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Quantification of ADF STEM Image Data for Nanoparticle Structure and Strain Measurements

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Annular dark-field (ADF) imaging in the scanning transmission electron microscope (STEM) is a well-established tool for atomic resolution characterization of materials. The key beneficial attributes of ADF STEM imaging are (i) its incoherence and (ii) its monotonic dependence on thickness and composition (for sufficiently high inner radius). Both these attributes have led to interest in the quantification of ADF STEM images for making physical measurements of materials (reviewed in Ref [1]). Here we explore the use of ADF STEM for the three-dimensional structural characterization of nanoparticles, and for measurements of lattice distortion and strain.

The incoherence of the ADF STEM imaging process means that for sufficiently thin samples, the image can be modelled as a convolution of the probe intensity with an object function that represents scattering by the sample. The convolution model means that integrating the intensity associated with a column in an image provides a useful value for quantification. If the detectors are carefully calibrated so that each image pixel value represents the fraction of the total incident fluence that is scattered to the detector, then the column integration can be shown to result in a cross-section value that represents the probability of scatter to the detector. As long as the columns are still clearly resolved, the cross-section quantity is highly robust to many imaging parameters [2]. The importance of this is that imaging parameters that can be challenging to measure, such as the degree of partial spatial coherence in the beam, have a negligible impact on the measured cross-section, allowing robust image quantification which has shown good agreement with calculation ever since the development of atomic resolution in STEM in the early 1970s. In contrast, conventional high-resolution TEM (HRTEM) imaging, being a coherent imaging mode, is highly sensitive to changes in coherence and other imaging parameters, and a persistent mismatch with simulations is found that has been referred to as the “Stobbs factor”.

An alternative approach to ADF STEM image quantification is to use the discrete nature of atoms and the resulting multimodal distribution of cross-sections measured from many atomic columns. The distribution of column intensities is decomposed into a small number of overlapping Gaussian distributions, where the optimal number of Gaussian components is selected using an Integrated Classification Likelihood (ICL) approach. Using this approach, potential errors in detector calibration are avoided, but sufficient electron fluence is required to enable to Gaussian distributions to be sufficiently resolved [3].

There are now a number of examples of these approaches being applied to various heterogeneous catalyst systems in order to reconstruct their 3D structure. The workflow is shown in Figure 1. To enable a sufficiently large number of nanoparticles within a sample to be reconstructed, tilt tomography is avoided, and an energy minimization approach used to find a likely particle structure [4].

Measurements of lattice distortions and strain in the STEM are hindered by systematic and nonsystematic scan distortions and noise, and these distortions can also affect the precision of measurements of cross-sections described above. There has been growing interest in the use of non-rigid registration methods applied to rapidly acquired sequences of images. These methods can be tailored for use with scanned images where the distortions within a single scan line can be assumed to be much smaller than distortions between separation scan lines [5]. Not only does this approach improve the precision of image quantification, but we also show how lattice distortions in nanoparticles can be imaged at atomic resolution [6].

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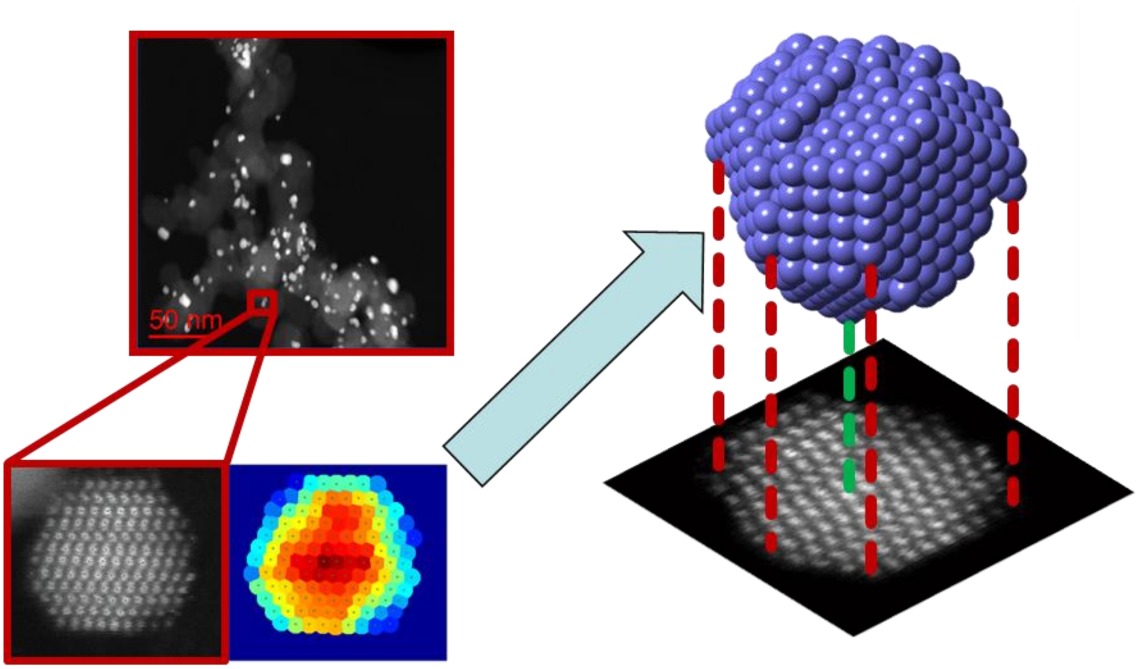

Figure 1. The workflow associated with determining nanoparticle structures through the quantification of ADF STEM images. Here the sample is a Pt supported on C heterogeneous catalyst. The intensity associated with each column is used to provide a count of the number of atoms in the column. The relative heights of columns is then determined through an energy minimization to reveal a candidate nanoparticle structure.

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