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Damage threshold of inorganic solids under free-electron-laser irradiation at 32.5 nm wavelength

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We exposed samples of B_4C, amorphous C, chemical-vapor-deposition (CVD)-diamond C, Si, and SiC to single 25 fs-long pulses of 32.5 nm free-electron-laser radiation at fluences of up to 2.2 J/cm^2. The samples were chosen as candidate materials for x-ray free electron laser (XFEL) optics. We found that the threshold for surface-damage is on the order of the fluence required for thermal melting. For larger fluences, the crater depths correspond to temperatures on the order of the critical temperature, suggesting that the craters are formed by two-phase vaporization [1].

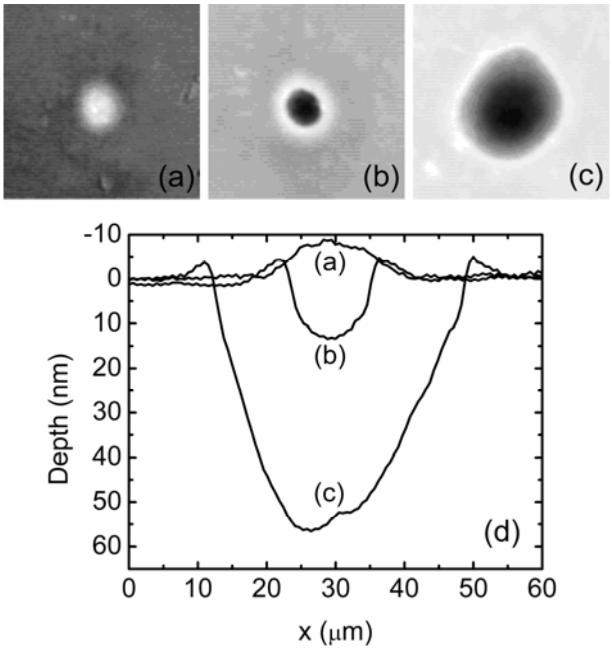

Fig. 1: FLASH-exposed SiC samples. (a) through (c) show depth profiles of spots exposed at fluences of less than 0.3 J/cm^2 ((a) and (b)) and 0.5 J/cm^2 (c). The range of the linear black (depression, positive) to white (extrusion, negative) scale is (a) 4 to -9 nm, (b) 13.5 to -5.5 nm, and 56.5 to -6 nm (c). The width of pictures (a) through (c) is 31.6 μm. (d) shows vertical lineouts through the center of the damaged

XFELs have the promise of producing extremely high-intensity ultrashort pulses of coherent, monochromatic radiation in the 1 to 10 keV regime. The expected high output fluence and short pulse duration pose significant challenges to the optical components, including radiation damage. It has not been possible to obtain direct experimental verification of the expected damage thresholds since appropriate x-ray sources are not yet available. FLASH has allowed us to study the interaction of high-fluence short-duration photon pulses with materials at the shortest wavelength possible to-date. With these experiments, we have come closer to the extreme conditions expected in XFEL-matter interaction scenarios than previously possible.

Fig. 1 (a) through (c) show surface profiles on a SiC sample exposed to different FLASH fluences from a ZYGO interferometer, and Fig. 1 (d) shows lineouts through the center. At low fluences (less than approximately 0.2 J/cm^2), the character of the damage was material-dependent, and we found highly diverse morphological changes at the irradiated surfaces. (i) In B_4C and SiC, we observed both craters and extrusions of a few nanometers in height; (ii) in Si, we observed only craters; and (iii) in a-C, we observed only extrusions. At larger fluences (up to 2.2 J/cm^2), we observed craters that were tens to hundreds of nanometers deep in all the samples.

In order to extract the threshold fluence for damage, it is necessary to study the onset of surface modifications at relatively low fluences. At these fluences, the pulse energy measurements were found to be very noisy. Assuming that the variation in the onset of damage is solely due to the statistical variation of the FEL output, we have developed a statistical method to analyze the data, based on the known pulse-energy distribution of SASE-based FELs. The measured energy fluctuations of high energy (weakly-attenuated) pulses can be described by a Gamma distribution with a shape parameter, describing the number of optical modes in the radiation source, of M=4.1. For 15-pulse exposure series at low energies (high attenuation), only the average pulse energy is known. For these low energy series we assume the same Gamma distribution function and assign the spots showing damage to the higher pulse energies out of the distribution. For example, the low-fluence exposures of the SiC samples were performed in two series with different average energies. In the first series with an average energy of 0.37 μJ, we observed surface modifications on 27 % of the exposed spots, which translates to a damage threshold of 0.46 μJ. In the second series with an average energy of 0.47 μJ, we observed surface modifications on 73% of the exposed spots, which translates to a damage threshold of 0.36 μJ. The error between these two independently-obtained damage thresholds for SiC is less than 25%, leading to a damage threshold fluence for SiC of 0.14 J/cm^2. Similar analyses result in measured damage threshold fluences of 0.060 J/cm^2 for a-C, 0.20 J/cm^2 for B_4C, 0.14 J/cm^2 for CVD-diamond, and 0.087 J/cm^2 for Si. We estimate the error in the damage threshold to be about 50% due to errors in the beam area, the energy measurements of the gas detector, and the small number of exposure per exposure series. Thresholds of inorganic materials [1] are at least one order of magnitude higher that the threshold found with the same radiation in an organic polymer [2]. This is not surprising because inorganic solids exhibit in general higher thermal as well as radiation resistance.

Low-fluence (threshold) damage is expected to be dominated by thermal processes, such as melting, occurring after the pulse, on timescales longer than 1 ps. One possible thermal damage mechanism is fluid motions that occur upon melting. The threshold damage fluence is expected to lie between the value required to reach the melting temperature and the value required to additionally supply the latent heat of melting. To calculate the melt thresholds, we use tabulated optical constants for cold materials, assuming that they do not change significantly during the pulse. This is supported by recent FLASH measurements of transmission of Al and Si and reflection of SiO_2 at fluences up to 2 J/cm^2. The calculated melt thresholds lie between 0.056 and 0.065 J/cm^2 for B_4C, 0.043 and 0.11 J/cm^2 for CVD-diamond, 0.077 and 0.18 J/cm^2 for Si, and 0.055 and 0.082 J/cm^2 for SiC. We find that the measured damage threshold fluences presented in the previous paragraph are on the order of the expected melting fluences. This result is very reassuring in that it supports the working assumptions made for determining the damage thresholds in LCLS optics.

We have recently extended this study from 32 nm wavelength radiation to 21.7 and 13.5 nm, getting closer to the actual XFEL operating parameters. We have also analyzed materials of wider interest for XFEL optics, such as thin films of high-Z materials considered for Kirkpatrick-Baez focusing optics and multilayer mirrors.

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References

[1] S. Hau-Riege et al., Appl. Phys. Lett. 90, 173128 (2007).
[2] J. Chalupsky et al., Opt. Express 15, 6036 (2007).