

# X-ray Imaging using Ultra-Small-Angle X-ray Scattering

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## Introduction

The analysis of small-angle scattering data provides quantitative volume-averaged information on materials microstructure. This analysis, however, requires *a priori* information on the shapes of the scattering objects within the sample. As a further complication, scattering from sample surfaces and double Bragg diffraction can introduce unanticipated scattering intensity that would invalidate the analysis. In addition, scattering data provides little information on the spatial arrangement of the scattering objects. We present here a new x-ray imaging technique, Ultra-small-angle X-ray Scattering (USAXS) imaging, that can provide information on the sizes, shapes, and three-dimensional arrangement of the scatterers. It also reveals the existence of double Bragg diffraction or surface scattering when those are present.

## Materials and Methods

USAXS imaging experiments were conducted using the UNICAT sector 33 USAXS instrument at the Advanced Photon Source.<sup>1</sup> This Bonse-Hart-type instrument is installed after a double-crystal monochromator (for photon energy selection) and two mirrors (for harmonic rejection). The size of the x-ray beam is controlled by incident beam slits, after which a six-reflection crystal

pair acts as a collimator. An ion chamber measures the photon flux incident on the sample (of the order of  $5 \times 10^{12}$  photons  $s^{-1}$   $mm^{-2}$ ). A second crystal pair (the analyzer) selects the scattering vector  $q$ . A USAXS scan is measured by rotating the analyzer and recording the scattered photons received by a photodiode detector.

Using the same instrument, USAXS images can be recorded by rotating the (nondispersive) analyzer to a selected  $q$  and replacing the photodiode detector with an imager (either an x-ray video camera or a nuclear emulsion plate). The x-rays that form the image are those produced by USAXS at this  $q$ . Small-angle X-ray cameras record scattering data over the entire  $q$ -range simultaneously and therefore cannot be used for USAXS imaging. USAXS imaging differs from diffraction-enhanced imaging (DEI) techniques in that USAXS images are recorded decades below the rocking curve of the analyzer crystals, whereas DEI images are recorded near the full-width at half-maximum of the analyzer rocking curve.<sup>2</sup> USAXS imaging also provides the capability of measuring the  $q$ -dependence of the scattering from individual objects within the sample, allowing size and shape information to be extracted. Finally, since rotating the sample about  $q$  does not affect the scattering contrast, such rotations can be used to produce stereo pairs.

The limitations on the spatial resolution of USAXS images are different in the directions perpendicular (defining  $q_{\perp}$ ) and parallel (defining  $q_{\parallel}$ ) to the analyzer crystals. In the  $q_{\perp}$  direction, the analyzer crystals have an angular acceptance determined by their rocking curve. For Si  $\langle 111 \rangle$  crystals and a photon energy of 8.94 keV, the half-width at half-maximum (HWHM) is approximately  $0.0007^{\circ}$ . The spatial resolution is proportional to the angular acceptance. For a sample-to-detector distance of 24 cm, the HWHM on the imager would be  $2.9 \mu m$ . The smallest sample-to-detector distance we have used so far is 5.8 cm, providing a HWHM resolution of  $0.7 \mu m$ . In the  $q_{\parallel}$  direction, the angular acceptance is determined primarily by the  $q = (q_{\perp}^2 + q_{\parallel}^2)^{1/2}$  dependence of the scattering. For spherical objects in the Porod-scattering regime, the  $q_{\parallel}$  angular acceptance at low  $q_{\perp}$  (around  $2 \times 10^{-4} \text{ \AA}^{-1}$ ) is comparable to the  $q_{\perp}$  acceptance. At much larger  $q_{\perp}$ , the angular acceptance increases significantly, thus degrading the quality of the images.

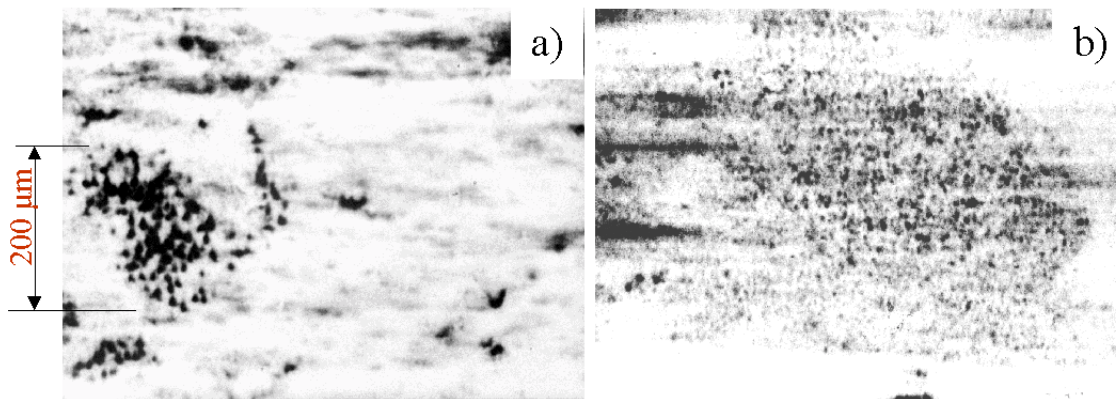


FIG. 1. USAXS images of the same region of the sample taken with a photon energy of 8.94 keV, a sample-to-detector distance of 24 cm and with (a)  $q = 1.3 \times 10^{-4} \text{ \AA}^{-1}$  and (b)  $q = 7.5 \times 10^{-4} \text{ \AA}^{-1}$ .

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## Results

USAXS imaging has been tested mainly on mildly deformed copper samples, in which creep cavities have formed on grain boundaries.<sup>3</sup> Figure 1 shows USAXS images from one of these samples; the images were obtained from the same sample volume at two different values of  $q$  ( $1.3 \times 10^{-4} \text{ \AA}^{-1}$  and  $7.5 \times 10^{-4} \text{ \AA}^{-1}$ ). The  $q = 1.3 \times 10^{-4} \text{ \AA}^{-1}$  image in Fig. 1(a) shows a cluster of spherical cavities that are approximately  $10 \mu m$  in diameter.

These cavities do not appear in the higher- $q$  image, Fig. 1(b). Instead, a large collection of much smaller objects is visible in an immediately adjacent region. Analysis of the  $q$ -dependence of the scattering from both collections of objects demonstrates that the shape factors of the cavities in Fig. 1(a) are consistent with spheres, whereas the objects visible in Fig. 1(b) are consistent with disk-shaped scatterers. Stereo images show that the disk-shaped objects appear only on the sample surfaces and therefore were produced during sample preparation.

## Discussion

Since most of the x-rays contributing to the image are those produced by USAXS, this imaging method is remarkably sensitive to microstructural features within a sample. It is complementary to absolute-calibrated USAXS scans, from which quantitative volume-averaged information on the same microstructural features can be determined. USAXS imaging should also prove invaluable as a tool for ensuring the validity of assumptions made during a USAXS analysis. It can be used on single-crystal, polycrystal, and amorphous samples.

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## References

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