Crystal optics as guard apertures for coherent x-ray diffraction imaging

Xianghui Xiao, Martin D. de Jonge, Yuncheng Zhong, Yong S. Chu, and Qun Shen

Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439

Received June 27, 2006; revised August 27, 2006; accepted August 30, 2006; posted August 31, 2006 (Doc. ID 72386); published October 11, 2006

A crucial issue in coherent x-ray diffraction imaging experiments is how to increase the signal-to-noise ratio when measuring relatively weak diffraction intensities from a nonperiodic object. A novel crystal guard aperture is described that makes use of a pair of multiple-bounce crystal optics to eliminate unwanted parasitic scattering background. This background is often produced by upstream optical elements such as a coherent-beam defining aperture. Recent experimental observation and theoretical analysis confirm the effectiveness of the crystal guard aperture method with coherence-preserved wave propagation through the crystal guard aperture and dramatically reduced scattering background in coherent x-ray diffraction images. © 2006 Optical Society of America


X-ray coherent diffraction imaging (CDI) is a recently developed microscopic technique.1 Compared with other microscopic techniques, it can image nonperiodic materials without lens optics and has the potential to reach nanometer spatial resolutions. Coherent diffraction imaging is done in two steps, first by recording a diffraction pattern from the nonperiodic specimen, using a coherent incident x-ray beam, and second by retrieving the specimen’s density function from the diffraction pattern with an iterative phasing method.3 Applications of x-ray CDI range from structural determinations of nanoparticles4,5 to reconstructions of x-ray optical wavefields5 and to high-resolution imaging of intact biological cells and other specimens.7,8

One of the main difficulties for achieving high spatial resolution using CDI is the ability to accurately measure weak diffraction intensities from a nonperiodic sample with high signal-to-noise ratios. Simulation results9 suggest that the signal-to-noise ratio needs to be at least 5 to reconstruct the original object by iterative phase-retrieval methods. Since a fully coherent x-ray beam is usually generated at existing synchrotron sources by selecting a coherent portion of an undulator x-ray beam by using a coherence-beam-defining aperture (CBDA), reduction of the unwanted parasitic scattering from the CBDA is key to achieving high signal-to-noise ratios in coherent diffraction images. To date, most x-ray diffractive imaging experiments have been done with soft x-rays,1,4,6,8 where a guard aperture (GA) is employed to remove parasitic and high-order scattering from the upstream CBDA. However, the usual guard edge or pinhole GAs have two failure modes: insufficient insertion leading to transmission of CBDA scatter or overinsertion resulting in the production of further scattering. Since the GA may itself produce scattering, sometimes additional GAs are employed, which in turn need to be positioned very carefully to minimize overall secondary scattering. This situation becomes worse for hard x-rays, since diffracted signals are further concentrated around the forward direction, and specular reflections may arise from the thicker CBDA edges that are needed at higher energies.

In this Letter, we describe a novel crystal guard aperture (CGA) that can essentially remove parasitic high orders and specular scattering from the upstream CBDA. The CGA is based on the fact that x-ray diffraction from a perfect crystal such as silicon can be used as an angular filter because x-ray reflectivity is extremely high only in a very narrow intrinsic angular width and decreases rapidly outside this narrow angular region. Although crystals are used in small-angle scattering experiments based on the original work of Bonse and Hart,10 to our knowledge, the work presented here represents the first time that crystal optics are employed to define a fully coherent x-ray beam in CDI experiments.

Our CDI experiment was performed using 8.1 keV x-rays at station 32-ID-B of the Advanced Photon Source at Argonne National Laboratory. The experimental setup is schematically shown in Fig. 1(a). A 10 μm pinhole was used as the CBDA, and an open-faced z-shaped channel-cut11 silicon (111) perfect crystal was employed as the CGA, which was located about 77 cm from the CBDA pinhole. We used a 5 μm electrodeposited lead microparticle12 as our test specimen, located 17 cm downstream from the channel-cut crystal. The coherent diffraction pattern from the specimen was collected by using a lens-coupled Roper ChemiPro charge-coupled device (CCD) area detector positioned 35 cm from the specimen. The CCD has 2048×2048 pixels, and the physical pixel size is 13 μm; with a 10× lens the effective pixel size was 1.3 μm. Polished CdW crystal was used as scintillator. We have adopted this setup to satisfy the oversampling requirement1,2 in the holographic regime, where a wavefront phase curvature can be used in the phase-retrieval process.13,14

To be useful in CDI experiments the crystal optic needs to preserve the wavefronts of the incident coherent x-ray beam. In particular, the phase curvature induced by the crystal must be quite simple and predictable. Otherwise, it may be difficult to distinguish...
between the incident illumination curvature and the phase structure of the sample. In the case of an incident plane wave, it is well known that the diffracted wave is also planar with a phase shift that depends on the angular deviation from the Bragg angle. When a coherent beam-defining aperture of a few micrometers in size is used, the wave after the aperture is not a planar wave. To evaluate the effect of crystal diffraction on this curved wave, a dynamic crystal diffraction theory\textsuperscript{15} that can be applied to curved wave-fields was used to calculate the diffracted wavefield from the crystals. The diffraction wave from a 10 μm pinhole was assumed to be the incident wave of the crystals, and double-bounce diffraction was calculated. The results of our calculation after propagating the crystal-diffraction wave to the sample plane and to the detector plane are shown in Figs. 1(b) and 1(c), respectively. By choosing the incident angle, we made the central peak of the pinhole diffraction the one being diffracted, and most of the high-order peaks were blocked.

As we expect, the phase curvature of the crystal-diffracted wave shown in Fig. 1(b) and 1(c) is very similar to that of the incident wave. The phase of a crystal-diffraction wave changes by π relative to the incident wave almost linearly as a function of the spatial coordinate across the crystal diffraction peak region. The phase shift makes the principal direction of the diffraction wave deflected by a tiny angle (~1 μrad), which would cause the whole diffraction pattern from the specimen to shift by only a small distance in the detector plane. Therefore the phase curvature after the CGA is essentially the same as that of the incident beam.

We started our experiment using the conventional CDI setup with a 10 μm pinhole as CBDA and a 25 μm pinhole as a guard aperture (PGA) to evaluate the background in the pinhole guarding arrangements. Figure 2(a) shows a typical Airy diffraction pattern from the 10 μm CBDA alone. Stray scattering from the pinhole edge is clearly visible in the high-scattering-angle region. Figure 2(b) shows a typical coherent diffraction pattern from the 5 μm lead microparticle with the CBDA-PGA setup. It is seen that the stray scattering has intensity comparable with the sample signal in the high-angle region. Although white-field correction, i.e., subtraction of the pattern without the specimen in the beam, may help to alleviate the stray-scattering problem, it is difficult to remove the stray scattering completely when it is strong.

We then proceeded to insert the channel-cut Si crystal optic into the setup, replace the PGA, tune to the vertically diffracting (111) reflection, and repeat the same measurements without and with the sample. Figure 2(c) shows the remnant of the Airy diffraction pattern from the CBDA after the crystal.
Compared with Fig. 2(a) the scatter in the vertical direction has been almost entirely eliminated by the crystal. In the horizontal direction the diffraction and scattering pass through the crystal. This could be removed by an additional horizontal-diffracting crystal or a PGA. Figure 2(d) shows the diffraction pattern of the lead microparticle sample that was used in Fig. 2(b). A 25 µm pinhole was used after the CBDA but before the crystal. The stray-scattering intensities in the radial seen in Fig. 2(b) have been entirely removed in Fig. 2(d).

Due to the existence of noise and stray scattering in the raw images, it is hard to evaluate the identity between Fig. 2(b) and 2(d) directly. One rough measurement of the identity between 2(b) and 2(d) is to calculate the cross correlation between them. Because the stray scattering in Fig. 2(b) is strong in the right-side region but weak in the left side, we used only the left half of those two images in the cross-correlation calculation, which gives a result of 0.99. This suggests that the two diffraction patterns obtained using the two different setups are almost identical. A more reliable evaluation of the quality of these images is to compare their reconstructions. This work is ongoing, and we hope to present results in a future paper.

It is known that Bragg diffraction from a crystal exhibits a diffuse-scattering streak along a direction perpendicular to the crystal surface, which is termed a crystal truncation rod. The intensity of the truncation rod in a single bounce diffraction decays as $\Delta \theta^{-2}$, where $\Delta \theta$ is the angular deviation from the exact Bragg peak. This effect can be seen upon close inspection of Fig. 2(c) and 2(d), as the weak background streaks in the vertical direction. Because we used double-bounce Bragg diffraction from a channel-cut crystal in our experiment, the truncation rod intensity is proportional to $\Delta \theta^{-4}$, which is comparable with the coherent scattered intensity from a nonperiodic sample in the small-angle regime. This problem can be easily solved by employing more than two reflections until the truncation rod intensity is reduced to a negligible level.  

In conclusion, we have shown that crystal optics can be used to remove unwanted scattering in coherent diffraction imaging experiments, overcoming the limitations of the traditional guarding scheme, which has so far presented a major problem for the application of CDI in the hard x-ray regime. Our calculation demonstrates that the phase curvature of a diffracted coherent wave from a crystal is almost identical to that of the incident coherent wave in the diffraction peak region. Our measurements demonstrate that the crystal guard aperture can indeed both preserve the incident-beam coherence and greatly reduce the stray-scattering background in coherent hard x-ray diffraction imaging experiments. This new method may also play an important role in the future developments of other coherent scattering studies such as proposed movable-aperture lensless transmission microscopy. We hope that the more effective crystal aperture will also generate interest in the rapidly evolving research area of coherent diffraction optics using coherent hard x-rays.

We are grateful to Hanfei Yan (Advanced Photon Source) for fruitful discussions on crystal diffraction theory. The use of the Advanced Photon Source is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under contract W-31-109-ENG-38. X. Xiao’s e-mail address is xhxiao@aps.anl.gov.

References

