Multi-objective Bayesian Optimization for High-resolution Electron Ptychography

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Abstract

Electron ptychography enables deep sub-angstrom spatial resolution of atomic structures by solving the inverse problem of electron scattering through the sample with the complete distribution of transmitted electrons recorded by 4D STEM. However, in practice, ptychography is computationally intensive and requires a delicate selection of parameters, prone to trails and errors from user experience. In this project, we design a new scheme targeting automated parameter tuning. Specifically,

- We demonstrate the additional objective of Fourier ring correlation (FRC) improves the performance by avoiding unphysical local minima.
- We adopt multi-objective Bayesian optimization to optimize the two metrics simultaneously and reach a Pareto front of equally feasible solutions that lead to the best reconstruction result.

Ptychography as an Inverse Problem

Ptychography solves an inverse problem that reconstructs images by processing coherent interference patterns scattered from an object of interest. In single slice ptychography, the physics is as simple as

$$I(\mathbf{k}) = |\mathcal{F}[\psi_e(\mathbf{r})]|^2 = \left|\int \psi_i(\mathbf{r}) T(\mathbf{r}) e^{2\pi i \mathbf{r}\cdot\mathbf{k}} d\mathbf{r}\right|^2$$

where $I(k)$ denotes the experimental measurement of the diffraction pattern intensity at real space scan position r, $\psi_i(r)$ denotes the incident probe function, $\psi_e(r)$ denotes the exit wave function, and $T(r)$ denotes the transmission function which contains information about the sample. We then solve

$$\min \sum_i \left\| \sqrt{I_j} - |\mathcal{F}[\psi_i(\mathbf{r}-\mathbf{r}_j)\cdot T(\mathbf{r})]| \right\|^2$$

where r_j denotes different scan positions. Multi slice ptychography furthermore considers the propagation of electrons through layers of the sample.

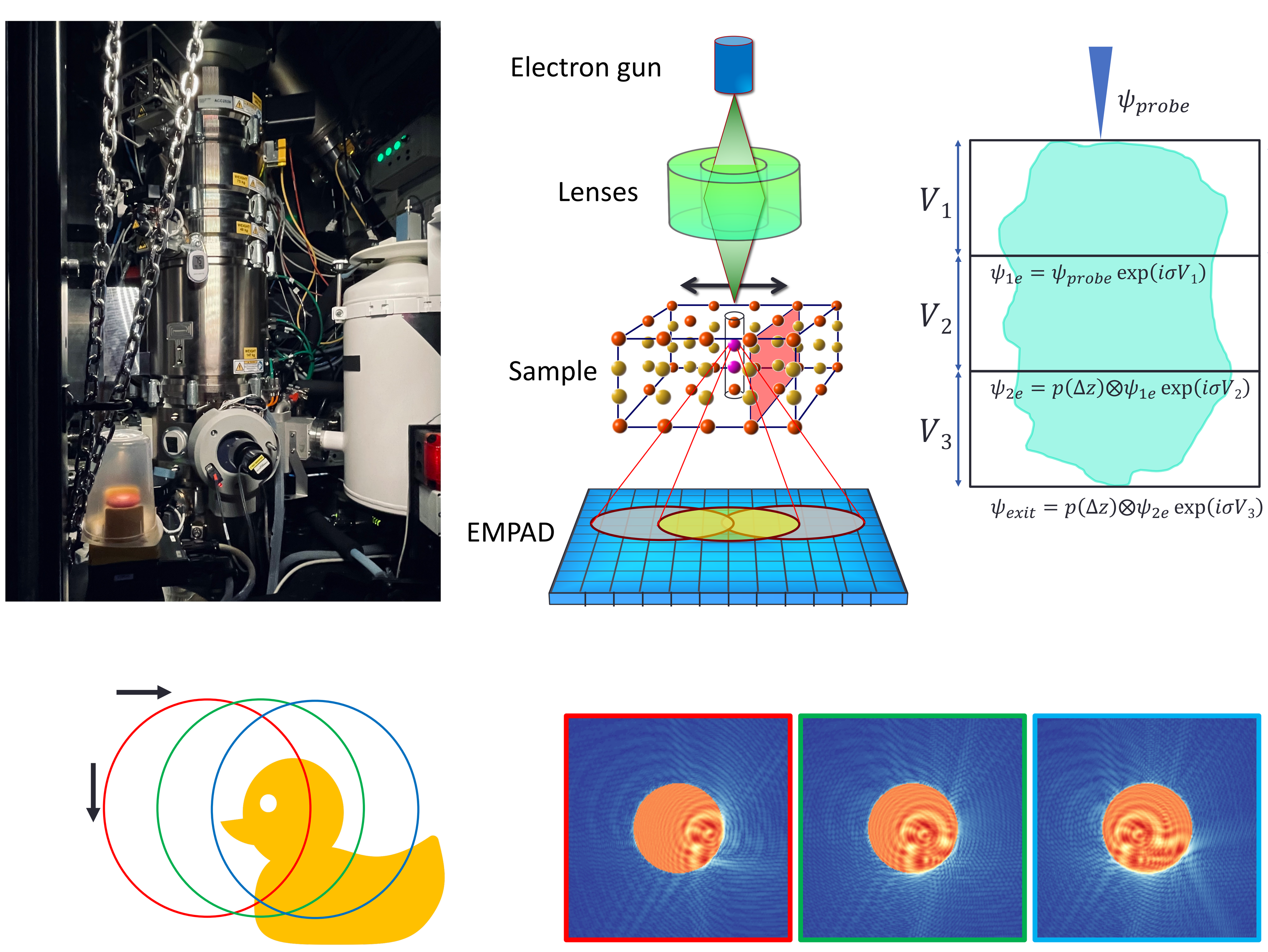


Fig. 1. 4D-STEM with a high dynamic range pixelated detector, EMPAD. With scanning transmission electron microscope (STEM), we scan an electron beam probe of width roughly a few angstroms through the sample of interest in real space along both x and y. For each scan position, a diffraction pattern forms afterwards and is recorded by EMPAD, thus 4D STEM data. We use a defocused probe for ptychography which allows for higher dose efficiency and increased depth of field. In multi slice ptychography, propagation of the wave through layers is considered.

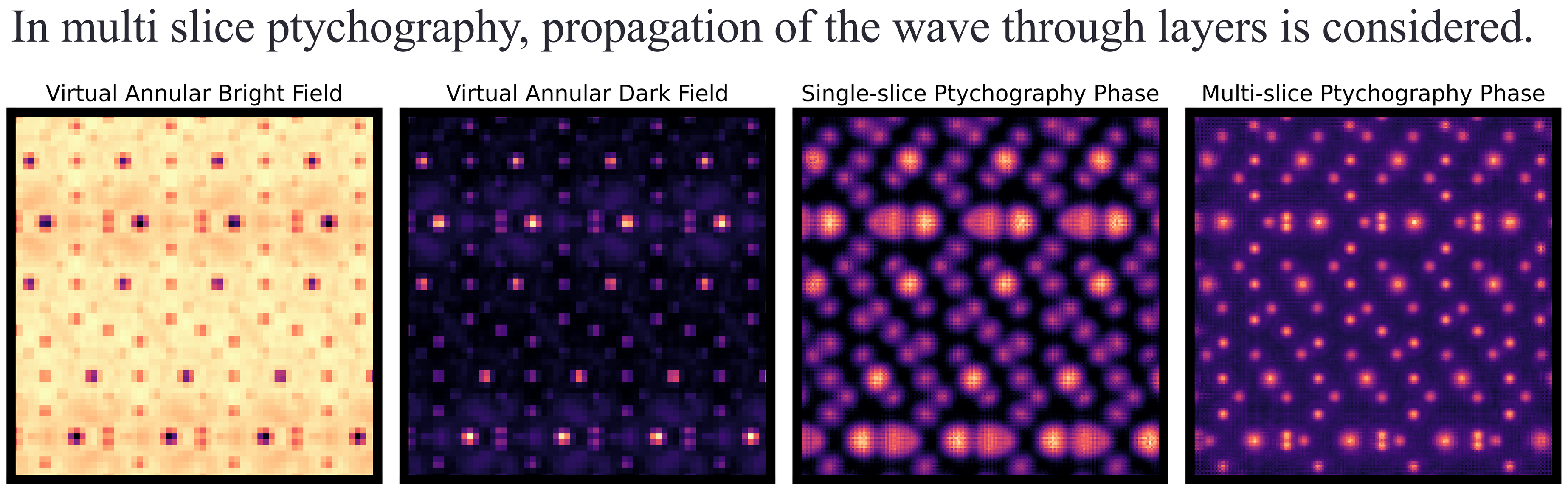
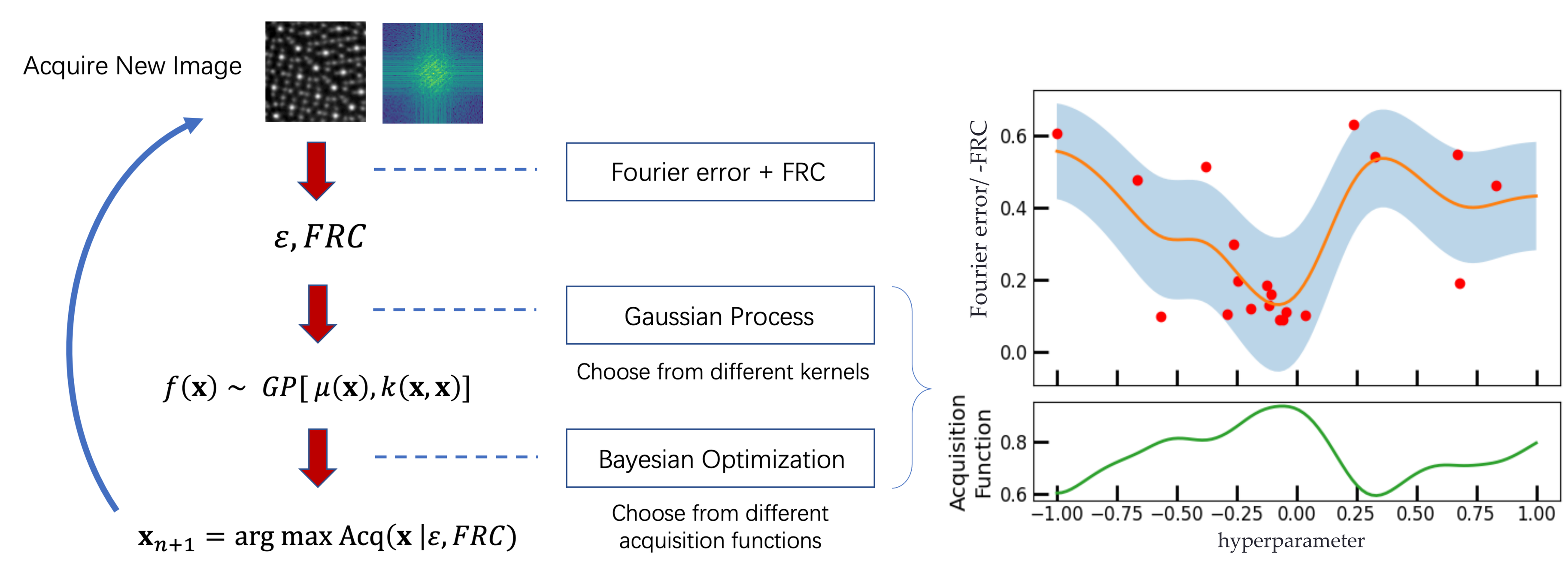


Fig. 2. Comparison of conventional imaging and ptychographic reconstructions of the $BaFe_{12}O_{19}$ for the [100] zone. (a) Annular bright field; (b) Annular dark field; (c) Single slice ptychography; (d) Multi slice ptychography. The 4D data set has dimensions of 64×64×105×105 taken on a simulated sample potential of size 20×20 Å and 12 nm thick. For ptychography the data is taken at defocus of 100 Å.

Fourier Ring Correlation as a New metric

In addition to the Fourier reconstruction error, we further introduce the Fourier ring/shell correlation coefficient commonly used in biological microscopic imaging to evaluate resolution limit as a second metric. The FRC is a cross correlation metric calculated in Fourier space where the input pair of images are Fourier transformed and then the correlation between the Fourier coefficients is calculated in each ring (for 2D images, hence "Fourier Ring Correlation").

$$\mathrm{FRC/FSC}_{12}(r_i) = \frac{\sum_{r\in r_i} \mathcal{F}_1(r)\cdot\mathcal{F}_2(r)^*}{\sqrt{\sum_{r\in r_i}\mathcal{F}_1^2(r)\cdot\sum_{r\in r_i}\mathcal{F}_2^2(r)}}$$



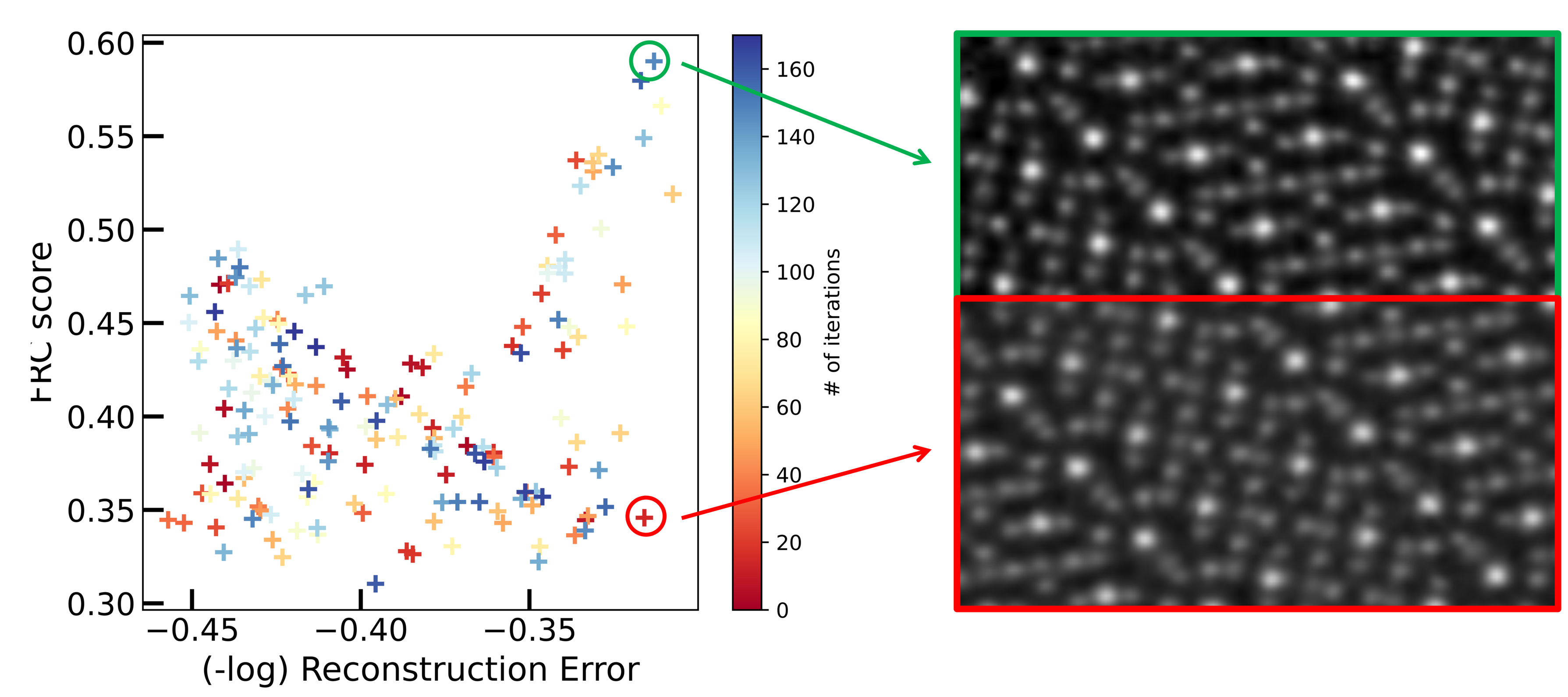


Fig. 3. Multi-objective Bayesian optimization simultaneously optimizes Fourier error and FRC score, which finds the subtle difference in resolution even at the same Fourier error.

Multi-objective Bayesian optimization

In addition to optimizing a single objective, Bayesian optimization (BO) can also be implemented to optimize multiple objectives at the same time, i.e.. multi-objective BO (MOBO) when there is a trade-off between the different objectives, and they cannot be optimized at the same time. The optimal set of solutions are found at the Pareto frontier after MOBO minimizes the hypervolume bounded by current parameter estimates and the reference points,

$$\mathrm{HV}(\mathcal{P}) = \lambda_d(\cup_{\mathbf{y}\in\mathcal{P}}[\mathbf{r},\mathbf{y}])$$

where λ_d is the Lebesgue measure on $\mathbb{R}^d$, $\mathbf{r}$ is the set of reference points and $\mathcal{P}$ is a finite approximation of the Pareto front.

$$\mathcal{P} = \{\mathbf{y}^{(1)}, \dots, \mathbf{y}^{(n)}\} \subset \mathbb{R}^d$$

Acknowledgement

This work was supported by the U.S. National Science Foundation under award PHY-1549132, the Center for Bright Beams.

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