Nanostructure analysis by coherent hard X-ray diffraction

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Abstract. We report our recent activities on x-ray diffraction microscopy. We have been developing hardware and software instruments for efficient recording of high-quality coherent diffraction data. By using the microscope system developed, we carried out coherent diffraction measurement for various samples in materials science and biology, and succeeded in three-dimensional reconstruction revealing the internal structure.

1. Introduction

X-ray diffraction microscopy is an innovative structural analysis method in nanoscience. In the method, the sample structure is reconstructed directly from the oversampled coherent diffraction data by using iterative phase retrieval methods. The reconstruction does not require a priori knowledge on the sample structure nor supplemental experimental data. Lenses are not necessary for the microscope, whence diffraction-limited resolution can be achieved in principle. The lensless feature is significant especially for hard x-rays, where it is difficult to fabricate a lens with a high numerical aperture. High penetration power of x-rays offers the unique possibility to achieve high-spatial-resolution imaging for relatively thick samples in a non-destructive manner, and can provide complementary information to other conventional microscopic methods, such as transmission electron microscopy and fluorescence microscopy.

We have been performing diffraction microscopy experiments by using hard x-rays from SPring-8 [1]. In this paper, we describe recent development of our microscope instruments and experimental results of three-dimensional reconstruction.

2. X-ray diffraction microscopy

2.1. Microscope instruments

Figure 1 shows the coherent diffraction microscope we developed. Synchrotron undulator radiation illuminates a sample particle, and the coherent diffraction patterns are recorded using a CCD detector.
covering a small-angle region around the forward scattering direction. In our measurement, the sample size is typically around 1 µm and is smaller than the transverse coherence area of the incident x-ray wave. Since coherent diffraction signals from a single micron-sized sample particle are weak, all optical components are put in a high vacuum in order to avoid the air scattering noise. A pinhole is installed about 1 m upstream of the sample to illuminate the targeted sample particle only even at the highest incident angle [2]. Guard slits are used to suppress parasitic scatterings from the pinhole and thus to allow high-quality data acquisition.

One of newly introduced features of our instruments is a CCD manipulator. The CCD manipulator can change the distance from the sample to the detector, $L$, by 700 mm. It helps efficient measurement of samples of different sizes at different incident x-ray energies. The change of the sample-to-detector distance is required to achieve a certain oversampling ratio, the number of the detector pixels in a single speckle, because the speckle size is approximately given by $aL/\lambda$. Here, $a$ is the sample size, and $\lambda$ is the x-ray wavelength, and we assumed that the sample size is smaller than the illuminating coherent beam size. This new feature is beneficial for e.g., element specific imaging using the anomalous scattering, where the incident x-ray energy is tuned near the absorption edge energy of a specific atom [3,4]. The CCD manipulator also helps to move the CCD detector to higher scattering angles to achieve higher spatial resolution.

We also developed semi-automated data acquisition software for efficient three-dimensional measurement at different incident angles. After each change of the incident angle, the software can automatically optimize the sample position to maximize the coherent diffraction intensity, and then start acquiring data. Although the sample is placed approximately on the rotation axis, the repeated sample position alignment helps to compensate the drifts of optics and the x-ray source. Careful alignment of optics is critical in recording high-quality data and in reducing the size of missing data near the forward scattering direction behind the beamstop. During the beamtime, we frequently check the quality of the measured data by manually reconstructing two-dimensional images.

**Figure 1.** X-ray diffraction microscope installed at BL29XUL in SPring-8.

### 2.2. Three-dimensional observation of nano-scale internal structures

Our microscope system allows us the acquisition of high-quality coherent diffraction data with a small missing-data size near the forward scattering direction [5], which is essential for faithful reconstruction of the sample structure. Three-dimensional structure can be reconstructed form a series of coherent diffraction data at different incident angles. In the first Born approximation, the three-dimensional reconstruction provides us an electron density map. In the following, we briefly describe two successful examples of our recent three-dimensional reconstructions of samples in materials science and biology.

In materials science application, we observed an aluminum alloy particle [6]. The sample 2014 aluminum alloy is known to contain submicron-sized S-phase ($\text{Al}_2\text{CuMg}$) precipitates, which play a
key role in controlling the hardness of the alloy. The values of the electron densities in literature show that the precipitate has 25% higher electron density than the aluminum matrix does. In our three-dimensional reconstruction, we observed an internal high-electron-density region inside a \(1 \mu m\) sized sample particle, and the high density indicates the region is rich in precipitates.

In biological application, we observed an unstained human chromosome [7]. Figure 2 shows the coherent diffraction pattern from a single chromosome. We obtained high-quality coherent diffraction data at 38 incident angles, and succeeded in reconstructing its three-dimensional structure. This is, to our knowledge, the first successful three-dimensional electron density mapping of a cell organelle by using hard x-rays. The two- and three-dimensional reconstruction shows that high electron density structure extends around the axes of the chromatids. The sample in the experiment was dried in air and is not in biologically ideal condition. We are now developing instruments to observe frozen-hydrated biological samples which are closer to the natural living state.

![Figure 2](image-url)

**Figure 2.** Coherent diffraction pattern from a single unstained human chromosome.

3. Summary

We developed coherent x-ray diffraction microscope system that allows us to record high-quality coherent diffraction data with a small size of the missing data near the forward scattering direction. The recently introduced CCD manipulator is effective especially in the measurement of samples of various sizes at various energies. With the X-ray diffraction microscope hardware together with the semi-automated control software, it became possible to perform three-dimensional data acquisition efficiently. As examples of three-dimensional observation using the microscope, we described successful results for an aluminum alloy and a human chromosome.

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