Three-dimensional coherent X-ray diffraction microscopy

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ABSTRACT

X-rays have been widely used in the structural analysis of materials because of their significant penetration ability, at least on the length scale of the granularity of most materials. This allows, in principle, for fully three-dimensional characterization of the bulk properties of a material. One of the main advantages of X-ray diffraction over electron microscopy is that destructive sample preparation to create thin sections is often avoidable. A major disadvantage of X-ray diffraction with respect to electron microscopy is its inability to produce real-space images of the materials under investigation — three are simply no suitable lenses available. There has been significant progress in X-ray microscopy associated with the development of lenses, usually based on zone plates, Kirkpatrick–Baez mirrors, or compound refractive lenses. These technologies are far behind the development of electron optics, particularly for the large magnification ratios needed to attain high resolution. In this article, the authors report progress toward the development of an alternative general approach to imaging, the direct inversion of diffraction patterns by computational methods. By avoiding the use of an objective lens altogether, the technique is free from aberrations that limit the resolution, and it can be highly efficient with respect to radiation damage of the samples. It can take full advantage of the three-dimensional capability that comes from the X-ray penetration. The inversion step employs computational methods based on over sampling to obtain a general solution of the diffraction phase problem.

Key words: Microscopy, Nanocrystal shapes, Strain, Three-dimensional coherent X-ray diffraction.

INTRODUCTION

This is sometimes but not always observed. When a nonsymmetric pattern is seen, it can be decomposed into symmetric and asymmetric parts. For an ideal crystal, the symmetric part can be considered as coming from the real part of the electron density, while the asymmetric part is associated with an imaginary density that may represent a component of strain projected onto the Bragg peak in use.16 Although they have not yet been demonstrated experimentally, important materials science applications involving local microstructure and defects can be expected in the future from this acute sensitivity to strain.

Imaging Self-Assembled Gold Nanoparticles

A good example of Bragg diffraction CXD is the 3D imaging of nanocrystals of gold.17 These experiments were carried out at the Advanced Photon Source (ASP) using undulator X-rays of 9.5 keV. The use of a Si(333) monochromator reflection ensured sufficient longitudinal coherence. The samples were prepared in situ by high-temperature annealing of a thin Au film previously evaporated onto the oxide of silicon wafer. A 1000 Å film was found to produce oval-shaped nanocrystals about 2 µm long, 1 µm wide, and slightly less than 1 µm thick, with well-developed facets, especially the (111) planes that the 3D diffraction patterns was recorded on a 22.5-µm-pixel CCD array located 2.8 m away by rotating the sample in steps of 0.002°.
A typical diffraction pattern at the centre of the rocking curve is shown in Figure 5s. This pattern has been summarized by averaging with a rotated copy to remove the small effects of strain discussed earlier. The two main fringe patterns are the long modulated diagonal streak, oriented earlier. The two main fringe patterns are the long modulated diagonal streak, oriented close to the (111) direction of the primary facets, and the concentric ring pattern, typical of any compact object. The size of the sample can be estimated directly from these fringe spacings, which allows us to postulate the “support” to be used as a real-space constraint in the iterative refinement of phases. Alternate cycles of Fienup’s hybrid input-output (HIO) and error-reduction algorithms were found to lead to a solution without apparent stagnation of the computation. The diffraction pattern calculated from the final image is shown in Figure 5b. The uniqueness of the solution was demonstrated experimentally by obtaining almost indistinguishable images, starting from different sets of random phase numbers used to “seed” the algorithm.

Several slices from the 3D image of a Au nanocrystal obtained from a full angular series are shown in Figure 6. The first characteristic feature, the bright spot in the central and nearby slices in Figure 6b, was anticipated from earlier experiments and theoretical considerations. A beam-line Be
Fig. 3: Reconstructed three-dimensional (3D) structure of the Ni sample in Figure 1 displayed in iso-surface rendering. The fine divisions on the vertical axis are 25 nm each; the x and y axes are in nm.

Fig. 4: Ewald construction showing how small tilts of the sample cause the detector plant to sweep through the 3D diffraction. The difference between incident and exit x-ray wave vectors, denoted $K_i$ and $K_f$, determines the momentum transfer, $Q$.

Fig. 5: (a) Measured diffraction pattern of a single Au nanocrystal. This pattern, symmetrized to remove the small effect of strain, represents the central slice of a 3D diffraction pattern (b) Simulated diffraction pattern of the same slice as in (a), from the phased 2D image of the projection of the nanocrystal.
window, 6 m in front of the sample, was found to introduce a second component to the mutual intensity function describing the coherence. The second component has a much shorter coherence length, which ultimately determines the size of the bright spot in the image. The second important feature in Figure 6b is that the contrast structure can be seen throughout the interior of the crystal. This mainly appears as stripes, oriented perpendicular to both the (111) and (111) directions, but also dark regions where the stripes merge. It is believed that the dark regions are not empty, but filled with Au with an orientation that is twinned with respect to the rest of the nanocrystal. Striped slip zones with approximately the observed spacing are known to form along (111) planes in fcc metals during deformation. It is surprising that so much internal structure is visible in a simple isolated metal crystal, grown in situ without further processing. This may indicate the presence of considerable residual stress associated with the separation of the grains during their formation.

CONCLUSION

The present paper describes 3D X-ray diffraction microscopy to single-molecule imaging. The oversampling requirement discussed earlier implies that the iterative Fourier transform methods will not work in general on crystallographic data. There are too many degrees of freedom for all of the density points in the unit cell of a crystal to be constrained by the amplitude-only data. This is usually called the crystallographic phase problem. However, if a small crystal is used instead, the diffraction extends away from the Bragg points; intensity measurements between the Bragg peaks can provide additional information that can allow a solution of the phase problem. The extreme limit of a single unit cell, for which the diffraction pattern is smooth continuous function with no Bragg peaks at all, is highly interesting for the study of molecules that do not crystallize readily, such as membrane proteins. Single-molecule imaging has already been demonstrated by electron diffraction from double-walled carbon nanotubes. To avoid radiation damage to the sample, the measurement of the diffraction pattern must be completed before the atomic nuclei and the accompanying core electrons within the molecule become displaced by more than a bond length. This time period, estimated to be around 50 fs, is within the range of accessibility of future X-ray free-electron lasers (XFELs) based on linear particle accelerators. The vital importance of the potential application to solve the structures of proteins is a strong driving force for the construction of XFEL sources in the coming decade.
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