Automated Crystalline Domain Segmentation in Polycrystalline TEM Images via Machine Learning-Based FFT Analysis

Woojin Bae[1], Shihyun Kim[1], Jinho Rhee[1]

[1]School of Chemical and Biological Engineering, Seoul National University, Seoul, Republic of Korea

Abstract

Fast Fourier Transform (FFT) analysis of high-resolution transmission electron microscopy (HRTEM) images is essential for crystallographic characterization, yet manual peak selection and domain delineation introduce substantial subjectivity and limit throughput. This challenge is acute in polycrystalline materials with weak crystallinity, such as solid electrolyte interphase (SEI) layers in lithium-metal batteries. We present an automated, GUI-based workflow integrating Gaussian Mixture Models (GMM) for FFT peak classification and DBSCAN for real-space domain extraction. This approach extends ML-based crystallographic segmentation—previously demonstrated in 4D-STEM—to conventional HRTEM imaging, democratizing automated domain analysis for the broader microscopy community.

Introduction and Methods

Traditional FFT-based crystallographic analysis requires manual distinction of crystalline peaks from noise, inverse transformation, grain boundary tracing, and area quantification—a workflow that is time-intensive and fundamentally subjective. Different researchers may select different peaks from identical FFT patterns, leading to inconsistent conclusions. For battery materials like lithium-metal SEI with weak crystallinity and nanoscale heterogeneity, this subjectivity becomes critical.

Recent advances in four-dimensional scanning transmission electron microscopy (4D-STEM) have demonstrated ML's power for automated crystallographic analysis [1]. By acquiring diffraction patterns at each scan position, 4D-STEM datasets can be clustered using k-means to automatically identify crystalline domains. However, 4D-STEM requires specialized instrumentation, generates massive datasets (hundreds of gigabytes), and demands substantial computational resources. Conventional HRTEM imaging remains far more accessible in most microscopy facilities. Our approach extends ML-based crystallographic segmentation to single HRTEM images, maintaining the speed and consistency advantages of 4D-STEM workflows while requiring only standard imaging capabilities.
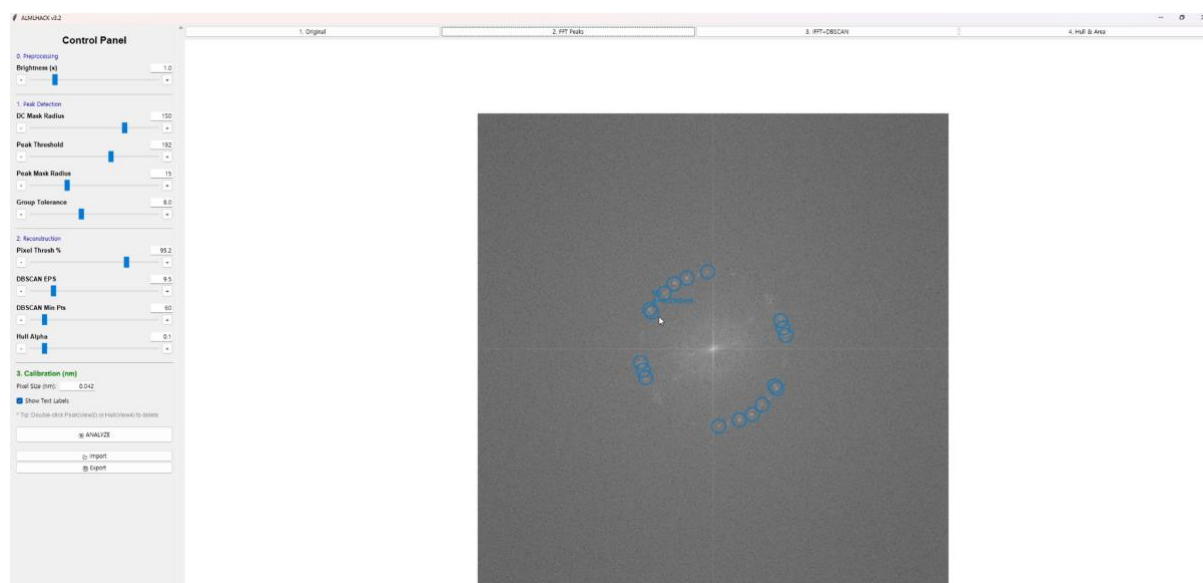
While previous approaches have applied DBSCAN clustering to inverse FFT reconstructions for domain segmentation, the critical bottleneck of manual FFT peak selection remained unaddressed [2 3]. Our key innovation lies in automating the entire upstream workflow: systematic FFT preprocessing to enhance peak visibility, automated individual peak detection, and ML-based classification to distinguish signal from noise. By integrating GMM-based peak classification with established DBSCAN domain clustering, we create a fully automated pipeline where the most subjective and time-consuming step—peak selection—is now handled consistently and reproducibly by machine learning [4]. This upstream automation is essential for enabling truly high-throughput crystallographic analysis.

Workflow: Our pipeline comprises four integrated stages. (1) GMM-Based Peak Classification: After computing the 2D FFT, we employ a two-component Gaussian Mixture Model to automatically classify peaks as signal-like (crystalline) or noise-like. GMM is ideal because FFT peak characteristics—intensity, sharpness, radial profile width—naturally form distinct distributions. Unlike supervised learning requiring labeled training data, GMM performs unsupervised clustering through expectation-maximization. Users adjust classification sensitivity via intuitive sliders, and non-physical peaks can be removed by double-clicking. (2) Automated d-Spacing Calculation: The tool automatically calculates d-spacing values from peak positions using user-provided pixel size calibration, enabling immediate phase identification. (3) DBSCAN-Based Domain Clustering: Selected peaks undergo inverse FFT to reconstruct real-space images showing crystallinity distribution. DBSCAN clustering groups spatially connected high-intensity regions while filtering isolated noise. DBSCAN's ability to discover arbitrarily shaped clusters without specifying cluster count makes it ideal for irregular grain boundaries. Two user-adjustable parameters (neighborhood radius ε and minimum sample threshold) control clustering behavior. (4) Visualization

and Quantification: Crystalline domains are color-coded and overlaid onto the original image. The tool automatically quantifies individual domain areas and total crystalline coverage. Non-physical regions can be excluded via double-clicks.
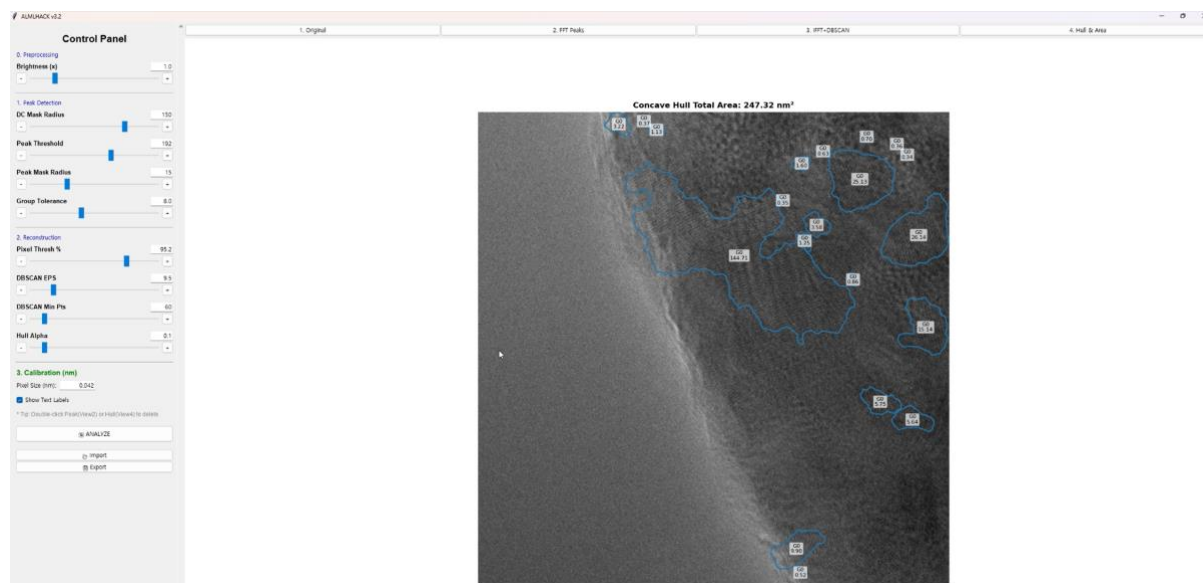
Results and Future Work

Our tool successfully analyzes polycrystalline HRTEM images from lithium-battery SEI layers and other complex systems where conventional analysis struggles with weak crystallinity and substantial phase heterogeneity. Automated GMM classification achieves robust distinction between crystalline signals and background noise across varying image quality conditions, with adjustable sensitivity accommodating materials ranging from highly crystalline to barely ordered. DBSCAN clustering effectively delineates grain boundaries even in images with irregular domain morphologies and significant amorphous fractions. The complete workflow executes in a fraction of the time required for manual analysis while producing reproducible results independent of analyst identity.

Figure 1. Automated FFT peak classification using Gaussian Mixture Models distinguishes signal-like crystalline peaks (blue) in polycrystalline SEI material, enabling reproducible peak selection.

Representative results from SEI analysis demonstrate successful identification of multiple coexisting crystalline phases with varying d-spacings, quantification of their spatial distributions, and measurement of individual crystallite sizes ranging from nanometers to tens of nanometers. The overlay visualization enables immediate assessment of crystallinity distribution relative to morphological features, facilitating interpretation of structure-property relationships.

Figure 2. DBSCAN-clustered crystalline domains overlaid onto original HRTEM image following inverse FFT reconstruction.

Future developments will focus on: (1) integrating crystallographic database matching for automatic phase suggestion; (2) correlating with EDS/EELS data for comprehensive structure-composition relationships.

By combining ML efficiency with flexible parameter control and expert correction, this tool addresses critical bottlenecks in HRTEM characterization. The GUI-based design ensures accessibility without extensive programming experience, democratizing advanced analytical capabilities across the microscopy community. We anticipate particular value for energy storage materials, catalysts, and structural ceramics where nanoscale crystalline heterogeneity understanding is essential.

References

[1] Ophus, C. Four-dimensional scanning transmission electron microscopy (4D-STEM): from scanning nanodiffraction to ptychography and beyond. Microsc. Microanal. 25, 563–582 (2019).

[2] https://github.com/woojinb1/masK-driven-Analysis-for-iNverse-fft-yield-evaluation

[3] Ester, M., Kriegel, H.-P., Sander, J. & Xu, X. A density-based algorithm for discovering clusters in large spatial databases with noise. in Proceedings of the Second International Conference on Knowledge Discovery and Data Mining (KDD'96) 226–231 (AAAI Press, 1996).

[4] Reynolds, D. Gaussian Mixture Models. in Encyclopedia of Biometrics (eds. Li, S. Z. & Jain, A.) (Springer, 2009).