Damage accumulation in thin ruthenium films induced by repetitive exposure to femtosecond XUV pulses below the single-shot ablation threshold

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The process of damage accumulation in thin ruthenium films exposed to multiple femtosecond extreme ultraviolet (XUV) free-electron laser (FEL) pulses below the critical angle of reflectance at the FEL facility in Hamburg (FLASH) was experimentally analyzed. The multi-shot damage threshold is found to be lower than the single-shot damage threshold. Detailed analysis of the damage morphology and its dependence on irradiation conditions justifies the assumption that cavitation induced by the FEL pulse is the prime mechanism responsible for multi-shot damage in optical coatings.

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1. INTRODUCTION

The development of high-peak-brilliance, high-repetition-rate free-electron laser (FEL) light sources operating in the extreme ultraviolet (XUV) and x-ray spectral regime, such as the FEL facility in Hamburg (FLASH), FLASH 2, European XFEL [1], and LCLS [2], leads to increased practical interest in the durability of thin films exposed to a large number of pulses. FEL optical elements such as mirrors or beam stoppers should be designed considering the limitation on durability to the FEL radiation together with requirements for high optical performance of the materials. Avoiding FEL-induced damage is also critical when long exposures of thin film samples are expected during FEL experiments. Examples of such experiments can be found in time-resolved studies of surface chemical reactions with pump–probe x-ray diffraction techniques [3].

Only recently were the first papers on the resistivity of metal films to multi-shot x-ray radiation published [4,5]. Earlier, Hau-Riege et al. [6] investigated B$_4$C coatings. In both cases
Table 1. Summary of Analyzed Exposures

| Angle of Incidence, deg | Numbers of Pulse Trains | Number of Pulses in One Pulse Train | Pulse-to-Pulse Repetition Rate within One Pulse Train, kHz | Average Absorbed Fluence, mJ/cm² |
|------------------------|-------------------------|-----------------------------------|----------------------------------------------------------|---------------------------------|
| 3.17                   | 1                       | 1, 50, 100, 200, 400              | 1000                                                     | Variable, to determine MSDT     |
|                        | 209                     | 100                               | 250, 500, 1000                                           |                                 |
|                        | 4                       | 1                                 | n.a.                                                     |                                 |
| 20                      | 23                      | 400                               | 1000                                                     | 12.5                            |

it was observed that the multi-shot damage threshold (MSDT) is slightly lower than the single-shot one, but no detailed analysis of the nature of multi-shot damage was yet reported. Within the XUV wavelength range, multi-shot damage studies of bulk Si samples were reported by Sobierajski et al. [7]. The prime damage mechanism was identified as crossing the melting threshold due to heat accumulation in the vicinity of the Si surface.

In this work we investigate the MSDT for Ru coatings exposed to different numbers of 100 fs pulses at a 13.5 nm wavelength generated at different repetition rates. We present the dependence of the MSDT on the repetition rate and the number of pulses. The influence of irradiation conditions—namely, the number of pulses and grazing incidence angle—on the morphology of the damaged spots is investigated. Mechanisms responsible for the observed damage phenomena are suggested, although detailed atomistic simulations are required to confirm the proposed mechanisms.

Understanding the multi-shot damage of Ru films has a practical relevance. Since Ru has a high reflection coefficient for the XUV and soft-x-ray wavelength ranges, and has a low oxidation rate, it is a very attractive material for reflective coatings. This work is a continuation of a series of studies of the durability of optics exposed to FEL radiation below the single-shot damage threshold [8] and a detailed analysis of the nature of single-shot damage discussed in [9].

2. EXPERIMENT

For this study, polycrystalline Ru coatings of 50 nm thickness were deposited on super-polished silicon substrates using magnetron sputtering in an Ar atmosphere. The thickness of the Ru layer was determined by x-ray reflectivity measurements. Exposure of the Ru films was performed at the beamline BL2 of the FLASH facility [10]. Details of the experiment can be found elsewhere [8,11].

For irradiations we used 100 fs XUV pulses with a wavelength of 13.5 nm. The light was p polarized with respect to the sample surface. We used the multi-shot irradiation mode where FLASH generated one pulse train per 0.1 s. [12]. The maximum duration of a pulse train was 400 μs, and the maximum repetition rate within the pulse train was 1 MHz. During the experiment we controlled the number of pulses within the pulse train by reducing the duration of the pulse train. It was also possible to reduce the repetition rate while keeping the pulse train duration constant, and thus reduce the number of pulses in the pulse train.

The determination of the MSDT was performed using Liu’s method [13–15] and the fluence scan method [11,16] based on measuring damaged areas with differential image contrast (also referred to as Nomarski) microscopy. The dependence of the damage threshold on the repetition rate and the number of pulses was studied using irradiations at 3.17° grazing incidence with an effective area of the beam [16,17] on the sample surface of 40,500 ± 2000 μm². The development of the damage morphology was studied through analysis of damage craters caused by different numbers of pulses arriving at 20° grazing incidence at a fixed average fluence per pulse using a focused beam with an effective area on the sample surface of 120 μm². The XUV reflectivity values at 3.17° and 20° were measured to be 95.5% and 68%, respectively [18]. In both cases, only the evanescent wave penetrated into the film, resulting in a comparable penetration depth for the different angles of incidence—2.5 nm and 3.5 nm, respectively. For Ru, the critical reflection angle for XUV radiation at a wavelength of 13.5 nm is 27°.

For a consistent comparison of the damage threshold values at different irradiation conditions, all analysis is performed in terms of absorbed fluence. This is calculated according to the formula \( F_{\text{abs}} = (1 - R) \times E/A \), where \( E \) is the total energy of the pulse, \( A \) is the effective area of the beam on the sample surface determined according to Ref. [17], and \( R \) is the reflectivity coefficient at the corresponding exposure conditions. The total energy of the pulse \( E \) was measured with a gas monitor detector as described by Tiedtke et al. [19]. The analyzed exposures are summarized in Table 1.

3. RESULTS AND DISCUSSION

First, we present the results of multi-shot damage experiments using 50 nm Ru films performed at a grazing incidence of 3.17°. The Ru films were exposed to single-pulse trains, consisting of a different number of pulses from 1 to 400 at a fixed repetition rate of 1 MHz. Another set of exposures was performed for a fixed number of pulses in a pulse train (100 pulses), but with a varying repetition rate (time interval between pulses in a train). These two sets of measured damage threshold values are summarized in Tables 2 and 3, respectively.

As one can see, the MSDT does not depend on the repetition rate within the error margin, which in our case was approximately 20%. This value represents the combined error of the pulse energy measurements and determination of the effective area of the beam. This suggests that heat accumulation is unlikely to be the damage mechanism for the given experimental conditions. This is in contrast to the previously reported conditions.
Table 2. Dependence of the Multi-Shot Damage Threshold on the Number of Pulses for the Exposure at 3.17° Grazing Incidence and 1000 kHz Pulse-to-Pulse Repetition Rate

| Number of pulses | 1  | 50 | 100 | 200 | 400 |
|------------------|----|----|-----|-----|-----|
| Damage threshold, mJ/cm² | 26.1 | 13.3 | 8.3 | 5.8 | 6.7 |

*The error in the threshold determination is approximately 20%.

Table 3. Pulse-to-Pulse Repetition Rate Dependence of the Multi-Shot Damage Threshold (Absorbed Fluence) Experimentally Determined for 100 Pulses Arriving at 3.17° Grazing Incidence

| Repetition rate, kHz | 250 | 500 | 1000 |
|----------------------|-----|-----|------|
| Damage threshold, mJ/cm² | 7.5 | 8.3 | 8.3 |

*The error in the threshold determination is approximately 20%.

work of Sobierajski et al. [7], where heat accumulation in bulk Si exposed to similar radiation conditions but at a normal incidence was found to be the main mechanism responsible for multi-shot damage caused by XUV femtosecond FLASH laser pulses. Ru, being a metal, has a high thermal conductivity, which guarantees that the energy that is absorbed at the surface diffuses through the entire Ru layer into the Si substrate, mostly before the next pulse in the train arrives.

Another observation is the decrease in the damage threshold upon increasing the number of pulses at a fixed repetition rate of 1 MHz, stabilizing at 200 pulses or more. This result suggests, together with the independence of the MSDT of the repetition rate described above, that there is an accumulation of an irreversible process. The increase in the MSDT with a decreasing number of pulses from 100 to 50 indicates that damage caused by a smaller number of pulses with low fluence is not observed, and that a high fluence of individual pulses is needed to cause detectable damage.

In order to understand the processes responsible for the multi-shot damage of Ru, we performed ex situ analysis of the damage morphology by means of high-resolution scanning electron microscopy (HR-SEM) and transmission electron microscopy (TEM). Additionally, we compared the damage morphologies obtained at 3.17° and 20° grazing incidence to study the influence of different angles of incidence. The HR-SEM image of a damaged spot exposed to a single pulse train that consisted of 50 pulses at a 1 MHz repetition rate at 3.17° grazing incidence is shown in Fig. 1(A). The mean value of the fluence per pulse was 15 mJ/cm², which is close to the measured damage threshold value in Table 2 of 13.3 mJ/cm². The damaged spot has a total area of ~320 μm² and can be characterized as a deep crater penetrating through the Ru layer into the Si substrate. The fact that the crater depth exceeds the thickness of the Ru layer is verified by imaging, using an energy-selective backscattered detector (not shown), which is sensitive to the elemental composition of a surface, confirming that Si is at the bottom of the crater. No other significant surface modifications around the craters, such as increased roughness or cracks, were detected by means of HR-SEM.

Figure 1(B) shows a crater resulting from irradiation of the Ru film with a much larger number of pulses, namely, 160,000 (400 pulse trains of 400 pulses each at a repetition frequency of 1 MHz), but performed at 20° grazing incidence with a mean fluence per pulse of 12.5 mJ/cm². From the color contrast we can conclude that the crater shown in Fig. 1(B) has a smooth silicon surface with Ru droplets on top. Figure 1 shows that although both exposures are carried out with a fluence close to the damage threshold, a much smaller number of pulses arriving at 3.17° causes comparable damage to that caused by pulses arriving at 20° grazing incidence.

To elaborate on the mechanism of accumulation of damage, we study the development of damage with an increasing number of pulses for irradiation at a 20° grazing incidence angle. The following exposures were analyzed: 209 individual pulses generated at a 5 Hz repetition rate, and 4 and 23 pulse trains, generated at a 10 Hz repetition rate. Each train in this experiment consisted of 400 pulses generated at a 1 MHz repetition rate. The damage threshold value, determined by Liu’s method, for one such pulse train at 2° was 14 ± 3 mJ/cm².

The mean value of the fluence per pulse in the irradiation was 12.5 mJ/cm², which is slightly below the MSDT. Only surface modifications of the Ru layer were detected.

Fig. 1. HR-SEM image of a damaged spot produced with (A) a pulse train consisting of 50 femtosecond XUV pulses with a 1 MHz repetition rate at a 3.17° grazing incidence angle and (B) 400 femtosecond XUV pulse trains at 20° grazing incidence. Each pulse train consisted of 400 pulses. The pulse train repetition rate was 10 Hz, while the pulse repetition rate within a pulse train was 1 MHz. The gray rectangular shadow visible in (B) is the trace of electron-induced C growth originating from SEM measurements carried out before the one displayed in the current figure.
TEM and atomic force microscopy (AFM) measurements of the damaged spots (not shown) confirmed that only the Ru layer was modified. All three damaged spots presented in Fig. 2 can be characterized as increased surface roughness, although the degree of roughening varies. Surprisingly, the surface roughness is considerably less after irradiation with 1600 pulses than after 209 individual pulses, although the opposite is expected.

The possible explanation for this contradiction is the random character of the pulse fluences within the irradiation. We should note that because of the self-amplified spontaneous emission operation of the FEL, the fluences of individual pulses varied from 1 to 30 mJ/cm². The histograms of the absorbed fluences per pulse for irradiations with 209, 9200, and 1600 pulses are shown in Fig. 3.

The solid lines in Fig. 3 show the number of pulses within the exposure sequence that have a fluence higher than the x coordinate value. Based on this analysis (see the magnified image in Fig. 3), one can see that there are more high-fluence pulses (more than 20 mJ/cm² of absorbed fluence) in the case of irradiation with 209 pulses than in the case of 1600 pulses. If we assume that there is a damage threshold in the range of 20–25 mJ/cm², this will lead to the conclusion that with 209 pulses, the Ru film was exposed to a larger number of pulses that can cause damage than in the 1600 pulse case, explaining the increased damage with 209 pulses.
In our recent investigations [9], we showed that the nature of single-shot XUV-induced ablation of Ru is photomechanical spallation in the stress confinement regime. This means that the heating of the lattice is faster than the acoustic relaxation time, which means that heating occurs at almost isochoric conditions. It was shown in damage studies of metals induced with optical lasers [20–22] that this situation leads to the generation of large thermo-induced stresses and, as a result, to spallation of the top part of the metal. The single-shot spallation threshold at a 20° grazing incidence was measured to be $F_{\text{spall}} = 64 \pm 13 \text{ mJ/cm}^2$ of absorbed fluence, while the melting threshold was calculated to be $F_{\text{melt}} = 13 \text{ mJ/cm}^2$ [9]. It is known from the literature [20] that spallation starts with nucleation of subsurface voids or cavities in a melted layer of irradiated metal, created as a result of propagation of a tensile stress wave. In a particular fluence range high enough to cause melting, but not sufficient to induce complete spallation, the cavities can remain frozen below the surface, which was proven experimentally [21–23] and with molecular dynamic simulations [21]. Therefore, for metals there is a cavitation threshold that is lower than the spallation threshold.

Within this damage mechanism, we suggest that individual pulses with the highest fluence in a pulse train are capable of not only melting the surface of Ru but causing cavitation below the surface, as described above. The existence of subsurface cavities can create significant roughness and swelling of the surface [21–23].

This hypothesis can also explain the difference in the damage caused by multiple pulses at 3.17° and 20° grazing incidence (Fig. 2). The angular dependence of the absorbed energy, simulated as the integral of the electromagnetic (EM) field in the top 5 nm of the film, is shown in Fig. 4. In the region of total external reflection, for Ru with 13.5 nm light ranging from 0° to 27° grazing incidence, only evanescent waves penetrate into the film [25]. The penetration depth in our case is about 3 nm. However, the density of the EM field increases with increasing angle of incidence, and therefore the absorption dose also increases. The increased absorbed fluence may eventually reach the spallation threshold, resulting in locally occurring spallation of Ru.

The difference in the damage caused at very grazing and close to critical angles of incidence can be explained by assuming the cavitation to be the onset of damage. The AFM image of the surface damaged by 209 pulses in Fig. 2(B) is shown in Fig. 5.

In a first approximation, the roughness observed in Fig. 5 can be considered to be surface areas inclined at a certain angle $\theta^*$ to the prime film surface. Therefore, the effective incident angle at the surface is not $\theta$ but $\theta + \theta^*$, and therefore the absorption dose is locally increased. According to Fig. 4, the same increment $\theta^*$ of the incidence angle will cause a much greater increase in the absorbed dose for less grazing angles of incidence. For a 3.17° prime incidence angle $\theta$, an increase in the angle of incidence, for example, by $\theta^* = 20°$, as shown in Fig. 5, will lead to an increase in the absorbed dose by a factor of 70, which for a fluence of 15 mJ/cm² means an increase locally to 1 J/cm², which is much more than the spallation threshold. However, for an incidence angle of $\theta = 20°$, a similar inclination will increase the absorbed dose by not more than 50%, leading to an effective maximum fluence of 45 mJ/cm² for the pulse with the highest fluence in the histogram in Fig. 3. This is still below the spallation threshold for a pristine Ru film.

4. CONCLUSIONS

We present an experimental study of the damage caused by multiple ultra-short XUV pulses in Ru coatings. We found that the MSDT does not depend on the repetition rate by comparing damage threshold values obtained for 100 pulses arriving at 3.17° with a repetition rate ranging from 250 KHz to 1 MHz.

Based on analysis of the development of the damage morphology caused by an increase in the number of pulses arriving at a 20° grazing angle with a fluence close to the damage threshold, we suggest that the prime cause of the multi-shot damage is roughening of the Ru surface, induced by a cavitation
process. Therefore, the MSDT should be equal to the cavitation threshold.

Comparing the morphology of the craters created by pulses arriving at 3.17° and 20° grazing incidence, we arrive at the conclusion that surface roughening should lead to a local increase in the absorption of radiation, which may subsequently lead to local spallation of the roughened areas in Ru films.

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**REFERENCES**

1. T. Tschentscher, C. Bressler, J. Grünert, A. Madsen, A. Mancuso, M. Meyer, A. Scherz, H. Sinn, and U. Zastrau, "Photon beam transport and scientific instruments at the European XFEL," *Appl. Sci.* 7, 592 (2017).
2. R. Schoenlein, S. Bouet, M. Minitti, and A. M. Dunne, "The Linac coherent light source: recent developments and future plans," *Appl. Sci.* 7, 850 (2017).
3. I. Inoue, Y. Inubushi, T. Sato, K. Tono, T. Katayama, Y. Kameshima, K. Ogawa, T. Togashi, S. Owada, Y. Amemiya, T. Tanaka, T. Hara, and M. Yabashi, "Observation of femtosecond X-ray interactions with matter using an X-ray–X-ray pump–probe scheme," *Proc. Natl. Acad. Sci. USA* 113, 1492–1497 (2016).
4. J. Krzywinski, R. Conley, S. Moeller, G. Gwalt, F. Siewert, C. Waberski, T. Zeschke, and D. Cocco, "Damage thresholds for blaze diffraction gratings and grazing incidence optics at an X-ray free-electron laser," *J. Synchrotron Radiat.* 25, 85–90 (2018).
5. J. Krzywinski, D. Cocco, S. Moeller, and D. Ratner, "Damage threshold of platinum coating used for optics for self-seeding of soft-x-ray free-electron laser," *Opt. Express* 23, 5397–5405 (2015).
6. S. P. Hau-Riege, R. A. London, A. Graf, S. L. Baker, R. Soufli, R. Sobierajski, T. Burian, J. Chalupsky, L. Juha, J. Gaudin, J. Krzywinski, S. Moeller, M. Messerschmidt, J. Bozek, and C. Bostedt, "Interaction of short x-ray pulses with low-Z x-ray optics materials at the LCLS free-electron laser," *Opt. Express* 18, 23933–23938 (2010).
7. R. Sobierajski, I. Jacyna, P. Dluzewski, M. T. Klepka, D. Klinger, J. B. Pelka, T. Burian, V. Hajkova, L. Juha, K. Saksil, V. Vozda, I. Makhhotkin, E. Louis, B. Faatz, K. Keitel, E. Plonjes, S. Schreiber, S. Toleikis, H. K. Nienhuys, G. Wacht, T. Mey, and H. Enkisch, "Experimental study of EUV mirror radiation damage resistance under long-term free-electron laser exposures below the single-shot damage threshold," *J. Synchrotron Radiat.* 25, 77–84 (2018).
8. I. Milov, I. A. Makhotkin, R. Sobierajski, N. Medvedev, V. Lip, J. Chalupsky, J. M. Sturm, K. Tiedtke, G. de Vries, M. Störmer, F. Siewert, R. van de Kruisj, E. Louis, I. Jacyna, M. Jurek, L. Juha, V. Hajkova, V. Vozda, T. Burian, K. Saksl, B. Faatz, B. Keitel, E. Plonjes, S. Schreiber, S. Toleikis, R. Loch, M. Hermann, S. Strobil, H-K. Nienhuys, G. Wacht, T. Mey, and H. Enkisch, and F. Blikker, "Mechanism of single-shot damage of Ru thin films irradiated by femtosecond extreme UV free-electron laser," *Opt. Express* 26, 19685–19685 (2018).
9. W. Ackermann, G. Asova, V. Ayvazyan, A. Azima, N. Baboi, J. Bahr, V. Balandin, B. Beutner, A. Brandt, A. Bolzmann, R. Brinkmann, O. I. Brovko, M. Castellano, P. Castro, L. Catani, E. Chiadroni, S. Choroba, A. Cianchi, J. T. Costello, D. Cubaynes, J. Dardis, W. Decking, H. D. Hashemi, A. Delserieux, G. Di Pirro, M. Dohlus, S. Dusterer, A. Eckhardt, H. T. Edwards, B. Faatz, J. Feldhaus, K. Flottmann, J. Frisch, L. Frohlich, T. Garvey, U. Gensch, C. Gerth, M. Gorler, N. Golubeva, H. J. Grabsch, M. Grecki, O. Grimm, K. Hacker, U. Hahn, J. H. Han, K. Honkavaara, T. Hott, M. Huning, Y. Ivanisenko, E. Jaceschke, W. Jalmuzna, T. Jezynski, R. Kammering, V. Katalev, K. Kavanagh, E. T. Kennedy, S. Khodyachyk, K. Klooe, V. Kucharyn, M. Kollerle, W. Kopeck, S. Korpanov, D. Koslin, M. Krassilnikov, G. Kube, M. Kuhlmann, C. L. S. Lewis, L. Lijle, T. Limberg, D. Lipka, F. Lohl, H. Luna, M. Luong, M. Martins, M. Meyer, P. Michelato, V. Mitchev, W. D. Moller, L. Monaco, W. F. M. Muller, O. Napieralski, O. Napoly, P. Nicolosi, D. Nolle, T. Nunetz, A. Oppelt, C. Pagani, R. Paparella, N. Pichalke, J. P. Gutierrez, B. Petersen, B. Petrosyan, G. Petrovian, D. Petroyan, J. Pfluger, E. Plonjes, L. Polletto, K. Poznai, E. Prat, D. Proch, P. Pucyk, P. Radcliffe, H. Redlin, K. Rehlich, M. Richter, M. Roehrs, J. Roensc, R. Romanuiu, M. Ross, J. Rossbach, V. Rybnikov, M. Sachwitz, E. L. Saldin, W. Sandner, H. Schlarb, B. Schmidt, M. Schmitz, P. Schmurr, J. R. Schneider, E. A. Schneidmiller, S. Schnepp, S. Schreiber, M. Seidel, D. Sertore, A. V. Shabunov, C. Simon, S. Simrock, E. Sombrowski, A. A. Sorokin, P. Spanknebel, R. Spesylsteve, L. Staykov, B. Steffen, F. Stephen, F. Stulle, H. Thom, K. Tiedtke, M. Tischer, S. Toleikis, R. Treusch, D. Trines, I. Tsakov, E. Vogel, T. Weiland, H. Weise, M. Welhoff, M. Wendt, I. Will, A. Winter, K. Wittenburg, W. Wurth, P. Yeates, M. V. Yurkov, I. Zagorodnov, and K. Zapfe, "Operation of a free-electron laser from the extreme ultraviolet to the water window," *Nat. Photonics* 1, 336–342 (2007).
10. R. Sobierajski, M. Jurek, J. Chalupsky, J. Krzywinski, T. Burian, S. D. Farahani, V. Hajkova, M. Harmand, L. Juha, D. Klinger, R. A. Loch, C. Ozkan, J. B. Pelka, K. Sokolowski-Tinten, H. Sinn, S. Toleikis, K. Tiedtke, T. Tschentscher, H. Wabnitz, and J. Gaudin, "Experimental set-up and procedures for the investigation of XUV free electron laser interactions with solids," *JINST* 8, P02010 (2013).
11. K. Tiedtke, A. Azima, N. von Bargen, L. Blittner, S. Bonfigt, S. Düsterer, B. Faatz, U. Frühling, M. Gensch, C. Gerth, N. Guerassimova, U. Hahn, T. Hans, M. Hesse, K. Honkavaara, U. Jastrow, P. Junaric, S. Kapitzki, B. Keitel, T. Kracht, M. Kuhlmann, W. B. Li, M. Martin, T. Núñez, A. Oppelt, C. Pagani, V. Häkow, S. Toleikis, R. Treusch, D. Trines, I. Tsakov, E. Vogel, T. Weiland, H. Weise, M. Welhoff, M. Wendt, I. Will, A. Winter, K. Wittenburg, W. Wurth, P. Yeates, M. V. Yurkov, I. Zagorodnov, and K. Zapfe, "Operation of a free-electron laser from the extreme ultraviolet to the water window," *Nat. Photonics* 1, 336–342 (2007).
14. J. M. Liu, “Simple technique for measurements of pulsed Gaussian-beam spot sizes,” Opt. Lett. 7, 196–198 (1982).
15. J. Chalupský, L. Juha, J. Kuba, J. Cihelka, V. Hájková, S. Koptyaev, J. Krása, A. Velyhan, M. Bergh, C. Caleman, J. Hajdu, R. M. Bionta, H. Chapman, S. P. Hau-Riege, R. A. London, M. Jurek, J. Krzywinski, R. Nietubyc, J. B. Pelka, R. Sobierajski, J. Meyer-ter-Vehn, A. Tronnier, K. Sokolowski-Tinten, N. Stojanovic, K. Tiedtke, S. Toleikis, T. Tschentscher, H. Wabnitz, and U. Zastrau, “Characteristics of focused soft X-ray free-electron laser beam determined by ablation of organic molecular solids,” Opt. Express 15, 6036–6043 (2007).
16. J. Chalupský, T. Burian, V. Hájková, L. Juha, T. Polcar, J. Gaudin, M. Nagasono, R. Sobierajski, M. Yabashi, and J. Krzywinski, “Fluence scan: an unexplored property of a laser beam,” Opt. Express 21, 26363–26375 (2013).
17. J. Chalupský, J. Krzywinski, L. Juha, V. Hájková, J. Cihelka, T. Burian, L. Vyšín, J. Gaudin, A. Gleeson, M. Jurek, A. R. Khorsand, D. Klinger, H. Wabnitz, R. Sobierajski, M. Störmer, K. Tiedtke, and S. Toleikis, “Spot size characterization of focused non-Gaussian X-ray laser beams,” Opt. Express 18, 27836–27845 (2010).
18. F. Scholze, C. Laubis, C. Buchholz, A. Fischer, S. Ploeger, F. Scholz, H. R. Wagne, and G. Ulm, “Status of EUV reflectometry at PTB,” Proc. SPIE 5751, 749–758 (2005).
19. K. Tiedtke, J. Feldhaus, U. Hahn, U. Jastrow, T. Nunez, T. Tschentscher, S. V. Bobashev, A. A. Sorokin, J. B. Hastings, S. Möller, L. Cibik, A. Gottwald, A. Hoehl, U. Kroth, M. Krumrey, H. Schöppe, G. Ulm, and M. Richter, “Gas detectors for x-ray lasers,” J. Appl. Phys. 103, 094511 (2008).
20. V. V. Zhakhovskii, N. A. Inogamov, and K. Nishihara, “New mechanism of the formation of the nanorelief on a surface irradiated by a femtosecond laser pulse,” JETP Lett. 87, 423–427 (2008).
21. C. Wu, M. S. Christensen, J.-M. Savolainen, P. Balling, and L. V. Zhigilei, “Generation of subsurface voids and a nanocrystalline surface layer in femtosecond laser irradiation of a single-crystal Ag target,” Phys. Rev. B 91, 035413 (2015).
22. S. I. Ashitkov, N. A. Inogamov, V. V. Zhakhovskii, Y. N. Emirov, M. B. Agranat, I. I. Oleinik, S. I. Anisimov, and V. E. Fortov, “Formation of nanocavities in the surface layer of an aluminum target irradiated by a femtosecond laser pulse,” JETP Lett. 95, 176–181 (2012).
23. J.-M. Savolainen, M. S. Christensen, and P. Balling, “Material swelling as the first step in the ablation of metals by ultrashort laser pulses,” Phys. Rev. B 84, 193410 (2011).
24. M. Born and E. Wolf, Principles of Optics, 7th ed. (Cambridge University, 2000).
25. M. J. Bedzyk, G. M. Bommarito, and J. S. Schildkraut, “X-ray standing waves at a reflecting mirror surface,” Phys. Rev. Lett. 62, 1376–1379 (1989).