

Improving Robustness of Electron Ptychography by Bayesian Optimization of Tilt and Thickness

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Meeting-report

Improving Robustness of Electron Ptychography by Bayesian Optimization of Tilt and Thickness

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From a buckling lamella of randomly oriented grains to randomly oriented nanoparticles, some samples are hard to be tilted on zone, and certain amount of mistilts are almost inevitable. Such mistilts blur the atoms in electron ptychographic reconstructions and impede their convergence. Although known mistilts can be added to the propagator function in multislice simulations [1, 2], finding tilt has been time- and computation-intensive. Conventionally, position averaged convergent beam electron diffraction (PACBED) patterns simulated with a range of roughly guessed thickness and tilt values have been compared to the experimental PACBED through least squares fitting. Recent studies sped up such a laborious process with convolutional neural networks (CNN) once the models were trained [3–5]. However, the training requires tens of thousands of simulated datasets per material system at a specific zone axis, if not more. This study proposes a Bayesian optimization approach that is universal to various material systems and requires a minimal number of simulations to determine thickness and tilt of 4D-Scanning Transmission Electron Microscopy (STEM) datasets, especially for the application of ptychography.

We find the thickness and tilt of the sample in the given 4D-STEM data following the workflow shown in Figure 1(a). Our code takes a crystallographic information file of the sample and the PACBED pattern of interest as inputs. A set of thickness and tilt values is suggested via Bayesian Optimization [6], which is then used to create a simulated PACBED pattern using abTEM [7] on the fly. Other experimental parameters, such as beam voltage, convergence angle, detector angle, and sampling, are kept constant during the process. The mean squared error between the experimental and simulated PACBEDs is used as the objective function. The code iterates and populates new sets of thickness and tilt values with the goal of minimizing the objective function. At the end of the iteration, the pair of thickness and tilt which gives the lowest mean squared error is chosen. Figure 1(b) shows the parameter space and corresponding mean squared errors of simulated PACBEDs when one of them chosen as the ground truth. All these simulations would have been required as training datasets for the CNN methods. On the other hand, although this objective map was not provided, the BO code successfully found the ground truth within 20 simulations. Only the datapoints indicated with circular markers were explored by the BO code. This simulation study shows that our BO approach is functional even with two orders of magnitude fewer number of simulations than the CNN based approaches.

The BO code was also applied to an experimental PACBED, as shown in Figure 2(a). The output tilt values were then applied during the multislice electron ptychographic reconstruction to compensate the sample tilt. Figures 2(b) and (c) show the sums of the reconstruction stacks without and with the tilt correction, respectively. The elongated atom columns due to the sample mistilt in (b) get straightened with the tilt correction in (c). For this dataset, the suggested tilt along the y-axis was 5 mrad. Applying trigonometry to the sample thickness of 13 nm, such tilt results in about 0.6 Å shift in atom position throughout the stack, just as shown in the depth profile in (b). Considering the lateral resolution of multislice electron ptychography being around 0.2 Å [8], such a shift due to sample tilt is worth fixing to achieve better atomic resolution images.

The preliminary result suggests that the BO approach can improve the robustness of ptychography by enabling reconstructions of wider range of datasets even with mistilts. In addition, without the need of training on massive datasets, our BO code for finding thickness and tilt could be a universal, time- and computation-effective solution to various material systems [9].

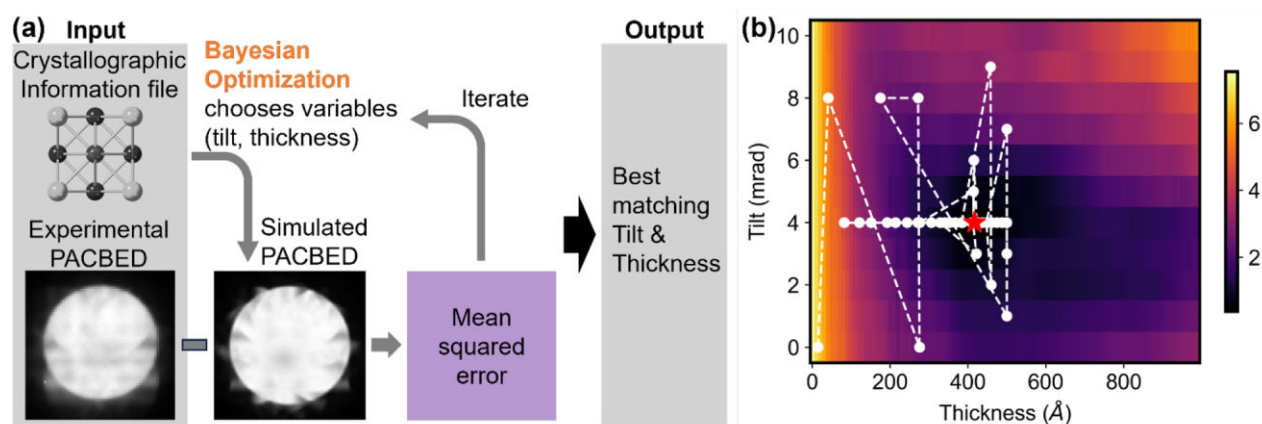


Fig. 1. (a) Workflow of finding thickness and tilt from a PACBED image via Bayesian optimization (BO). (b) Objective map of 1100 simulated PACBEDs (100 thicknesses & 10 tilts), evaluated with respect to the ground truth simulated PACBED (red star marker). Note that only 50 images of these were invoked via BO, which are shown as white points.

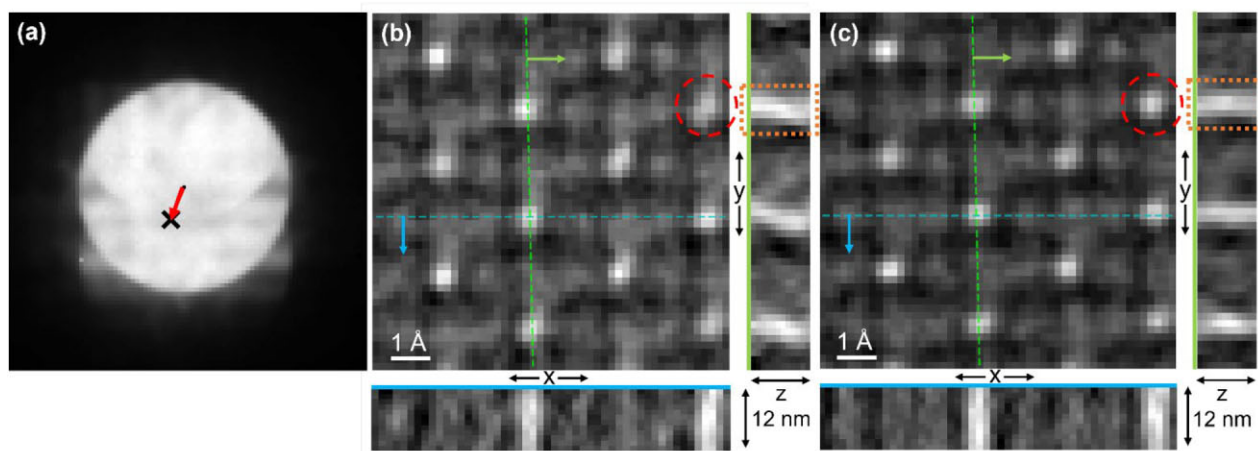


Fig. 2. (a) Experimental PACBED. Red arrow indicates the tilt, and the cross marks the suggested center by the BO code. Results from multislice electron ptychographic reconstructions of the same dataset (b) without and (c) with the tilt correction, using the values from the BO code. (b) The atom column circled in red appears elongated due to the sample tilt, while the equivalent atom column appears rounder in the tilt corrected reconstruction in (c). Depth profile along the y-axis shows slant atom columns (one enclosed in orange box) due to the uncorrected tilt, which is straightened in (c).

References

1. E J Kirkland in "Advanced Computing in Electron Microscopy: Second Edition", (Springer New York, NY) p.151.
2. H Sha *et al.*, *Science Advances* 8 (2022), eabn2275. <https://doi.org/10.1126/sciadv.abn2275>
3. W Xu and J M LeBeau, *Ultramicroscopy* 188 (2018), p. 59–69. <https://doi.org/10.1016/j.ultramic.2018.03.004>
4. C Zhang *et al.*, *Ultramicroscopy* [Online] 210, 112921 (2020), <https://doi.org/10.1016/j.ultramic.2019.112921> (accessed Oct. 29 2023).
5. M Oberaigner *et al.*, *Microscopy and Microanalysis* 29(1) (2023), p. 427–436.
6. P I Frazier, *arXiv preprint* (2018), <https://doi.org/10.48550/arXiv.1807.02811>.
7. J Madsen and T Susi, *Open Research Europe* 1 (2021), 24.
8. Z Chen *et al.*, *Science* 372 (2021), p. 826–831. doi:10.1126/science.abg2533
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