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Thermal stability of tungsten zone plates for focusing hard x-ray free-electron laser radiation

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Abstract. Diffractive Fresnel zone plates made of tungsten show great promise for focusing hard x-ray free-electron laser (XFEL) radiation to very small spot sizes. However, they have to withstand the high-intensity pulses of the beam without being damaged. This might be problematic since each XFEL pulse will create a significant temperature increase in the zone plate nanostructures and it is therefore crucial that the optics are thermally stable, even for a large number of pulses. Here we have studied the thermal stability of tungsten zone-plate-like nanostructures on diamond substrates using a pulsed Nd:YAG laser which creates temperature profiles similar to those expected from XFEL pulses. We found that the structures remained intact up to a laser fluence of 100 mJ cm^{-2} , corresponding to a 6 keV x-ray fluence of 590 mJ cm^{-2} , which is above typical fluence levels in an unfocused XFEL beam. We have also performed an initial damage experiment at the LCLS hard XFEL facility at SLAC National Accelerator Laboratory, where a tungsten zone plate on a diamond substrate was exposed to 10^5 pulses of 6 keV x-rays with a pulse fluence of 350 mJ cm^{-2} without any damage occurring.

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1. Introduction

Hard x-ray free-electron lasers (XFELs) are novel sources of x-rays, providing unprecedented brightness, high coherence and ultra-short pulses. These sources enable numerous new and exciting scientific experiments [1]. Many of these rely on the ability to focus or shape the beam. One option for this task is the use of diffractive optics of various kinds and a particular area where diffractive optics can play an important role is in focusing XFEL beams to very small dimensions. Fresnel zone plates can provide diffraction-limited focusing of x-rays to focal spot diameters of 50 nm and below [2–4] with aberration-free wave fronts [5], which is a crucial requirement for, e.g., coherent imaging. The goal of this paper is to experimentally verify the thermal stability of nanofocusing tungsten zone plates on diamond substrates. Zone plate optics made in this way could provide a pathway towards XFEL focal spot sizes in the 10 nm regime [2].

Because of the high brightness of XFEL sources any optics that absorb even a fraction of the beam risk being fatally damaged. Possible damage mechanisms include radiation damage, mechanical failure caused by thermal stress/strain and thermally induced effects such as melting and recrystallization. The amount of absorbed radiation depends on the material of the optic and the wavelength of the radiation. Unfortunately, typical materials for hard x-ray diffractive optics, such as gold, iridium and tungsten, exhibit significant absorption. As a result, the temperature rise in a single pulse is as high as several hundred kelvin [6] and, depending on the repetition rate of the XFEL, there can also be a significant temperature increase over time. In such cases it becomes increasingly important to cool the optics effectively, for instance by the use of a diamond substrate with excellent heat conduction properties. Recent experiments performed at the LCLS hard XFEL facility at SLAC National Accelerator Laboratory showed that gold zone plates on a thin silicon substrate did not survive in the beam for long, whereas iridium zone plates on a diamond substrate as well as purely diamond-based zone plates were undamaged [7]. It is clear that good thermal stability of the optical material and substrate is crucial for use at XFEL sources.

In this work we investigate the thermal stability of tungsten zone plates on a thick diamond substrate. To mimic the pulsed nature of the XFEL radiation, zone-plate-like nanostructures were exposed to a pulsed Nd:YAG laser. Even though the absorption length of the green light

(532 nm) and the longer pulse length (ns) differ significantly from the XFEL case they still create similar temperature gradients in the optic. The technique has been used in a similar way to successfully determine the damage threshold for x-ray mirrors for the LCLS [8], in agreement with later measurements at LCLS [9]. In order to better control the absorption of laser light in nanostructures we used linear gratings with the same period as the outer parts of a zone plate instead of a real zone plate structure. These gratings should be a good model system for zone plates from a damage perspective since we expect the outer parts of the zone plate, where the period is smallest, to be most susceptible to damage. The fabricated tungsten gratings were exposed to a large number of pulses with peak temperatures ranging from a few hundred kelvin to temperatures high enough to melt the structures. We evaluated the damage using scanning electron microscopy (SEM) and x-ray diffraction measurements, where we compared the grating diffraction efficiency before and after exposure. Finally, we performed initial damage experiments at LCLS, where tungsten zone plates on diamond substrates were exposed to 10^4 and 10^5 pulses of 6 keV x-rays at the XPP end station without any observable damage.

2. Fabrication of tungsten nanostructures and zone plates

Zone plates for hard x-rays typically consist of a suitably patterned optical material which is placed on a substrate. Patterning of the optical material is usually done by electron beam lithography in combination with either etching or electro-plating or a combination of both. The choice of optical material is dependent on the energy of the x-rays and the fabrication possibilities. Ideally, the material should provide a strong phase shift and have minimal absorption. For x-ray energies between 5–15 keV this can be achieved by low Z elements such as silicon and carbon (diamond) if the thickness of the diffracting structures can be made thick enough (several μm). However, this is a significant fabrication challenge, especially for optics with high resolution that requires narrow structures. An alternative is to use high Z elements such as gold, iridium, tantalum, platinum or tungsten which provide an adequate phase shift at much lower thicknesses, but at the cost of increased absorption.

In this study we used tungsten zone plates and linear gratings on $100\ \mu\text{m}$ thick CVD diamond substrates. Tungsten is an excellent material for diffractive hard x-rays optics (around 5–15 keV), providing high diffraction efficiency and straightforward fabrication using standard nanofabrication techniques [10]. The high melting point (3695 K) and high thermal conductivity are also favorable properties for XFEL applications. A diamond substrate thickness of $100\ \mu\text{m}$ is a good compromise between thermal properties and high x-ray transmission. However, it should be noted that for low repetition rate sources, such as the LCLS, thinner substrates of a few μm offer sufficient heat conduction, with the additional benefit of higher transmission for x-rays in the region around 5 keV. A thicker substrate becomes important for high repetition sources, such as the European XFEL.

The gratings and zone plates were made in a multi-step process where the pattern was defined by electron beam lithography in a resist and thereafter transferred to a chromium layer that was used as an etch mask for the final pattern transfer into an underlying tungsten layer by reactive ion beam etching. As a last step the chromium layer was removed. The fabrication process is identical to that described in [11], except that the diamond etch step was omitted here. The resulting gratings had a period of 200 nm, a thickness of approximately 350 nm and a size of $200\ \mu\text{m} \times 200\ \mu\text{m}$. An SEM image as well as an illustration of one such grating is presented in figure 1. The zone plates used in the LCLS experiment were fabricated in the

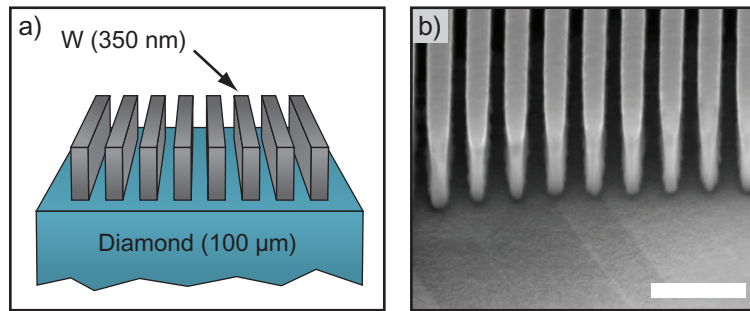


Figure 1. (a) Illustration showing a 350 nm thick tungsten grating with a period of 200 nm on top of a 100 μm thick diamond substrate. (b) SEM image of one of the fabricated gratings used in the study. The scale bar is 500 nm long.

same way and had a diameter of 75 μm , an outermost zone width of 50 nm and a thickness of 250 nm.

3. Heating with nanosecond visible-light laser pulses

3.1. Methods

The aim of this experiment was to emulate, by using a pulsed Nd:YAG laser, the temperature fluctuations in a tungsten zone plate when illuminated with an XFEL beam. We first consider how a tungsten nanostructure is affected by XFEL illumination at 6 keV, the same energy that was used in the LCLS experiments. Radiation from an XFEL consists of extremely short pulses in the order of tens of femtoseconds with energies of a few mJ and a typical beam diameter of 500 μm to 1 mm. Depending on the absorption coefficient of the zone plate material the energy in a single pulse can be enough to completely destroy the optic. However, we are interested in the case where the absorbed dose is below the single pulse damage threshold. In this case the absorbed XFEL pulse can be treated as an instantaneous heat source and classical thermodynamics can be used to describe the temperature evolution in the system. It has been suggested and experimentally verified that this approach is valid if the absorbed dose is below the melting dose of the material [8, 9].

In this way we have calculated the temperature in a tungsten grating after irradiation by one XFEL pulse with parameters chosen to match the LCLS source and the experimental conditions described in section 4. For more information regarding the simulation details see [6, 12]. Figure 2 shows the resulting temperature in the hottest region in the middle of the beam where the temperature increase in the tungsten is about 750 K. Because of the much lower absorption coefficient of diamond the substrate is only heated a few degrees. After the pulse, heat will flow down into the substrate. The temperature in the tungsten structures decreases and reaches ambient temperature within 10 μs .

It is important to point out that the absorption coefficient of tungsten depends on the photon energy and therefore the heating will do, too. However, 6 keV is in the lower energy range where we think our tungsten zone plates will be used and for higher energies the absorption coefficient is only lower, meaning that if the zone plates are undamaged at 6 keV they are not likely to be damaged at higher energies.

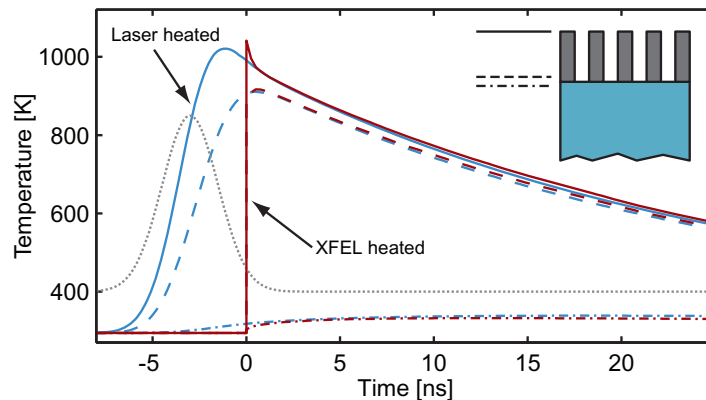


Figure 2. Results of transient thermal calculations for a 350 nm thick tungsten grating on a 100 μm thick diamond substrate comparing heating by a 1 mJ XFEL pulse of 6 keV photon energy (350 mJ cm^{-2}) (red) and heating by a 3.5 ns long Nd:YAG laser pulse (57 mJ cm^{-2}) (blue). The temporal profile of the laser pulse is shown as a dotted gray line and the temperature is plotted versus time for the top surface of the grating (solid line), the bottom of the grating (dashed line) and the top of the substrate (dash-dotted line).

If we now consider heating of tungsten nanostructures by a visible (532 nm) Nd:YAG laser with 3.5 ns long pulses the temperature evolution is slightly different, which is shown in figure 2. The laser fluence has been adjusted such that the resulting temperature increase matches the LCLS case. The main differences between laser and XFEL heating are that the laser pulse is much longer and only heats the surface of the tungsten, whereas the XFEL radiation is absorbed almost uniformly throughout the tungsten structure. Nevertheless, comparing the two cases, it can be seen that shortly after the peak temperature of the laser-heated sample the temperature profiles are very similar, both in the tungsten itself and in the diamond substrate. The two methods should therefore create comparable stresses in the structures even though the heating mechanisms are different.

For the experiments we used a Quantel Brilliant Q-switched frequency-doubled Nd:YAG laser producing 3.5 ns long pulses at 532 nm wavelength with a repetition rate of 20 Hz. Figure 3 shows an illustration of the experimental arrangement including the most important components. The sample was placed inside a vacuum chamber and the laser beam was passed through a 3 mm diameter aperture and a 500 mm focal length lens which created a 100 μm large (FWHM) focal spot on the sample. A beam splitter was placed before the lens and one part of the beam was used to measure the laser power, which could be varied by neutral density filters.

Since most of the structures of a hard x-ray zone plate are smaller than the wavelength of the laser light they will act as wire grid polarizers [13], transmitting light polarized perpendicular to the structure orientation. For this reason the experiment described would not be possible in a controlled way with a real zone plate since the amount of absorbed light would vary over the zone plate area. We therefore used linear tungsten gratings with a period of 200 nm instead, similar to the period in the outer part of the zone plates, in order to control the absorption in the structures. The orientation of the gratings was chosen such that the lines were parallel with the polarization of the incident laser light. In this way the radiation is either reflected or absorbed on the surface of the gratings, essentially acting as a flat surface but with a slightly

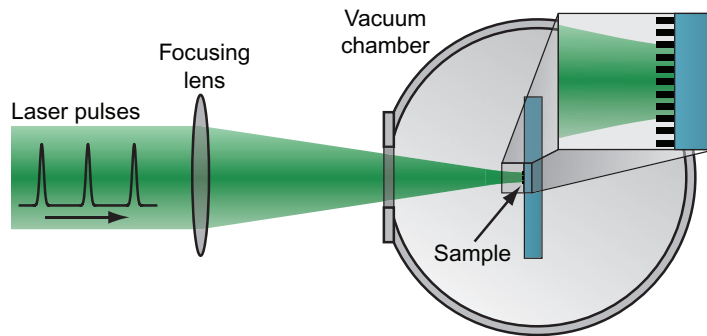


Figure 3. Schematic experimental arrangement for the laser heating experiments. The grating samples were mounted inside a vacuum chamber (pressure $< 5 \times 10^{-6}$ mbar) and the 3.5 ns long pulses from a Nd:YAG laser (532 nm, 20 Hz) were focused to a spot size of $100 \mu\text{m}$ (FWHM) using a 500 mm focal length lens. The orientation of the grating lines was parallel with the polarization of the laser light.

reduced reflectivity. The reflectivity was measured to be 39%, which is in good agreement with the theoretical value of 40% obtained by a rigorous coupled wave simulation using the software GD-Calc⁴. The slight deviation from the simulated value is probably due to an uncertainty in the line-to-space ratio of the fabricated gratings and deviations from an exact rectangular shape of the grating lines used in the simulations.

Gratings were exposed to a varying number of pulses and fluences and the damage was evaluated using SEM and by comparing the x-ray diffraction efficiencies of the gratings before and after laser exposure. The change in diffraction efficiency of the first and minus first order was measured at 9 keV using a liquid-jet gallium microfocus source [14] and a Photonic Science x-ray camera. The temperature in the gratings during exposure was estimated from the incident laser fluence, the laser absorption in the gratings and a transient thermal calculation as described above. This temperature could then be used to calculate the corresponding x-ray fluence.

3.2. Results and discussion

Figure 4 shows gratings exposed to three different fluence levels with results ranging from no visible damage to almost completely melted structures. The corresponding x-ray diffraction patterns can be seen in figure 5. For gratings exposed to a fluence level of 100 mJ cm^{-2} for 1.72×10^6 pulses no damage was observed in the SEM pictures and the diffraction efficiency was unchanged. Calculations give a maximum temperature of about 1500 K in the tungsten structures. When the fluence was increased to 180 mJ cm^{-2} the temperature rises to around 2500 K and cracks in the grating lines appear. As a result of this, small patches of the gratings melted and the diffraction efficiency was reduced by roughly 30%. For the highest level of fluence, 360 mJ cm^{-2} , the estimated temperature increase is as high as 4300 K and the structures are almost completely destroyed in as few as 1200 pulses. However, as can be seen in the SEM picture some of the structure still remain intact but the diffraction efficiency is close to zero.

⁴ Grating Diffraction Calculator (GD-Calc®) <http://software.kjinnovation.com/GD-Calc.html>.

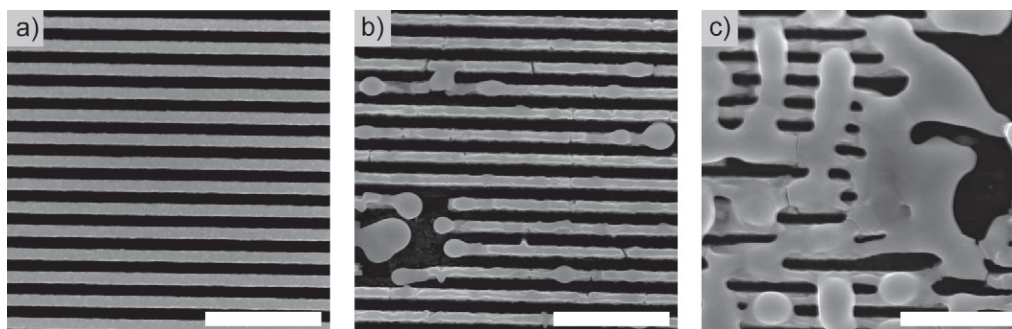


Figure 4. SEM images of tungsten gratings with 200 nm period after laser exposure to (a) 100 mJ cm^{-2} for 1.72×10^6 pulses, (b) 180 mJ cm^{-2} for 7.2×10^4 pulses and (c) 360 mJ cm^{-2} for 1.2×10^3 pulses. The scale bar is $1 \mu\text{m}$ long.

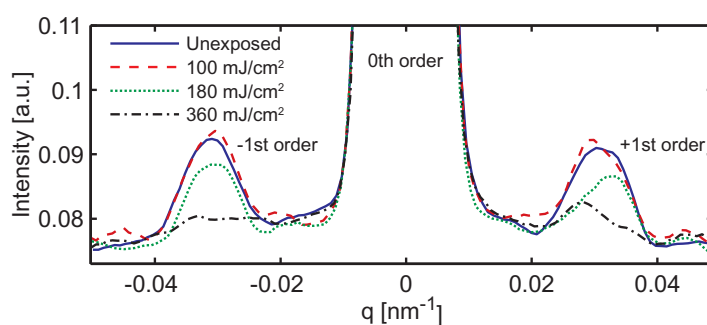


Figure 5. Measured diffraction patterns from tungsten gratings after exposure to 100 mJ cm^{-2} for 1.72×10^6 pulses, 180 mJ cm^{-2} for 7.2×10^4 pulses and 360 mJ cm^{-2} for 1.2×10^3 pulses.

The damage appears as small cracks in the grating lines, followed by increased heating and melting. Furthermore, if the contact between the tungsten structures and the diamond substrate is weakened, the heat flow into the substrate is reduced, which further accelerates the heating of the tungsten structures.

It is now possible to calculate an x-ray fluence for which no damage is expected. Using a laser fluence of 100 mJ cm^{-2} in our theoretical model gives a maximum surface temperature of 1500 K and a maximum temperature of 1300 K in the base of the tungsten structures. Comparing this to calculations for an XFEL pulse, similar temperature profiles are expected from an x-ray fluence of 590 mJ cm^{-2} at 6 keV photon energy.

It is interesting to compare this fluence to measured fluence levels at existing XFELs and expected levels at future sources. The fluence is determined by the pulse energy and the beam size at the position of the optics. To reduce the heat load on optics, many experimental stations are located several hundred meters from the source where the beam size is large and the fluence level is, in most cases, below the expected damage threshold for our tungsten zone plates. As an example, a beam size of 1 mm and pulse energy of 2 mJ results in an x-ray fluence of 175 mJ cm^{-2} , which should be compared to the expected damage threshold above 590 mJ cm^{-2} .

4. LCLS damage experiment

4.1. Methods

In addition to the visible laser damage experiments, described in section 3, we have also performed damage experiments at the XPP end station at LCLS [15]. Two tungsten zone plates on diamond substrates were exposed to 10^4 and 10^5 pulses of 6 keV x-rays, at 120 Hz, with an average pulse energy of about 1 mJ and a beam size of $500 \mu\text{m}$ (FWHM), corresponding to an x-ray fluence of 350 mJ cm^{-2} in each pulse, which is in the region where the visible-light experiments show no signs of damage. To monitor any decrease in diffraction efficiency during the experiment a scintillator-based x-ray camera was used to record the diffracted light, for each pulse, at a position approximately 50 cm behind the focus of the zone plate. After the experiments were completed both zone plates were examined by SEM, atomic force microscopy (AFM) and x-ray energy dispersive spectroscopy (EDS).

4.2. Results and discussion

The tungsten zone plates survived the LCLS beam with no signs of damage to the nanostructures in the SEM images. Furthermore, we did not observe any change in the diffraction efficiency throughout the course of the experiment. These results are in good agreement with the predictions from the visible-light experiments in section 3. The only sign of exposure to the beam was a slight discoloration of the tungsten surface in exposed areas, when viewed in a visible light microscope and SEM. Further analysis with EDS and AFM confirmed that the cause of the discoloration was a thin layer of carbon, which is typical for exposure to x-rays with high intensity and for the relatively rough vacuum (10^{-2} mbar) the zone plates were placed in [16].

Estimating the temperature in the zone plate using the method described in 3.1 yields a maximum temperature of 1040 K and temporal temperature profiles almost identical to figure 2. The only difference is that the zone plates in the x-ray experiment were slightly thinner than the structure simulated in figure 2. However, this has only a minor impact on the expected temperatures.

5. Summary

To summarize, tungsten zone plates on a thick diamond substrate show promise for withstanding the thermal fluctuations caused by exposure to an unfocused hard XFEL beam. Systematic damage studies with visible laser pulses suggest that tungsten zone plates should withstand x-ray fluences in the order of 590 mJ cm^{-2} or higher. In experiments structures were exposed to 1.72×10^6 Nd:YAG pulses with a fluence of 100 mJ cm^{-2} (equivalent to 590 mJ cm^{-2} of 6 keV x-ray fluence) and temperature increases of roughly 1200 K in each pulse without any sign of damage.

In addition, a tungsten zone plate was exposed to 10^5 LCLS pulses at 6 keV x-ray energy and 350 mJ cm^{-2} pulse fluence (1 mJ pulse energy and $500 \mu\text{m}$ FWHM beam diameter) without showing signs of damage.

Although the results of this study are very promising it is important to remember that the list of possible damage mechanisms also include long-term radiation damage. To investigate

this, longer experiments with exposures to hard x-ray radiation are required. One should also remember that the temperature profiles created from visible laser heating, even though similar, are not identical to those resulting from XFEL pulses and it is therefore desirable to perform additional XFEL experiments at higher fluences. At last, the higher repetition rate of the European XFEL might result in additional problems due to the much higher average power deposited in the zone plate.

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