# Multislice electron ptychography enables lattice vibration-limited resolution and linear phase-contrast imaging in thick samples

Zhen Chen[1], Yi Jiang[2], Yu-Tsun Shao[3], Megan Holtz[4], Michal Odstrcil[5], Manuel Guizar-Sicairos[5], Isabelle Hanke[6], Steffen Ganschow[6], Darrell Schlom[7] and David Muller[8]

[1]School of Applied and Engineering Physics, Cornell University, Ithaca, NY, USA, New York, United States, [2]Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439, USA, United States, [3]School of Applied and Engineering Physics, Cornell University, Ithaca, NY, USA, Ithac, New York, United States, [4]Department of Materials Science and Engineering, Cornell University, Ithaca, NY, USA, United States, [5]Paul Scherrer Institut, 5232 Villigen PSI, Switzerland, Switzerland, [6]Leibniz-Institut für Kristallzüchtung, Max-Born-Str. 2, 12489 Berlin, Germany, Berlin, Germany, [7]Department of Materials Science and Engineering, Cornell University, Ithaca, NY, USA, Ithaca, New York, United States, [8]School of Applied and Engineering Physics, Cornell University, Ithaca, NY, USA, Ithaca, New York, United States

The ultimate limit to spatial resolution in an electron microscope is set by the thermal vibrations of the atoms themselves, which are on the order of 10-20 pm at room temperature [1]. Using multislice electron ptychography, we are surprisingly close to this limit. Although conventional annular dark-field imaging has reached spatial resolutions better than 50 pm [2,3], such performance can only be realized in thin samples, usually only a few nanometers thick and after the correction of high-order aberrations. In thicker samples, strong multiple scattering changes the probe shape and causes dechannelling effects between neighboring atomic columns which reduces the interpretable resolution [4]. Multiple scattering also leads to a complicated image contrast that is nonlinearly or even nonmonotonically dependent on the sample thickness, especially for phase-contrast imaging methods.

Ptychography uses scanning diffraction and 4D-STEM datasets to iteratively reconstruct the electrostatic potential and has reached a resolution of 39 pm in thin 2D materials [5]. However, conventional ptychography (single-slice) assumes that the exit-surface wave function can be expressed as a multiplication of the incident probe and a single complex transmission function whose phase represents the sample projected potential. This approximation usually fails for samples thicker than a few nanometers in the presence of strong multiple scattering. For thick samples, attempts at phase retrieval from both multi-slice electron ptychography [6-8] and Bloch wave based scattering matrix [9] approaches have been reported. However, to date, none of the experimental demonstrations have been widely adopted due to limited resolution or image quality.

Here, we demonstrate a robust experimental realization of inversion of the multiple scattering using a regularized implementation of multislice electron ptychography [10]. This approach provides ultra-high-resolution reconstructions for samples thicker than tens of nanometers (Figure 1(b)). More importantly, the contrast maintains a linear dependence on thickness over a wide thickness range (Figure 1(c)). In contrast, conventional phase-contrast imaging techniques such as single-slice ptychography or high-resolution TEM (HRTEM) have much worse resolution and provide image contrast not directly interpretable for samples thicker than a few nanometers (Figure 1(b) and (d)). The linear phase-contrast can also greatly widen the applicable sample thickness and resolution for phase-contrast tomography. Furthermore, three-dimensional structural information such as locations of single dopants can also be obtained, since multislice ptychography retrieves the potential of the sample at different depth positions separately.

The potential reconstructed from multislice ptychography from an experimental dataset acquired from a 21-nm-thick $PrScO_3$ sample (Figure 2(a)), similar to that from a simulated dataset (Figure 2(d)), shows only a slightly additional blurring compared to the potential at 300 K (Figure 2(b)). All sub-lattices including the Pr-Pr dumbbells with a separation of only 0.59 Å, pure oxygens and O-Sc-O triple atoms are resolved. The

diffractogram of the phase image shows an isotropic information transfer up to 4.39 Å$^{-1}$, corresponding to 23 pm in real space. Quantitative analysis of the atomic column width reveals that the blurring from the instrument is smaller than 20 pm, which is smaller than the intrinsic broadening from thermal fluctuations. The method also allows for direct measurements of local Debye-Waller factors of atoms near defects or interfaces [11].
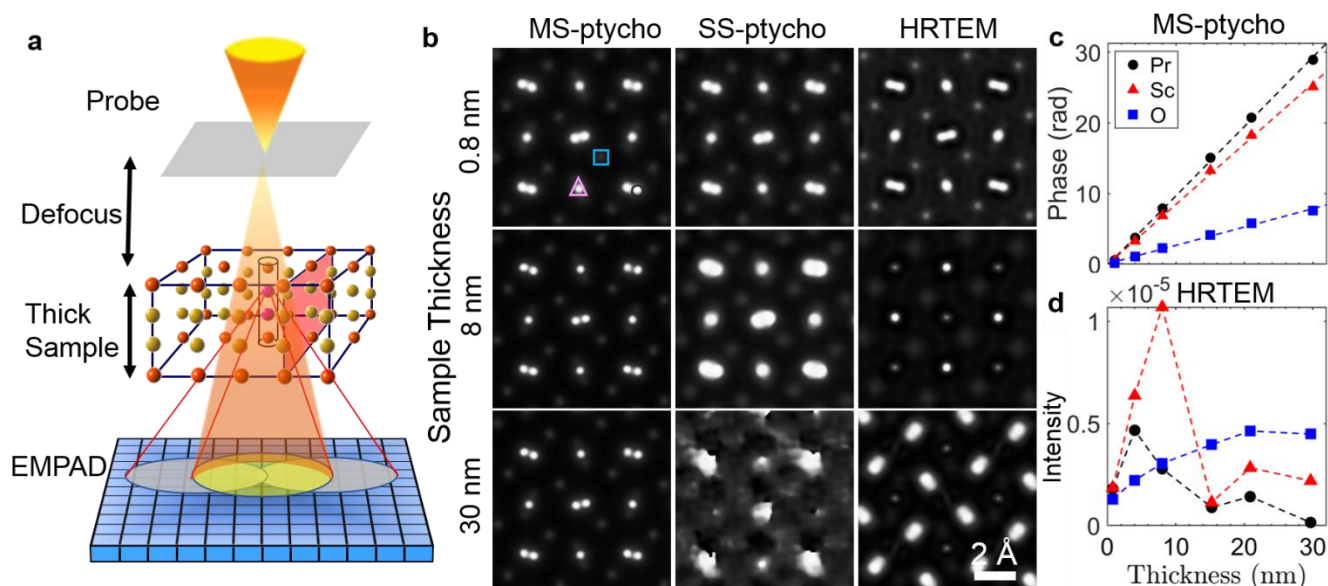


**Figure 1.** Linear phase-contrast imaging using multi-slice electron ptychography. (a) Setup of 4D-STEM experiments using a defocused probe illumination. (b). Thickness dependent contrast from different phase-contrast imaging methods via simulations: Multislice ptychography (MS-ptycho), single-slice ptychography (SS-ptycho), and conventional high-resolution TEM (HRTEM) with an information limit of 1.25 Å-1. (c). Thickness evolution of the intensity at atomic columns from mult-islice ptychography and HRTEM.
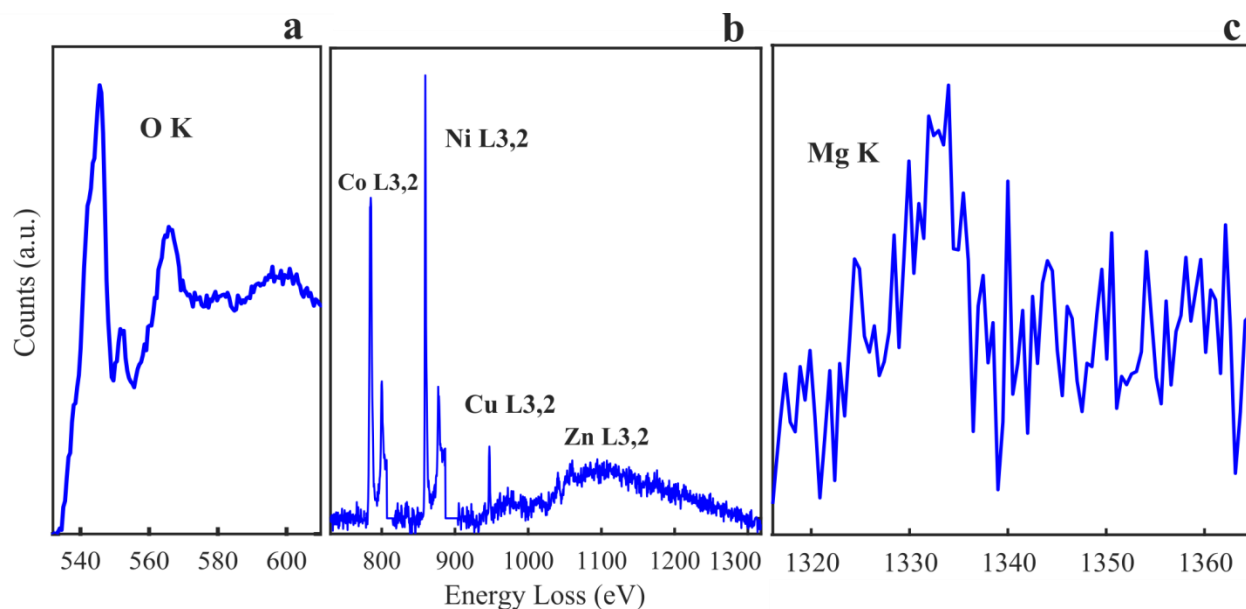


**Figure 2.** Lattice-vibration-limited resolution from multislice ptychography. (a). Multislice ptychographically-reconstructed phase image from an experimental dataset acquired from a 21 nm thick $PrScO_3$ sample. (b)-(d). Simulated projected potential of $PrScO_3$: (a) static potential neglecting zero-point fluctuations, (b) at 300 K, and (c) reconstructed from multislice ptychography using simulated diffraction data at 300 K including thermal

fluctuations for 21 nm thick PrScO3. (e) and (f). Diffractogram of the phase image from experiment: (e) 2D and (f) a line profile along the white dashed line on (e). Phase images in (a) and (d) are normalized to a unit-cell thickness of 8.03 Å.

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