Towards Atomic Resolution 3-D X-ray Diffraction Microscopy

Due to the fact that X-ray wavelengths are on the order of the size of atoms, scientists have long dreamed of atomic resolution X-ray microscopes which could visualize arrangement of atoms in three dimensions. However, X-rays are much more difficult to focus than electrons. The smallest X-ray focal spot currently achievable is around 30 nm [1].

This limitation can be overcome by using coherent X-ray diffraction and the oversampling phasing method. When a finite specimen is illuminated by coherent X-rays, the weakly scattered X-ray photons form a continuous diffraction pattern in the far field. This continuous pattern can be sampled at spacings finer than the Nyquist frequency (*i.e.* the inverse of the specimen size), which we call "oversampling". Oversampling a diffraction pattern corresponds to surrounding the electron density of the specimen with a no-density region. The higher the sampling frequency, the larger the no-density region. When the no-density region is larger than the electron density region, the phase information is in principle available from the diffraction pattern itself and can be directly retrieved by using an iterative algorithm [2]. The first demonstration experiment of this form of microscopy was carried out by using coherent soft X-rays in 1999 [3]. More recently, it has been extended to image the shapes of nano-crystals by using hard X-rays [4]. However, the experiments that have been carried out thus far have been limited to imaging 2-D samples, and the highest resolution achieved to date is around 70 nm [3,4].

Here, we describe the first experiment to image the 3-D structure of a non-crystalline material at 50-nm resolution, which was reported in Phys. Rev. Lett. [5]. The experiment was carried out at the undulator beamline **BL29XU**. Figure 1 shows a schematic layout of the experimental instrument in which all the components are in vacuum with a pressure of ~10^{-6} torr. The sample, fabricated by electron beam lithography, consists of two single-layered Ni patterns (each with a size of 2.5 × 2 × 0.1 µm) rotated 65° relative to each other in-plane and separated by a distance of 1 µm. The sample is supported by a thin silicon nitride membrane window. Figure 2(a) shows a scanning electron microscopy (SEM) image of the sample. Due to the 1 µm separation of the two layers, the SEM image shows the pattern in the top layer, and the pattern in the bottom layer is visible only as a soft blur.

Figure 2(b) shows a 2-D diffraction pattern at a resolution of 8 nm recorded from the sample by using coherent X-rays with a wavelength of 2 Å. The total exposure time of the diffraction pattern is about 45 minutes using unfocused X-rays from the undulator beamline. By using the oversampling phasing method, the diffraction pattern was directly converted to the high-resolution image shown in Fig. 2(c). The top and bottom layered patterns are clearly seen as overlapped in this 2-D image projection, and the variation of the electron density at the nanometer scale is also visible. To obtain the 3-D structural information, a series of thirty-one 2-D

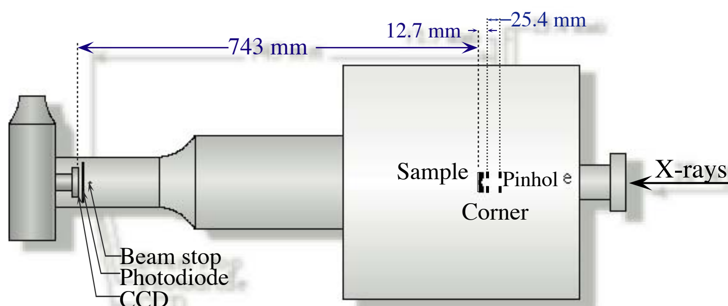

Fig. 1. Schematic layout of the experimental instrument used to record the coherent X-ray diffraction patterns.

diffraction patterns were recorded from the sample with the rotation angles ranging from –75° to 75° in 5° increments. The 2-D diffraction patterns were then assembled to produce a 3-D diffraction pattern. By using a 3-D phase retrieval algorithm [5], the 3-D structure of the non-crystalline material was successfully reconstructed at a resolution of 50 nm. Figure 2(d) shows a 3-D iso-surface rendering of the reconstructed image. The finest division in the z-axis corresponds to 25 nm and the distance between two patterns is about 1 μm, which is consistent with the known characteristics of the sample.

We anticipate that this form of microscopy will have wide applications in both materials and biological sciences. For materials science samples, which are less sensitive to radiation damage, this form of microscopy can, in principle, achieve atomic resolution in three dimensions. In biology, this form of microscopy can be applied to image the 3-D structures of whole cells, cellular organelles and supramolecular structures at high resolution, while the resolution will be mainly limited by radiation damage to the specimens.

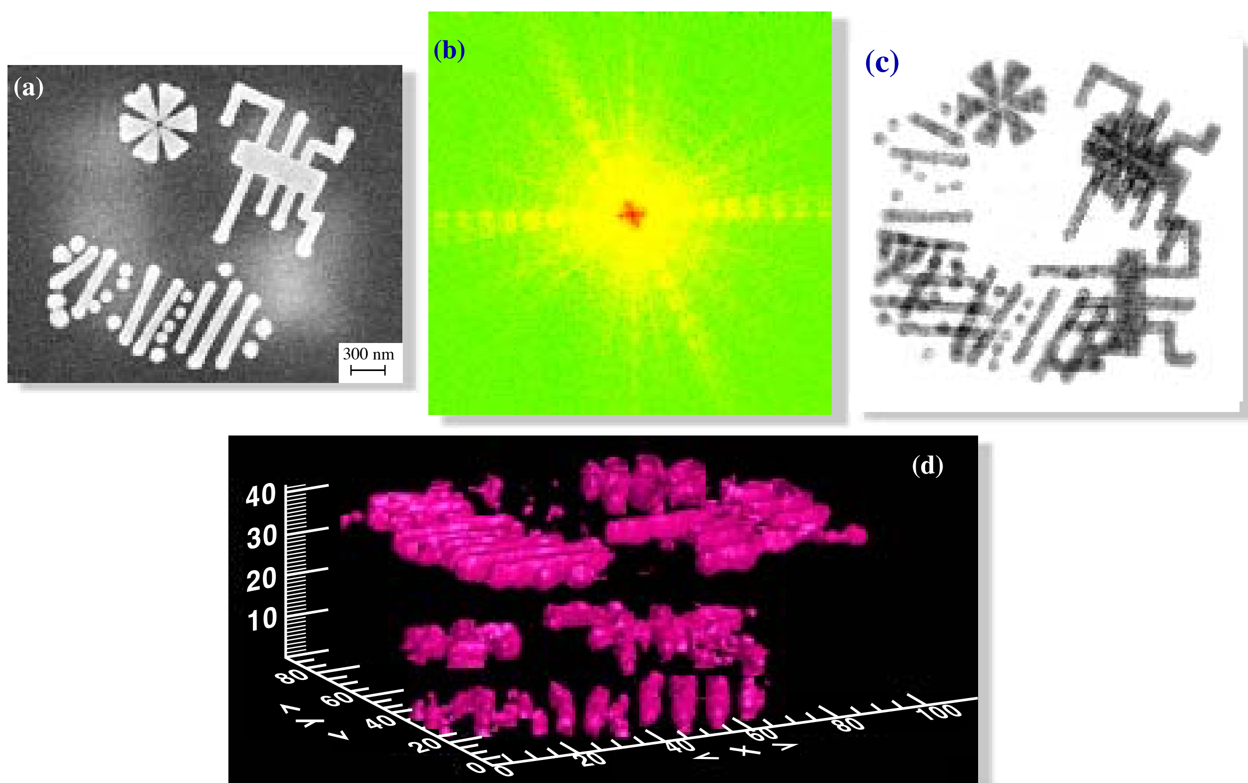

*Fig. 2. **(a)** An SEM image of an Ni sample with buried structures. **(b)** A high-resolution diffraction pattern recorded from the sample. **(c)** A high-resolution image reconstructed from **(b)**. **(d)** The reconstruction of a 3-D nanostructured material at 50 nm resolution (displayed in iso-surface rendering). [Reproduction from ref. 5.]*

Jianwei Miao[a], Tetsuya Ishikawa[b] and Keith O. Hodgson[a,c]

(a) Stanford Synch. Rad. Lab., Stanford Univ., USA
(b) SPring-8 / RIKEN
(c) Stanford University, USA

E-mail: Miao@SLAC.Stanford.EDU

References

[1] J. Kirz *et al.*, Q. Rev. Biophys. **28** (1995) 1.
[2] J. Miao *et al.*, J. Opt. Soc. Am. A **15** (1998) 1662.
[3] J. Miao *et al.*, Nature **400** (1999) 342.
[4] I. K. Robinson *et al.*, Phys. Rev. Lett. **87** (2001) 195505.
[5] J. Miao, T. Ishikawa, B. Johnson, E. H. Anderson, B. Lai and K. O. Hodgson, Phys. Rev. Lett. **89** (2002) 088303.