavelengths of (a) 13.5 nm with a fluence of 294 mJ/cm^2, and (b) 10.3 nm with a fluence of 107 mJ/cm^2, respectively [9].

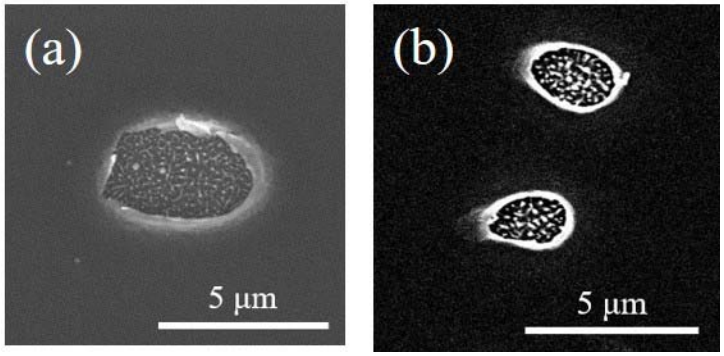

Figure 2. SEM images of damage structures on aluminum surfaces by the irradiation of (a) femtosecond soft x-ray laser pulse with the wavelength of 13.5 nm, pulse width of 70 fs, and a fluence of ~25 mJ/cm^2, and (b) picosecond soft x-ray laser pulse with the wavelength of 13.9 nm, pulse width of 7 ps, and a fluence of ~20 mJ/cm^2, respectively.

is the condition, when the laser fluence exceeds the melting threshold, significant damage structure appear. Figure 2(a) and 2(b) show damage structures on Al surfaces induced by the femtosecond and the picosecond soft x-ray laser pulse irradiations, respectively. Experimental damage thresholds of femtosecond and picosecond pulse irradiations were the same to be 14–15 mJ/cm^2, also when the irradiation fluence was equivalent, the formed damage structures were the same. Both of irradiation experiments obtained the same result. The experimental result mentioned above clearly show the fact that damage structures on Al surface, also Au surface, appeared when the irradiation fluence exceeded melting thresholds as same as the Si case. In the relatively low fluence irradiation, it is considered that the first change of the surface structures on Al and Au surfaces are probably due to splash of the molten layer.

We also evaluated damage thresholds of silica glasses (SiO_2), which had different concentrations of impurities, by use of femtosecond and picosecond soft x-ray laser pulses [11]. Three types of silica glasses, ED-A, ES, and HR, were used as irradiation samples, since each sample contained different concentrations of impurities [12]. The ED-A grade was a synthetic silica glass and had the lowest concentration of impurities. The ES grade was synthetic silica and had the highest concentration of the hydroxyl group (O–H). The HR grade was fused silica and provides the highest concentration of impurities. Figures 3(a) shows AFM image of the ablation crater formed on ED-A sample by the femtosecond soft x-ray laser pulse irradiation. The graph presented under the image represents cross-sectional depth profile of the damage, as presented by the line A–B in AFM image. Figure 3(b) presents ablation volumes for each SiO_2 sample derived from AFM measurements as a function of the irradiation fluence. Damage threshold values (shown as LIDT in Figure) were evaluated

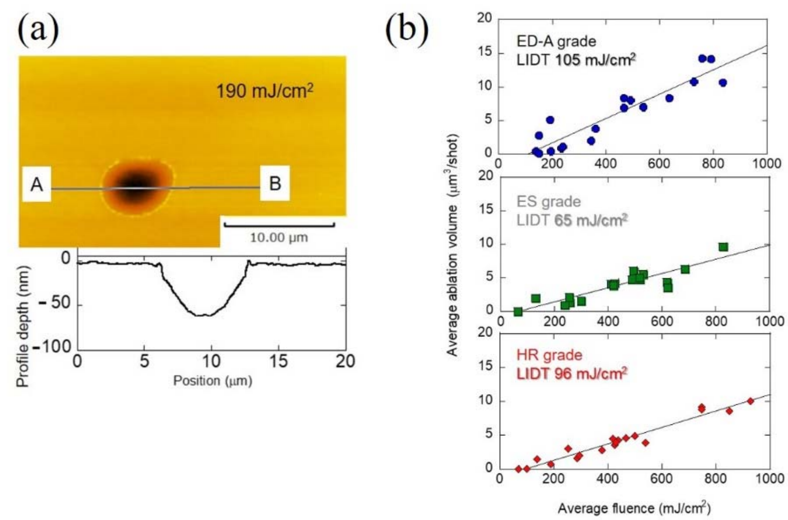

Figure 3. (a) AFM image of ablation craters on ED-A samples formed by femtosecond soft x-ray laser pulse with a fkuence of 190 mJ/cm^2 and (b) fluence dependence on the ablation volume for silica glass samples.

to be 105 mJ/cm^2 for ED-A, 65 mJ/cm^2 for ES, and 96 mJ/cm^2 for HR grades. The damage threshold of the ES grade was slightly lower than others. This means that the metallic impurities do not become a damage initiator in the soft x-ray region. The ES grade includes significant amounts of oxygen (O) atoms as a hydroxyl group (O–H), and oxygen absorbs soft x-rays around 13 nm. This means that the volume of the O–H in sample would influence the damage threshold obtained by the femtosecond soft x-ray laser. Damage thresholds of ED-A and HR grades obtained by the picosecond soft x-ray laser pulse were also evaluated. Damage thresholds were evaluated to be 82 mJ/cm^2 for ED-A and 71 mJ/cm^2 for HR samples. The damage threshold values estimated by the femtosecond and the picosecond soft x-ray laser pulses were similar. In addition, damage structures induced by the picosecond soft x-ray laser pulse did not have unexpected structures, such as rims or cracks as same as by the femtosecond laser pulse. The absence of unexpected structures suggested that damage structures formed without thermal affection.

Experimental results obtained above clearly show that the ablation phenomena, i.e. damage thresholds and modification structures, obtained by the picosecond soft x-ray laser provided the same results as the femtosecond soft x-ray laser. This means that picosecond soft x-ray laser irradiation experiments can provide a good benchmark for femtosecond soft x-ray laser irradiation experiments and also take data for the discussion of theoretical ablation mechanisms.

4. SUMMARY

We show material surface reactions induced by short pulse soft x-ray laser irradiation experiments (see also Sects. 1-4 [13]). Experimental result for Si at 13.5 nm wavelength irradiation shows that there is a condition where the melting threshold becomes the damage threshold. Ablation experiments for Al and SiO_2 samples revealed that damage thresholds induced by the femtosecond and the picosecond soft x-ray lasers are equivalent, so that the picosecond laser ablation will be a good benchmark for the femtosecond laser ablation. We hope that our experimental results can contribute to the understanding of the damage formation process generated by ultra-short soft x-ray pulses.

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REFERENCES

[1] Faenov, A. Ya , Inogamov, N. A., Zhakhovskii, V. V., Khokhliv, V. A., Nishihara, K., Kato, Y., Tanaka, M., Pikuz, T. A., Kishimoto, M., Ishino, M., Nishikino, M., Nakamura, T., Fukuda, Y., Bulanov, S. V., and Kawachi, T., App. Phys. Lett. **94**, 231107 (2009).

[2] Shibuya, T., Takasashi, T., Sakaue, K., Dhin, T.-H., Hara, H., Higashiguchi, T., Ishino, M., Koshiba, Y., Nishikino, M., Ogawa, H., Tanaka, M., Washio, M., Kobayashi, Y., and Kuroda, R., Appl. Phys. Lett. **113**, 171902 (2018).

[3] Ishino, M., Faenov, A. Y., Tanaka, M., Tamotsu, S., Hasegawa, N., Nishikino, M., Pikuz, T. A., Kaihori, T., and Kawachi, T., Appl. Phys. A **110**, 179 (2013).

[4] Inogamov, N. A., Faenov, A. Ya., Khokhlov, V. A., Zhakhovsky, V. V., Petrov, Yu. V., Skobelev, I. Yu., Nishihara, K., Kato, Y., Tanaka, M., Pikuz, T. A., Kishimoto, M., Ishino, M., Nishikino, M., Fukuda, Y., Bulanov, S. V., Kawachi, T., Anisimov, S. I., and Fortov, V. E., Contrib. Plasma Phys. **49**, 455 (2009).

[5] Inogamov, N. A., Zhakhovsky, V. V., Faenov, A. Ya., Khokhlov, V. A., Shepelev, V. V., Skobelev, I. Yu., Kato, Y., Tanaka, M., Pikuz, T. A., Kishimoto, M., Ishino, M., Nishikino, M., Fukuda, Y., Bulanov, S. V., Kawachi, T., Petrov, Y. V., Anisimov, S. I., Fortov, V. E., Appl. Phys. A **101**, 87 (2010).

[6] Starikov, S. V., Faenov, A. Ya., Pikuz, T. A., Skobelev, I. Yu., Fortov, V. E., Tamotsu, M., Ishino, M., Tanaka, M., Hasegawa, N., Nishikino, M., Kaihori, T., Imazono, M., Kando, M., and Kawachi, T., Appl. Phys. B **116**, 1005 (2014).
[7] Norman, G., Starikov, S., Stegailov, V., Fortov, V., Skobelev, I., Pikuz, T., Faenov, A., Tamotsu, S., Kato, Y., Ishino, M., Tanaka, M., Hasegawa, N., Nishikino, M., Ohba, T., Kaihori, T., Ochi, Y., Imazono, T., Fukuda, Y., Kando, M., and Kawachi, T., J. Appl. Phys. **112**, 013104 (2012).
[8] Owada, S., Togawa, K., Inagaki, T., Hara, T., Tanaka, T., Joti, Y., Koyama, T., Nakajima, K., Ohashi, H., Senba, Y., Togashi, T., Tono, K., Yamaga, M., Yumoto, H., Yabashi, M., Tanaka, H., and Ishikawa, T., J. Synchrotron Rad. **25**, 282 (2018).
[9] Dinh, T. -H., Medvedev, M., Ishino, M., Kitamura, T., Hasegawa, N., Otobe, T., Higashiguchi, T., Sakaue, K., Washio, M., Hatano, T., Kon, A., Kubora, Y., Inubushi, Y., Owada, S., Shibuya, T., Ziaja, B., and Nishikino, M., Commun. Phys. **2**, 150 (2019).
[10] Medvedev, N., Tkachenko, V., Lipp, V., Li, Z. & Ziaja, B., 4open **1**, 3 (2018).
[11] Mikami, K., Ishino, M., Dinh, T. -H., Motokoshi, S., Hasegawa, N., Kon, A., Inubushi, Y., Owada, S., Kinoshita, H., and Nishikino M., Opt. Lett. **45**, 2535 (2020).
[12] Kuzuu, N., Yoshida, K., Yoshida, H., Kamimura, T., and Kamisugi, N., Appl. Opt. **38**, 2510 (1999).
[13] D. Bleiner, The Science and Technology of X-ray Lasers: A 2020 Update Proc. SPIE 11886, 1188602 (2021)