Human-in-the-loop interface for Automated experiments in Electron Microscopy

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Abstract

Scanning transmission electron microscopy (STEM) is a powerful tool for exploring structure, composition, and chemical properties on nanometer and atomic level and hence is a powerful tool for materials discovery. However, the STEM paradigm is based on use of classical scanning relying on point and grid based spectroscopies We report the development of the tool for direct control of operational STEM that enables materials discovery using imaging and spectroscopic modes, following either predefined policies of myopic discovery workflows. The latter are illustrated by human-in-the-loop automated experiments **(hAE)** with bayesian optimisation (BO), that allows dynamic control of reward functions while running the microscope. This is a new paradigm of STEM where we dynamically tune the behaviour of ML agent rather then directly controlling the instrument. While several studies have highlighted the potential of human-in-the-loop automated experiments (hAE), there remains a need for a software interface to facilitate these experiments effectively. In this work, we developed a web tool that integrates Bayesian optimization with human intervention capabilities, allowing users to modify scalarizers (also referred to as reward or target properties) and adjust experimental policy dynamically to favour exploration or exploitation. The tool also provides flexibility to manipulate descriptor sizes, thereby capturing the complex structure-property relationships at nanoscale/atomicscale. We have also provided a simulator version on pre-acquired data as an educational interface for experimentalist new to automated experiments. The code is provided as the supplementary material. The simulator can be accessed live at `https://tiny.utk.edu/hAE`

1 Introduction

The exploration of materials at the nano and atomic scales has been revolutionized by recent advancements in scanning transmission electron microscopy (STEM)[Williams and Carter, 1996, Crewe, 1974, Stephen J., 2011] combined with electron energy-loss spectroscopy (EELS)[Wu et al., 2018, Kalinin et al., 2021, Lovejoy et al., 2018], and Energy-dispersive X-ray spectroscopy (EDX)[Thermo

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Fisher Scientific, n.d.]. These technologies enable researchers to probe the structural, chemical, and electronic properties [Wu et al., 2018, Kim et al., 2023, Bosman et al., 2006, Varela et al., 2012, Gázquez et al., 2017, Roccapriore et al., 2023, Noircler et al., 2021], of materials with unprecedented spatial resolution. Despite the power of these techniques, traditional data acquisition methods in STEM-EELS-EDX rely on static [Pratiush et al., 2024a, Kalinin et al., 2023] sampling approaches, often constrained by predefined parameters and limited by the inability to adapt dynamically to the evolving landscape [Liu et al., 2023a,b] of the experiment. The need for real-time decision-making in STEM-EELS-EDX experiments has spurred the development of human-in-the-loop automated experiments (hAE) [Pratiush et al., 2024a, Kalinin et al., 2023, Liu et al., 2023a,b]. In these setups, human operators intervene during the automated process, guiding the exploration based on observed data patterns. This approach bridges the gap between fully automated systems and human expertise, allowing for adaptive experimentation that can pivot between exploration of unknown regions and exploitation of known areas of interest. There have been work on Bayesian optimization [Frazier, 2018, Rasmussen, 2004] particularly deep kernel learning [Ziatdinov et al., 2022a, Valleti et al., 2024, Ziatdinov et al., 2022b, Kalinin et al., 2023] to conduct automated experiments. However, currently there is no implementations of hAE[Pratiush et al., 2024a, Kalinin et al., 2023, Liu et al., 2023a] on live instrument. There is a need of user-friendly interface that seamlessly integrates automated decision-making with human inputs. The challenge lies in developing a flexible software environment that allows real-time adjustments to experimental parameters, such as scalarizers (target properties) and descriptor sizes, while maintaining robust optimization pathways. In this work, we present a web-based tool designed to facilitate hAE in STEM-EELS-EDX. By leveraging Bayesian optimization, the tool empowers users to dynamically adjust experimental policies, shifting between exploratory and exploitative actions based on evolving data. Users can manipulate scalarizer to target specific material properties and fine-tune descriptor sizes, enhancing the tool's ability to capture complex structure-property relationships at the nanoscale. This approach paves the way for a paradigm shift that redefines the role of human intuition and machine learning in scientific discovery by incorporating human-machine interaction.

Contribution of this work:

- Developed a tool for Bayesian optimization with intervention in target property, policy, and other hyperparameters.
- Use case shown on pre-acquired data.
- Use case with live instrument control.

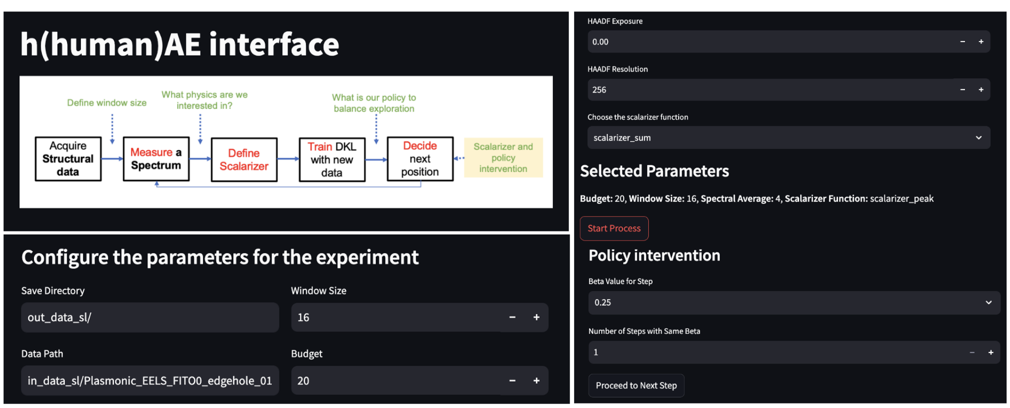

Figure 1: The human-in-the-loop automated experiment (hAE) interface.

2 Methodology

Core Bayesian optimization algorithms, implemented using the gpax [Ziatdinov, n.d.] library, utilize Gaussian Processes to efficiently explore and exploit the experimental parameter space. The tool integrates various Python libraries to manage the experiments, with Streamlit [Streamlit, n.d.] handling the user interface, enabling real-time configuration of experimental parameters and dynamic interventions. To complement live instrument operation and account for intermittent accessibility and for operator training, we provide a Python-based microscope simulator which supports the testing and demonstration of the tool's functionality, allowing for the development and validation of optimization strategies using simulated data, thereby avoiding the need for constant access to the actual microscope. Pre-acquired datasets16 enable replication and validation of optimization processes under controlled conditions. For **live experiments**, the tool interfaces with Autoscript software developed by ThermoFisher, Inc. and Gatan server [Pratiush et al., 2024b] software to achieve direct control of the microscope hardware, providing real-time feedback and adjustments to bridge software optimization and physical data acquisition. This interface can be easily extended to other manufacturers. The experimental workflow begins with the configuration of initial parameters, such as descriptor size, scalarizer function, and budget, while supporting dynamic interventions that allow users to modify policies like the exploration-exploitation balance, step count, and physical properties of interest. Bayesian optimization guides the selection of measurement points, adapting continuously to the evolving data landscape and offering visual feedback to facilitate decision-making. More detailed methodology can be found in paper [Kalinin et al., 2023, Pratiush et al., 2024a] introducing hAE in electron microscopy.

3 Results: The hAE tool in action

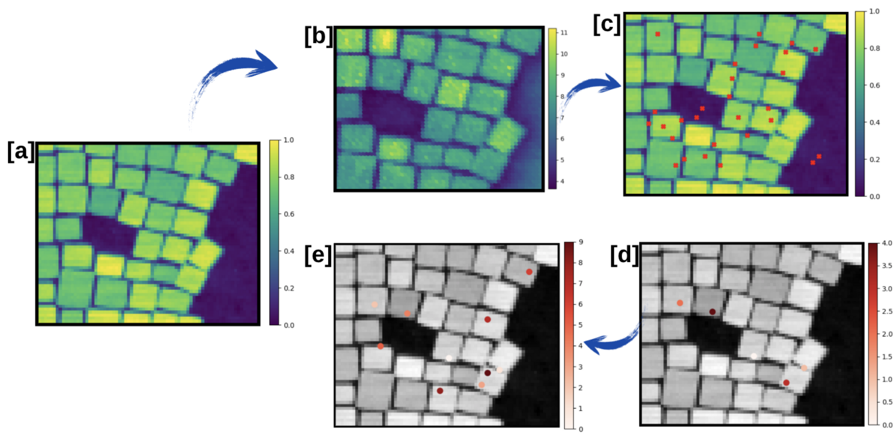

Figure 2: hAE workflow on Pre-Acquired Data: The figure illustrates the workflow of an hAE experiment conducted on pre-acquired data. Panel (a) shows the overview image of the nanoparticles under investigation. Panel (b) displays the target property, or scalarizer, selected by the domain expert based on the EELS spectrum. Panel (c) depicts the initial seed points chosen to initiate the workflow. Panel (d) presents the first five points selected using a Bayesian optimization approach with a beta parameter set to 0.25. Finally, panel (e) demonstrates a human intervention in the experimental policy, where the beta value is adjusted from 0.25 to 1 to alter the exploration-exploitation balance.

Conducting live experiments is costly, especially when the parameter space for exploration is extensive. To optimize the process, researchers typically start by acquiring a spectral image and analysing it within the hAE framework before initiating a live experiment. This preliminary analysis helps to determine the optimal descriptor size, scalarizer (target property), and the appropriate policy interventions that should be employed during the live experiment. As illustrated in Figure 2, the workflow begins with selecting seed points and choosing initial parameters based on pre-acquired data, allowing for informed decision-making and efficient use of live microscopy resources. In Figure 3 we show the hAE workflow on live microscope.

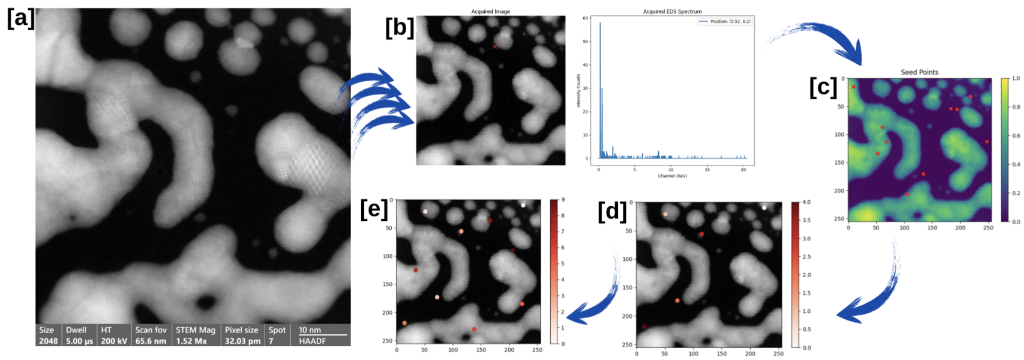

Figure 3: The hAE workflow on a live microscope with gold nanoparticles is shown as follows: Panel (a) provides an overview of the nanoparticles, while panel (b) shows the EDX spectrum at a seed point. Panel (c) marks the initial seed points for the workflow, and panel (d) presents the first five points selected using Bayesian optimization with a beta of 0.25. Panel (e) demonstrates a human adjustment in experimental policy, changing the beta from 0.25 to 1 to modify the exploration-exploitation balance.

4 Relevant Material use cases for the Tool

In this section we will decribe the tool usecase. We are unable to try it on different samples due to time constraint. In the tool one can set the desired material property (peak in the EDX or EELS spectrum). So in next iteration the BO chooses a point based on this target property. The policy can be set by the user to being more exploratory or exploitative based on beta parameter tuning.

4.1 EELS peaks

In **lithium battery materials**, the Li K-edge at around 55 eV is critical for identifying lithium compounds, though its low energy requires higher incident electron energies to reduce the background signal from plasmon excitations. The N K-edge at 400 eV is used to **detect nitrogen in steel**, with techniques like iterative averaging and second-differential processing enhancing weak nitrogen signals. In **biological systems**, where X-ray analysis risks causing radiation damage, EELS can map sulfur, phosphorus, and calcium by analyzing their L-shell ionization edges (135 eV for sulfur, 165 eV for phosphorus). For **heavier elements in biological specimens** like aluminum , K-edges in the 1000-3000 eV range are advantageous due to their strong signal-to-background ratios and reduced plural scattering, allowing analysis of thicker specimens.

4.2 EDX peaks

EDX is suitable for detecting elements heavier than Na (Sodium), while EELS is more sensitive to lighter elements Egerton [2011]. By combining EDX and EELS, one can obtain a more comprehensive understanding of a material. For instance, in the analysis of biological samples, EDX can be used to detect common elements like Na, K, Mg, Cl, P, and S, while EELS can be used to analyze lighter elements or investigate specific features like phosphorus distribution in DNA. In materials science, EDX can provide overall elemental composition, while EELS can be used to study local bonding environments or valence states at interfaces or defects.

5 Future work

To enhance the versatility and effectiveness of our tool for physics discovery, we plan to implement several key upgrades. First, by incorporating **multi-objective Bayesian optimization**, we can simultaneously optimize multiple material properties, broadening the tool's flexibility for uncovering new insights. Additionally, we aim to support **real-time tuning** of instruments, allowing for dynamic and responsive adjustments during experiments. Finally, given the broad applicability of our tool's core principles, we plan to extend its use to **other microscopy techniques**, such as atomic force microscopy (AFM) and scanning tunneling microscopy (STM), to further broaden its reach and potential impact.

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