

## Phase Imaging beyond the Diffraction Limit with Electron Ptychography

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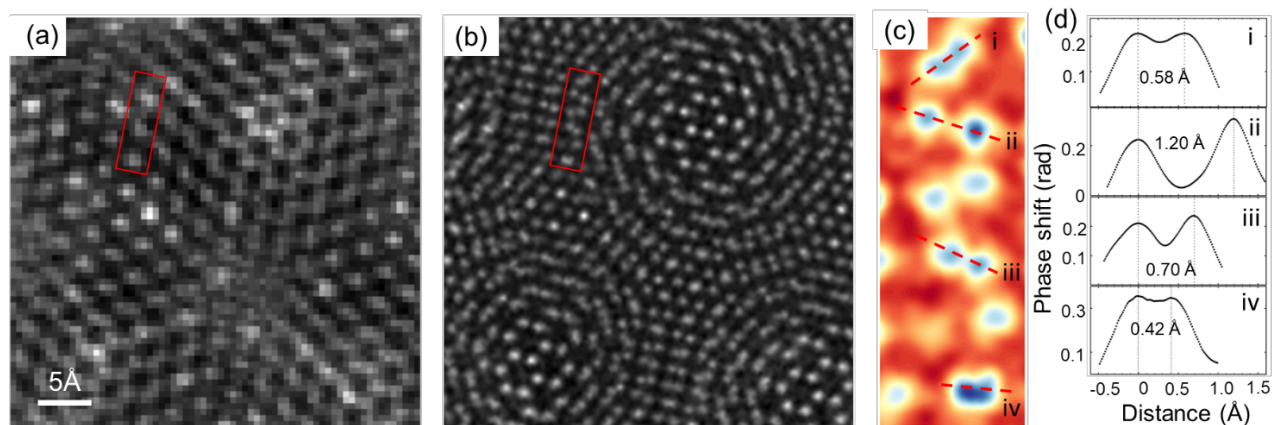
The past three decades have seen the rapid development and maturation of aberration-corrected electron lenses. With the recent advances in detector technology and reconstruction algorithms, the resolution limits are now dominated by counting noise through the maximum allowable dose, either by radiation damage to the sample, or by recording times. Ptychographic phase retrieval algorithms offer an approach to using all of the scattered electrons – potentially enhancing both the resolution and dose-efficiency. Here we show how in-focus ptychography enables imaging at more than double the diffraction limit of the lens, and how out-of-focus ptychography improves the dose efficiency compared with ADF STEM, simultaneously providing a four times faster acquisition, double the information limit and double the precision.

Image resolution is dominated by the energy (or wavelength) of the electron beam and the quality of the lens. Two-dimensional materials are imaged with low beam energies to avoid damaging the samples, limiting spatial resolution to  $\sim 1$  Å. By combining our new design of electron microscope pixel array detector (EMPAD) [1] which has the dynamic range to record the complete distribution of transmitted electrons at every beam position, and a ptychographic phase retrieval algorithm [2] to process the data, we have been able to increase the spatial resolution well beyond the traditional lens limitations reaching a 0.39 Å resolution for MoS<sub>2</sub>, at 80 keV, the same dose and imaging conditions where conventional imaging modes reach only 0.98 Å [3]. The improved resolution, dose efficiency and robustness to environmental noise enabled by ptychography make it easy to identify defects such as sulfur monovacancies, as well as subtle structural arrangements and tilts on the sulfur sublattice that are undetectable by conventional imaging modes. For twisted bilayers we are able to resolve the shear distortions and interactions between the layers (Figure 1). [3]

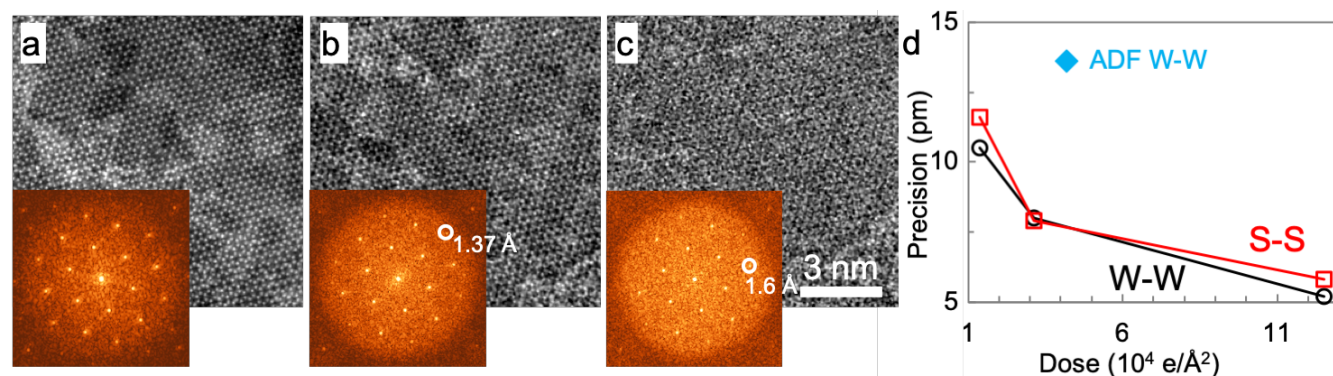
In-focus ptychography, like conventional STEM, is not well suited to imaging large areas at high spatial resolution – as the resolution is increased, the number of samples required grows quadratically in dwell time or dose. Operating out-of-focus decouples the resolution and real-space sampling requirements [4], provided the detector has sufficient dynamic range and pixels. While simulations have shown the out-of-focus reconstructions to have better convergence at low dose, compare to in-focus [3], in practice this was not the case when simple initial probe estimates were used. With improved probe diversity, we have been able to image 120 nm fields of view with 0.69 Å resolution at 80 keV producing 6000x6000 pixel images. Figure 2 shows the dose dependence and resolution of out-of-focus ptychography on WS<sub>2</sub> monolayers. Figure 2d shows the precision with which we can measure S-S or W-W bond lengths, both in ptychography and ADF STEM, showing a roughly factor of two advantage for ptychography over ADF at the same dose.[5]

## References:

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 [3] Y. Jiang *et al*, *Nature* **559**, (2018), p343.  
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 [5] We thank Jiwoong Park, Saien Xie, Hui Gao, Ming-Hui Chiu, and Lain-Jong Li for samples. Research supported by US National Science Foundation (grants DMR-1539918, DMR-1429155, DMR-1719875) & US Department of Energy Basic Energy Sciences (DE-SC0005827).



**Figure 1.** Real space resolution demonstration of full-field, in-focus ptychography using a twisted bilayer MoS<sub>2</sub>. (a) Synthesized ADF image; (b) Phase image from ptychographic reconstruction; (c) Enlarged phase image of the marked area in (b); (d) Intensity line profile of corresponding positions labeled with dashed red lines in (c). The peak separation distances are overlaid on (d). Data adapted from [3]



**Figure 2.** Dose-dependence of out-of-focus Ptychographic reconstructions of a monolayer WS<sub>2</sub> sample: a. 58000 e/Å<sup>2</sup>; b. 3300 e/Å<sup>2</sup>; c. 790 e/Å<sup>2</sup>. These are extracts from a larger field of view. Scan step size is 0.85 Å. The dose-dependence of the precision with which the W-W and S-S bond lengths can be measured from ptychography, and ADF imaging (only W-W was measurable at low dose) is shown in (d).