



Coherent Diffraction Imaging (CDI): Bragg CDI

Monography: FI089

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1 Introduction

Coherent X-ray Diffraction Imaging (CXDI) is a lensless imaging technique, where the object (sample) is illuminated by a coherent laser-like beam and then the scattered light is collected by a detector. The image of the object can be recovered with an advanced phase retrieval algorithm. The object can be reconstructed since the incident wave is coherent and then the diffracted wave is proportional to the Fourier transform of the object. Because of the large coherent fraction needed (> 70%) CXDI methods were recently applied at FEL facilities (fully coherent). But CXDI experiments are planned for the first 4th generation synchrotrons, such as SIRIUS, where the coherent fraction reaches the required level. Common CXDI methods developed are: plane-wave CXDI, Bragg CXDI, Fresnel CXDI and ptychographic CXDI. The result of the CXDI experiment is a 2D image of the sample. In order to obtain a 3D image, the sample need to be rotated and a 2D image is acquired at each sample orientation. This work presents in more detail the Bragg CXDI and the plane wave CXDI.

2 CXDI methods [7]

2.1 Plane-wave CXDI



Figure 1: Concept of plane-wave CXDI [6]

The object is illuminated by a plane wavefront and the direct beam is blocked on the detector by a beam stop. Plane-wave CXDI is insentitive to sample drift and vibration and achieves therefore a spatial resolution of down to 2 nm (2D).

The SIRIUS beamline Caterete is designed for using plane-wave CXDI. The beamline optics consists of a double channel-cut monochromator and two side deflecting mirrors accepting only the coherent part of the beam. The horizontal focusing mirror has a magnification of approximately 1:1 and the vertical focusing mirror demagnify the beam by a factor of 2. The depth of focus in this geometry amounts a few meters and in consequence it supplies a plane wave at focal position. The beam size at focus position is about 40 μ m (h) x 30 μ m (v) with a coherent fraction of greater than 70% and a coherent flux in the order of 10¹³ Ph/s/100mA.



Figure 2: Concept of Bragg CXDI [6]

In Bragg CXDI a nanocrystal is illuminated by a coherent wavefront, and the diffraction pattern surrounding the Bragg peak is measured. The phase of the complex 3D image, obtained from inversion of the diffraction pattern, is related to displacement and strain field of the crystal lattice.

At SIRIUS, the CARNAÚBA beamline is prepared to use Bragg CXDI at NanoProbe endstation. The beam in NanoProbe focus is almost diffraction limited and amounts approximately 120 nm at 2 keV and 30 nm at 14 keV.

2.3 Fresnel CXDI



Figure 3: Concept of Fresnel CXDI [6]

In Fresnel CXDI, the sample is illuminated by a curved wavefront created by a Fresnel Zone Plate (FZP). The sample is positioned in front of or behind the focal spot of the FZP. The Fresnel diffraction pattern I smeasured by the detector. Because of the curved wavefront the phase retrieval algoritm converges faster. The resolution is higher compared to Ptychographic and Bragg CXDI.





2.4 Ptychographic CXDI



Figure 4: Concept of Ptychographic CXDI [6]

In ptychographic (scanning) CXDI the extended sample (object) is scanned on a 2D grid over the nano beam (or small aperture, which chooses only the coherent part of the beam). The complete 2D image is then constructed from the diffraction patterns of partially overlapping regions. The sample can be scanned either in reflection or transmition mode with sub-nm resolution. The iterative phase retrieval algorithm is further used not only to solve the complex amplitude of the object, but also of the incoming beam (probe). With this it is possible to corrent for errors of the scanning stage.

3 Image reconstruction

The iterative phase retrieval algorithm (IPRA) is used for image reconstruction in CXDI. The algorithm consists of four steps and requires iterating between the real and reziprocal space. It starts with a random guess of the initial phase.



Figure 5: Iterative phase retrieval algorithm





Applying an inverse FFT on the measured data set together with the estimated phase gives an initial image. Based on the oversampling of the diffraction intensity a boundary slightly larger than the sample envelope can be estimated from the initial image. The electron density outside this boundary is reduced, while the negative electron density inside is modified. This is the second step. In the third step this a FFT is applied to the modified image, which generates a new Fourier transform. In the last step the magnitude of the new Fourier transform is replaced with the measured data. This will give a better estimate of the Fourier transform and can be used for the next iteration.

4 Bragg CXDI

4.1 Example 1: 3D Bragg CXDI on an extended ZnO crystal

In this work the strain field of ZnO tetrapod crystal samples are measured and characterized. ZnO is widely used as industrial material for piezoelectric and pyroelectric applications.

The here described experiment was performed at the APS beamline 34-ID-C. The ZnO sample was located in the focus of a KB mirror system illuminated by a 1.5 µm x 1.5 µm beam of 11.5 keV coherent radiation. The vertical coherence length maintained from the secondary source slits amounts 72 µm. The beam in the focus is then considered fully coherent in vertical direction. The horizontal coherence length amounts 330 nm, which is slightly smaller than the sample width. The longitudinal coherence length was measured to be 660 nm. Because of the small horizontal coherence length, the assumption of fully coherent illumination is not correct and an algoritm for the data reconstruction, which handles partial coherence need to be implemented. The ZnO tetrapod sample was mounted in the rotation center of the diffractometer. The 3D Bragg diffraction pattern (around the 101 Bragg peak) was collected by rotating the sample from -0.25° to 0.25° in 0.01° step size along the vertical direction. At each point 50 measurements with 0.1 s exposure time were accumulated. In order to map the 3D diffraction data over the long dimension of the ZnO tetrapod crystal the sample was translated along the vertical direction in six steps with a step size less than 0.5 µm and at each point the 3D Bragg diffraction pattern was acquired as described above. This allows the analysis of strain field changes along the ZnO crystal. The scanning method combines the Ptychographic with the Bragg CXDI.

The data set of each ptychographical position is reconstructed indivially using regular phase retrieval algorithm. As individual reconstructed images are a product of the object (sample section) and probe (complex incident beam), they can give a good estimation of the length for the illuminated section and each scan position. The X-ray beam intersects most in center of the ZnO tripod arm and moves off it at the two ends. This calculated section length can be used to compensate for the positioning accuracy of the vertical step motor at each individual step. The product of object and probe ($\Psi_i = O_i P$) from the individual reconstruction process can replace the product-updating process in the ptychographic algorithm because of its equilence. The first reconstruction step assumes a plane wave illumination and a random guess of the object. The n-th step is updated as

$$P'_{n+1} = \sum_{i} O^*_{n,i} \Psi_{n,i} / \sum_{i} |O_{n,i}|^2$$





where i denotes the i-th scan spot and * the complex conjugate. As the sample thickness in this experiment was smaller than the depth of focus of the KB mirrors, the incident beam can be approximated as 2D wavefront and then the variation through the sample is negligible. P'is then converted to a 2D probe (incident beam).

$$P_{n+1}(x, y) = \frac{\sum_{z} P'_{n+1}(x, y, z) |P'_{n+1}(x, y, z)|^2}{\sum_{z} |P'_{n+1}(x, y, z)|^2}$$

The 3D Probe P_{n+1} is then recreated by setting each frame in z-direction as P_{n+1} . The next step is updating the object as:

$$O_{n+1} = \sum_{i} P_{n+1}^* \Psi_{n,i} / \sum_{i} |P_{n+1}|^2$$

The reconstruction process applied in this work differs from original ptychographic algorithm in this way that the product of object and probe is updated from independent reconstructions of individual data sets.

The size of the ZnO arm obtained from the reconstructed image was 2.57 μ m (length) x 0.37 μ m (width), which is consistent with the before measured SEM images. The obtained magnitude in the central cut plane (figure 6b) is smoother than in a previous study [4] performed with regular Bragg CXDI. The combination of ptychographic and Bragg CXDI removes artefacts by the beam and improves the magnitude fluctuations at the sample edges. In the same image one also observes "hot spots", which might be caused by incomplete recovery of the probe in the horizontal direction (remember the horizontal coherence length is slightly smaller than the sample width).



Figure 6: Ptychographic reconstructed magnitude and phase distributions of the ZnO arm

The small phase range of ± 0.6 radians indicates that the ZnO arm is only slightly strained. The strongest phases (strain) are observed on bottom of the arm, which is caused by mismatch of the lattice near the interfaces.

The CXDI phase retrieval algorithm assumes fully coherent illumination, but in this work a partially coherent bema from a 3rd generation synchrotron was used. In consequence,





the partial coherent illumination causes artefacts in the reconstructed image, which could be mitigated by a newly developed method implementing partial coherence analysis within the reconstruction process [5]. Using the redundant information from overlapping scans (ptychographic CXDI) the incident X-ray beam can be recovered and characterized. In this way the verical focus was found 4.04 mm downstream the sample plane.

4.2 Example 1: Bragg CXDI of epitaxial nanostructures using focused hard X-ray ptychography

This work shows the measurement of coherent Bragg volumes measured from an individual β - and δ -Bi₂O₃ epitaxial nanocrystal epitaxially grown on single-crystal oxide substrate using Bragg CXDI. The experiment was performed at the APS NanoProbe beamline (CNM) and aimed to characterize the local strain state of the growth behavior of the Bi₂O₃ β - and δ -phases. Such nanostructures are good oxygen conductors that are stable in bulk only under limited environmental conditions.

The sample was prepared by growth (CVD) of mixed $Bi_2O_3 \beta$ - and δ - pyramidal nanostructures on single-crystal oxide substrate (STO). The edges of the pyramids are parallel to STO in-plane [100] directions, which are parallel to Bi₂O₃ [110] directions. A 500 µm x 500 µm Ni grid was patterned on the substrate by oiptical laser lithography and sputter desposition in order to better define and find the nanostructure regions of interest. In the experiment the sample was illuminated by a 90 nm beam of 10.4 keV X-rays in the focus of a zone plate. The position of the beam can be maintained with an accuracy of 5 nm during the measurements, also due to simultaneously performed fluorescence mapping (imaging by constrast modes). The substrate was oriented with 12.4° incidence angle to satisfy the (002) Bragg condition for the β - and δ -Bi₂O₃, where the similarity of the c lattice and the 0.15° divergence of the focused beam allows the (002) diffraction of both phases to be seen simultaneously with a CCD camera. In contrary to the previous example (chapter 4.1.) there is no rocking around the Bragg peak necessary. A raster scan over the sample combined with a fluorescence map identify individual nanocrystals to be measured. The phase specific map identifies and distinguishes both phases at individual nanocrystals on the substrate. At this experiment only the nanocrystals with β -phase were measured by Bragg CXDI.



Figure 7: Reconstructed Bragg volume b) in focus and c) 225 nm upstream the focus





The pyramidal nanocrystal has a size of abou 1-1.5 μ m. With the 90 nm X-ray beam the nanocrystal is only partly illuminated and therefore the reconstructed Bragg volume not only depends on crystal shape and strain, but also on position and defocus of the incident beam. Figure 7c) with the focus 225 nm upstream the sample shows more dramatic changes of the diffracted speckles depending on beam position on the nanocrystal. To overcome this problem, the diffraction images were acquired at finely spaced angular increments along the crystal's rocking curve (2 θ fixed, θ with 1.5° range and 0.006° steps, starting from 11.5°). The crystal Bragg volume was reconstructed from the diffraction images by using a ptychographical algorithm. The magnet hutch endstation at the EMA beamline at the new 4th generation synchrotron SIRIUS can improve drastically this limitation of the small focused beam size. The focused coherent beam at the magnet hutch endstation amounts 1.2 μ m x 0.4 μ m at 10 keV radiation and therefore would cover almost the full nanocrystal.

4.3 Example 3: 3D reconstruction of size and shape of protein microcrystals by using Bragg CXDI

The aim of this work was to improve the quality of 3D protein structure of a Lysozyme crystal reconstructed from the diffraction images and to better understand the fundamental process that drives radiation damage. The results were validated against RSM and transmission electron microscopy.

The here described experiment was also performed at the APS beamline 34-ID-C. The crystals, mounted on a sample stage on a goniometer able to rotate over all three diffraction angles, were illuminated with 9 keV X-rays with a focal beam size of 1.7 μ m x 1.3 μ m FWHM (focus of a KB mirror system). The data were collected by a Timepix (Medipix family) photon counting detector located 1.4 – 1.77 m from the sample. A closer distance was used to align the crystal (sample) and to find the right Bragg peak. As usual in Bragg CXDI the diffraction images were acquired rocking with fine steps (0.01°) around the Bragg peak (0.3° range). The Bragg angle for the Lysozyme crystals are located between 1° and 5°. The Lysozyme crystals were prepared in a similary way as for protein crystallography.

The crystal size was determined by TEM mesurments and amounts 1.2 μ m x 1.2 μ m, which fits within the requirements for Bragg CXDI experiments (fully coherent beam larger than sample, oversampling). The here used reconstruction algorithm is similar to the ones used in the other two examples and combines error reduction (ER) algorithm with the standard phase retrieval algorithm. In the diffraction image the crystal size and shape are related to structures away from the center of the Bragg peak. And strain in the crystal lattice reflects in asymmetry of crystals' electron density. The phase maps recoverd from the crystal size as result of the Bragg CXDI measurement was 1.1 μ m x 0.8 μ m x 1.8 μ m ±0.2 μ m, which is consistent for the 2D-size measured by TEM. The radiation dose applied to the crystals after 10 rocking curves was 652.8MGy. Reconstructing the crystal volume after each step relates the applied dose with changes in shape and size of the crystal (radiation damage). In the experiment after the 10 rocking curves, the Bragg peak had faded to 2.5% of the initial total integrated intensity.



Figure 8: Measurment and reconstruction of the Lysozyme cristal. a) measured Bragg peak in reziprocal space, b) and d) reconstructed volume of the crystal in different orientation (containing phase information from reconstruction), c) and e) slide related to b) and d)

Errors in reconstructions and related artefacts (sligh phase gradients at the crystal edges) in the reconstructed volume are caused by assuming a plane wave illumination of the sample. But indeed, such artefacts can be caused by a curved wave. The artefacts could be minimized by recovering the incident focused beam profile as done in the first example (chapter 4.1.), which was done at the same beamline.

5 Conclusions

Bragg volume and strain distributions can be investigated by Bragg CXDI. The SIRIUS beamlines CARNAUBA and EMA will use this technique and the focused coherent beam is larger than in the here described studies. The homogenous coherent illumination of the nanostructures will reduce the measurement and reconstruction time and minimize the artefacts discussed in the three examples.

Different problems related to a curved incident wavefront can be overcome by the new CATERETE [9] beamline at SIRIUS synchrotron. There, plane wave CXDI can be used to quantify the 3D morphology and structure of internal cellular organells in cells, bacteria or even viruses. Also, the protein structure of a small virus like the HIV virus (~150 nm) might be solved at CATERETE beamline without the need for crystallization, if the resolution can be improved approximately to 1 nm. In that case the radiation damage will be drastically reduced due to short illumination.





6 References

- [1] X. Huang, Three-dimensional Bragg coherent diffraction imaging of an extended ZnO crystal, Journal of applied crystallography 45, 778- (2012)
- [2] S.O. Hruszkewycz, Bragg coherent diffraction imaging of epitaxial nanostructures using focused hard X-ray ptychography, AIP conference proceedings 1365, 235 (2011)
- [3] H.D. Coughlen, Three-dimensional reconstruction of the size and shape of protein microcrystals using Bragg coherent diffraction imaging, Journal of optics 18, 054003 (2016)
- [4] G. Xiong, Coherent X-ray diffraction imaging of ZnO nanostructures under confined illumination, New journal of physics 13, 033006 (2011)
- [5] J.N. Clark, High-resolution three-dimensional partially coherent diffraction imaging, Nature communications 3, 993 (2012)
- [6] E. Welch, Coherent diffraction imaging (lensless imaging), SLAC, presentation, Aug. 2015
- [7] J. Miao, Beyond crystallography: Diffractive imaging using coherent X-ray light sources, Science 348, 6234 530- (2015)
- [8] EMA beamline PDR report, CNPEM (2017)
- [9] CATERTE beamline PDR report, CNPEM (2017)
- [10] CARNAUBA beamline PDR report, CNPEM (2017)