X-Ray Dosimetry During Low-Intensity Femtosecond Laser Ablation of Molybdenum in Ambient Conditions

Michal J. Wesolowski, Christopher C. Scott, Benjamin Wales, Ali Ramadhan, Sahr Al-Tuairqi, Surajith N. Wanasundara, Karim S. Karim, Joseph H. Sanderson, Carl A. Wesolowski, and Paul S. Babyn

***Abstract*— The absorbed X-ray dose was measured during 35 fs pulsed laser ablation of a high purity molybdenum target in ambient conditions for optical intensities from 1.8×10^{14} to 1.6×10^{15} W/cm^2 (0.5–4.5 mJ per pulse) at a repetition rate of 1 kHz. During 1 s of ablation, at a distance of 6 cm from the source, appreciable X-ray dose was observed at all optical intensities, ranging from 0.08 μGy, at the minimum intensity, to 0.97 μGy at the maximum intensity, corresponding to equivalent dose rates of 0.3–3.5 mSv/h. At the highest optical intensity, an absorbed dose of 0.29 μGy was measured at a distance of 12 cm from the source, corresponding to an equivalent dose rate of 1.1 mSv/h. Characterization of the laser plasma emissions showed X-rays with energies approaching 20 keV at higher laser pulse energies. In Canada, the annual equivalent dose limit for nonradiation workers is 1 mSv. Our findings suggest that under certain conditions it is possible to exceed this dosage in as little as an hour of fs-laser material processing in ambient conditions. As such, workers in these environments may need to be trained in radiation safety, equipped with personal dosimeters, and provided proper radiation shielding.**

***Index Terms*— Femtosecond (fs) laser ablation, molybdenum, X-ray dosimetry.**

I. Introduction

THE introduction of chirped pulse amplification (CPA) in the 1980s allowed for the development of the current state-of-the-art, high-power, and ultrashort pulsed laser systems [1]. Since that time, the availability and use of compact CPA-based femtosecond (fs) laser systems in research, industrial, and medical fields have increased significantly. These lasers can produce pulses capable of microtexturing [2], cutting [3], welding [4], nanomaterial synthesis [5]–[7], tissue ablation [8], [9], and surgery [10]. With this growth in applicability, it is becoming increasingly important to understand and manage the many safety aspects associated with the operation and use of fs-laser systems.

The most obvious hazards associated with the operation of Class IV fs lasers involve exposure of the eyes or skin to the direct laser beam or its reflections. Such exposure can cause serious eye injury or skin burns. It is therefore suggested that users be mindful of the beam position, keep reflective materials away from the beam, and wear proper personal protective equipment at all times when occupying a laser controlled area. While these simple steps can help to minimize the risk of injury associated with the most well-known fs-laser hazards, there are other potential hazards that are often overlooked in laboratories that use fs lasers in materials research.

It has been well established in the literature that the interaction of very high optical intensity ($> 10^{17}$ W/cm^2) fs-laser pulses with matter can create plasma that emits high brightness ionizing radiation with energies up to the MeV range [11], [12]. In fact, such X-ray sources have been studied thoroughly due to their potential use in medical imaging applications [13], [14]. Dosage rates as high as 10^{11} Gy/min have been reported for such systems [15].

Much less attention has been paid to the emissions from plasmas created by the fs-laser processing of materials in ambient conditions and at lower optical intensities (10^{14}–10^{16} W/cm^2), a situation which is far more common in laser materials processing laboratories. Pikuz *et al.* [16] and Thogersen *et al.* [17] have shown that hard X-rays can be emitted from plasmas created at these laser intensities; this knowledge does not appear to have widely been disseminated in laser safety guidelines. As a result, radiation safety and monitoring are not typically addressed in fs-laser materials processing laboratories.

In a preliminary effort to address this knowledge gap, the dosimetry and spectral data of plasma X-ray emissions associated with the fs ablation of molybdenum in ambient conditions are presented.

II. Materials and Methods

A regeneratively amplified Ti: sapphire tabletop laser system with a 1 kHz repetition rate was used to generate 35 fs, horizontally polarized laser pulses with a central wavelength

Manuscript received July 20, 2017; revised July 31, 2017; accepted August 1, 2017. Date of publication August 14, 2017; date of current version September 14, 2017. This work was supported in part by the Natural Sciences and Engineering Council of Canada and in part by the Sylvia Fedoruk Canadian Center for Nuclear Innovation.

M. J. Wesolowski, S. N. Wanasundara, C. A. Wesolowski, and P. S. Babyn are with the Department of Medical Imaging, University of Saskatchewan, Saskatoon, SK S7N 0W8, Canada (e-mail: mike.wesolowski@usask.ca; nalantha@gmail.com; carl.wesolowski@gmail.com; paul.babyn@saskatoonhealthregion.ca).

C. C. Scott and K. S. Karim are with the Department of Computer and Electrical Engineering, University of Waterloo, Waterloo, ON N2L 3G1, Canada (e-mail: chris.scott@uwaterloo.ca; kkarim@uwaterloo.ca).

B. Wales, A. Ramadhan, S. Al-Tuairqi, and J. H. Sanderson are with the Department of Physics and Astronomy, University of Waterloo, Waterloo, ON N2L 3G1, Canada (e-mail: bwales@uwaterloo.ca; ali.ramadhan@uwaterloo.ca; soso-f20@hotmail.com; j3sanderson@uwaterloo.ca).

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TNS.2017.2739156

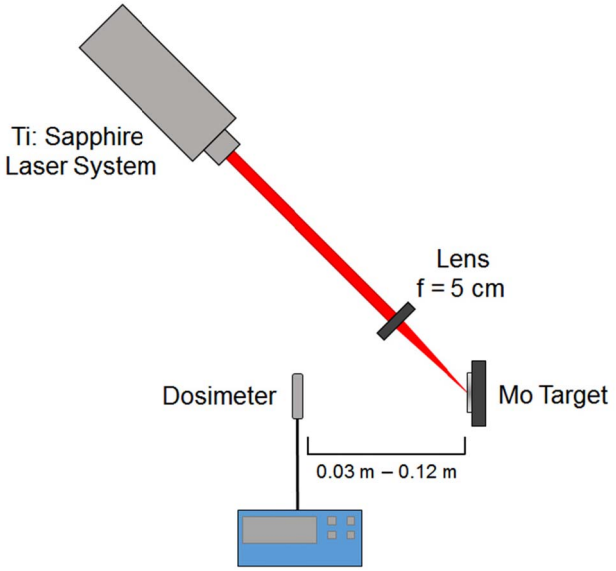

Fig. 1. Schematic of the experimental setup. A 35 fs-pulsed laser beam was focused onto a high purity molybdenum target. The absorbed dose was measured as a function of distance from the target as well as energy per laser pulse.

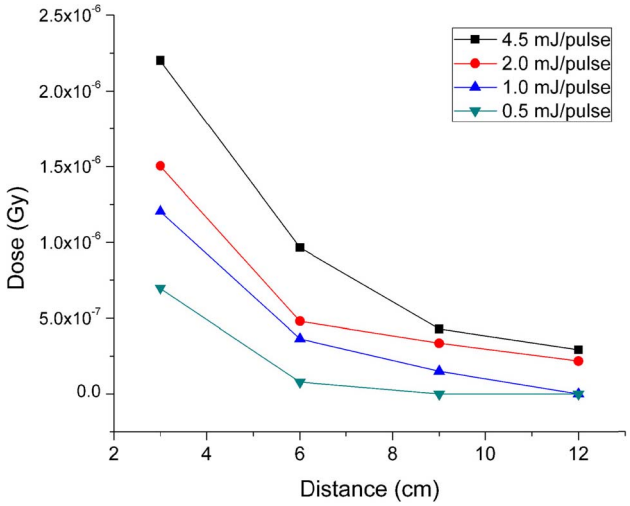

Fig. 2. Absorbed dose in air as a function of distance from the molybdenum target after 1 s of ablation, for multiple laser pulse energies.

of 800 nm. Pulses were focused onto the surface of a high purity molybdenum (Mo) target, using a 5 cm focusing lens to a spot size of 100-μm full width-half max diameter (knife edge technique). Experiments were carried out at standard temperature and pressure. During the experiments the energy per pulse was varied from 0.5 to 4.5 mJ, which resulted in focal intensities from $I = 1.8 \times 10^{14}$ to 1.6×10^{15} W/cm^2 at the target surface. The angle of incidence between the laser beam and plane of the target was kept constant during the experiments at 45° to maximize absorption of laser energy at the target [18]. Absorbed dose information was collected using a Solidose 300 Digital Dosimeter which was calibrated in terms of air kerma and sealed against ambient light. A schematic of the experimental setup is found in Fig. 1.

To determine the relationship between the absorbed dosage and laser pulse energy the dosimeter was positioned perpendicular to the Mo target at a fixed distance away from the surface and the dosage was measured while the target was ablated, with increasingly higher pulse energies (0.5, 1, 2, and 4.5 mJ per pulse), for one second (1000 pulses) at each energy. The horizontal position of the Mo target was changed between exposures to ensure fresh surface material; this was repeated three times at each pulse energy and averaged to account for microscopic variability of the target's surface structure. The position of the dosimeter was also altered from 3 to 12 cm away from the target surface in 3 cm steps, at each pulse energy, to determine the drop off in absorbed dose with distance. The absorbed dose was also measured as a function for angle from the target surface using a pulse energy of 2 mJ per pulse, at a distance of 12 cm from the target, to determine if the emissions were anisotropic. To gain insight into the spectral characteristics of the emitted radiation, X-ray spectral data were collected perpendicular to the Mo target during ablation using a CdTe spectrometer (AmpTek X-123).

TABLE I
DOSIMETRY DATA

	Pulse Energy (mJ)			
	0.5	1.0	2.0	4.5
Distance (cm)	Dose (μGy)			
3.0	0.70	1.20	1.50	2.20
6.0	0.08	0.36	0.48	0.97
9.0	0	0.15	0.34	0.43
12.0	0	0	0.22	0.29

Values of absorbed dose measured after 1 s of ablation of molybdenum by 35 fs-laser pulses at various source to detector distances and pulse energies. Values are averaged from 3 ablations, each at a new position on the target surface.

III. RESULTS AND DISCUSSION

The measured absorbed doses resulting from one second of target ablation as a function of distance from the detector for multiple laser pulse energies are summarized in Table I, and shown in Fig. 2. Each value is averaged from three separate ablations. At the lowest pulse energy, 0.5 mJ, the measured dose at 3 and 6 cm from the detector was 0.70 and 0.08 μGy, respectively, and below the detection limit of the dosimeter (0.01 μGy) at a distance of 9 cm. Increasing the pulse energy increased the dosage at each measured distance. For example, at a pulse energy of 1 mJ, the dose at 3, 6, and 9 cm was measured to be 1.20, 0.36, and 0.15 μGy, respectively, and below the detection limit of the dosimeter at 12 cm. At a pulse energy of 2 mJ the dose at 3, 6, 9, and 12 cm from the source was measured to be 1.50, 0.48, 0.34, and 0.22 μGy, respectively. As expected, the largest dose measured during the experiment was observed at the highest pulse energy and the closest distance to the dosimeter. At 4.5 mJ per pulse the measured dose from one second of target ablation at 3, 6, 9, and 12 cm from the source was on average 2.20, 0.97, 0.43, and 0.29 μGy, respectively.

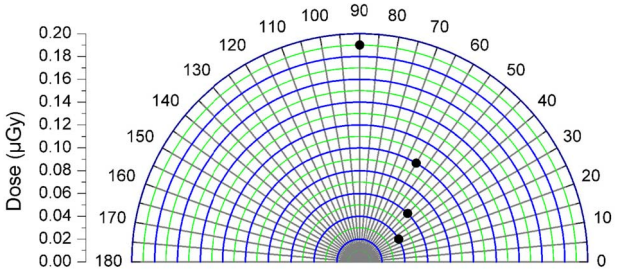

Fig. 3. Absorbed dose measured in air as a function of angle from the molybdenum target surface with 2-mJ laser pulse energies at a distance from the target of 12 cm.

The coefficient of variation for this data set was 20%, suggesting that X-ray production is somewhat variable from ablation to ablation. The maximum measured absorbed dose for a single ablation was 24 μGy observed at 3 cm from the target at the highest pulse energy.

In all cases the absorbed dose appears to obey a nonlinear relationship with distance. At the highest pulse energy used in this paper, 4.5 mJ/pulse, this relationship can be represented with a power function, $D = 9.86 \times 10^{-6} x^{-1.36}$ Gy, where D is the absorbed dose in gray and x is the distance from the source to the dosimeter measured in centimeters. The exponent of the power function is greater than -2 suggesting that the emissions may be anisotropic. To determine if this is the case the dose as a function of angle was observed. As is shown in Fig. 3, the dose decreases from 0.19 μGy perpendicular to the target to 0.04 μGy at 30° from the target surface. This result is similar to those of previous work showing anisotropic emissions from the fs-laser ablation of Cu, suggesting that this could be a general phenomenon [19].

It is interesting to note that at the lowest pulse energies used in this paper there was no detectable dose at 12 cm from the source, while appreciable dose was measured at this distance during higher energy ablation. This suggests an overall increase in the X-ray energy with increasing laser pulse energy, in addition to the observed increase in X-ray yield. To gain some understanding of the energy distribution, X-ray spectra observed in air at two pulse energies, 2 and 0.5 mJ are shown in Fig. 4(a) and (b), respectively. X-rays with energies approaching 20 keV were observed from 2 mJ/pulse ablation, however, characteristic emissions from Mo were not conclusively observed, likely due to the fact that the cross section for k-shell interaction is low for electron energies that are only slightly higher than the photopeak. At the lower laser pulse energy no X-ray energies greater than 15 keV were observed in the spectrum. It is also well known that the attenuation of X-rays in air is nonlinear with energy, which may also contribute to the dose behavior first outlined in Fig. 2. This nonlinear attenuation has extensively been studied and energy specific mass attenuation data are freely available from the National Institute for Standards and Technology [21].

Importantly, at the highest pulse energy used here, 4.5 mJ, which corresponds to an optical intensity of 1.6×10^{15} W/cm^2, the equivalent dose rate ranged from 1.1 to 7.9 mSv/hr at a source to dosimeter distance varying from 3 to 12 cm, with the lowest dose rate measured at the furthest distance from the source. Previously, Thogersen *et al.* [17] showed a dose rate as high as 10 mSv/h at a distance of 13 cm during the ablation of metals with 120 fs pulses and comparable optical intensities to those used here. The annual public radiation dose limit in Canada is 1 mSv [20]. The results presented here and those previously presented by Thogersen *et al.* [17] suggest that this value could easily be exceeded during fs-pulsed laser material processing lasting more than an hour. This suggests that laser materials processing experiments that are carried out in ambient conditions using similar laser parameters should be shielded to protect against X-rays with energies up to at least 20 keV, and workers in these environments should likely receive radiation safety training in addition to monitoring their radiation exposure. At a minimum, workers should not have their hands near a high-z target during fs-laser machining, should further investigate the radiation safety aspects of operation of their lasers and if X-rays are detected should likely wear ring dosimeters. Pending further developments, for the reduction of radiation exposure our recommendation is to increase distance from the source, limit exposure time, and add shielding to experimental spaces; in alignment with the as low as reasonably achievable principle.

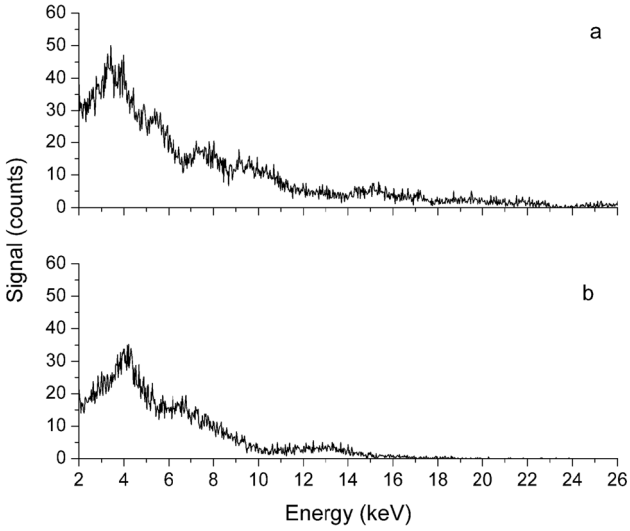

Fig. 4. X-ray plasma emissions spectra observed in air during the ablation of a molybdenum target with (a) 2 mJ and (b) 0.5 mJ laser pulse energies and a target to detector distance of 12 cm.

IV. Conclusion

The ablation of molybdenum with 35 fs-laser pulses having pulse energies that ranged from 0.5 to 4.5 mJ was found to produce high-energy X-rays and equivalent dose rates as high as 1.1 mSv/hr at 12 cm from the source. Laboratories in which laser materials processing takes place are currently unregulated for X-ray exposure and it is not generally appreciated that X-rays are emitted during the low-intensity fs-laser ablation of solids in air environments. Pending further investigation of the radiation safety aspects of the operation of these lasers for materials processing it is suggested that at the very least

workers keep their hands far from a high-z ablation target and consider monitoring their X-ray exposure with personal dosimeters.

REFERENCES

[1] D. Strickland and G. Mourou, "Compression of amplified chirped optical pulses," *Opt. Commun.*, vol. 56, no. 3, pp. 219–221, 1985.

[2] R. A. Delgado-Ruiź *et al.*, "Femtosecond laser microstructuring of zirconia dental implants," *J. Biomed. Mater. Res. B, Appl. Biomater.*, vol. 96, no. 1, pp. 91–100, 2011.

[3] W. Zhang *et al.*, "Ti:sapphire femtosecond laser direct micro-cutting and profiling of graphene," *Appl. Phys. A, Solids Surf.*, vol. 109, no. 2, pp. 291–297, 2012.

[4] H. Huang, A. Hu, P. Peng, W. W. Duley, and Y. Zhou, "Femtosecond laser-induced microwelding of silver and copper," *Appl. Opt.*, vol. 52, no. 6, pp. 1211–1217, 2013.

[5] M. J. Wesolowski, B. Moores, Z. Leonenko, R. Karimi, J. H. Sanderson, and W. W. Duley, "Synthesis of polymer-like hydrogenated amorphous carbon by fs-pulsed laser induced plasma processing of solid hexane," *Plasma Process. Polym.*, vol. 9, no. 7, pp. 701–708, 2012.

[6] M. J. Wesolowski *et al.*, "Polyyne synthesis and amorphous carbon nano-particle formation by femtosecond irradiation of benzene," *Carbon*, vol. 49, no. 2, pp. 625–630, 2011.

[7] M. J. Wesolowski, S. Kuzmin, B. Wales, J. H. Sanderson, and W. W. Duley, "Self-assembly of thin carbon micro-shells through pulsed laser irradiation of a ferrocene/benzene solution," *J. Mater. Sci.*, vol. 48, no. 18, pp. 6212–6217, 2013.

[8] F. H. Loesel *et al.*, "Non-thermal ablation of neural tissue with femtosecond laser pulses," *Appl. Phys. B, Lasers Opt.*, vol. 66, pp. 121–128, Jul. 1998.

[9] N. Nishimura, C. B. Schaffer, E. H. Li, and E. Mazur, "Tissue ablation with 100-fs and 200-ps laser pulses," in *Proc. 20th Annu. Int. Conf. IEEE Eng. Med. Biol. Soc.*, vol. 20. Nov. 1998, pp. 1703–1706.

[10] R. R. Krueger, J. Kuszak, H. Lubatschowski, R. I. Myers, T. Ripken, and A. Heisterkamp, "First safety study of femtosecond laser photodisruption in animal lenses: Tissue morphology and cataractogenesis," *J. Cataract Refractive Surg.*, vol. 31, no. 12, pp. 2386–2394, 2005.

[11] J. D. Kmetec, "Ultrafast laser generation of hard X-rays," *IEEE J. Quantum Electron.*, vol. 28, no. 10, pp. 2382–2387, Oct. 1992.

[12] J. D. Kmetec, C. L. I. Gordon, J. J. Macklin, B. E. Lemoff, G. S. Brown, and S. E. Harris, "MeV X-ray generation with a femtosecond laser," *Phys. Rev. Lett.*, vol. 68, pp. 1527–1530, Apr. 1992.

[13] B. Hou *et al.*, "Dependence of hard X-ray yield on laser pulse parameters in the wavelength-cubed regime," *Appl. Phys. Lett.*, vol. 84, no. 13, pp. 2259–2261, 2004.

[14] R. Toth *et al.*, "Evaluation of ultrafast laser-based hard X-ray sources for phase-contrast imaging," *Phys. Plasmas*, vol. 14, no. 5, p. 053506, 2007.

[15] C. Tillman *et al.*, "Survival of mammalian cells exposed to ultrahigh dose rates from a laser-produced plasma X-ray source," *Radiology*, vol. 213, no. 3, pp. 860–865, 1999.

[16] S. A. Pikuz *et al.*, "X-ray source for microstructure imaging under air conditions based on fs laser plasma," *J. Phys. Conf. Ser.*, vol. 244, no. 4, pp. 042030–042034, 2010.

[17] J. Thogersen, A. Borowiec, H. K. Haugen, F. E. McNeill, and I. M. Stronach, "X-ray emission from femtosecond laser micromachining," *Appl. Phys. A, Solids Surf.*, vol. 73, no. 3, pp. 361–363, 2001.

[18] M. Schnürer *et al.*, "Hard X-ray emission from intense short pulse laser plasmas," *Phys. Plasmas*, vol. 2, no. 8, pp. 3106–3110, 1995.

[19] Y. Hironaka, K. G. Nakamura, and K.-I. Kondo, "Angular distribution of X-ray emission from a copper target irradiated with a femtosecond laser," *Appl. Phys. Lett.*, vol. 77, no. 25, pp. 4110–4111, 2000.

[20] *Radiation Protection Regulations Consolidation: SOR/2000–203 Canada, Radiation Dose Limits Interpretation*, document 12–13, 2016.

[21] J. Hubbell and S. Seltzer, *Tables of X-Ray Mass Attenuation Coefficients and Mass Energy-Absorption Coefficients 1 keV to 20 MeV for Elements $Z = 1$ to 92 and 48 Additional Substances of Dosimetric Interest*, document NISTIR 5632, NIST, Gaithersburg, MD, USA, 1995.