doi:10.1017/S1431927622002240
Microsc. Microanal. 28 (Suppl 1), 2022

Lattice-Vibration Limited Resolution, 3D Depth Sectioning and High Dose-Efficient Imaging via Multislice Electron Ptychography

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Multidimensional data generated from modern aberration-corrected electron microscopes offers many new opportunities to explore the rich structural information in materials and devices. With the access to both real and diffraction space, four-dimensional scanning transmission electron microscopy (4D-STEM) has been demonstrated a wide impact [1]. Electron ptychography is such an example. It uses 4D-STEM datasets to iteratively reconstruct the electrostatic potential of the sample and greatly benefits from the advancements of direct electron detectors [2]. It works well for samples with weak scattering [3] or thicknesses below a few nanometers [4]. For thick bulk samples, however, conventional ptychography has a degraded performance due to strong multiple scattering. In fact, multiple scattering affects all conventional imaging techniques, usually detrimentally, such as a reduction of the interpretable resolution [5] and nonlinear or nonmonotonic image contrast with sample thickness [6]. In those cases, intensive image simulations and modelling are usually required for a reliable interpretation of atomic-resolution images. Phase retrieval methods to solve the inverse problem of multiple scattering can circumvent these problems [7-9].

Here, we demonstrate a robust experimental realization of the inversion for multiple scattering using a regularized implementation of multislice electron ptychography [10]. This approach can reconstruct the structure of thick samples layer by layer (Figure 1(b)), following the idea of Cowley-Moodie multislice theory [11]. The phase images from different layers are linearly additive, which avoids the complicated contrast reversals in conventional phase-contrast imaging techniques such as high-resolution TEM. The inversion of multiple scattering together with the corrections of scan distortions, residual aberrations, and partial coherence of the probe achieves a spatial resolution close to the intrinsic atomic size, i.e., the lattice-vibration limit. More ‘hidden’ atoms, such as the Pr-Pr dumbbells in Figure 1(c) can be unambiguously resolved, while the conventional HAADF images cannot (Figure 1(d)). High contrast and signal-to-noise of all atoms including light elements such as oxygen (Figure 1(c)) allows for measuring the atomic positions at a sub-pm precision.

The robust multislice electron ptychography method also provides some new imaging capabilities: We demonstrate that a depth resolution of better than 3 nm can be achieved from only one projection measurement, without sacrificing any lateral resolution [9]. This has many potential applications, such as locating individual atomic dopants and precipitates in crystalline matrix. Furthermore, simulations show that multislice electron ptychography from relatively thick samples can use more than 10 times lower dose to obtain similar contrast and resolution, compared to other low-dose imaging techniques, such as integrated differential phase contrast (iDPC / iCoM) images [12].

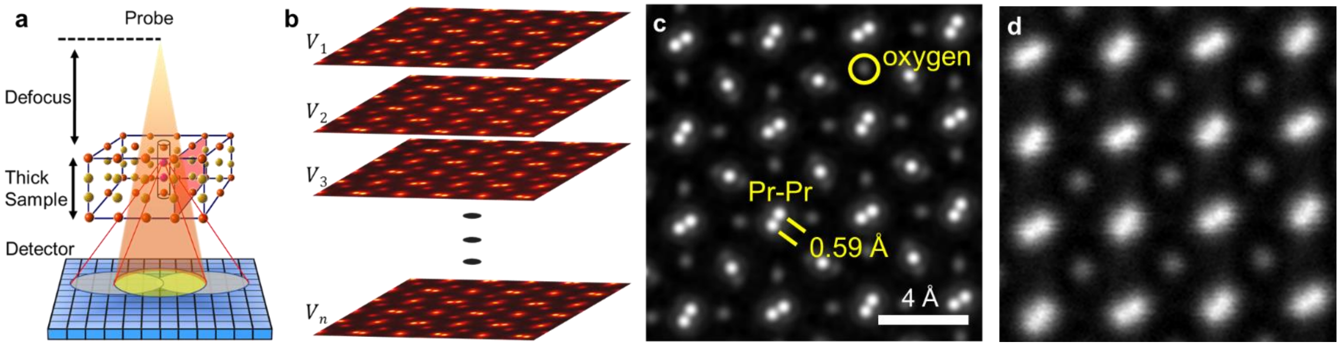

Figure 1. Phase retrieval of thick samples using multislice electron ptychography. (a) Schematic of 4D-STEM experiments for ptychography. (b). Layer-by-layer atomic structures reconstructed from multislice electron ptychography. (c). Projected phase image of $PrScO_3$ [001] at a sub-20 pm resolution from multislice electron ptychography and (d) one conventional HAADF image from the same microscope.

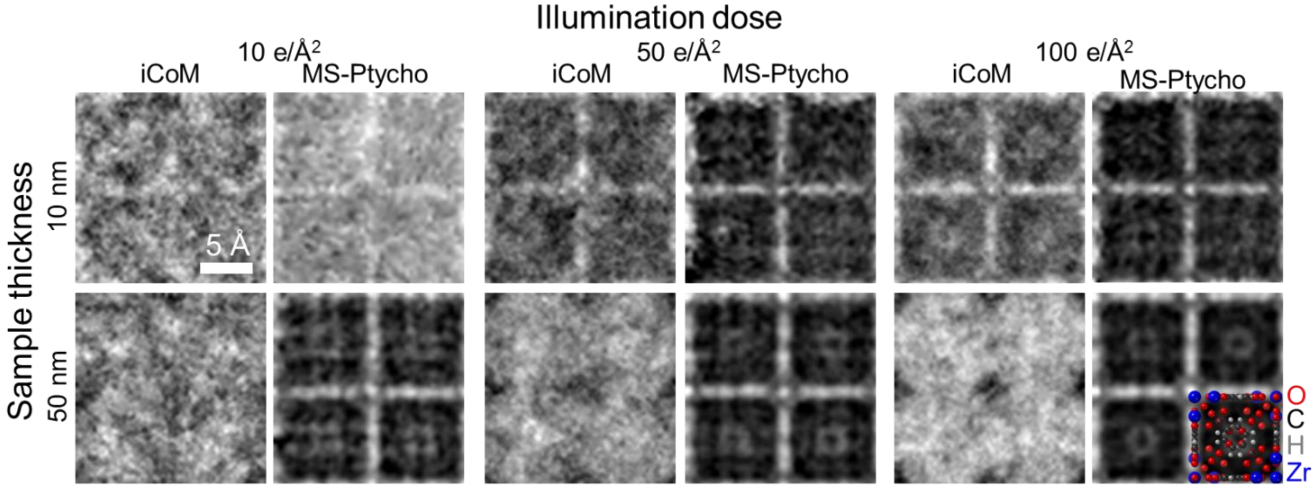

Figure 2. Low-dose imaging of thick samples from multislice electron ptychography (MS-ptycho) and integrated center-of-mass (iCoM). The results are from simulated data using a probe-forming semi-angle of 15 mrad and a defocus of 100 nm at 300 keV for a model sample of metal-organic framework, UiO-66.

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[12] Research supported by US NSF (grants DMR-2039380 and DMR-1719875).