Reconstruction of 3-D Coherent X-ray Diffraction Patterns

G. J. Williams,[1] M. A. Pfeifer,[1] I. K. Robinson[2]
[1]Department of Physics and [2]Materials Research Laboratory (MRL),
University of Illinois at Champaign-Urbana (UIUC), Urbana, IL, U.S.A.

Introduction

In the special case of diffraction where the coherence volume of the incoming x-ray beam is larger than the spatial extent of the diffracting object, the intensity distribution around each Bragg point is the Fourier transform of the object. In order to reconstruct the real space density distribution, we must find the relative phases of the diffracted photons. We solve this "phase problem" by utilizing the redundant information in our intensity measurement in an iterative fitting routine.

Methods and Materials

The samples began with 100-nm-thick Au films grown on a Si(100) substrate. Individual samples were baked in air at various temperatures between 850° and 1050°C for periods of about 20 h. Some of the samples baked at 1050°C were then annealed at lower temperatures, between 700° and 850°C.

The result of this procedure was the formation of micrometer-sized single crystals. Scanning electron microscopy (SEM) images after preparation are shown in Fig. 1. The polyhedral shapes are expected if the crystals are in equilibrium with their vapor. These shapes can be predicted by using the Wulff construction, which relies on minimization of free energy with the constraint that the total volume of the crystal is conserved. The crystallographic faces with the lowest free energy have the largest area, and their centers are closest to the origin of the crystal.

We conducted the diffraction experiment at the University-National Laboratory-Industry Collaborative Access Team (UNI-CAT) 33-ID beamline at the APS. A direct-read charge-coupled device (CCD) array was mounted on the detector arm of a four-circle diffractometer.

For our experiment, the detector arm of the diffractometer was extended to satisfy the far field condition. A pair of custom slits was used to isolate a single crystal in the beam. The coherence length of the beam, defined by beamline parameters, was a few micrometers. At the selected energy of 10 keV, it was expected that the penetration depth in Au would be large enough to allow us to diffract from the entirety of a single crystal.

Results

The 3-D coherent x-ray diffraction (CXD) pattern was captured by rocking the incident angle of the diffractometer while capturing images. This resulted in a series of images containing nearly parallel slices through the 3-D pattern. Each of these 2-D images corresponded to the density projection of the crystal onto a plane described by the position of the incident angle at which it was captured.

Even before the inversion of the data in Fig. 2, we can recover some information from the characteristics of the pattern. For example, the flares are caused by the faces of the crystal, while the modulation of the flares is caused by interference between these faces.

In 2-D, we can only recover the projection of the crystal's density [1]; however, if we can invert a full 3-D

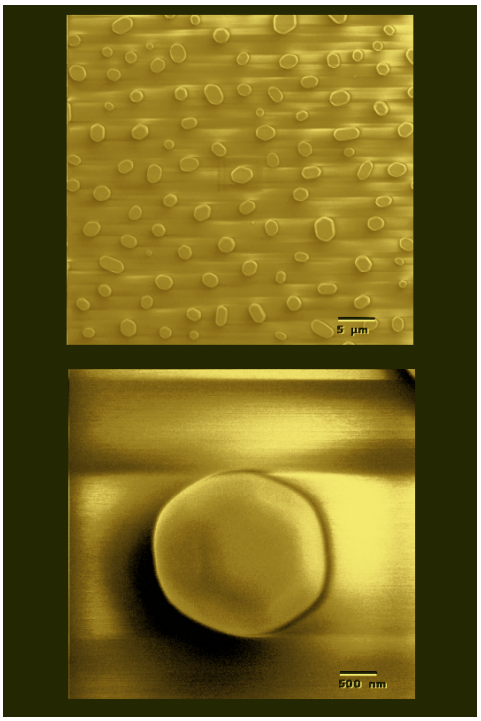

FIG. 1. SEM images of small Au single crystals. Samples are prepared by heating in air at 1000℃. The crystals are small enough to illuminate coherently.

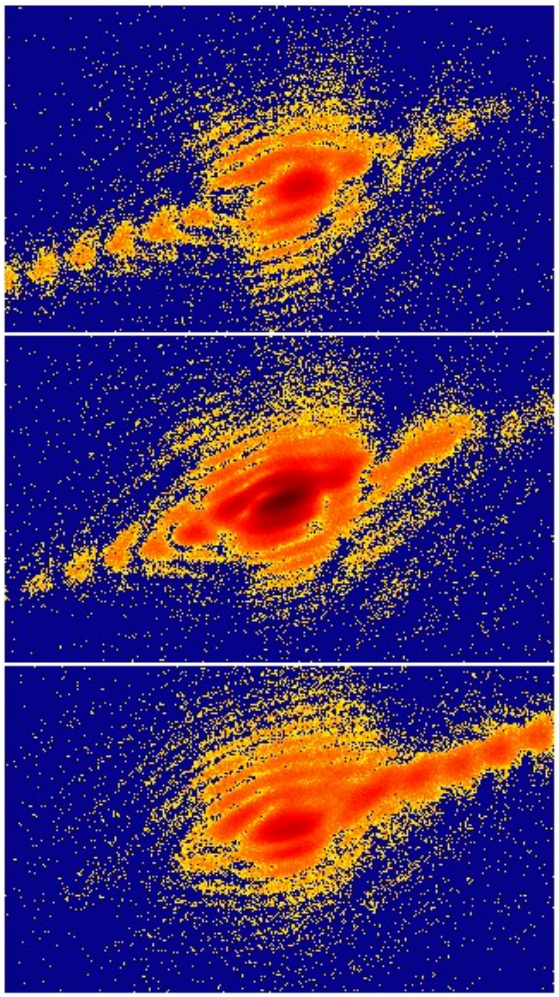

FIG. 2. These are 2-D slices through the 3-D CXD pattern. Many such slices are collected and stacked to recover the full 3-D pattern prior to inversion.

image, we can recover the *shape* of the crystal. By stacking our collected 2-D images, we form the necessary 3-D image, which is used in an iterative fitting method. Part of such a series is shown in Fig. 2. Slices through the reconstructed real space object are shown in Fig. 3. The error in the 3-D calculation is less than 20% per point.

Discussion

We have successfully inverted 3-D CXD patterns from micrometer-sized structures. Some features of our reconstructions are not fully understood, and, although we have arrived at a consistent crystal shape, we have not achieved point to point uniqueness in our reconstructed crystal. It is our hope that in future experiments, we will refine the collection of 3-D data and thereby improve our reconstructions.

This technique offers the possibility of nonintrusively imaging small crystalline objects, including domains in ceramics and buried interfaces. In fact, one can show that the CXD pattern from a strained crystal will be slightly deformed, allowing the possibility to image strain within such a structure.

Acknowledgments

Use of the APS was supported by the U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. The UNI-CAT facility is supported by the UIUC MRL (DOE, State of Illinois Board of Higher Education, Higher Education Cooperation Act [IBHE-HECA], and National Science Foundation), Oak Ridge National Laboratory (DOE under contract with UT-Battelle, LLC), National Institute of Standards and Technology (U.S. Department of Commerce), and UOP LLC.

Reference

[1] I. K. Robinson, I. A. Vartanyants, G. J. Williams, M. A. Pfeifer, and J. A. Pitney, Phys. Rev. Lett. **87**, 195505 (2001).

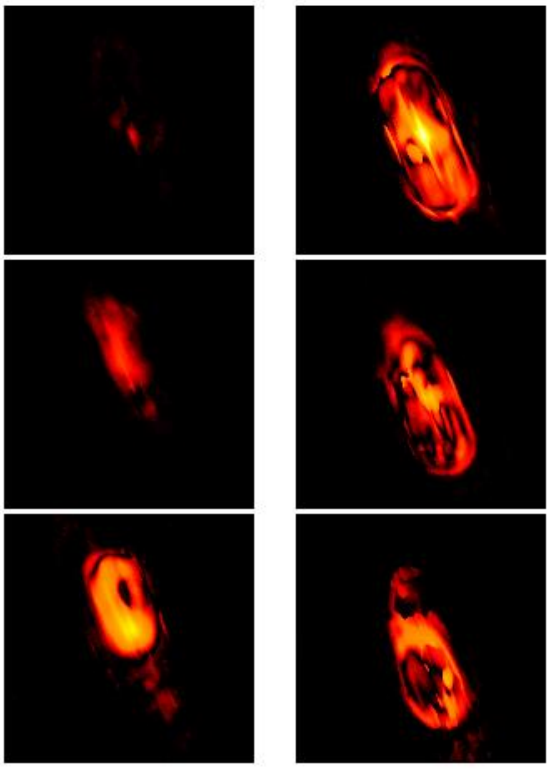

FIG. 3. These are 2-D slices through the reconstructed 3-D diffracting crystal. Artifacts due the partial coherence of the beam are believed to be present.