# Damage characteristics of platinum/carbon multilayers under X-ray free-electron laser irradiation

Jangwoo Kim[*a], Takahisa Koyama[b], Hirokatsu Yumoto[b], Ayaka Nagahira[a], Satoshi Matsuyama[a, c], Yasuhisa Sano[a], Makina Yabashi[d], Haruhiko Ohashi[b, d], Tetsuya Ishikawa[d], Kazuto Yamauchi[a, c, e]

[a] Department of Precision Science and Technology, Graduate School of Engineering, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan

[b] Japan Synchrotron Radiation Research Institute/SPring-8, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5198, Japan

[c] Japan Science and Technology Agency, CREST, 4-1-8 Honcho, Kawaguchi, Saitama 332-0012, Japan

[d] RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan

[e] Research Center for Ultra-Precision Science and Technology, Graduate School of Engineering, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan

## ABSTRACT

We evaluated the irradiation damage induced to platinum/carbon multilayers induced by hard X-ray free-electron lasers (XFELs). In this study, in order to test the use of the platinum/carbon multilayers for future XFEL focusing applications, we evaluated the structures in almost exactly the condition in which they would actually be used. The X-ray reflectivity of the multilayers was measured by using XFELs, and a cross-section of the multilayer that was irradiated by an XFEL was observed by transmission electron microscopy. We used a non-monochromatic beam at a photon energy of 10 keV. We confirmed that the intensity of the conditions under which the multilayers are to be used is sufficiently lower than the breakdown threshold of platinum/carbon multilayers.

**Keywords:** multilayer, damage characteristics, platinum/carbon, XFEL irradiation

## 1. INTRODUCTION

X-ray free-electron lasers (XFELs)[1, 2] can now be used to produce intense femtosecond pulses in the hard X-ray region. The use of these X-ray sources in conjunction with analysis methods has unique advantages, and their applications in exploring new frontiers of science may be possible by using focusing optics.

There are various types of X-ray focusing optic systems, such as those using refractive, diffractive, and reflective optics[3–9]. The use of reflective optics for intense X-ray focusing is effective because it is a grazing-incidence optical system. Sub-100-nm focusing under the diffraction-limited condition has already been achieved using total-reflection mirrors. The use of reflective optics for reducing the focal spot size requires a large grazing incidence angle in order to increase the numerical aperture. A multilayer mirror provides for a sufficiently large grazing incidence angle to accommodate spot size reduction, which cannot be achieved using total-reflection mirrors[10].

Since intense X-ray beams damage multilayer structures[8, 11], in this study, we evaluated the irradiation damage caused to platinum/carbon multilayers as induced by hard XFELs. In order to use the platinum/carbon multilayers for XFEL focusing in the future, we evaluated those in almost exactly the condition of intended actual use. Two types of multilayer were prepared: one with a short period and one with a long period. The X-ray reflectivity of the multilayers was measured by using XFELs. A cross-section of the multilayer that was irradiated by an XFEL was observed by transmission electron microscopy in order to evaluate the damage induced by XFEL irradiation. We used a non-monochromatic beam at a photon energy of 10 keV.

*kim@up.prec.eng.osaka-u.ac.jp; phone 81 6 6879-7286; fax 81 6 6879-7286

Advances in X-Ray/EUV Optics and Components VIII, edited by Ali Khounsary,
Shunji Goto, Christian Morawe, Proc. of SPIE Vol. 8848, 88480S · © 2013 SPIE
CCC code: 0277-786X/13/$18 · doi: 10.1117/12.2022735

## 2. EXPERIMENTS

### 2.1 Fabrication of multilayers

We have analyzed two platinum/carbon (Pt/C) multilayer samples, which were fabricated by a DC magnetron sputtering system. These samples were deposited on silicon (100) wafer substrates, which had thicknesses of 500 μm. We employed highly pure 2"-diameter targets made respectively of platinum and carbon, and we alternated between using the platinum target and the carbon target. All the layers were deposited under an argon gas pressure of 0.1 Pa and a flow rate of 35 sccm. A feedback system was used to maintain the electric power at 20 W and at 120 W for the platinum and carbon targets, respectively.

The details of the samples evaluated in this research are shown in Table 1. The measured thicknesses are shown in Table 1 along with the nominal thicknesses. The thicknesses of the fabricated samples were estimated by using the X-ray reflectometer method, which is a non-destructive and non-contact method for thickness determination of thin films. The X-ray reflectivities of these samples were measured by X-ray reflectometry (Rigaku SmartLab). The experimental setup consisted of a Cu-Kα X-ray source ($\lambda = 0.154$ nm) and a $\theta$–$2\theta$ goniometer stage for changing the grazing incidence angle on the sample surface. The intensity of the X-ray reflected by a sample was monitored at grazing incidence angles.

Table 1. Details of the Pt/C multilayer samples. The thicknesses of the fabricated samples were measured by using the Rigaku SmartLab. *N* is the number of bilayers, *d* is the multilayer period, and $\gamma$ is the thickness ratio of the platinum to the multilayer period.

| Sample | *N* | Design | | Measured (Rigaku SmartLab) | |
|---|---|---|---|---|---|
| | | *d* (nm) | $\gamma$ | *d* (nm) | $\gamma$ |
| A | 45 | 5.0 | 0.5 | 4.78 | 0.508 |
| B | 45 | 2.5 | 0.5 | 2.45 | 0.527 |

### 2.2 Sample evaluation

Characterization of the Pt/C multilayers using the XFEL was conducted in beamline 3 at SACLA[2]. In order to ensure that it functions as a multilayer film in a condition where the multilayer film is actually used, A and B multilayer films were evaluated by using the non-monochromatic beam XFELs at the photon energy of 10 keV. The detailed parameters of beam are shown in Table 2. In the first Bragg angle, the change of reflectivity of the B multilayer was determined under X-ray irradiation for a duration of 5 hours. Irradiation damage is expected to be induced by the XFEL to the B multilayer, because the first Bragg angle of the multilayer is very great. After the irradiation, the cross-section of the B multilayer was compared with the non-irradiated section by transmission electronic microscopy.

Table 2. Beam parameters. FWHM is full-width at half-maximum.

| Photon energy | Pulse duration | Pulse repetition | Mean pulse energy | Diameter (FWHM) |
|---|---|---|---|---|
| 10 keV | 20 fs | 10 Hz | 130 μJ | 300 μm |

## 3. RESULTS AND DISCUSSION

Figure 1 shows the X-ray reflectivities of samples A and B measured by using the XFEL as a function of grazing incidence angle. The solid line represents the measured reflectivity, and the dashed line shows the results of curve fitting. The experimental data from samples A and B shows that the reflectivities of the first Bragg peak were 78.3% and 51.8%, respectively. Sufficiently high reflectivities were obtained, and these samples function as multilayer films. The periods of

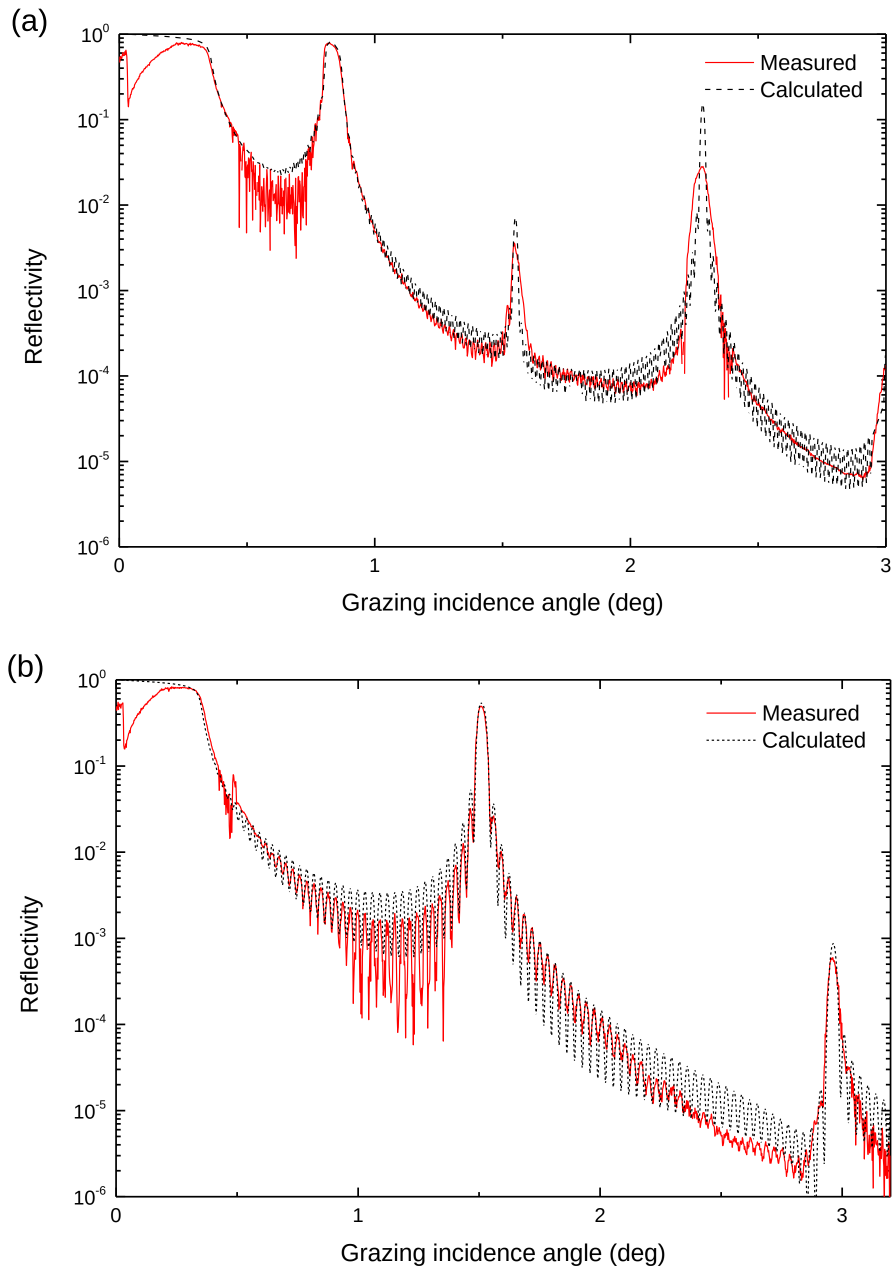


Figure 1. X-ray reflectivity of the Pt/C multilayers (a: sample A, b: sample B) as a function of grazing incidence angles. The solid line and dashed line show the experimental data and the calculated curve fitting data, respectively. The X-ray reflectivities of the first Bragg peak are (a) 78.3% and (b) 51.8%.

the multilayer and the ratios of the platinum layer thickness to the multilayer period were estimated by using the X-ray reflectometer method and are shown in Table 3. The calculated multilayer periods were in good agreement with the results obtained by using the Rigaku SmartLab, and the difference was less than 1%.

Table 3. Multilayer period and the ratio of the platinum layer thickness to the multilayer period of the Pt/C multilayer samples. The thicknesses of the fabricated samples were measured by using the Rigaku SmartLab and the XFEL. *d* is the multilayer period, and *γ* is the thickness ratio of the platinum to the multilayer period.

| Sample | Measured (Rigaku SmartLab) | | Measured (XFEL) | | Deviation of multilayer period (%) |
|---|---|---|---|---|---|
| | *d* (nm) | *γ* | *d* (nm) | *γ* | |
| A | 4.78 | 0.508 | 4.74 | 0.513 | 0.84 |
| B | 2.45 | 0.527 | 2.42 | 0.528 | 1.22 |

The change of the reflectivity of the B multilayer in the first Bragg peak was shown in Figure 2. During the 5 h of measurements, the reflectivity was not changed. After 50, 100, 200, and 300 min, the angle of the first Bragg peak was measured, and the results are shown in Figure 2. The angle of the first Bragg peak also did not change.

Figure 3 shows the transmission electron microscope cross-section image of non-irradiated and irradiated sections of the B multilayer. The dark and bright layers correspond to the platinum and carbon layers, respectively. The surface and the substrate sides of the non-irradiated and irradiated sections were observed. No evidence of irradiation damage caused by the XFEL was found. The multilayer period and the ratio of the platinum layer thickness to the multilayer period were estimated by using the transmission electron microscopy images. These values were a little bigger than the results from the Rigaku SmartLab and the XFEL; one explanation for this is that there is a possibility that the cross-section is slightly inclined.

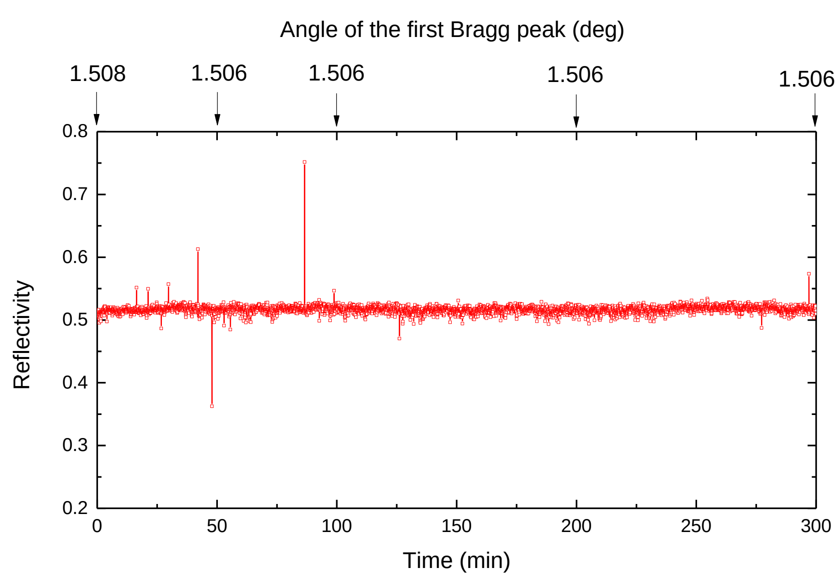


Figure 2. Change of the X-ray reflectivity versus time, with annotations indicating the measured angle of the first Bragg peak of sample B at several times.

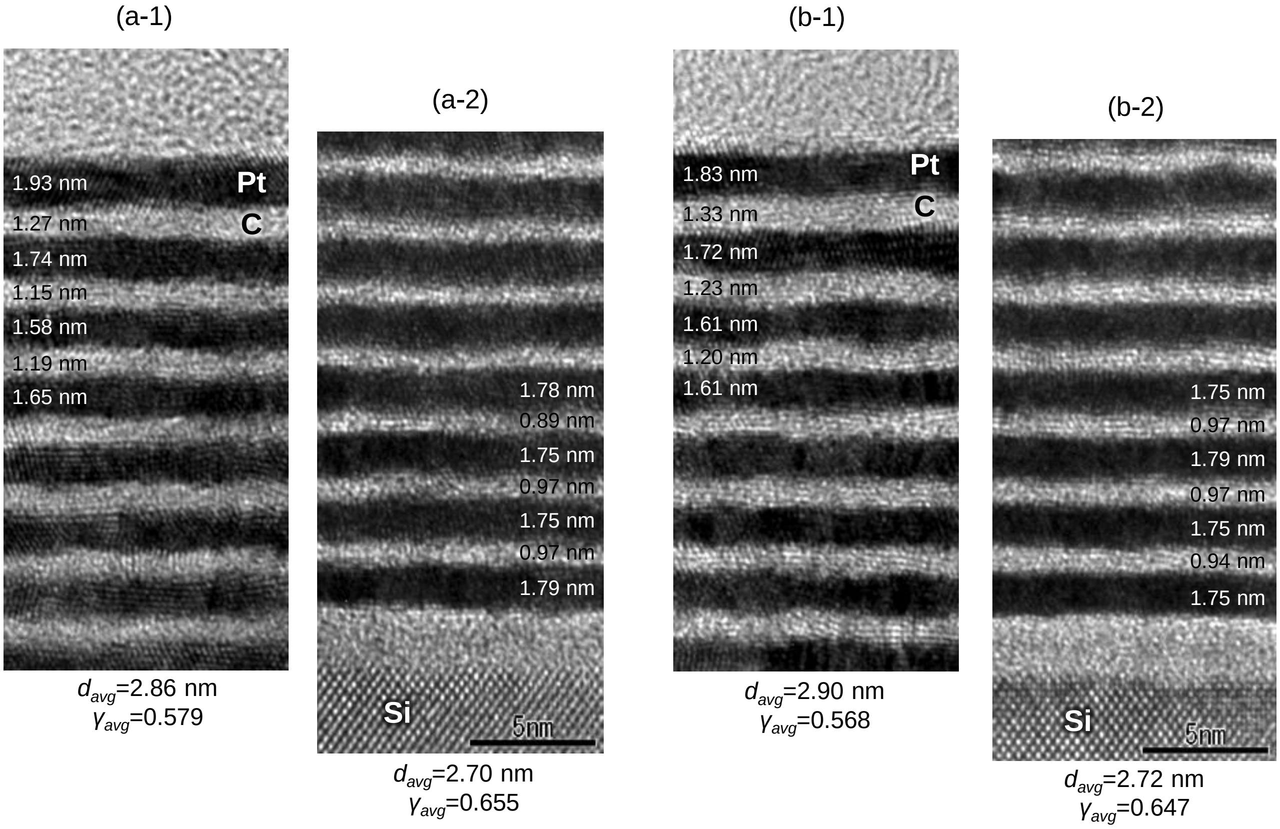


Figure 3. Cross-sectional bright-field transmission electron microscopy image of the B multilayer. (a) and (b) show non-irradiated and irradiated sections, where the notations “-1” and “-2” in the figure part names denote the surface side and the substrate side, respectively. The dark and bright layers correspond to platinum and carbon layers, respectively. $d_{avg}$ is the average multilayer period, and $\gamma_{avg}$ is the average ratio of the platinum layer thickness to the multilayer period.

During the 5 h of irradiation, the reflectivity and the transmission electron microscopy image were not changed. It was confirmed that serious damage to the multilayer film is not generated by unfocused XFEL irradiation, based on the above results.

A previous work found the breakdown threshold of a single layer of platinum under normal incidence[11]. When irradiated with a beam that has not been focused, single-layer platinum is not destroyed. However, it is expected that the breakdown mechanisms of grazing incidence and of the multilayer are different from those of normal incidence and of a single layer, respectively. In this study, it was not possible to find the threshold of the Pt/C multilayer, but it was found that the Pt/C multilayer functions correctly under the condition that we plan to actually use it. Further, in the Pt/C multilayers, it has been reported that X-ray reflectivity was improved by carbon doping of the platinum layers[12, 13]. Using this technique, high-efficiency focusing of the XFEL is expected.

## 4. SUMMARY

We evaluated the use of an XFEL with Pt/C multilayers for future XFEL focusing applications. Irradiation damage caused by intense X-rays was expected. The X-ray reflectivity and cross-section of the multilayer were measured to observe the irradiation damage. However, damage was not observed in the condition we plan to actually use it. We confirmed that the intensity under the condition of intended actual use is sufficiently lower than the breakdown threshold of the Pt/C multilayers. High-efficiency focusing of the XFEL using the Pt/C multilayer is expected.

## ACKNOWLEDGEMENTS

This research was mainly supported by a Grant-in-Aid for Scientific Research (S) no. 23226004 and a Grant-in Aid for JSPS Fellows no. 25129 from the Ministry of Education, Sports, Culture, Science and Technology, Japan. It was also partially supported by CREST from the Japan Science and Technology Agency. The use of BL3 at SACLA was supported by RIKEN and the Japan Synchrotron Radiation Research Institute.

## REFERENCES

[1] Emma, P., Akre, R., Arthur, J., Bionta, R., Bostedt, C., Bozek, J., Brachmann, A., Bucksbaum, P., Coffee, R., Decker, F.-J., Ding, Y., Dowell, D., Edstrom, S., Fisher, A., Frisch, J., Gilevich, S., Hastings, J., Hays, G., Hering, Ph., Huang, Z., Iverson, R., Loos, H., Messerschmidt, M., Miahnahri, A., Moeller, S., Nuhn, H.-D., Pile, G., Ratner, D., Rzepiela, J., Schultz, D., Smith, T., Stefan, P., Tompkins, H., Turner, J., Welch, J., White, W., Wu, J., Yocky, G. and Galayda, J., "First lasing and operation of an ångstrom-wavelength free-electron laser," Nature Photon. 4, 641–647 (2010).

[2] Ishikawa, T., Aoyagi, H., Asaka, T., Asano, Y., Azumi, N., Bizen, T., Ego, H., Fukami, K., Fukui, T., Furukawa, Y., Goto, S., Hanaki, H., Hara, T., Hasegawa, T., Hatsui, T., Higashiya, A., Hirono, T., Hosoda, N., Ishii, M., Inagaki, T., Inubushi, Y., Itoga, T., Joti, Y., Kago, M., Kameshima, T., Kimura, H., Kirihara, Y., Kiyomichi, A., Kobayashi, T., Kondo, C., Kudo, T., Maesaka, H., Maréchal, X. M., Masuda, T., Matsubara, S., Matsumoto, T., Matsushita, T., Matsui, S., Nagasono, M., Nariyama, N., Ohashi, H., Ohata, T., Ohshima, T., Ono, S., Otake, Y., Saji, C., Sakurai, T., Sato, T., Sawada, K., Seike, T., Shirasawa, K., Sugimoto, T., Suzuki, S., Takahashi, S., Takebe, H., Takeshita, K., Tamasaku, K., Tanaka, H., Tanaka, R., Tanaka, T., Togashi, T., Togawa, K., Tokuhisa, A., Tomizawa, H., Tono, K., Wu, S., Yabashi, M., Yamaga, M., Yamashita, A., Yanagida, K., Zhang, C., Shintake, T., Kitamura, H. and Kumagai, N., "A compact X-ray free-electron laser emitting in the sub-ångstrom region," Nature Photon. 6, 540–544 (2012).

[3] Suzuki, Y., Takeuchi, A., Takano, H. and Takenaka, H., "Performance Test of Fresnel Zone Plate with 50 nm Outermost Zone Width in Hard X-ray Region," Jpn. J. Appl. Phys. 44, 1994–1998 (2005).

[4] Schroer, C. G., Kurapova, O., Patommel, J., Boye, P., Feldkamp, J., Lengeler, B., Burghammer, M., Riekel, C., Vincze, L., Hart, A. van der and Küchler, M., "Hard x-ray nanoprobe based on refractive x-ray lenses," Appl. Phys. Lett. 87, 124103 (2005).

[5] Kang, H. C., Maser, J., Stephenson, G. B., Liu, C., Conley, R., Macrander, A. T. and Vogt, S., "Nanometer Linear Focusing of Hard X Rays by a Multilayer Laue Lens," Phys. Rev. Lett. 96, 127401 (2006).

[6] Morawe, Ch., Guigay, J.-P., Mocella, V. and Ferrero, C., "An analytical approach to estimating aberrations in curved multilayer optics for hard x-rays: 2. Interpretation and application to focusing experiments," Opt. Exp. 16, 16138–16150 (2008).

[7] Mimura, H., Handa, S., Kimura, T., Yumoto, H., Yamakawa, D., Yokoyama, H., Matsuyama, S., Inagaki, K., Yamamura, K., Sano, Y., Tamasaku, K., Nishino, Y., Yabashi, M., Ishikawa, T. and Yamauchi, K., "Breaking the 10 nm barrier in hard-X-ray focusing," Nature Phys. 6, 122–125 (2010).

[8] David, C., Gorelick, S., Rutishauser, S., Krzywinski, J., Vila-Comamala, J., Guzenko, V. A., Bunk, O., Färm, E., Ritala, M., Cammarata, M., Fritz, D. M., Barrett, R., Samoylova, L., Grunert, J. and Sinn, H., "Nanofocusing of hard X-ray free electron laser pulses using diamond based Fresnel zone plates," Sci. Rep. 1, 57 (2011).

[9] Yumoto, H., Mimura, H., Koyama, T., Matsuyama, S., Tono, K., Togashi, T., Inubushi, Y., Sato, T., Tanaka, T., Kimura, T., Yokoyama, H., Kim, J., Sano, Y., Hachisu, Y., Yabashi, M., Ohashi, H., Ohmori, H., Ishikawa, T. and Yamauchi, K., "Focusing of X-ray free electron laser with reflective optics," Nature photon. 7, 43–47 (2013).

[10] Barbee. Jr., T. W, "Multilayers for x-ray optics," SPIE Proc. 563, 2–28 (1985).

[11] Koyama, T., Yumoto, H., Senba, Y., Tono, K., Sato, T., Togashi, T., Inubushi, Y., Katayama, T., Kim, J., Matsuyama, S., Mimura, H., Yabashi, M., Yamauchi, K., Ohashi, H. and Ishikawa, T., "Investigation of ablation thresholds of optical materials using 1-μm-focusing beam at hard X-ray free electron laser," Opt. Express 21, 15382–15388 (2013).

[12] Kim, J., Yokoyama, H., Matsuyama, S., Sano, Y. and Yamauchi, K., "Improved reflectivity of platinum/carbon multilayers for X-ray mirrors by carbon doping into platinum layer," Curr. Appl. Phys. 12, S20–S23 (2012).

[13] Kim, J., Matsuyama, S., Sano, Y. and Yamauchi, K., "Improvement of interface roughness in platinum/carbon multilayers for X-ray mirrors," Key Eng. Mater. 523–524, 1076–1079 (2012).