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

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Samples of B4C, amorphous C, chemical-vapor-deposition-diamond C, Si, and SiC were exposed to single 25 fs long pulses of 32.5 nm free-electron-laser radiation at fluences of up to 2.2 J/cm². The samples were chosen as candidate materials for x-ray free-electron-laser optics. It was found that the threshold for surface damage is on the order of the fluence required for thermal melting. For larger fluences, the craters depths correspond to temperatures on the order of the critical temperature, suggesting that the craters are formed by two-phase vaporization. © 2007 American Institute of Physics. [DOI: 10.1063/1.2734366]

X-ray free-electron lasers (XFELs) have the promise of producing extremely high-intensity ultrashort pulses of coherent, monochromatic radiation in the 1–10 keV regime,1,2 enabling many research endeavors. The expected high output fluence and short pulse duration pose significant challenges to the optical components required to utilize XFEL beams, including radiation damage. Theoretical work on the design of robust optics has been discussed in the literature.3–7 It is expected that high-melting-point, low-atomic-number materials will be most resistant to damage. It has been suggested that the fundamental mechanism that determines the damage threshold for single-pulse exposures in insulators is thermal melting.3,6 For multiple-pulse exposures, the damage threshold is potentially lower due to fatigue effects associated with thermomechanical stresses,7 chemical changes, or phase transitions.8

It has not been possible to obtain direct experimental verification of the expected damage thresholds since appropriate x-ray sources are not yet available. Recently, an extreme-ultraviolet free-electron laser in Hamburg (FLASH) has been built at DESY.9 This source 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. In this letter we describe our experimental studies of the onset of single-pulse damage in materials such as B4C, C, Si, and SiC.

We exposed different materials to the focused FLASH beam at normal incidence in November 2005. FLASH was operated at a wavelength of 32.5±0.5 nm, with pulse energies up to 5.8 µJ, and pulse durations of 25±5 fs. A gas attenuator was used to adjust the pulse energy and a nearly transparent gas monitor detector (GMD) was used to measure each pulse energy.10 The beam was then focused onto the sample using a grazing incidence ellipsoidal mirror to a maximum energy fluence of approximately 2.2 J/cm². At each attenuator setting we exposed 15 positions of each sample with single pulses. Coupled with the natural pulse-to-pulse variation of the energy, this allowed us to span a relatively fine set of exposure fluences.

We studied five different materials, which are likely candidates for XFEL optics: (i) SiC slabs were fabricated using chemical-vapor deposition and had an average grain size of 7.5 µm. The root-mean-square (rms) roughness of the sample surface measured with atomic-force microscopy (AFM) was 1.8 Å. (ii) B4C slabs were fabricated by hot pressing of B4C powder, had an average grain size of 5 µm, and a rms surface roughness of 5 Å. (iii) Si samples were cleaved from a 0.28-mm-thick-boron-doped silicon (001) wafer with a surface roughness of less than 4 Å. (iv) 46-nm-thick amorphous-carbon (a-C) samples with a surface roughness of less than 3 Å were made by magnetron deposition on a silicon wafer.11 (v) 1-mm-thick polycrystalline...
chemical-vapor-deposition (CVD)-diamond slabs were fabricated using chemical-vapor deposition on a Si substrate. We irradiated the smooth back side of the diamond after removing the Si substrate.

Prior to exposure, we verified the cleanliness of the samples using Nomarski differential interference contrast (DIC) microscopy, which is sensitive to phase differences across the sample, allowing us to visualize depth gradients. After FLASH exposure, we analyzed the exposed areas of the Si, SiC, and B4C samples using a white light interferometer and the exposed areas of the a-C and CVD-diamond samples using the Nomarski DIC microscopy. Some exposed areas were also analyzed with an AFM.

Figures 1(a)–1(c) show surface profiles on a SiC sample exposed to different FLASH fluences from the interferometer, and Fig. 1(d) shows line outs through the center of the damaged regions. We extracted the peak crater depths from the surface profiles. Figure 2 shows the peak depth as a function of flashed laser fluence for SiC. The negative depth values refer to surface extrusions. Fluences below 0.3 J/cm² are noise dominated.

The GMD is based on the photoionization of a low density noble gas or N₂, creating electrons and ions that are detected with Faraday cups. The ion signal was averaged, typically over 25 s. It was calibrated at a synchrotron storage ring. The measurement uncertainties are about 4%. The electron signal was used to get individual pulse energies. The electron signal was calibrated by comparing its average to the averaged ion signal. The relative uncertainty of the pulse energy is about 10%, dominated by the inherent statistical fluctuations of the FEL pulse intensities during the calibration. At the time of these experiments, the readout electronics (electrometer, integrators, and analog-to-digital converters) of the GMD were not optimized for pulse energies below 1–2 μJ, and we found that the signal was too much noisy to determine individual pulse energies below 1 μJ.

In order to extract the threshold fluence for damage, it is necessary to study the onset of surface modifications at relatively low fluences. 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 for pulse energies below 1 μJ, based on the known pulse energy distribution of self-amplified spontaneous-emission-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. As shown in Fig. 3, 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%. Using the measured focal spot areas, the damage threshold fluence for SiC was found to be with the GMD and were found to be very noisy at pulse energies below 1 μJ (fluence below 0.4 J/cm²). For fluences greater than 0.2 J/cm², we found that the peak depth generally increases with beam energy. For fluences less than 0.2 J/cm², the trend was often not clear, likely due to the large noise in the pulse energy measurements.

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![Figure 1](image1.png)

**FIG. 1.** FLASH-exposed SiC samples. (a)–(c) show depth profiles of spots exposed at fluences of less than [(a) and (b)] 0.3 J/cm² and (c) 0.5 J/cm². 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 (c) 56.5 to −6 nm. The width of pictures [(a)–(c)] is 31.6 μm. (d) shows vertical line outs through the center of the damaged regions.

![Figure 2](image2.png)

**FIG. 2.** Peak crater depth as a function of nominal laser fluence for SiC. The negative depth values refer to surface extrusions. Fluences below 0.3 J/cm² are noise dominated.
In summary, we exposed amorphous C, B$_4$C, CVD diamond, Si, and SiC materials to extremely short FLASH pulses at 32.5 nm. We found that the single-pulse damage threshold as detected by white light interferometry and confirmed by AFM is comparable to the conventional melt threshold, substantiating the claim that the limit for the maximum permissible fluence for LCLS optics can be based on this threshold.

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9FLASH was formerly known as VUV-FEL and TTF2 FEL. See http://vuv-fel.desy.de and references therein.
12We used the ZYGO white light interferometer fabricated by ZYGO Corp., Middlefield, CT.
17J. Krzywinski (unpublished).