

A New Approach for 3D Quantitative STEM Using Defocus Corrected Electron Ptychography

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Two-dimensional (2D) imaging from nanoparticles with atomic resolution is routinely performed using aberration corrected scanning transmission electron microscopy (ac-STEM) and ac-TEM. Three-dimensional (3D) imaging from a nanoparticle, however, is not straightforward in TEM. Electron tomography is a technique which is widely used to reconstruct a 3D morphology of a sample with nanometer-scale resolution in TEM. In electron tomography, several 2D TEM images are acquired from a sample at different viewing angles and then a 3D morphology of the sample is reconstructed from those 2D images. Recent developments in electron tomography have pushed its resolution to the atomic scale [1]. Nevertheless, as the acquisition of several images at different tilt takes time and therefore mobile atoms on the surface of the specimen can move during the acquisition process, it is not feasible to determine the exact position of atoms on the surface of the specimens using this technique. Recent advances in quantitative annular dark-field (ADF) STEM provided new approach to reconstruct a 3D model of an object with the atomic resolution from only one ADF image [2]. In this technique, first, the number of atoms in each atom column in an ADF image is quantified to create atomistic models from the object and then an energy minimization is applied to relax the object's 3D structure [2]. The long exposure time for electron tomography and the energy minimization after atom counting for the ADF-STEM can dramatically affect the accuracy of the reconstructed 3D model in the above mentioned techniques. In addition, these techniques cannot (in the most cases) be applied on beam sensitive materials or compounds containing both light and heavy atoms.

Electron ptychography is a powerful technique which can be exploited to study the atomic structure of materials including those containing both light and heavy elements. Furthermore, recent developments in the hardware of the electron microscope's detectors significantly reduces the electron dose as well as time required for ptychographic data acquisition which pave the way for characterization of various beam sensitive materials. In electron ptychography, first, a series of electron diffraction patterns (i.e. a 4D STEM dataset) are collected by scanning an electron beam (probe) across a specimen. Then, some mathematical algorithms such as the single side-band (SSB) method [3], Wigner distribution deconvolution (WDD) [4] and extended ptychographical iterative engine (ePIE) [5] are used to deconvolve the probe and object transfer functions from the 4D STEM dataset. The key assumption in these algorithms is that the probe function is constant for all the probe positions since the aberrations of the microscope's electromagnetic lenses are almost constant during the very short time of data acquisition. Although the lens aberrations can be assumed to be constant for each probe position in a dataset, the probe function is not unique since the geometry of the specimen at each probe position across the imaging area alter the defocus value of the probe (Figs. 1). Thus, we have to use a defocus-corrected probe function for each probe position to calculate the object transfer functions.

Our simulation results (not shown here) show that it is possible to calculate the probe's defocus value

using electron ptychography. In order to do that, a series of ptychographic phase images are reconstructed using different assumed defocus values of the probe and then for every atom column in the reconstructed phases, we find the defocus at which that particular column exhibits its maximum phase (i.e. the apparent defocus). Moreover, we demonstrate that 3D models of nanoparticles can be obtained from 4D-STEM datasets acquired simultaneously with HAADF images. Here, we calculated the number of Pd atoms for each atom column observed in a HAADF image from a Pd nanocube (Fig. 2(a)), and then we measured the height of those columns from their absolute defocus extracted from the WDD ptychographic phase reconstructed from a 4D-STEM dataset acquired simultaneously with the HAADF image. Finally, the 3D model of the Pd nanocube were simply reconstructed as we had the number of atoms in each column as well as the height of those columns (Fig. 2(b-d)). We expect this approach to be applicable to reconstruct not only an accurate ptychographic phase but also a 3D model of any other nanostructure.

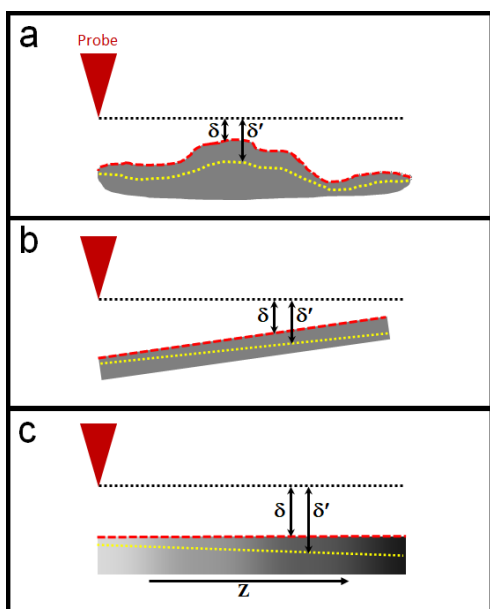


Figure 1. Absolute defocus (δ) and apparent defocus (δ') levels for (a) a sample with an uneven surface, (b) tilted sample and (c) sample with compositional variation.

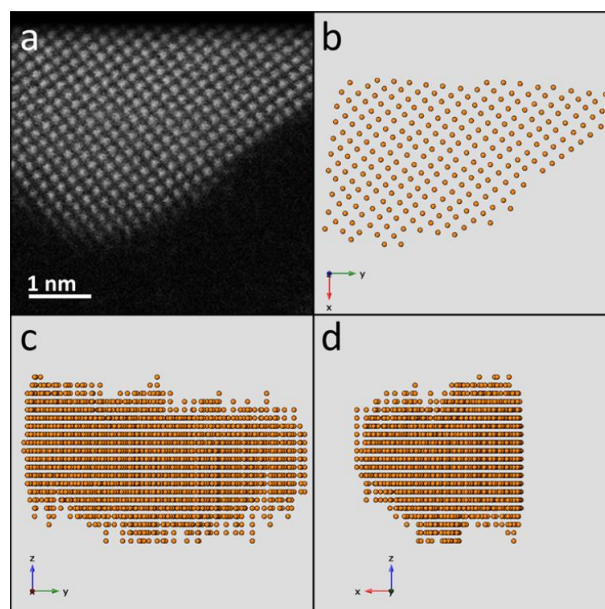


Figure 2. (a) ADF image obtained from a Pd nanocube. (b-d) 3D reconstructed model in three different viewing direction for the nanocube shown in (a).

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