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Intelligent and Automatic Parameter Optimization for High-resolution Electron Ptychography

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Electron ptychography is a phase retrieval method in scanning transmission electron microscopy (STEM) with deep sub-angstrom resolution [1] and high dose-efficiency [2]. Advances in pixelated detectors [3] allowing rapid collection of diffraction patterns at every scanning point of a STEM experiment has significantly made electron ptychography more practical and available. However, creating a high-quality ptychographic reconstruction requires careful selection of both reconstruction parameters as well as experimental parameters. While experienced experts can develop some intuition between parameters and reconstruction quality, finding the most optimal parameters for new datasets or applications remains a subjective and time-consuming process. This reduces the overall throughput and prevents electron ptychography from wider adaptation.

Here, we propose a framework for intelligent automatic parameter selection using Bayesian optimization (BO) with Gaussian processes, which is well-suited for finding extrema in unknown functions that are expensive to evaluate. The technique has been used in many research areas such as machine learning [4] and autonomous microscopy experiments [5]. For electron ptychography, we utilized BO with state-of-the-arts algorithms to simultaneously optimize 8 different types of reconstruction parameters. Without additional prior knowledge, our workflow automatically produces high-resolution ptychographic reconstruction of an experimental dataset of $MoSe_2/WS_2$ [5]. The result optimized by BO (Fig. 1a&c) agrees well with the expert-chosen reconstruction. For comparison, parameters that are chosen by random search often lead to significantly worse quality as many experimental errors are not corrected, as shown in Fig. 1b&d.

We further used BO to investigate several complex tradeoffs between key experimental parameters, including scan step size and probe defocus, and search for optimized conditions under a fixed total electron dose. Figure 2 illustrates ptychographic reconstructions of simulated bilayer MoS_2 datasets at various dose levels. Comparing to a fixed set of parameters that were used in previous work [2], BO can find parameters that produce more superior reconstruction (Figure 2a-d), especially at ultra-low dose where the physical requirements for good reconstructions are more stringent. The comprehensive study provides valuable guidance for future experimental designs.

Automatic parameter selection via BO provides an easier workflow for creating high-quality electron ptychography reconstructions without requiring expert intuition. The algorithm is useful for optimizing both reconstruction and experimental parameters. With increasing hardware and software capabilities, it may be possible to perform parameter optimization in-line with real-time experiments and create more autonomous experiments [6].

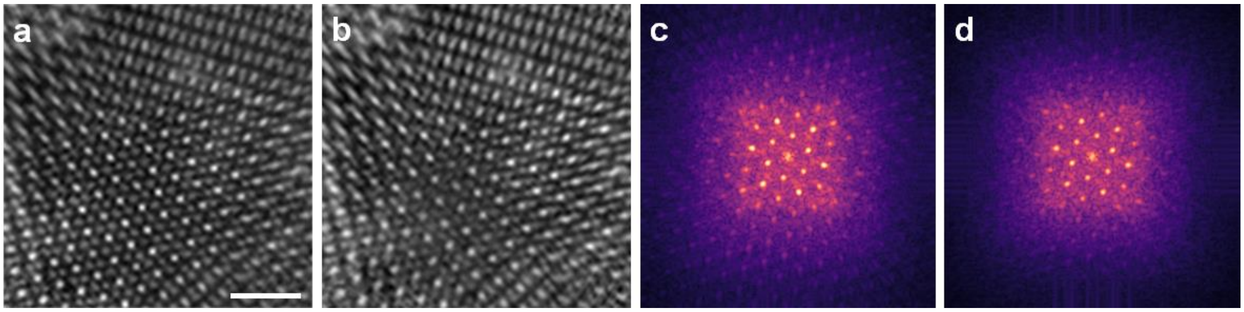

Figure 1. Ptychographic reconstructions of an experimental dataset of $MoSe_2/WS_2$. (a) Reconstructed phase using parameters optimized by BO. (b) Reconstruction using the parameters from random search. (c,d) Fourier magnitude of reconstructions in (a) and (b), respectively. Scale bar is 1 nm.

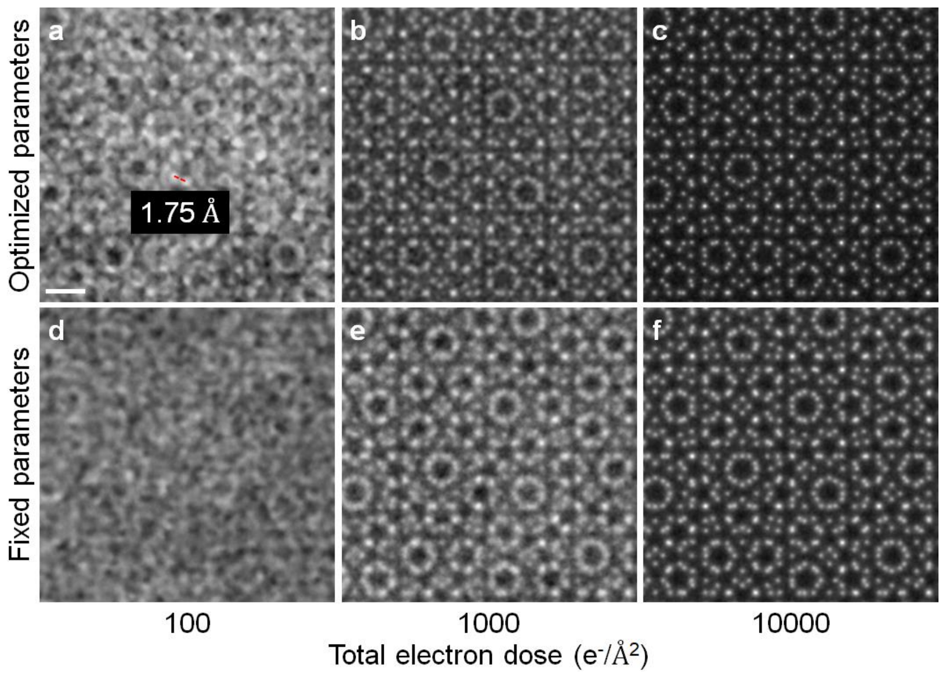

Figure 2. Ptychographic reconstructions of simulated datasets of bilayer MoS_2 at different electron dose levels. (a,b,c) The experimental parameters were optimized by BO for each dose level. (d,e,f) Fixed experimental parameters that are similar to previous work [2]. While the expert parameters match well with BO at the highest dosage, it suffers at lower doses. At the lowest dose, the expert parameters fail to resolve any distinguishable structures in contrast with the optimized parameters, which can still resolve atoms that are 1.75 Å apart. Scale bar is 5 Å.

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