

Real-world uses of machine learning for automated experiment: upgrading synthesis and characterization tools!

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Monday September 30th, 2024
Ferris Hall, UTK

ORNL is managed by UT-Battelle, LLC for the US Department of Energy



Acknowledgements



- Sergei Kalinin (UTK)
- Ye Cao (UT/Arlington)
- Jan-Chi Yang (NCKU Taiwan)
- S. Jesse (ORNL)
- P. Ganesh (ORNL)
- Anahita Khojandi (UTK)
- G. Duscher (UTK)
- Many other students, postdocs, and staff at CNMS and ORNL

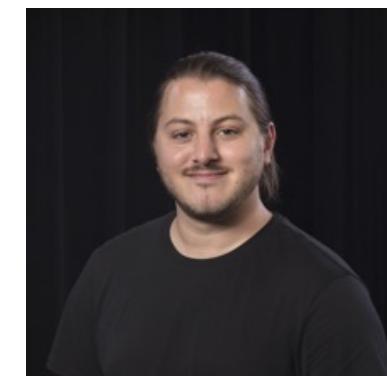
Kevin
Roccapriore



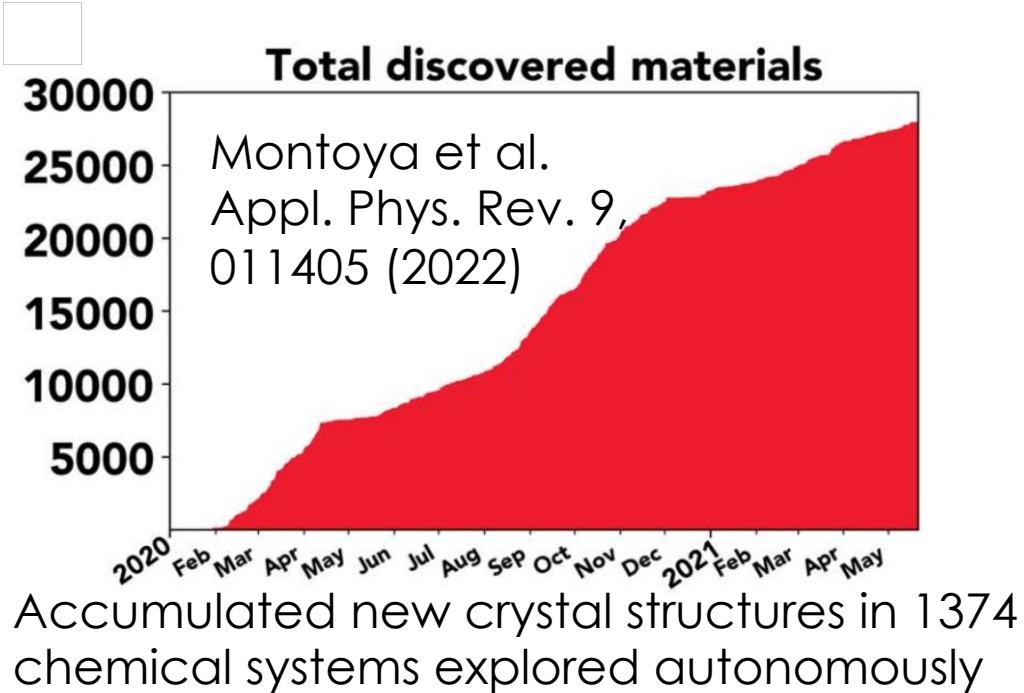
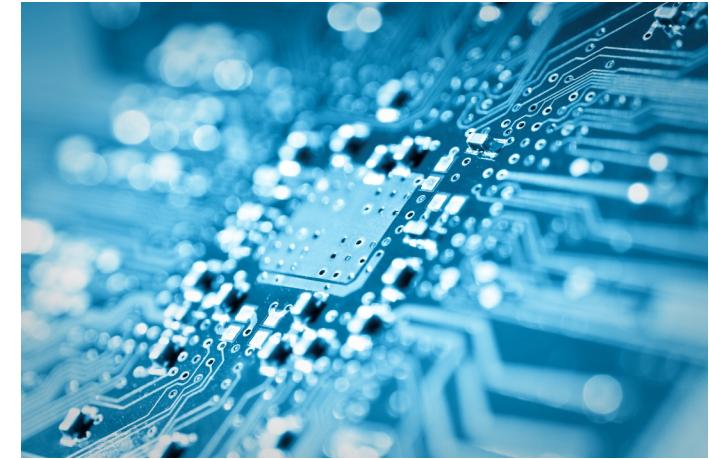
Yongtao Liu



Sumner Harris



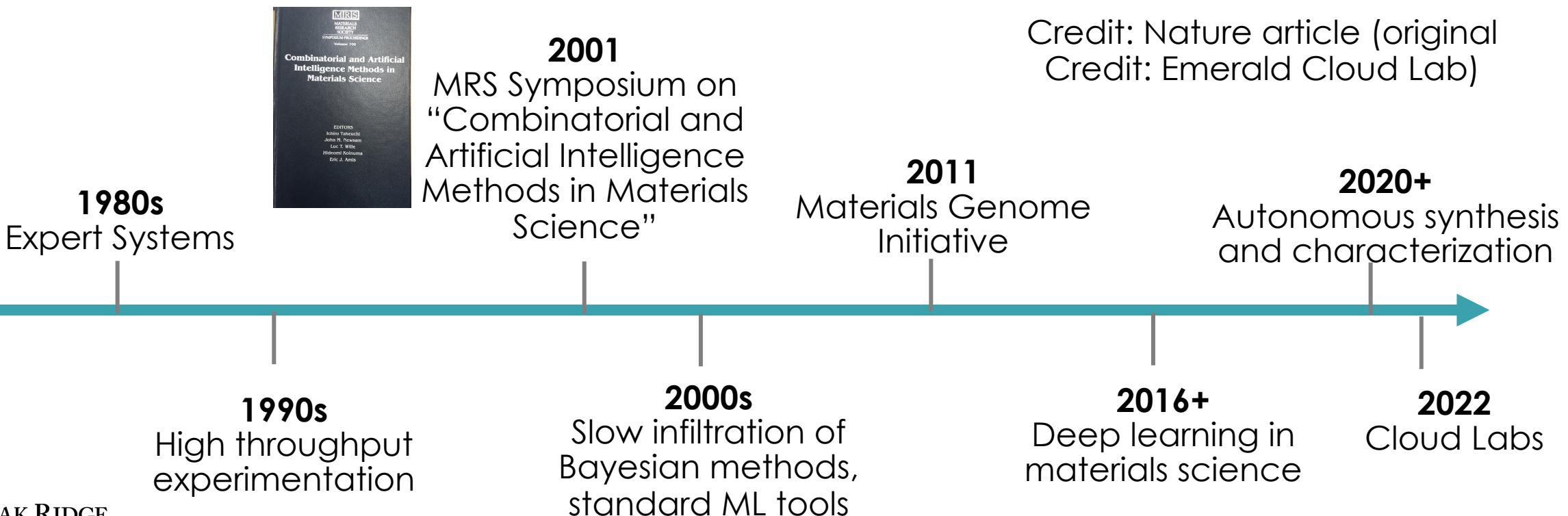
Today's materials challenges: autonomous labs?



- Renewable energy, self-driving cars, transparent displays, new memory technologies, energy harvesting and generation
- Materials become more complex. We can predict many new materials. But we need to (a) synthesize them and (b) understand them, not just optimize for properties.

Machine Learning (in Materials) History

- Compute
- Algorithms
- Connections
- Automation/Abstractions



"Cloud Labs"



Credit: Nature article (original)
Credit: Emerald Cloud Lab

Autonomous = Automated + ‘Smart’

Automated System



+



- Turns on every morning at 6AM
- No ‘intelligence’: preset time.
- Cannot change.

Autonomous System



+



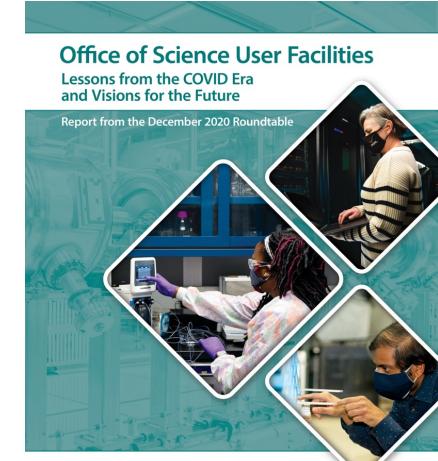
+



- Watering dependent on sensors
- Adaptable
- Reduced wastage

We need autonomous experiments for ...

- New capabilities – new experiments
- Increased efficiency
- Better able to incorporate theory – improves both aspects
- New pathways for synthesis
- Scalability of research
- Autonomous discovery
- Improved User Experience



Article

Autonomous chemical research with large language models

<https://doi.org/10.1038/s41586-023-06792-0> Daniil A. Boiko¹, Robert MacKnight¹, Ben Kline² & Gabe Gomes^{1,3,4,✉}

nature chemical engineering

Article

Autonomous reaction Pareto-front mapping with a self-driving catalysis laboratory

Received: 29 July 2023

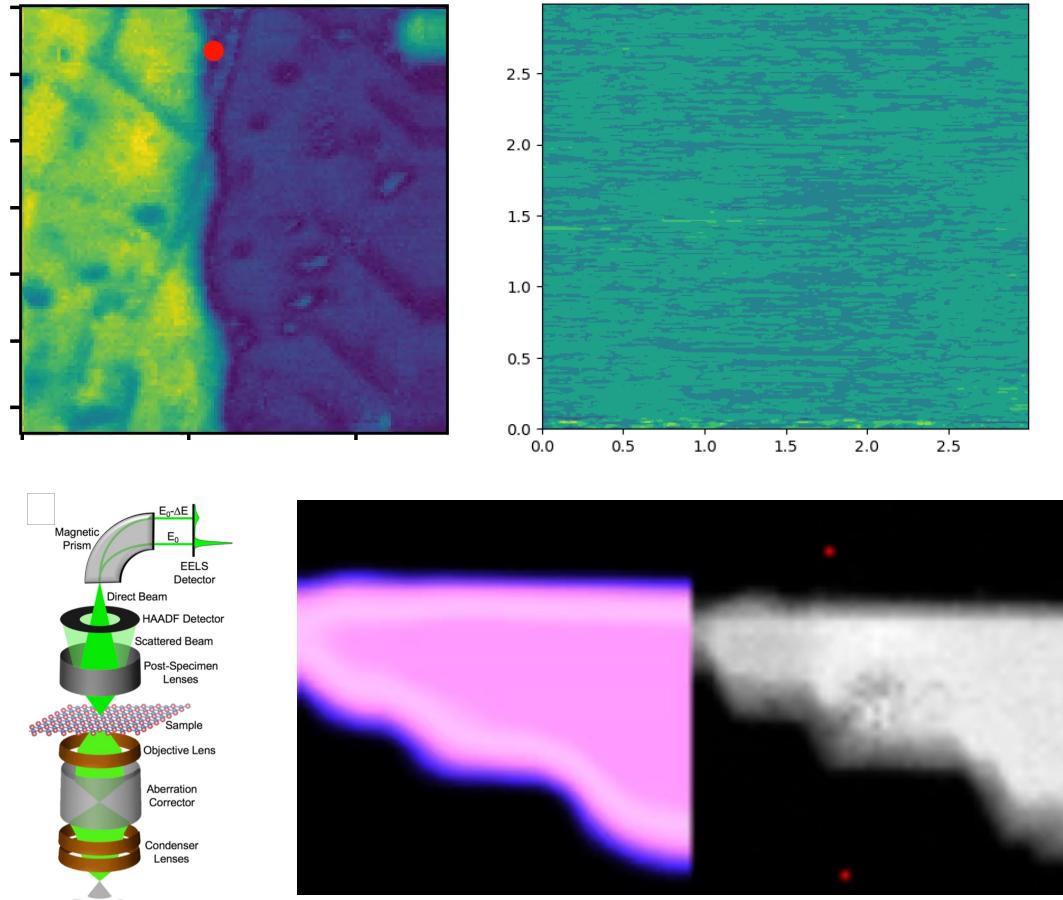
Accepted: 25 January 2024

J. A. Bennett¹, N. Orouji¹, M. Khan¹, S. Sadeghi¹, J. Rodgers² &

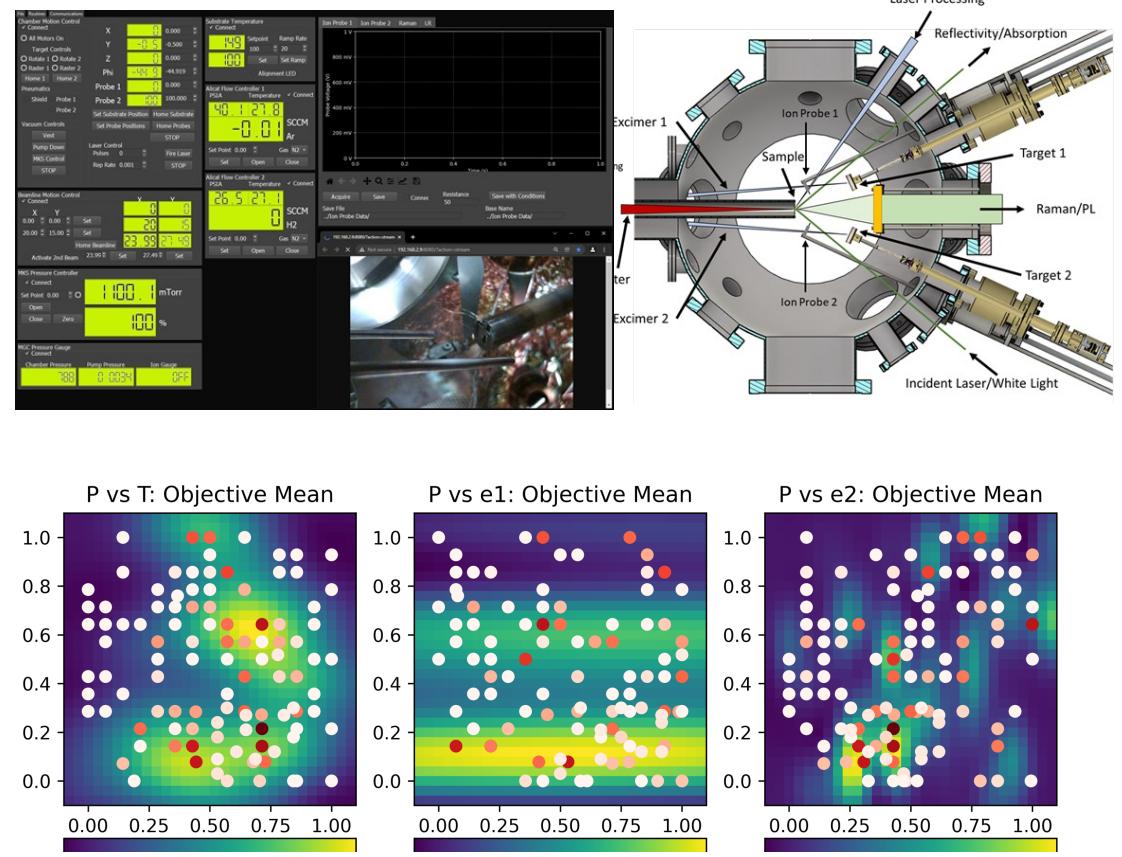
M. Abolhasani¹✉

Outline

Autonomous Microscopy

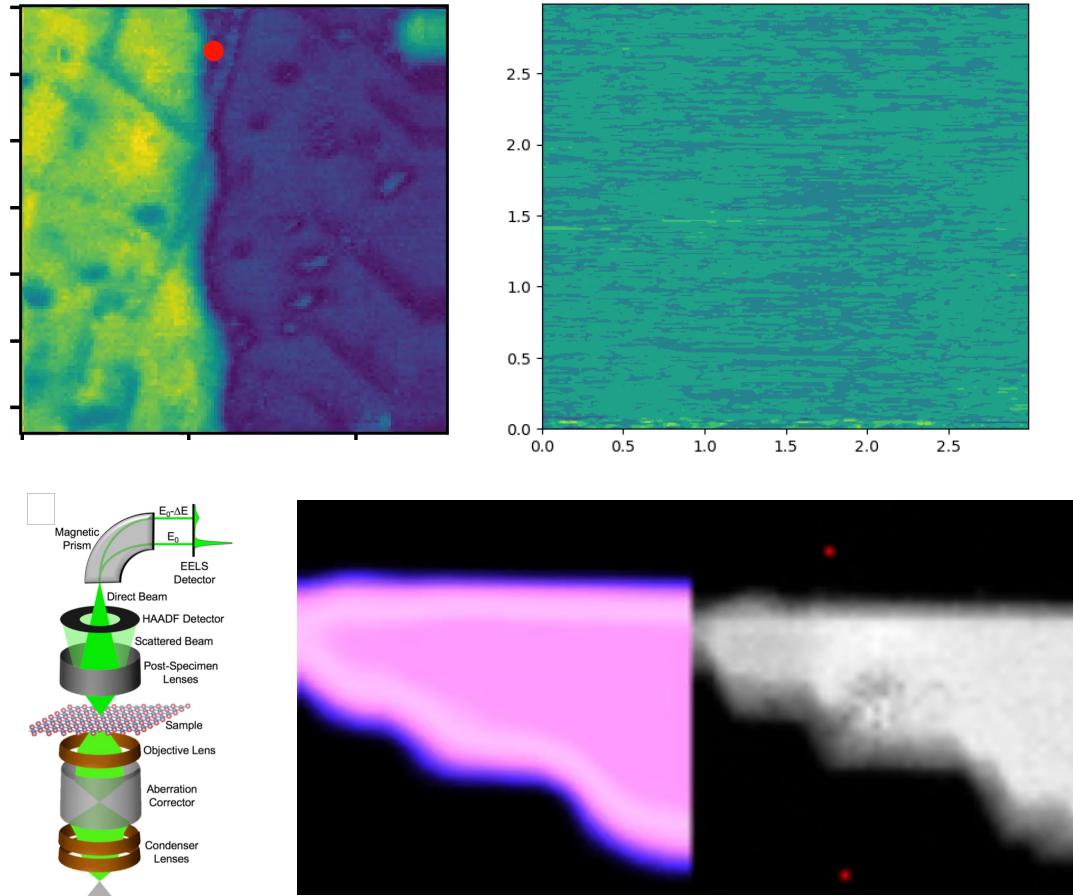


Autonomous Synthesis

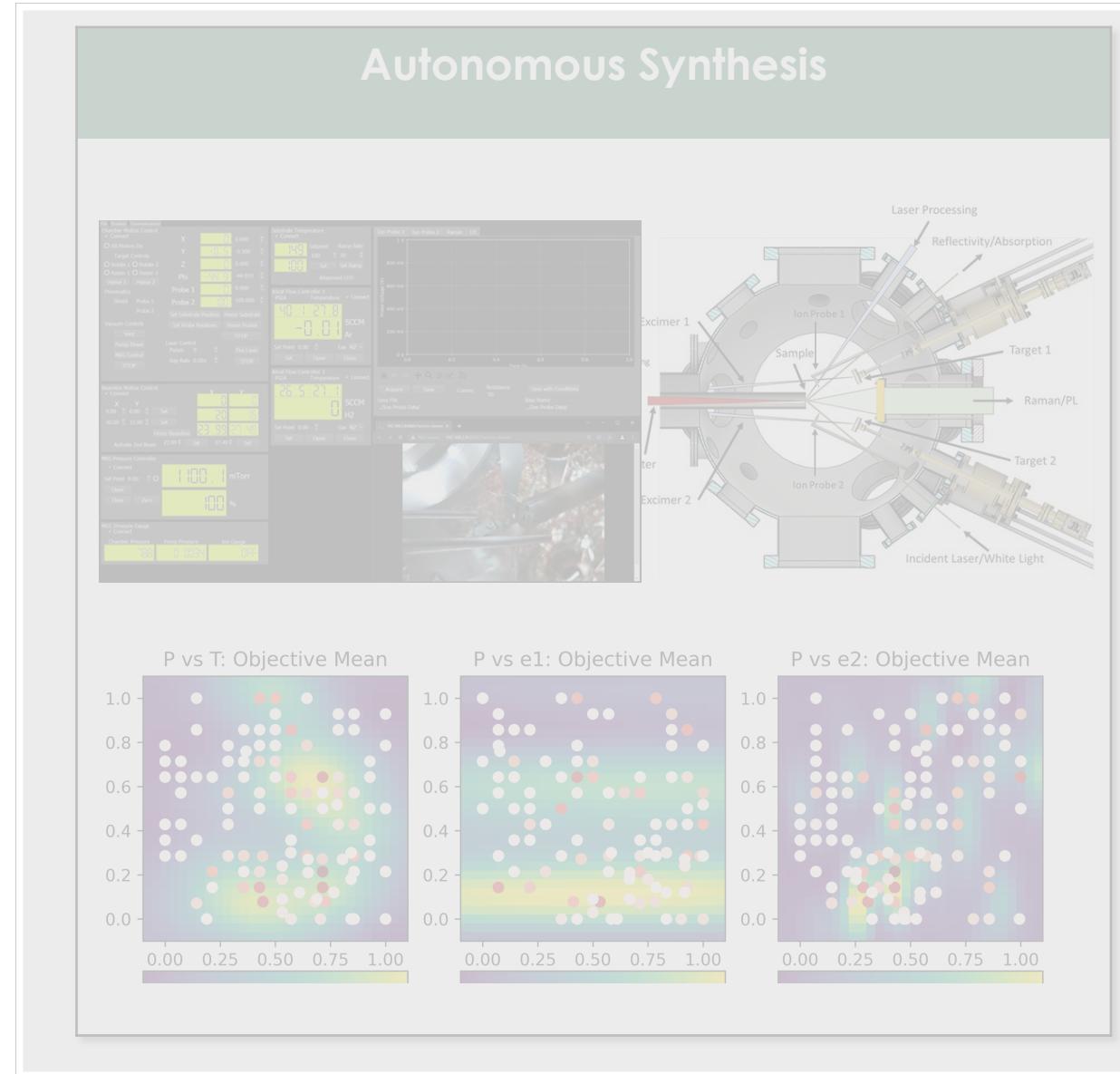


Outline

Autonomous Microscopy



Autonomous Synthesis



Automating SPM: AEcroscopy

Software Infrastructure



Welcome to AEcroscopy

Get Started

Get Started

Experiment

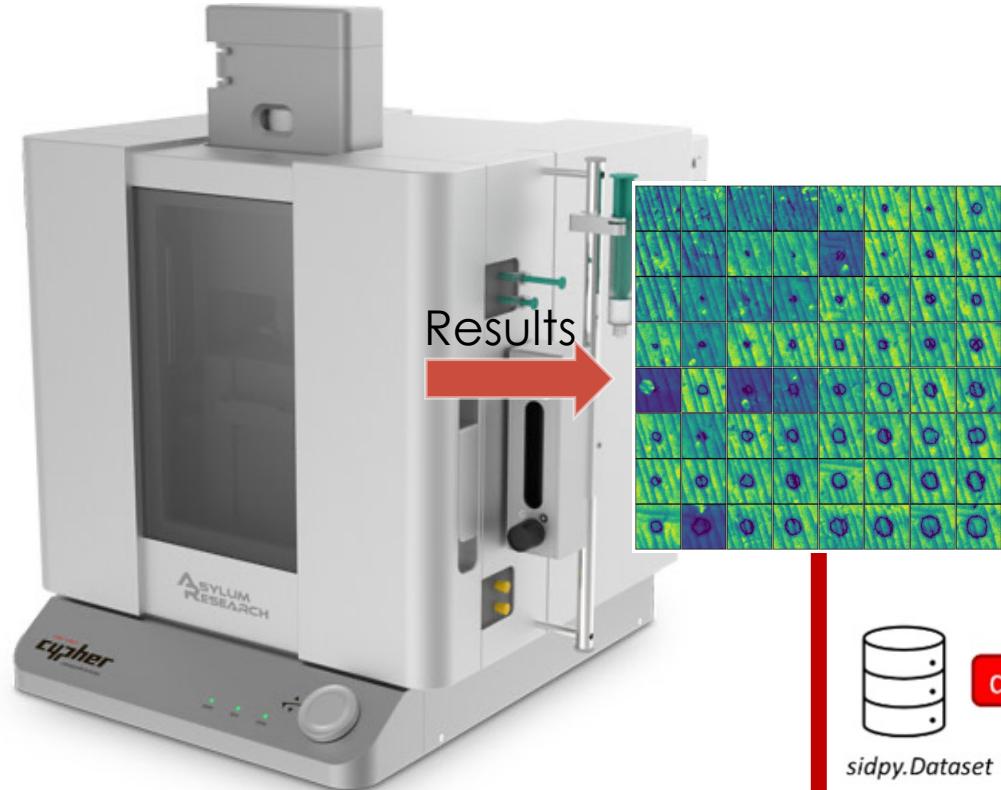
Experiments

```
=  
Step 4. Do a BEPFM at the whole experiment area  
  
dset_pfm, dset_chns, dset_cs = newexp.raster_scan(raster_parms_dict = {"scan_pixel": 32, "scan_x_start": -1.0, "scan_y_start": -1.0, "scan_x_stop": 1.0, "scan_y_stop": 1.0}, file_name = "pfm_whole", ploton = False)  
  
f, (ax1, ax2, ax3, ax4, ax5, ax6) = plt.subplots(1, 6, figsize = (30, 5), dpi = 100)  
ax1.imshow(dset_pfm[:, :, 0])  
ax2.imshow(dset_pfm[:, :, 1])  
ax3.imshow(dset_pfm[:, :, 2])  
ax4.imshow(dset_pfm[:, :, 3])  
ax5.imshow(dset_chns[0, :, :])  
ax6.imshow(dset_chns[1, :, :])  
plt.show()
```

Y. Liu et al. Small Methods 2301740 (2024)

Deploy

Results



sidpy.Dataset

An ecosystem for microscopy data ingestion, analytics and visualization

pycroscopy

A general-purpose package for microscopy imaging and spectroscopy data analytics, including registration, image cleaning, unmixing, etc.

scifireaders For ingesting a variety of microscopy files for output to sidpy dataset objects

pyusid Python package for reading and visualizing our universal spectral imaging dataset format

pynsid Python package for reading and visualizing our N-dimensional spectral imaging dataset format

sidpy Python utilities for storing, visualizing and fitting Spectroscopic Imaging Data

bglib Utilities to analyze, fit and visualize Band - Excitation and G - mode imaging data primarily for CNMS SPM Users

atomai Deep learning toolkit for analysis of atomically resolved imaging and spectroscopy datasets

stemtools Python based codes for analysis of 4D-STEM and aberration - corrected vanilla STEM datasets

pytemlib Python tools for simulation, registration, analysis and visualization of TEM datasets

sidpy 0.11.2

Search docs

SIDPY Installation Getting Started Tutorials on Basics Guidelines for Contribution Upgrading from Matlab Contact us sidpy EXAMPLES Basic usage Parallel computing Visualization HDFS Tools

sidpy.hdf.hdf_utils.get_attr

sidpy.hdf.hdf_utils.get_attr(h5, object, attr_name)

Returns the attribute from the h5py object

Parameters: h5: object (h5py.dataset, h5py.Group or h5py.File) – object whose attribute is desired attr_name (str) – Name of the attribute of interest

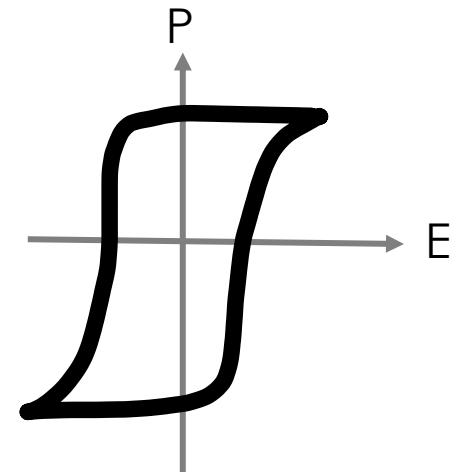
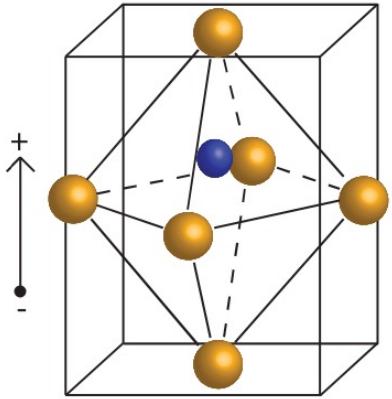
Returns: att_val – value of attribute, in certain cases (byte strings or list of byte strings) reformatted to readily usable forms

Return type: object

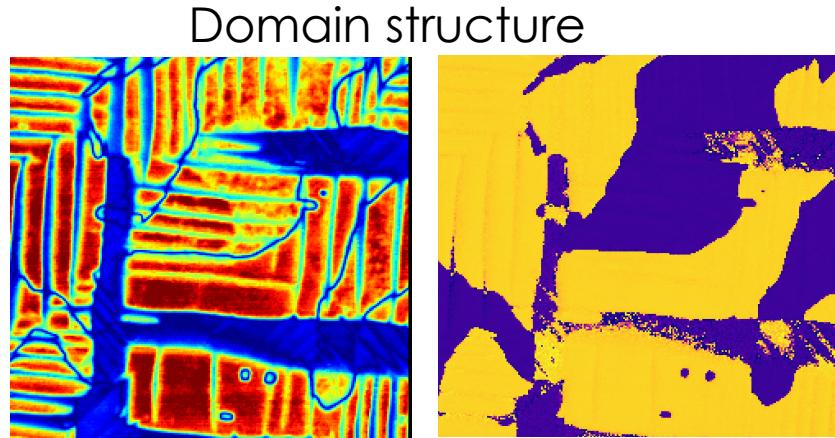
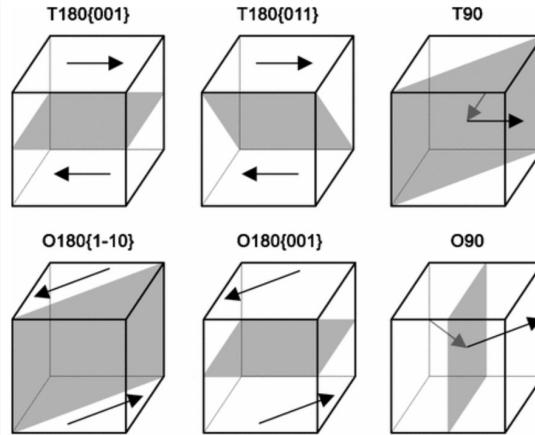
Vasudevan et al. Advanced Theory and Simulations 6, 2300247 (2023)

- Standardized data model
- In-built processing and viz utilities

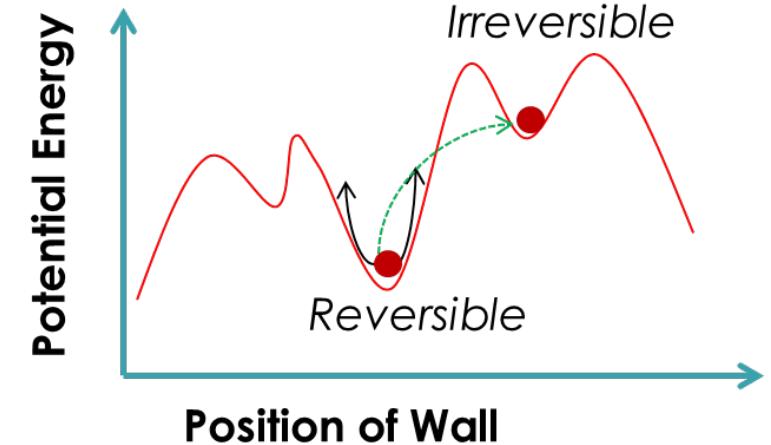
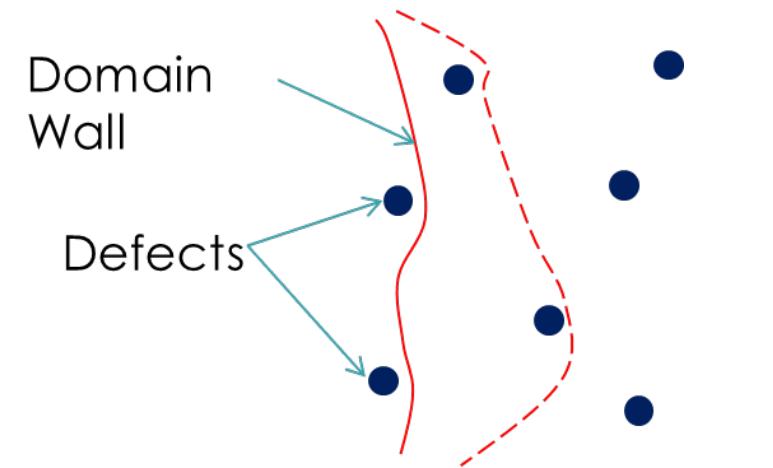
Science Problem: Domain wall dynamics in ferroics



Some wall types



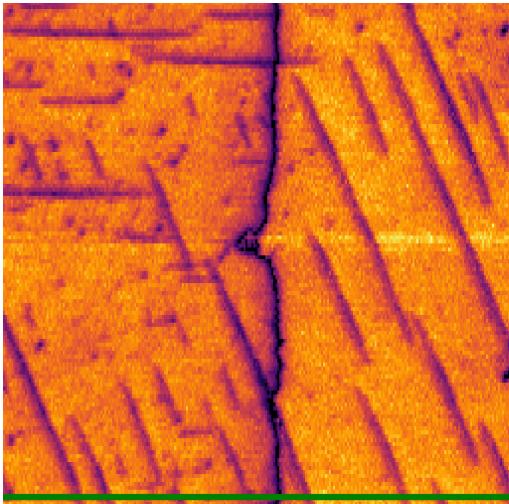
- Spontaneous electric polarization which can be reoriented by electric field (P-E "loop")
- Usually contain domains separated by domain walls



To what extent are the pinning centers mobile/static? What exactly is sufficient to pin the wall? How can we measure pinning strength? Are these transitions visible?

Automated experiment for probing domain wall energetics

PFM Amplitude (2um x 2um)



Problem:

How does the local domain structure impact the wall pinning?

Design of experiment:

Scan individual lines and step up the voltage applied to the domain wall. Move onto next line. Repeat

Mani Valetti
(UT-K)



Experimental Workflow

Pole a domain wall in the center of the image

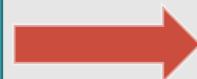


Image a single line (SSA disabled)

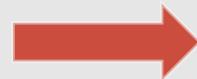
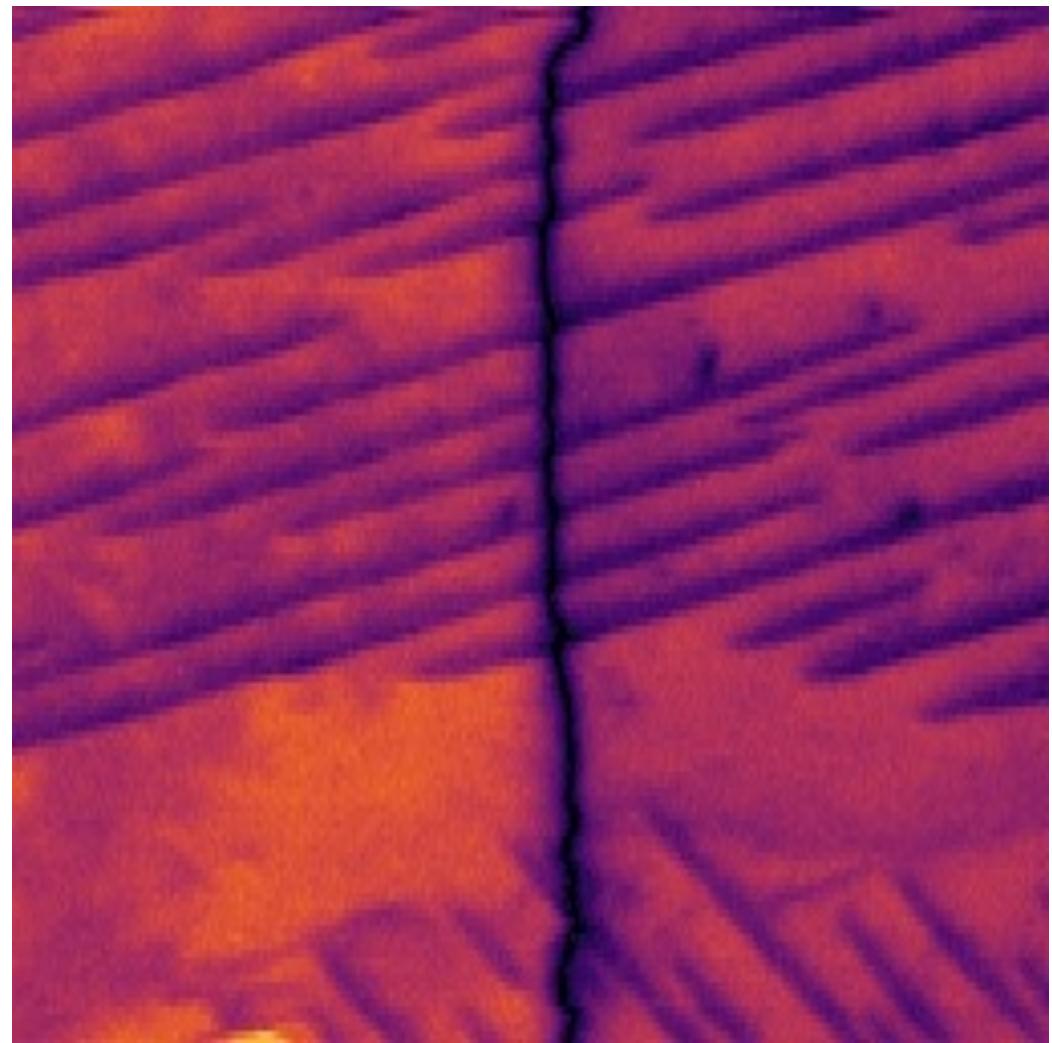
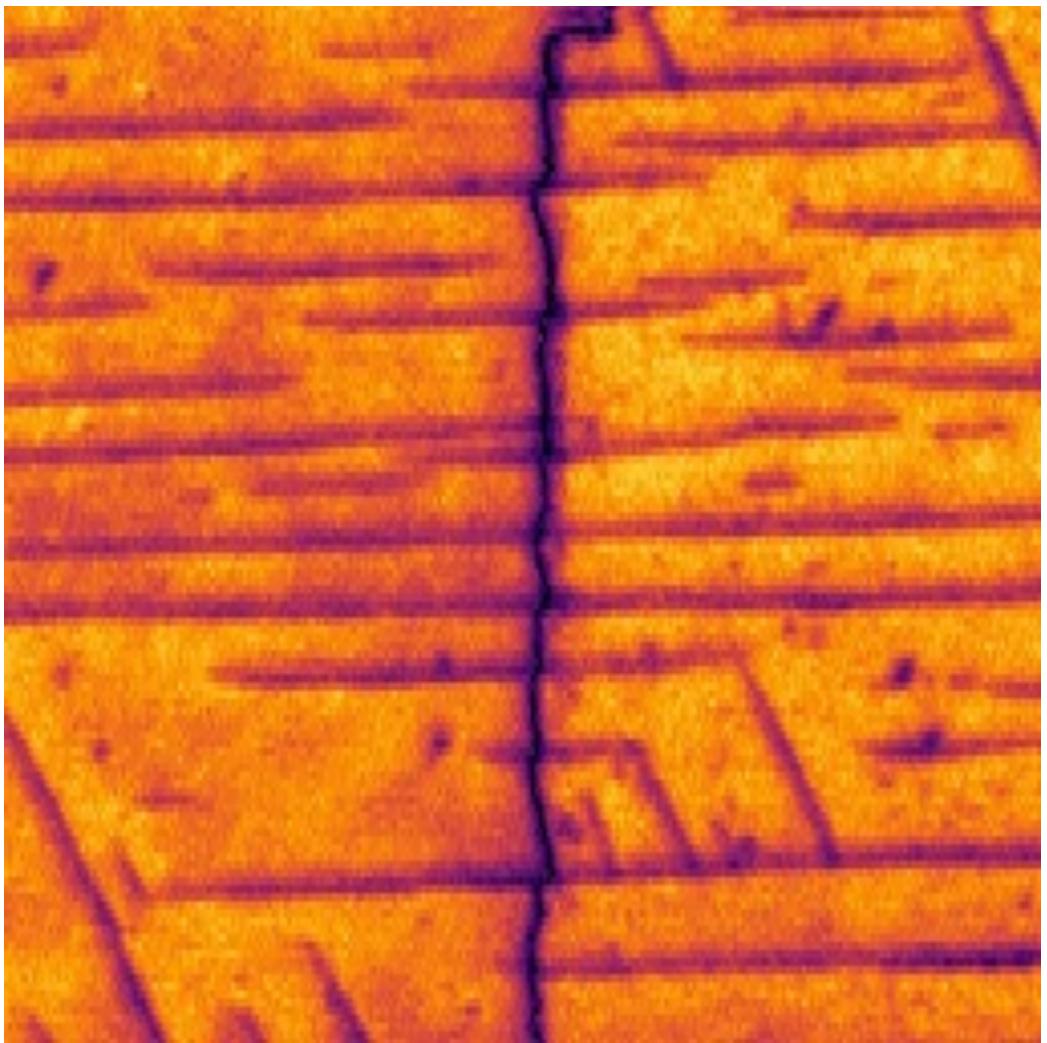


Image the line, **locate wall**, apply bias pulses at the wall. Repeat.

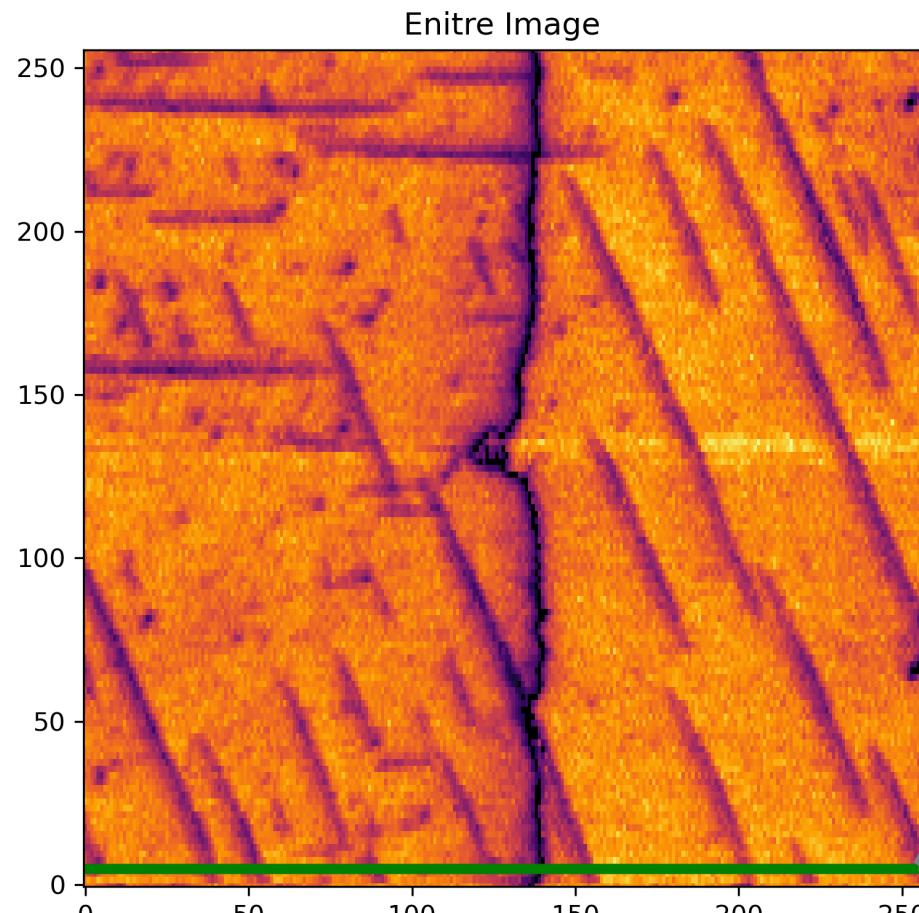


Move up to the next line

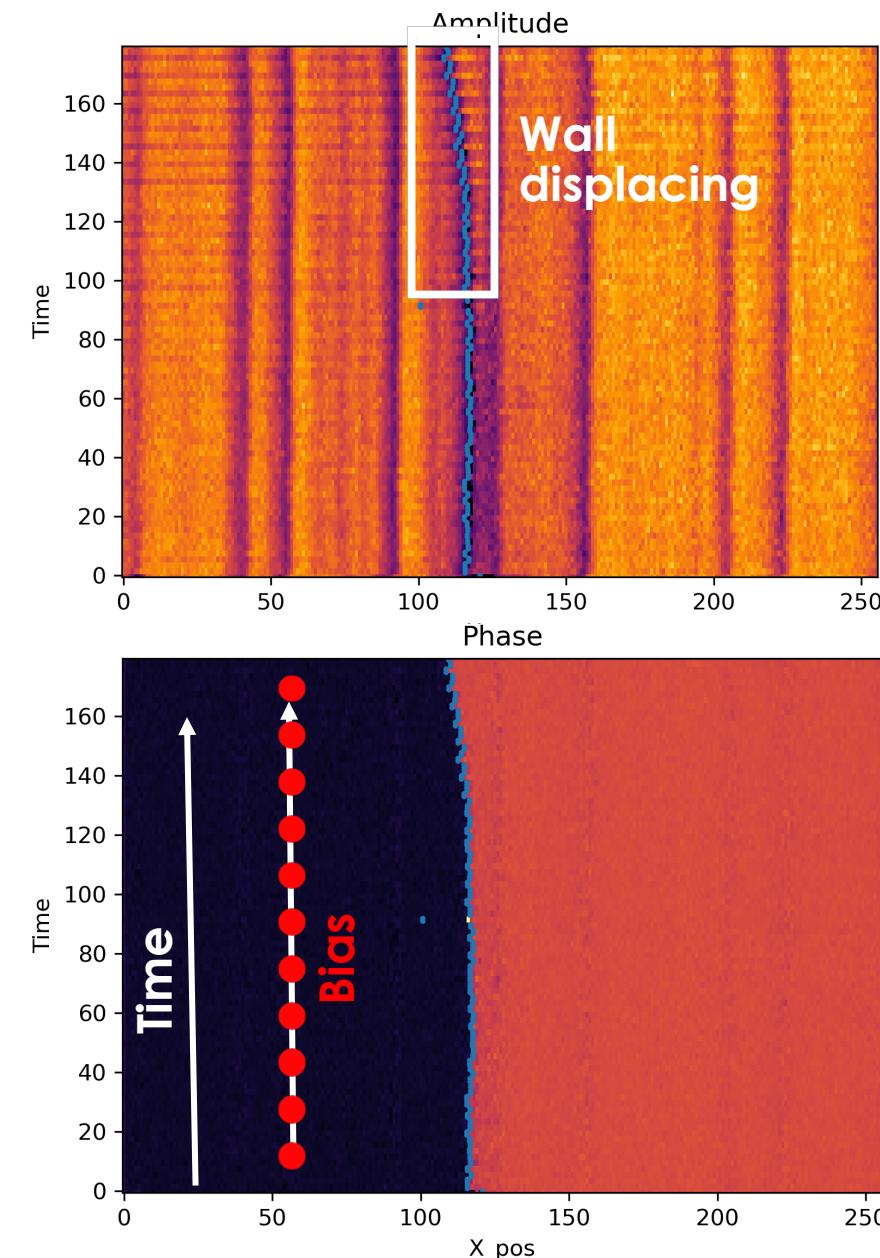
Example datasets



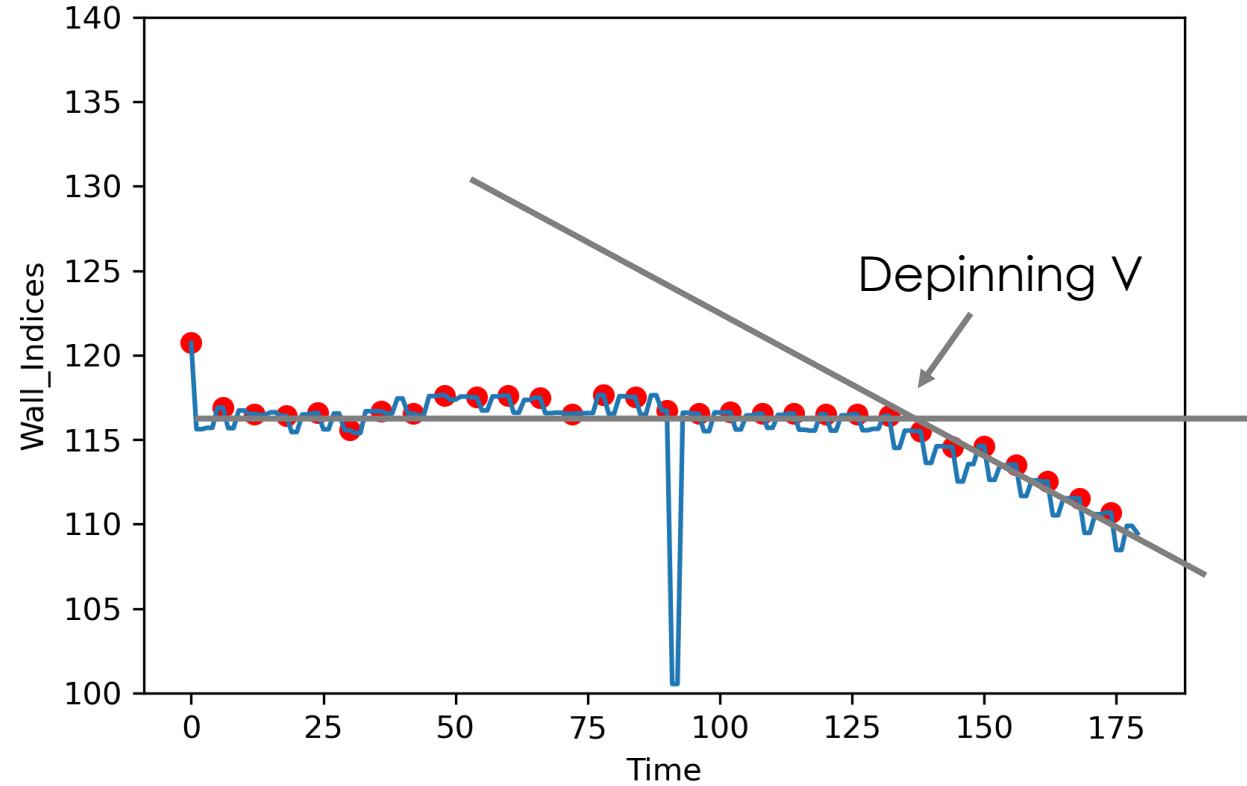
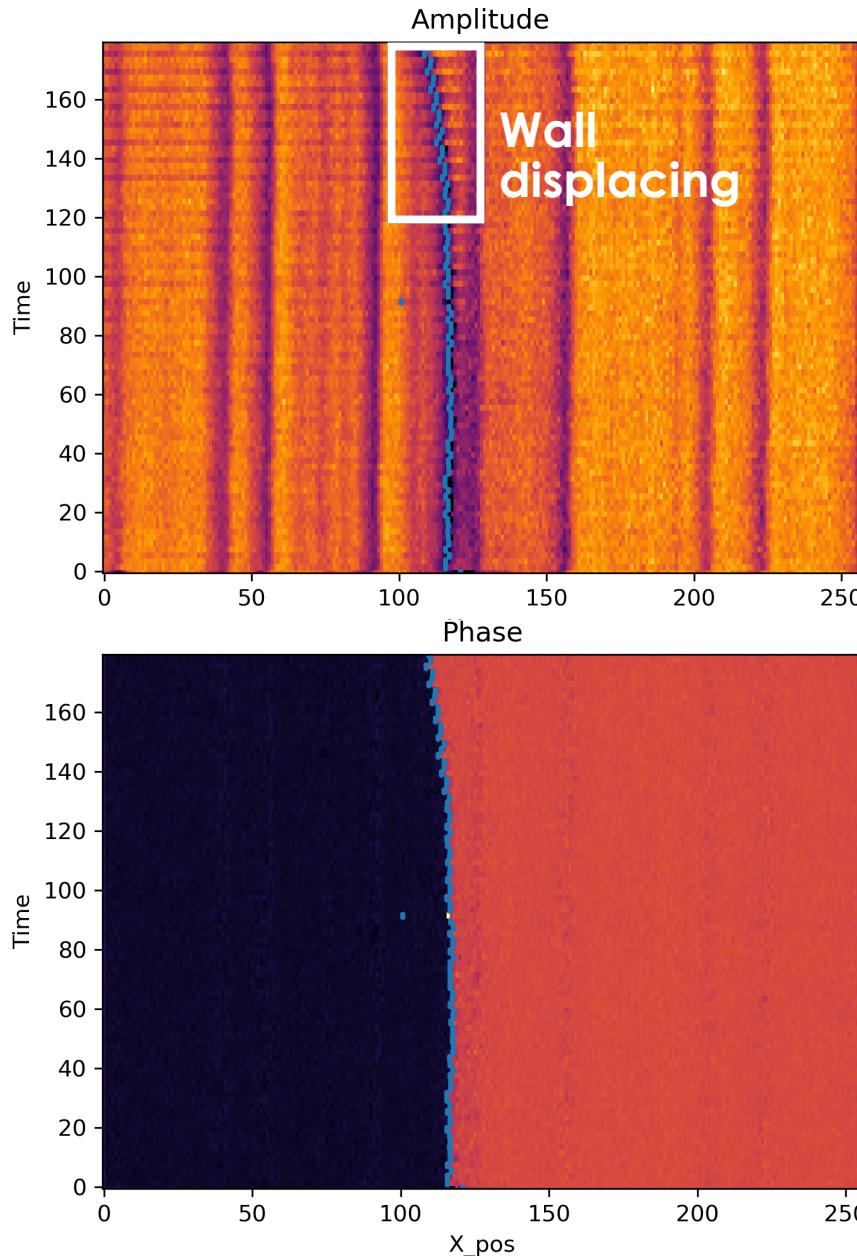
Automated experiment for probing domain wall energetics



Sample from J. C. Yang (NCKU/Taiwan)
Work by S. Mani Valleti (UTk /ORNL)

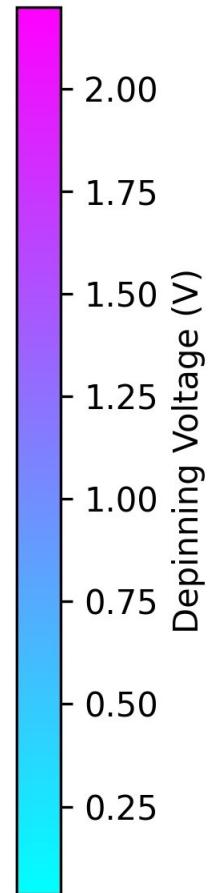
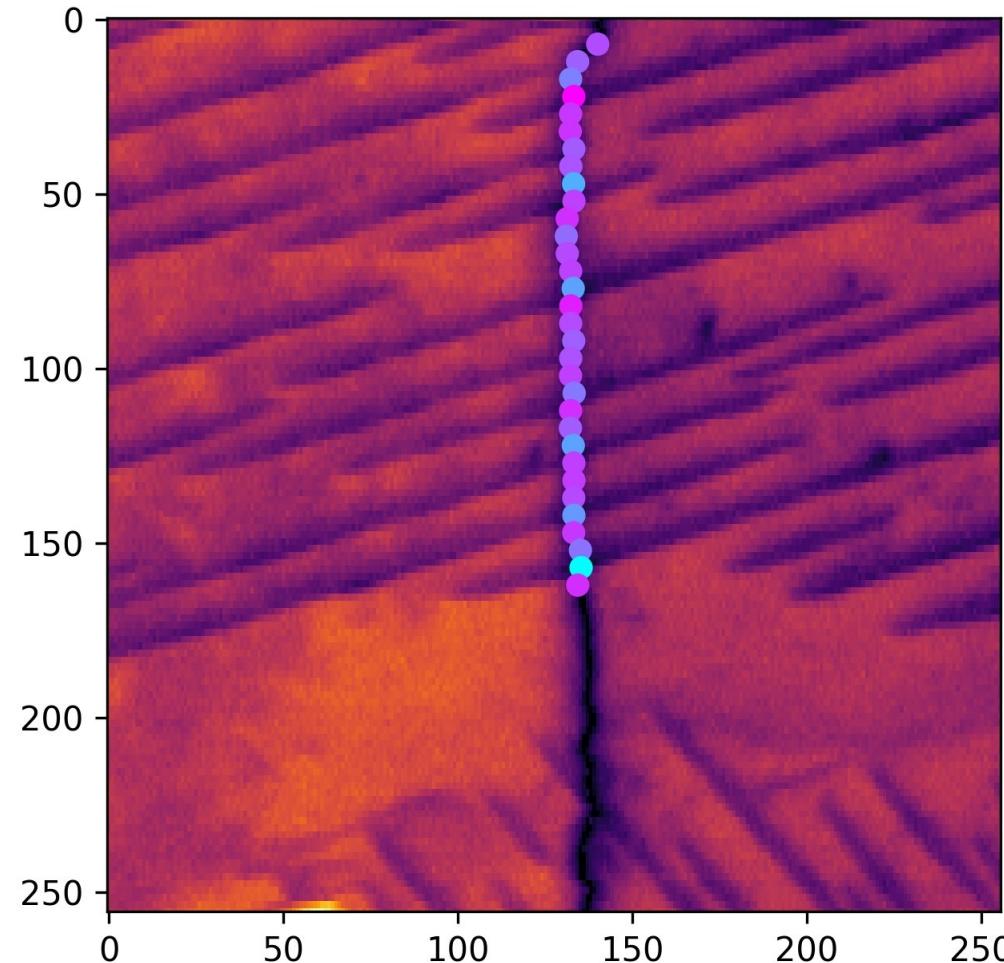
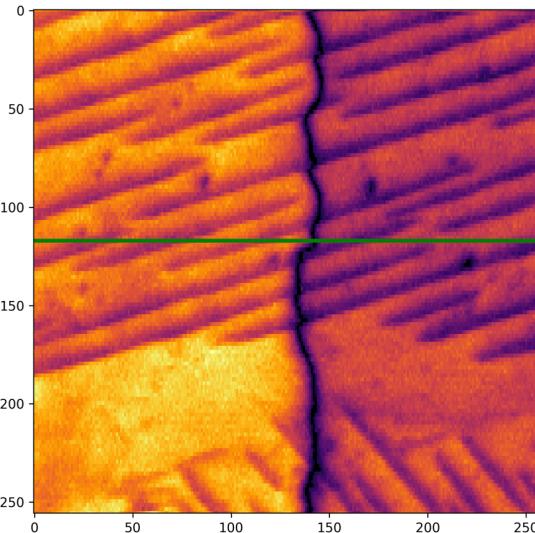
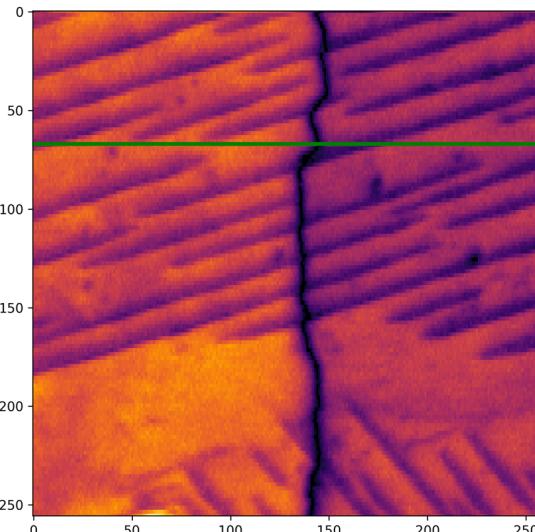


Automated experiment for probing domain wall energetics



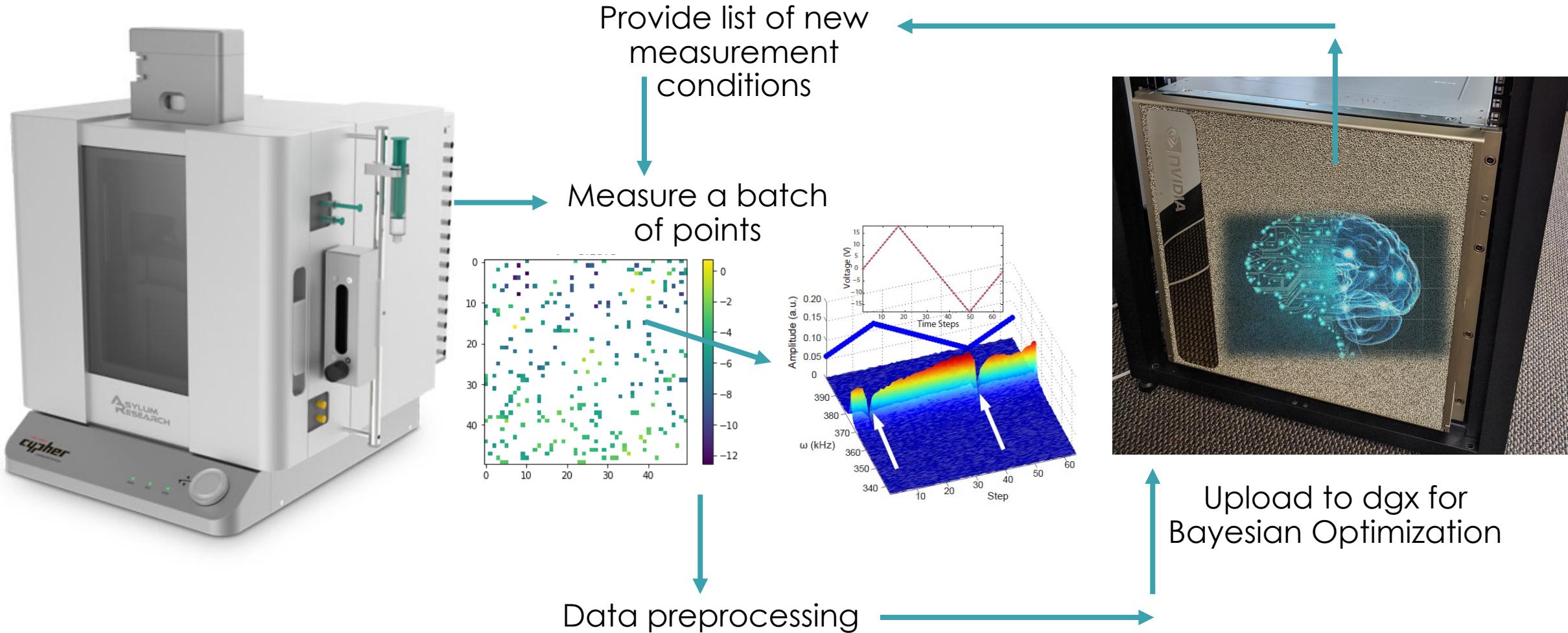
- Snap back is indicative of wall tension
- Working on statistics of snap distances
- Depinning voltage clearly visible

Individual Lines



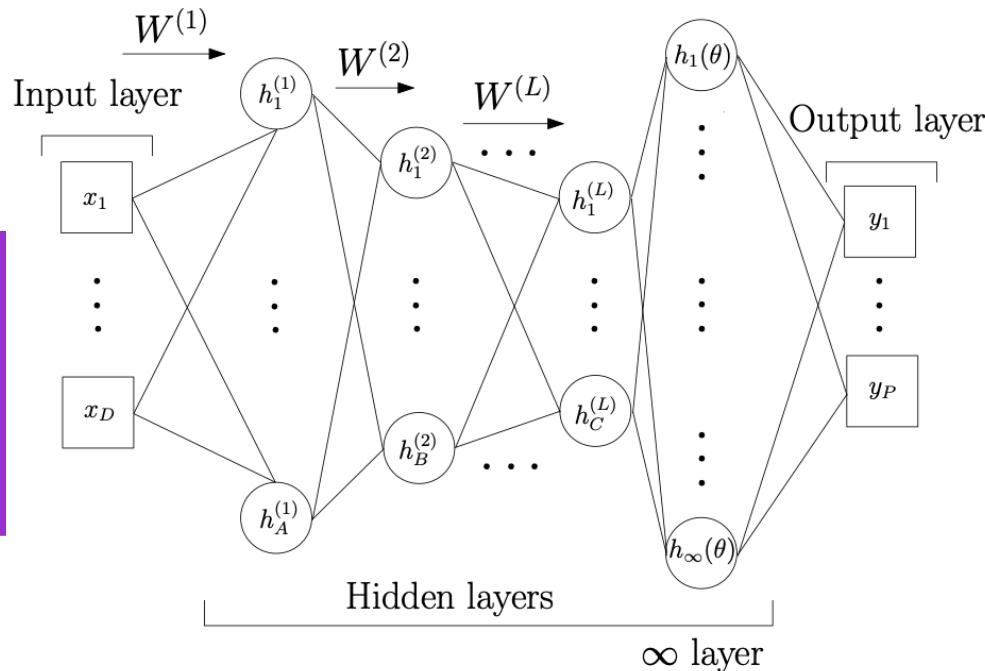
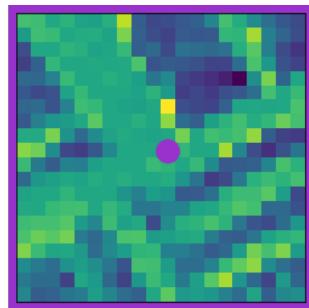
Beyond simple automation: Bayesian Optimization

- Large spectroscopic datasets take too long to capture: efficiency in sampling required. Can be done via Bayesian optimization.

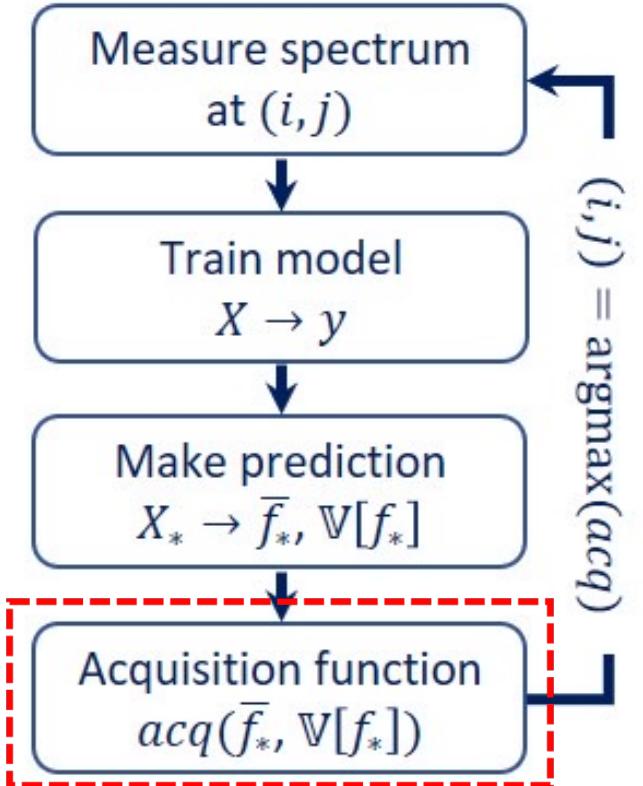
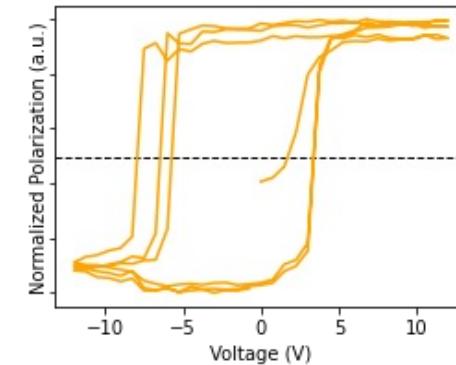


Deep Kernel Learning: More images, better kernels

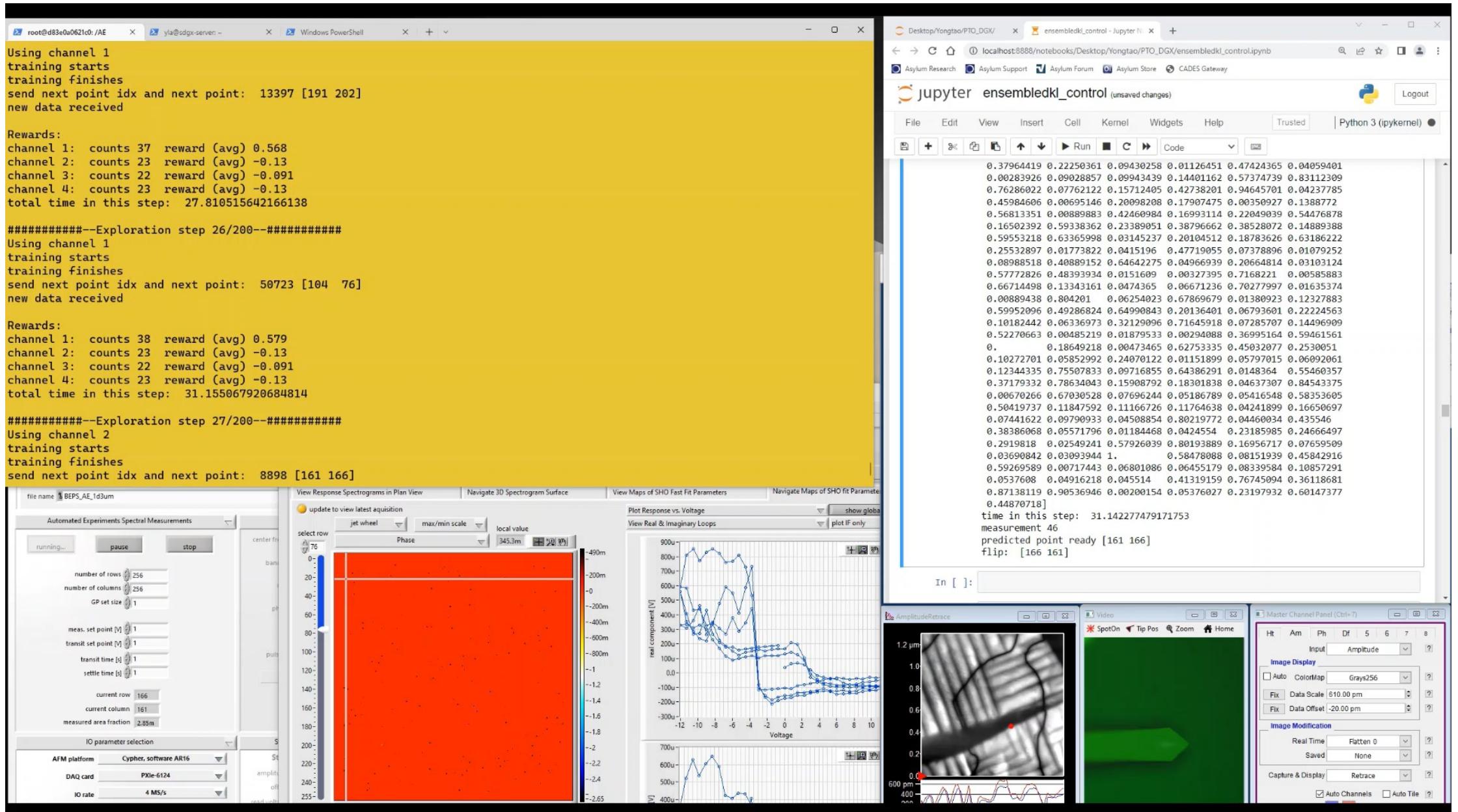
Deep Kernel Learning
(A.G. Wilson, 2015)



Combine a neural network with GP, learn parameters jointly

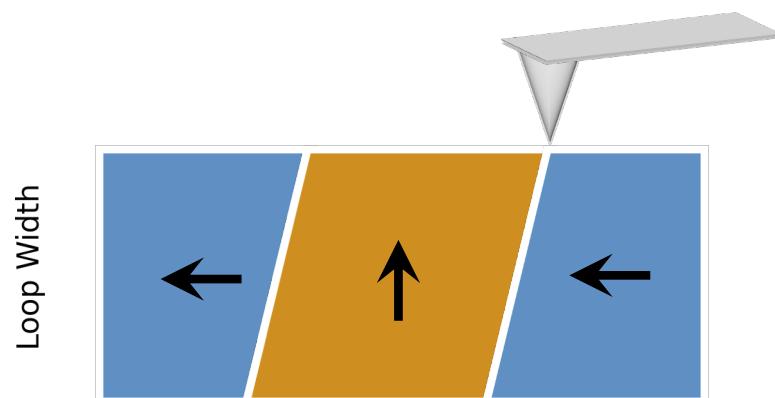
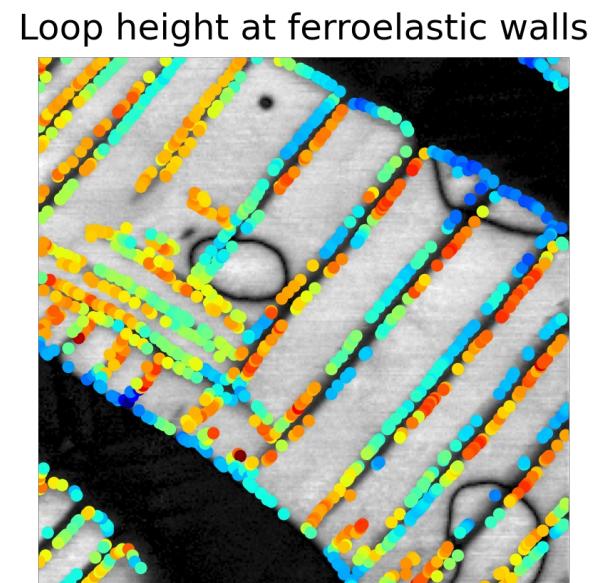
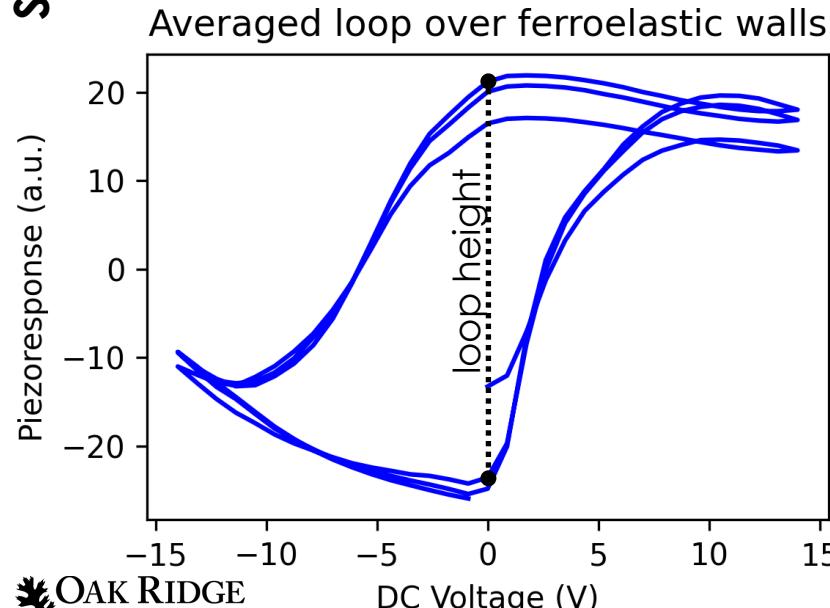
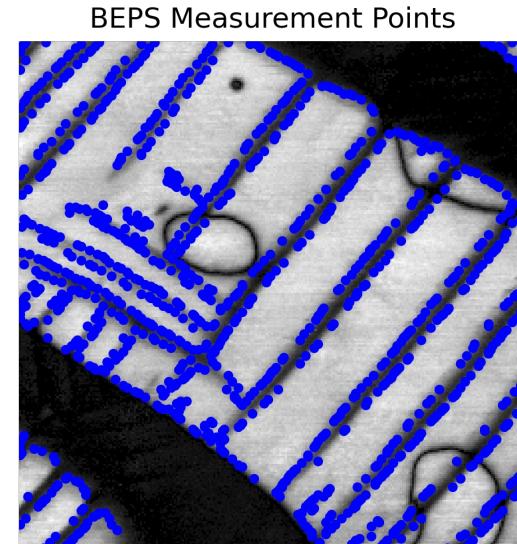
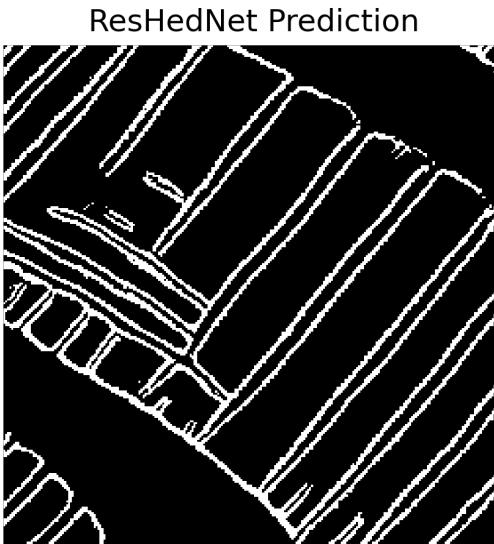
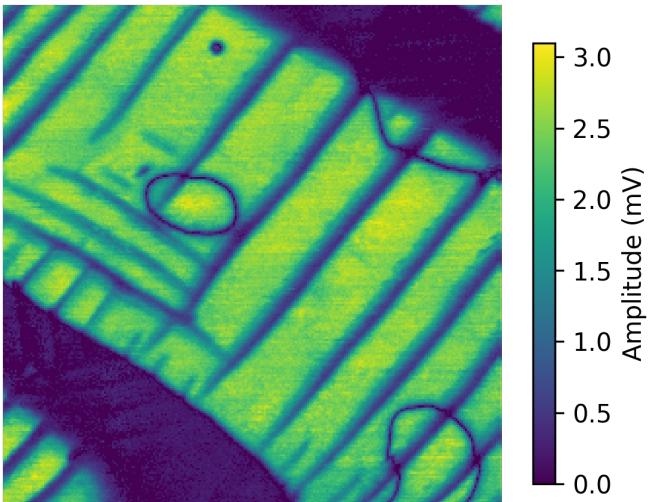


Liu, Yongtao, Kyle P. Kelley, Rama K. Vasudevan, Hiroshi Funakubo, Maxim A. Ziatdinov, and Sergei V. Kalinin. Nature Machine Intelligence 4, 4 (2022): 341-350.

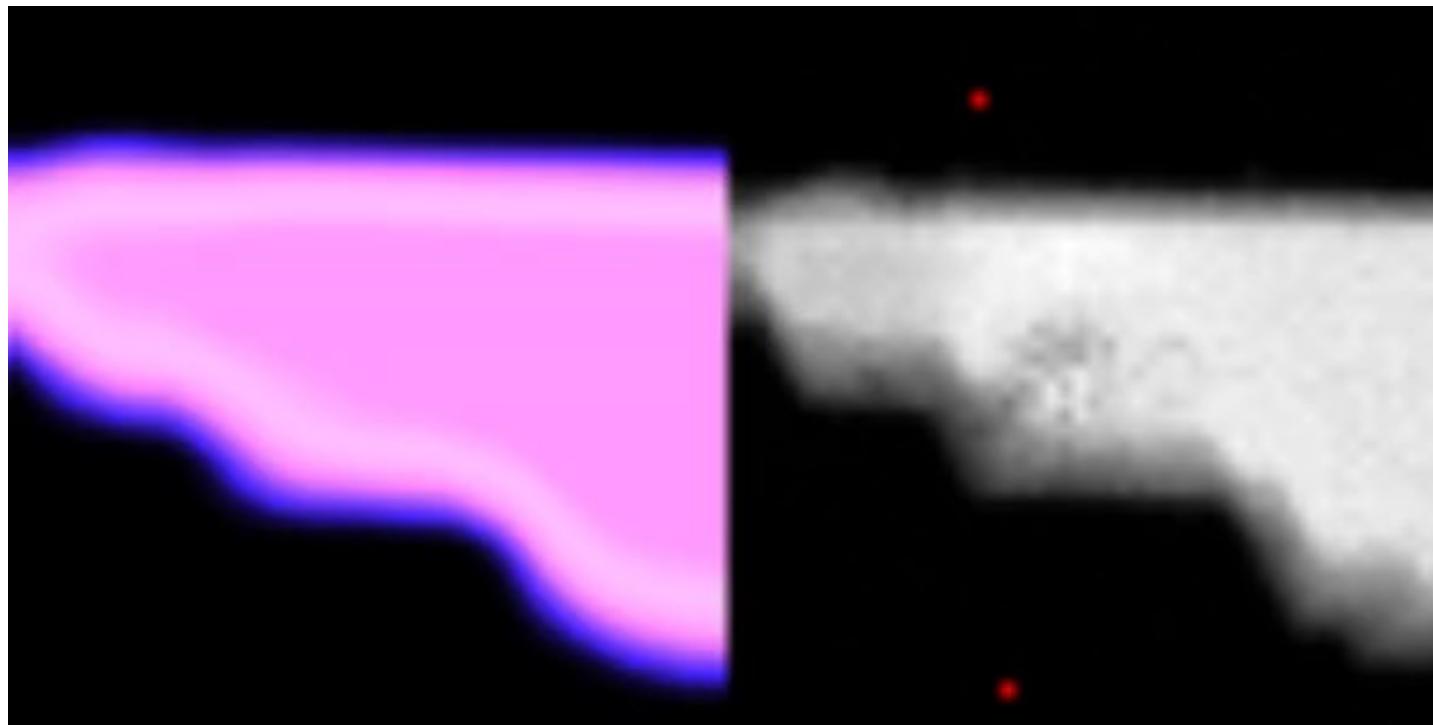


Automated SPM—Objects of Interest

Slide by Yongtao Liu ORNL



STEM Bayesian Optimization



- Bulk and edge plasmons in MnSP₃ investigated via deep kernel active learning
- Optimize for ratio of peaks to find where edge plasmon is strongest
- Automated structure-property relationship determination

K. Roccapriore et al., Adv. Sci. 2203422 (2022)

Beyond Simple Bayesian Optimization

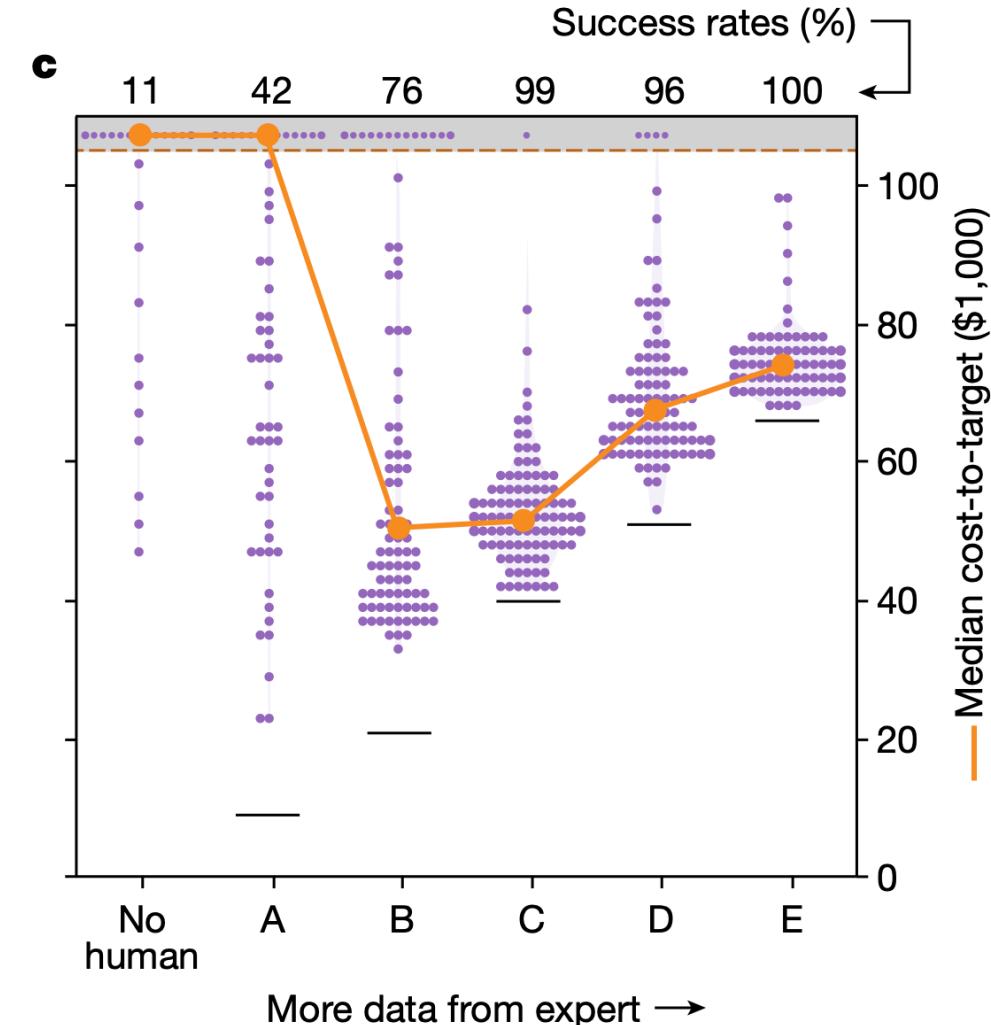
- There is a significant issue with traditional BO based methods
 - We need a scalarizer function to be known *apriori*
- Two solutions
 - Use recommender system to learn the scalarizer
 - Use the notion of curiosity and optimize for it

Algorithms: Human in the Loop

- Human-AI systems work better together than either alone
- Prior knowledge injection can be used to shape targets, reduce wastage, can be informed by simulations
- Increase AI alignment, monitor autonomous progress. Develop better metrics for monitoring autonomous experiments, and better methods for human input

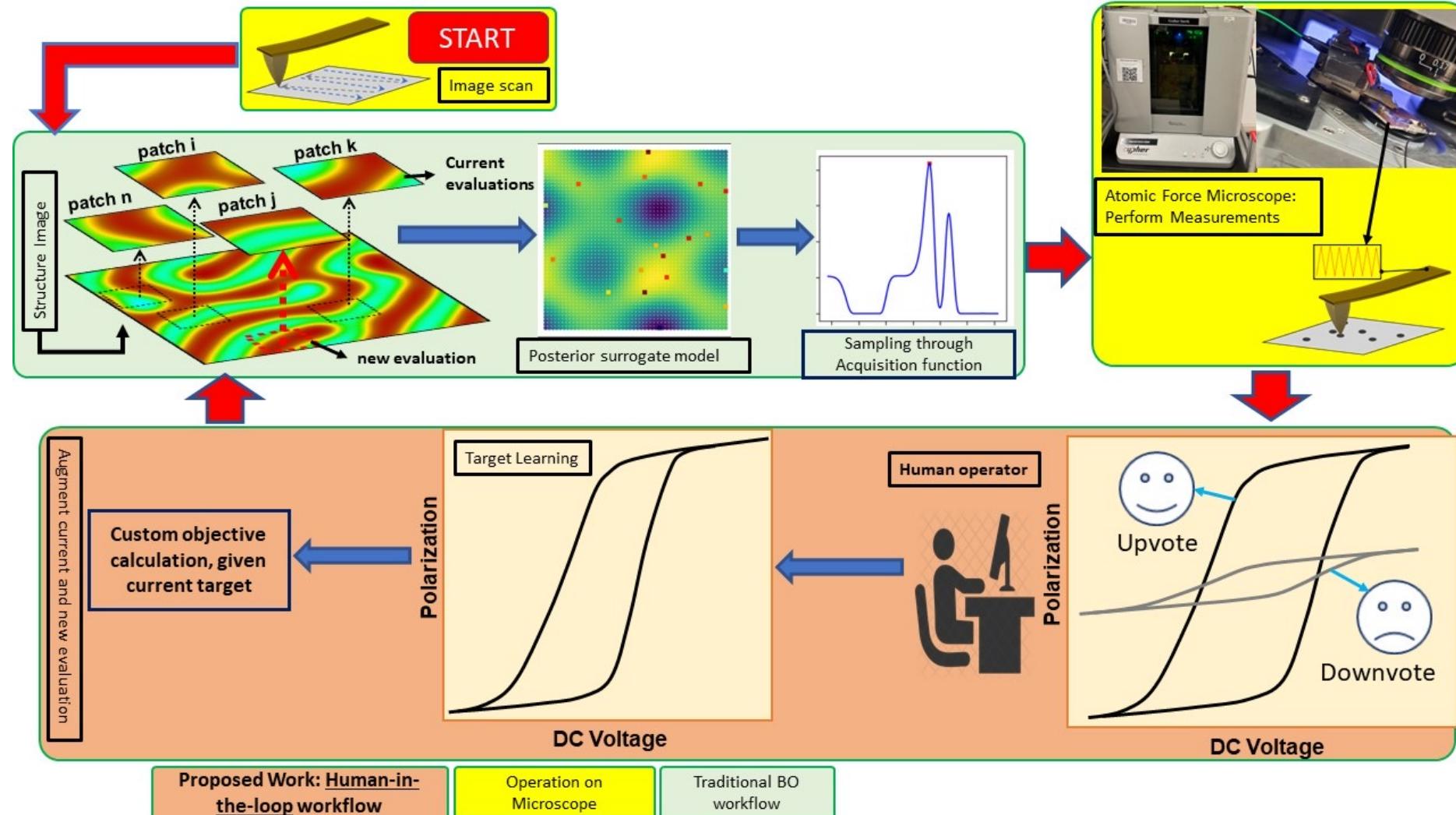
Article

Human-machine collaboration for improving semiconductor process development



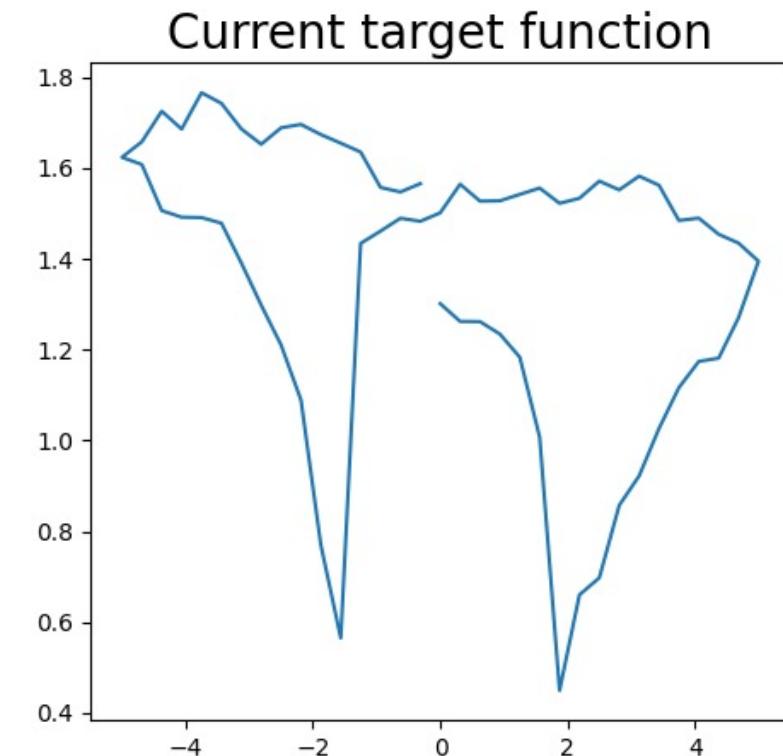
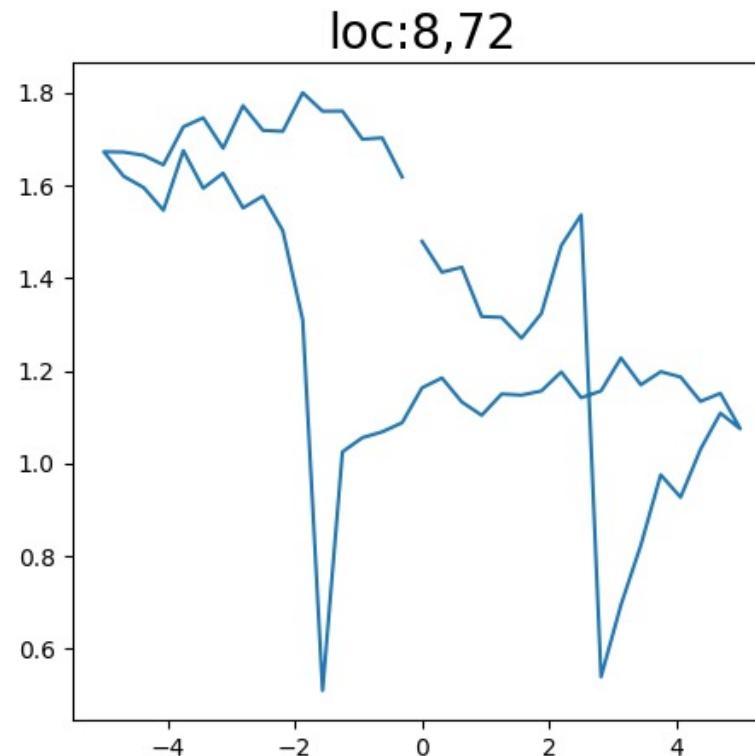
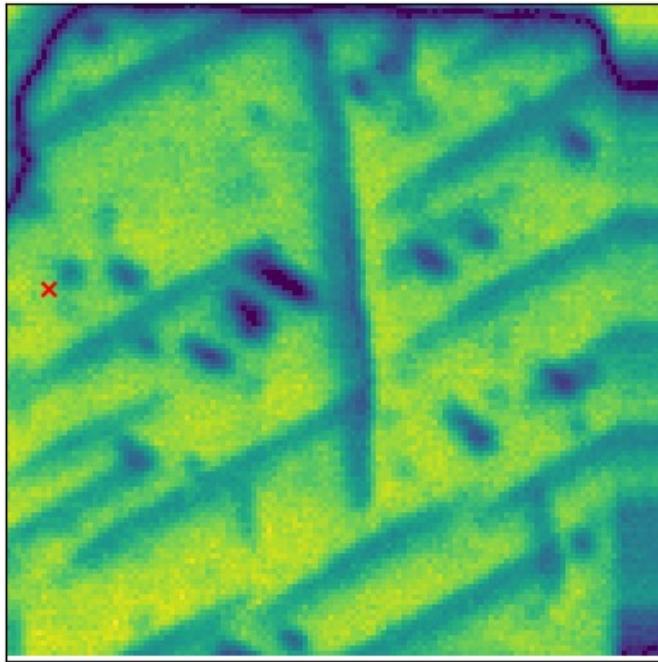
Kanarik et al. Nature **616**, 707 (2023)

Learning what to optimize: reccomender systems



A. Biswas et al. npj Comp. Mater. 10, 29 (2024)

Spectral Recommender: When no Target will suffice



User gets to rate spectra (in this case 15 spectra were rated)

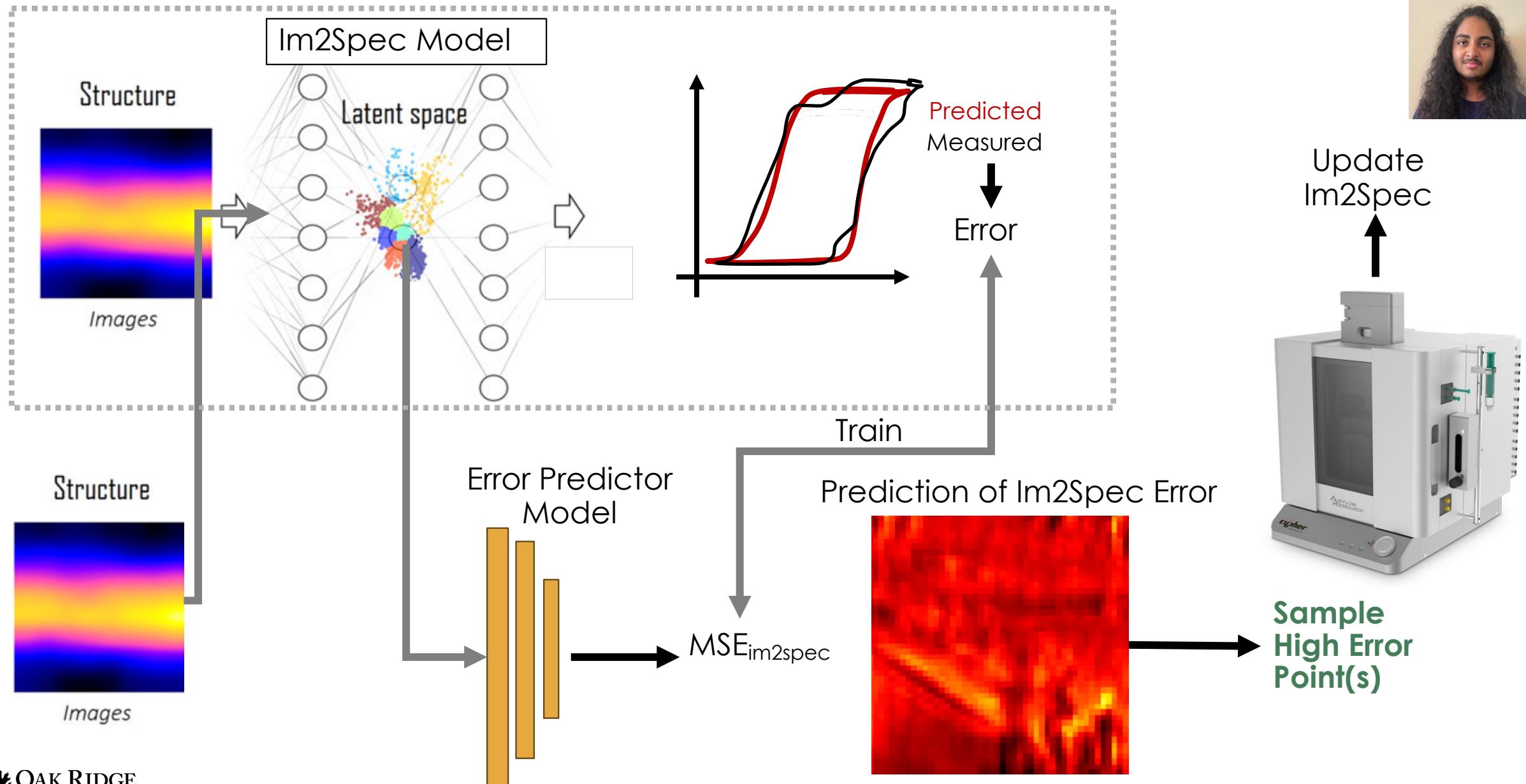
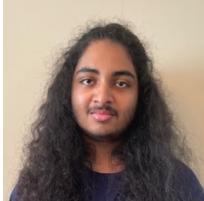
Sample by J. C. Yang (NCKU/Taiwan)

Spectral Recommender system

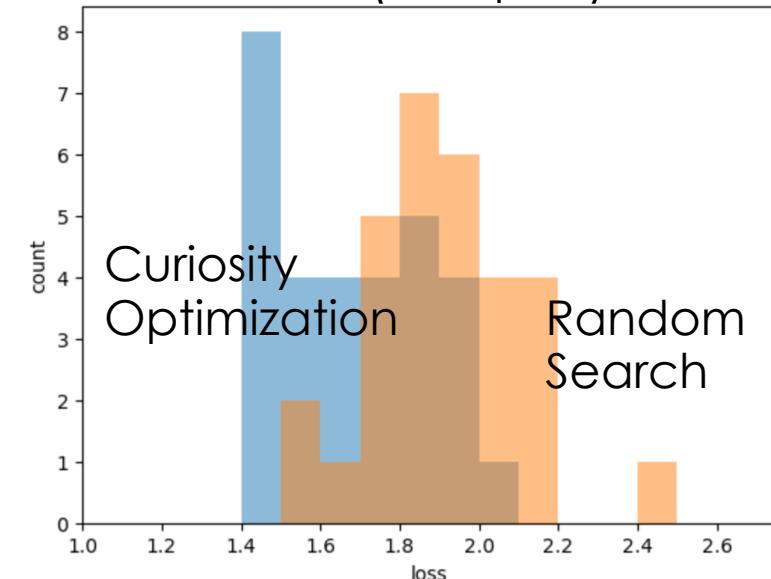
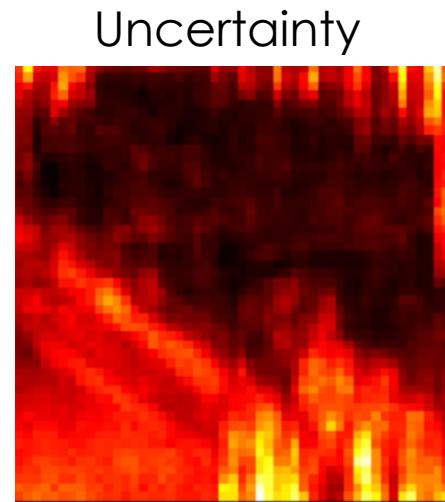
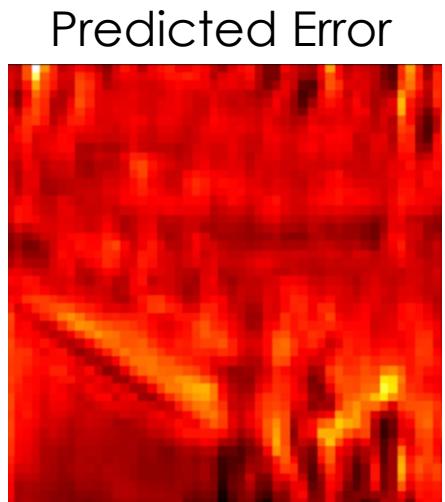
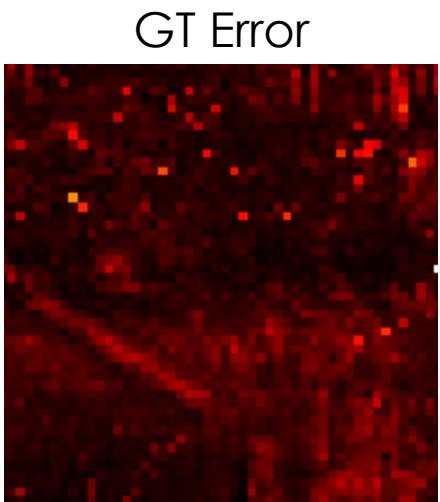
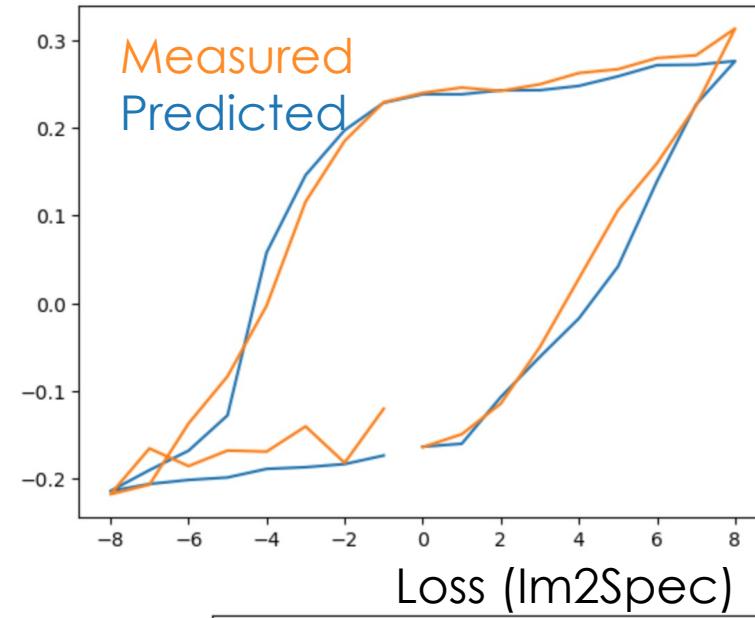
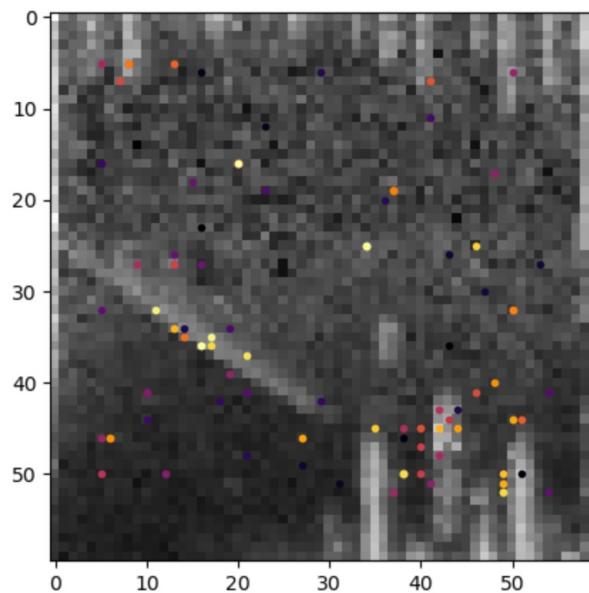
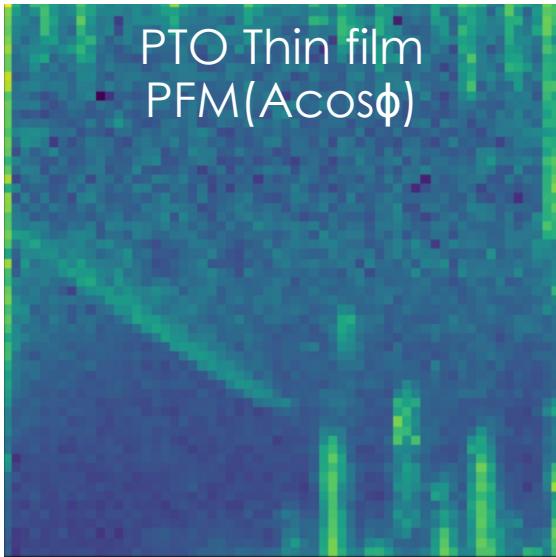


Another alternative: “Curiosity” optimization

Adi Vatsavai
UNC

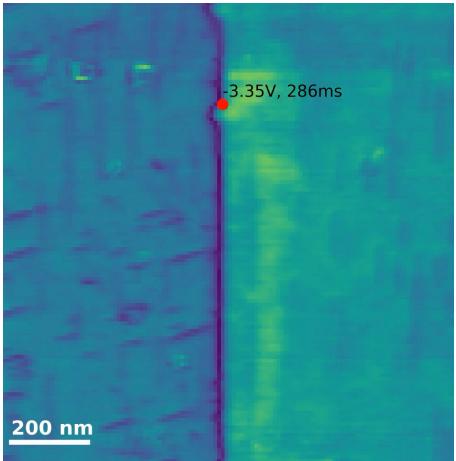


Error prediction model enables efficient sampling!



AI Improves Mesoscopic and Atomic Manipulation

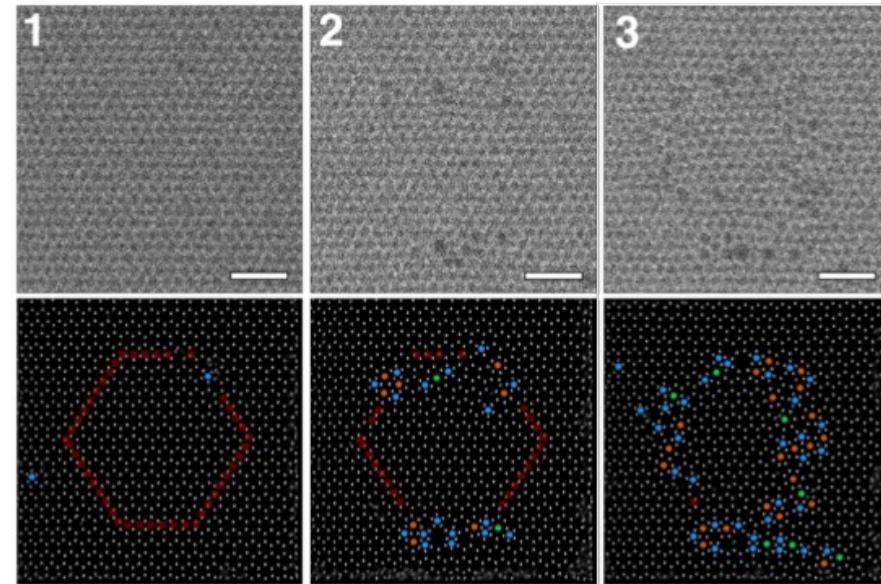
SPM: Autonomous Wall Manipulation



B. Smith et al. RSC Dig. Disc. (2024)

- Automated transition acquisition: write and perturb domain walls with voltages from the SPM tip
- Train physics-guided neural networks for prediction and digital twin, for arbitrary wall configurations and pulse parameters

STEM: Workflows for Defect Writing

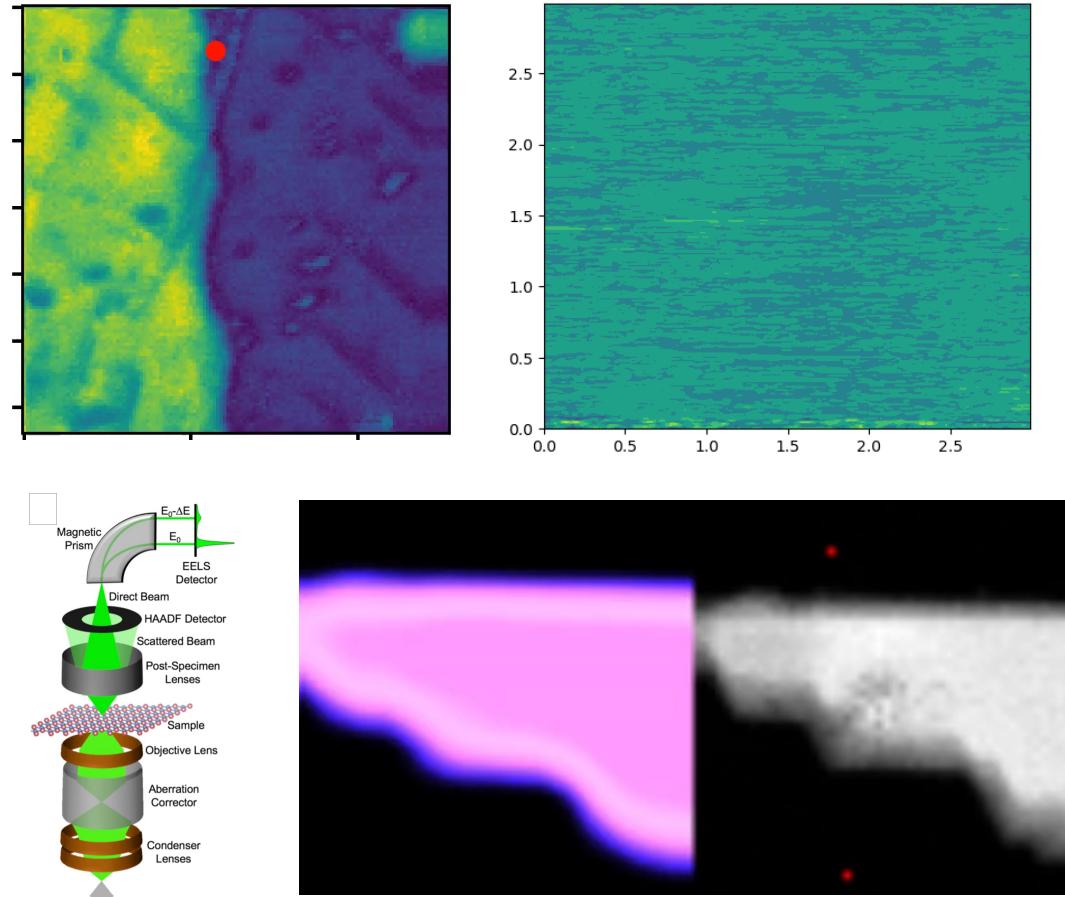


K. Roccapriore et al. ACS Nano 16, 17116 (2022)

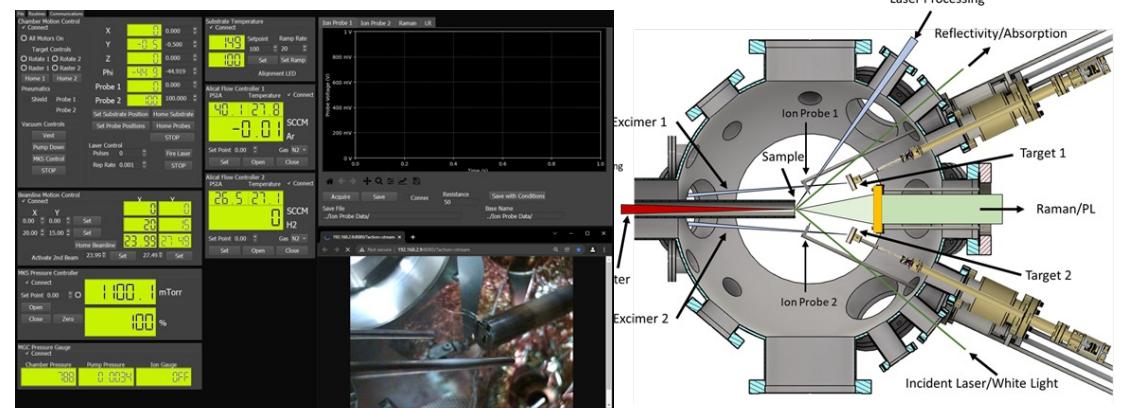
- Graphene imaging – steer beam in desired path, image again, find defects with computer vision, avoid defective areas, repeat
- Fully automated ‘avoidance patterning’ workflow to create defective regions on demand

Outline

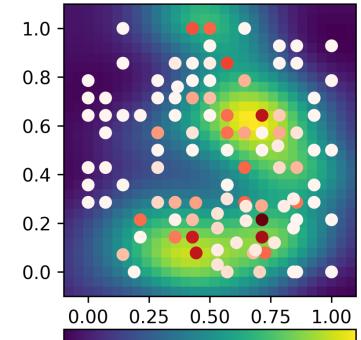
Autonomous Microscopy



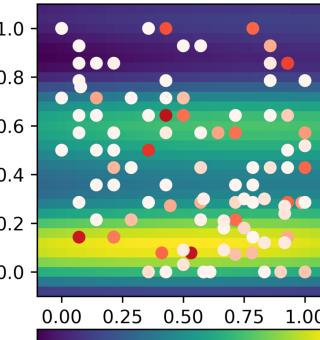
Autonomous Synthesis



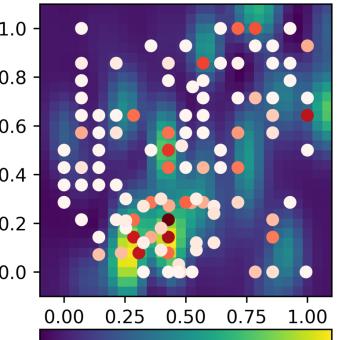
P vs T: Objective Mean



P vs e1: Objective Mean

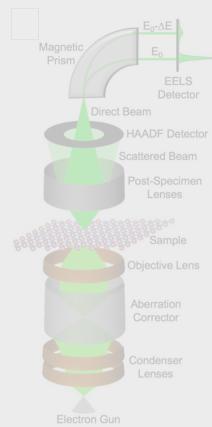
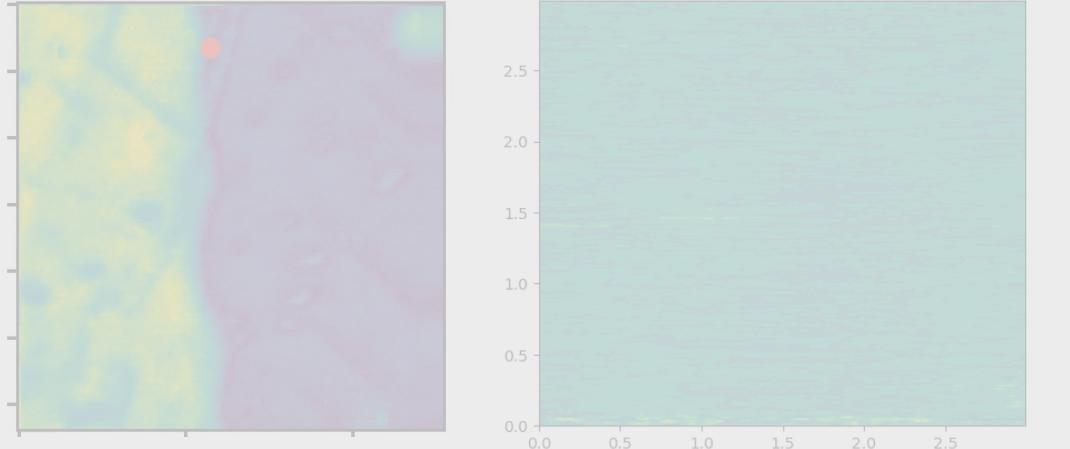


P vs e2: Objective Mean

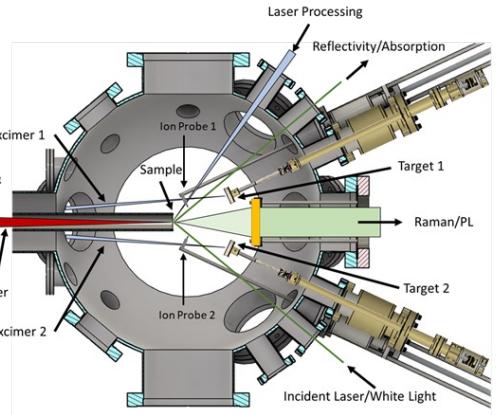


Outline

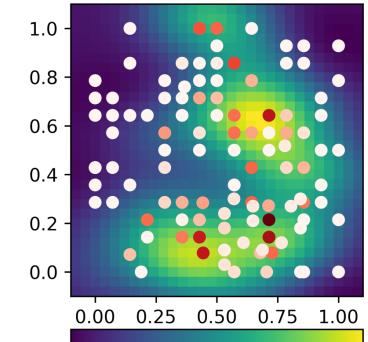
Autonomous Microscopy



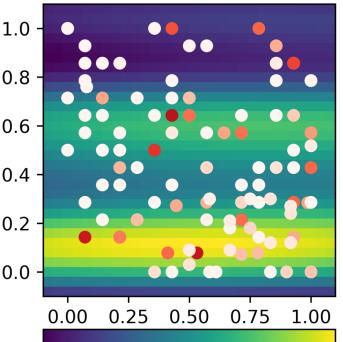
Autonomous Synthesis



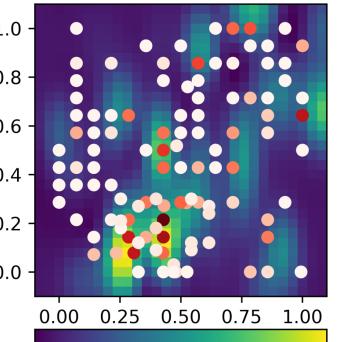
P vs T: Objective Mean



P vs e1: Objective Mean



P vs e2: Objective Mean



Autonomous Pulsed Laser Deposition

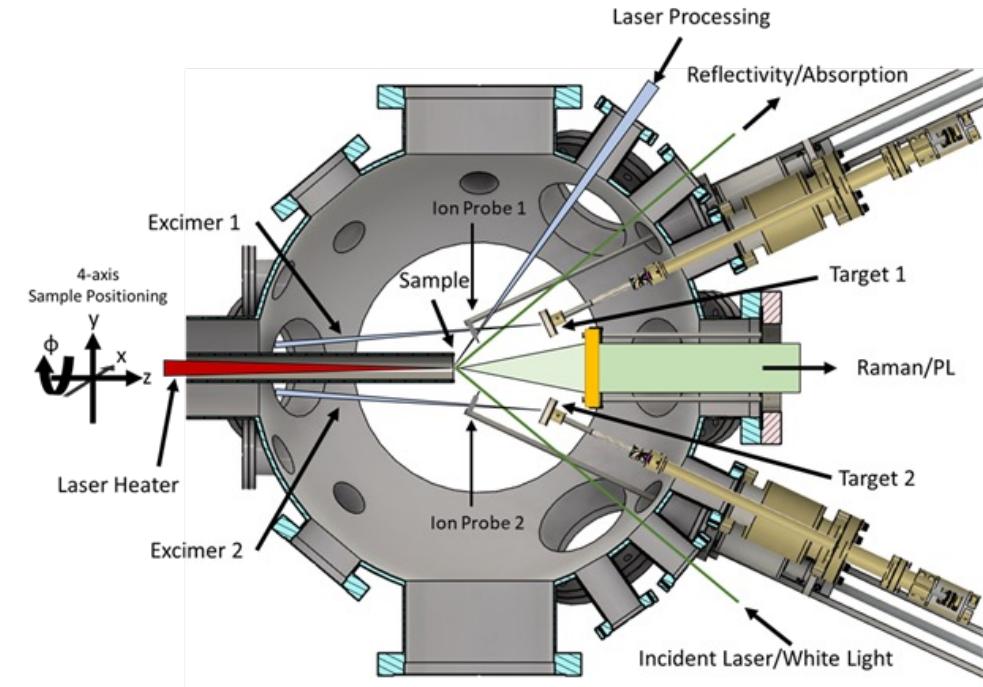
- ✓ Full process automation
- ✓ Automated material characterizations

- PLD Chamber Design

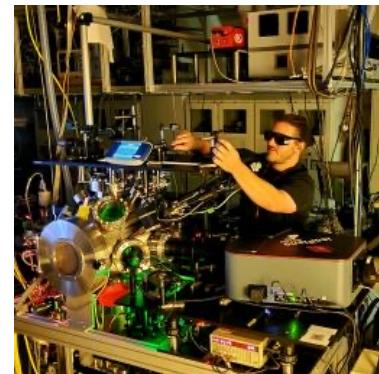
- Two PLD targets enable **simultaneous deposition** of different materials – tune stoichiometry of deposition
- **Plasma diagnostics:** ICCD Imaging, ion probe, gated plume spectroscopy.
- **Sample diagnostics/characterization:** Raman, PL, laser reflectivity, white light absorbance.
- **Laser Processing:** x,y nanosecond excimer laser processing of the sample

- **Full automation** of chamber processes

- Allows in-person and **remote/programmatic** control of all PLD processes and diagnostics via Python-> **enables machine autonomy and seamless integration with ML libraries**
- **Automated control** of: excimer laser energy, spot size, pulse number, and rep rate; background gas pressure, flow rates, mix of 2 gases; processing laser energy, pulses, rep rate, sample position; etc...



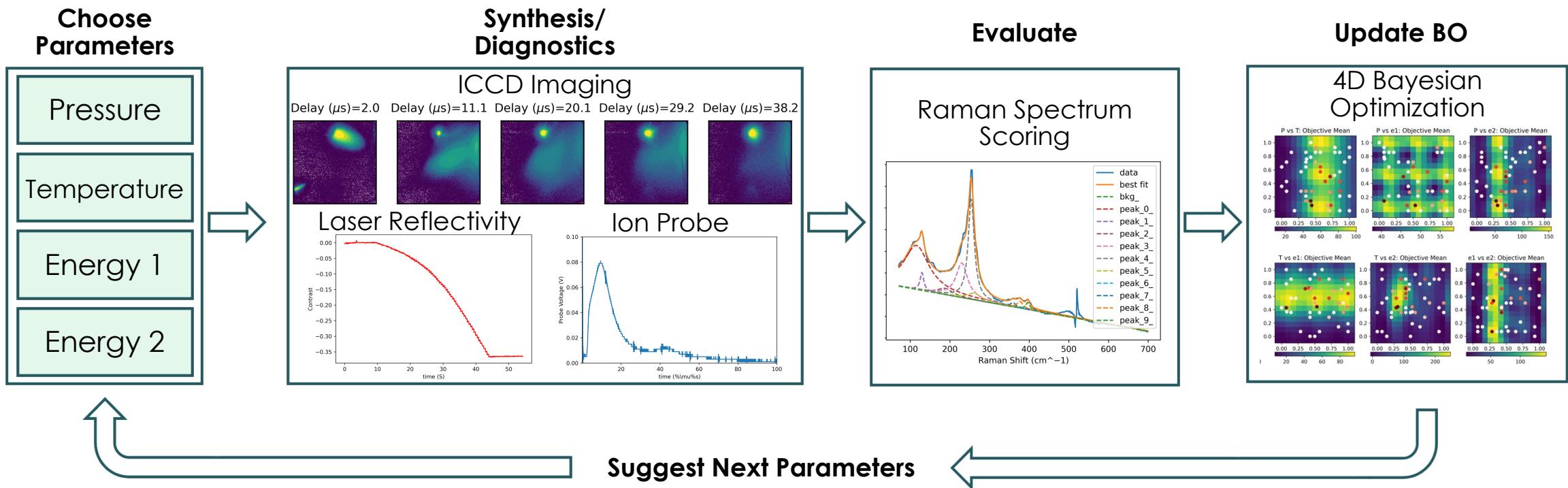
Sumner Harris



Autonomous synthesis of WSe₂

Goal: Autonomously search a broad parameter space to identify good growth conditions for ultrathin (<3 monolayers) WSe₂ by PLD.

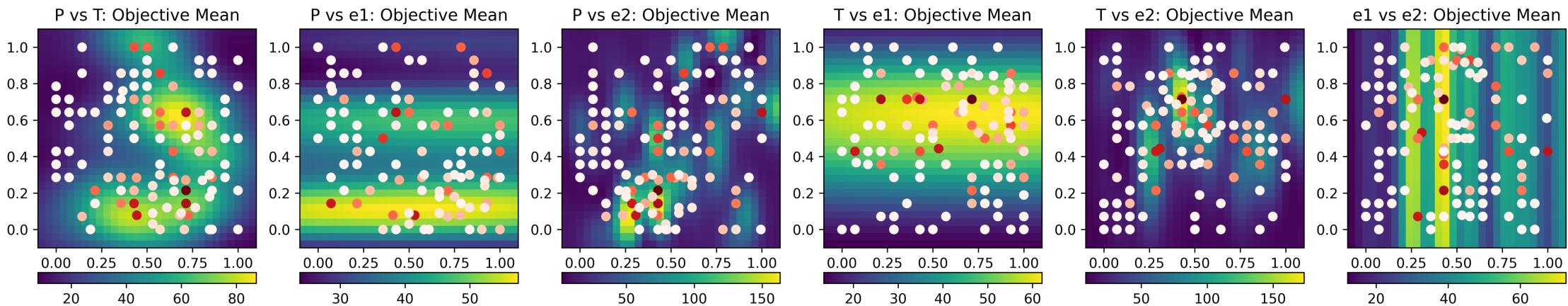
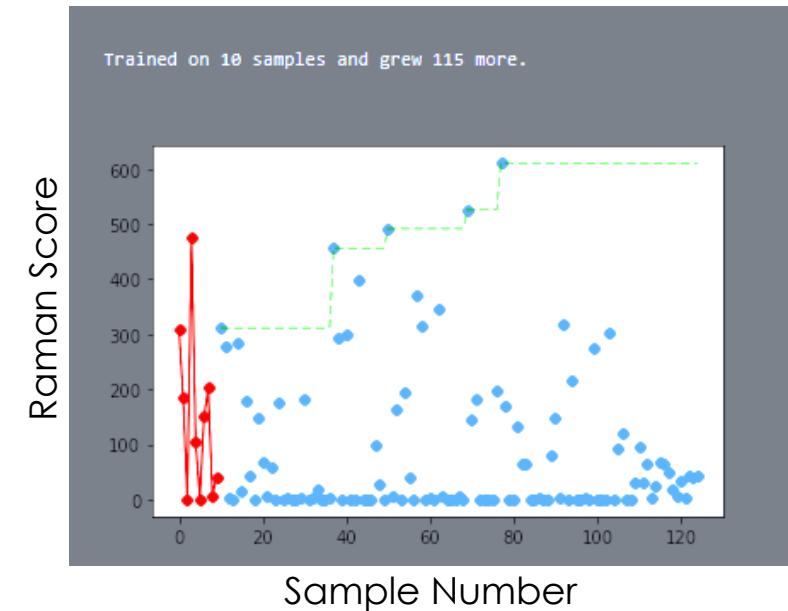
- 4D Bayesian optimization to search for optimal Raman “Score”
- Co-deposition PLD, general Workflow:



Autonomous synthesis of WSe₂

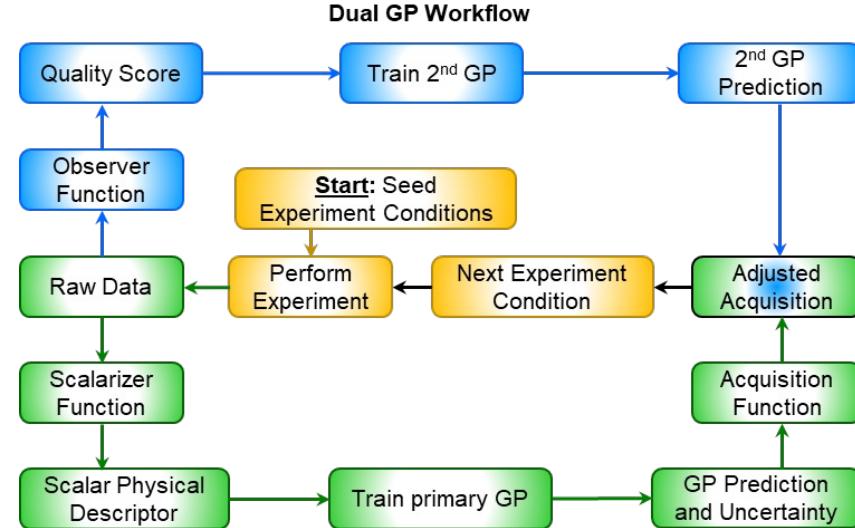
Started with 10 initial samples that were previously grown, autonomously grew 115 more with Bayesian optimization.

- Result after 115 samples (0.25% of space explored)
 - Showing averaged **surrogate function** projected into each 2D parameter plane
 - Seems **insensitive to WSe₂ laser energy**
 - Se laser energy seems critical -> **Needs Se compensation**
 - Potential identification of multiple growth regimes

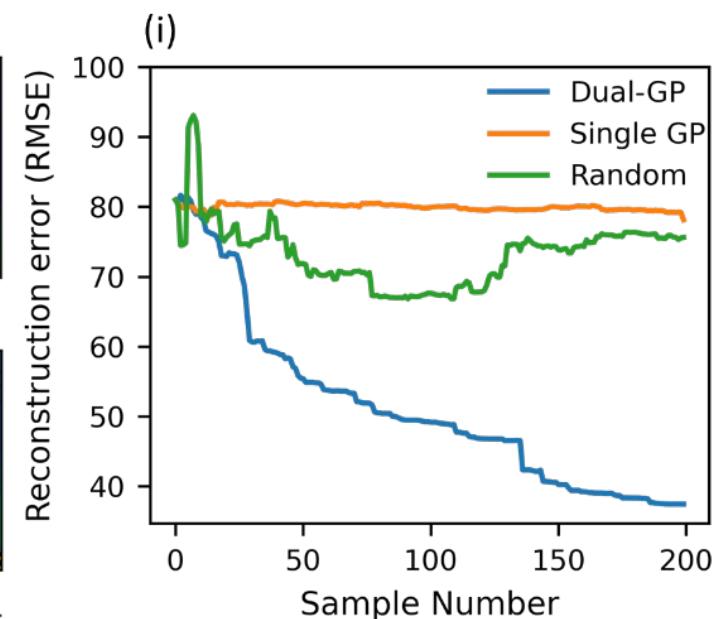
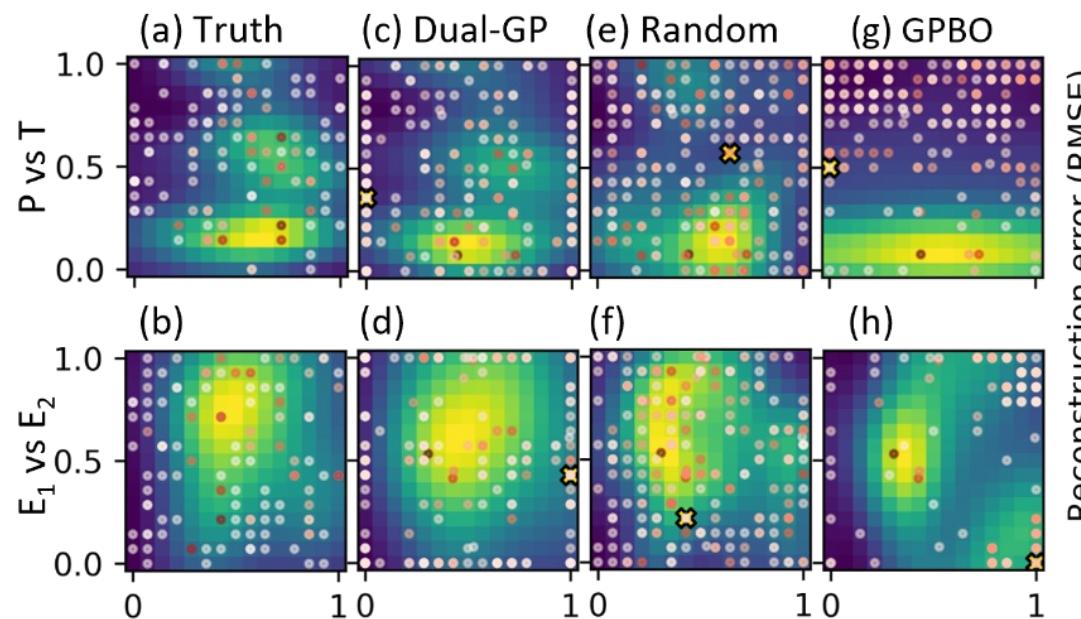


Colormap of predicted Raman spectrum score projected into each 2D plane.

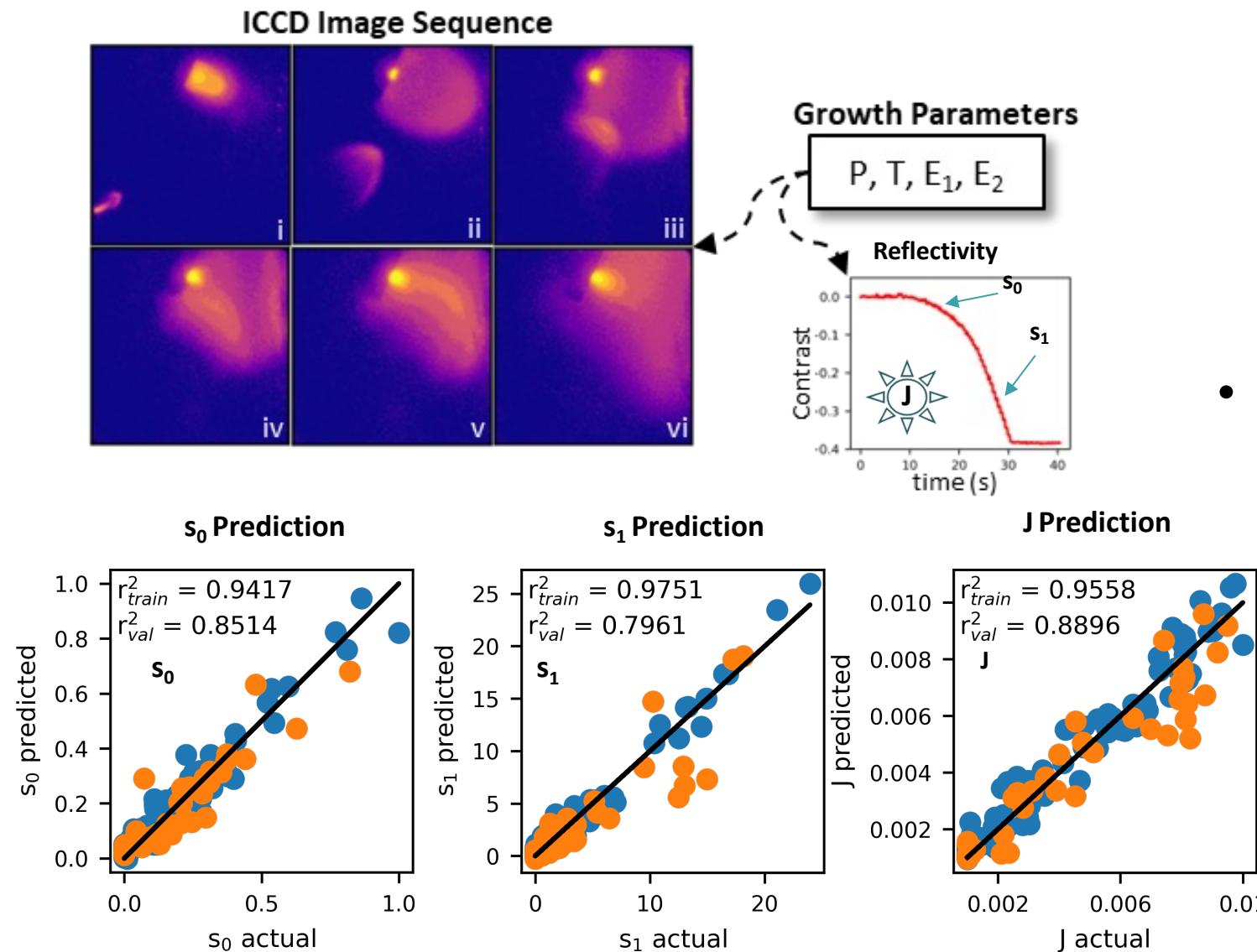
Dual GP method to reduce ‘bad’ samples



- One of the problems is that samples are wasted if the quality of the spectra is not sufficient for the scalarizer
- A dual GP method is proposed to alleviate this, that forces exploration only in areas where the quality of the score is sufficiently high



In-situ plume imaging



- By imaging the PLD plume, we can predict values of pressure, temperature, and laser fluences
- We can also estimate parameters of a kinetic model!

To realize control, we need to predict state dynamics

- The “**state process**” is the state of a dynamical system at time instant n where h is the “state model”, θ is a vector of parameters and w is random noise $\sim \mathcal{N}(0, \sigma)$

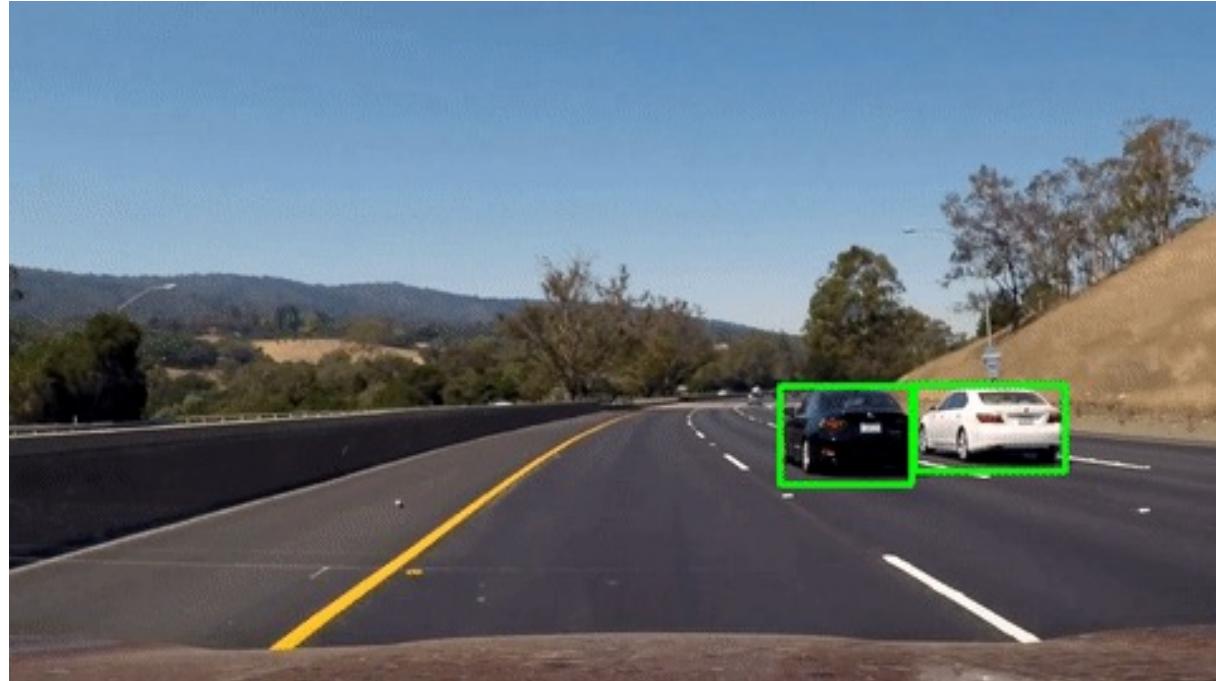
$$X_{n+1} = h(X_n, \theta) + w_n$$

- The “**observation process**” is a measurement related to the state X_n where H is some matrix and ξ is random noise $\sim \mathcal{N}(0, \gamma)$

$$Y_{n+1} = HX_{n+1} + \xi_{n+1}$$

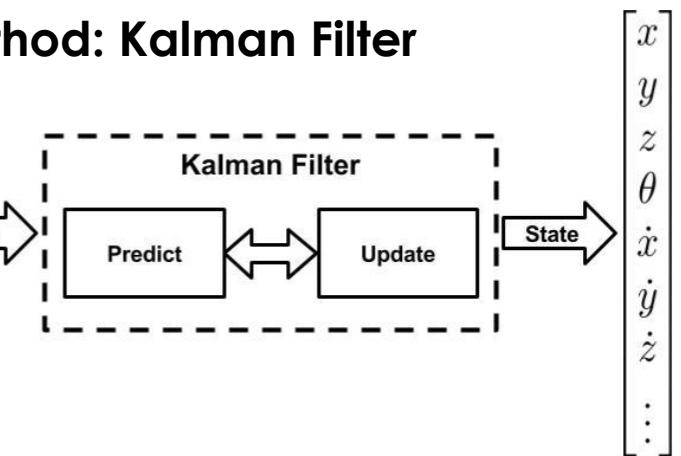
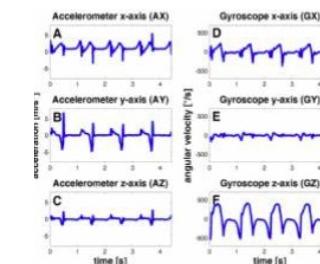
Goal: estimate the parameters θ of the state model from $\{Y_n\}$

- Solve this problem with **particle filters**. Use the recursive predict-update equations to generate a cloud of particles to approximate the posterior pdfs



<https://realitybytes.blog/2017/08/15/state-estimation-kalman-filters/>

Traditional method: Kalman Filter



Simple two-step kinetic model for growth of thin film

Nucleation and Growth

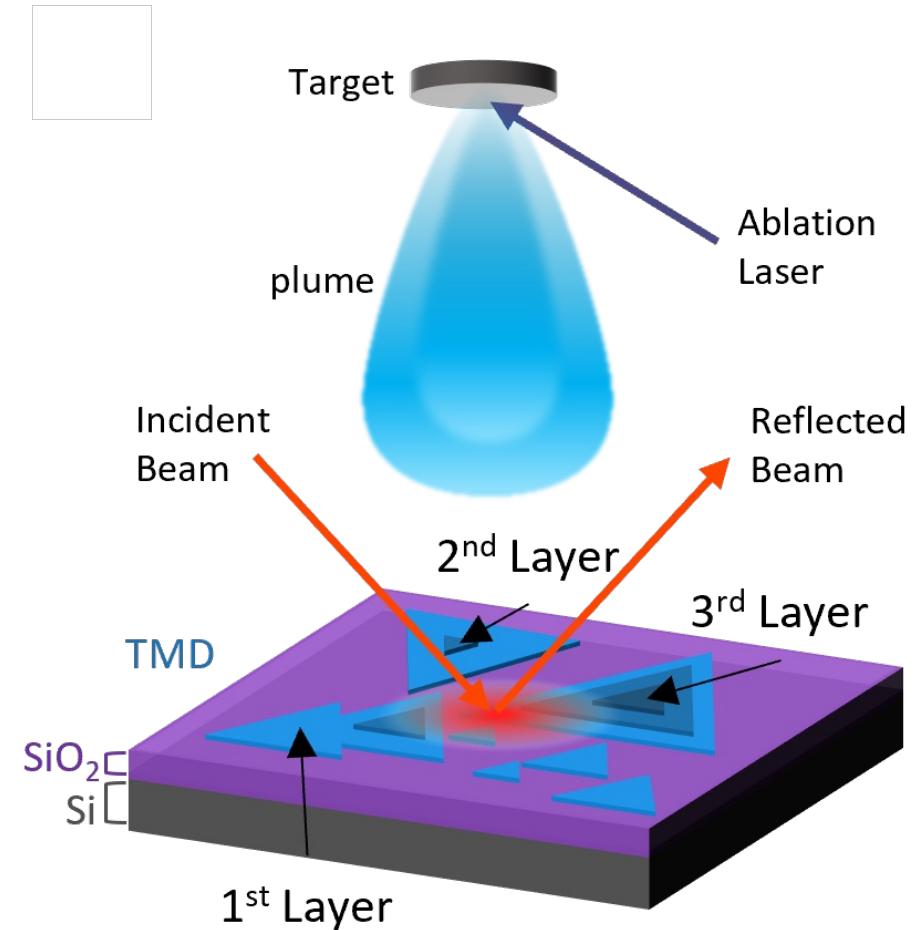


$$\frac{df_i}{dt} = k_{ni}(f_{i-1} - f_i) + k_{gri}(f_{i-1} - f_i)f_i$$

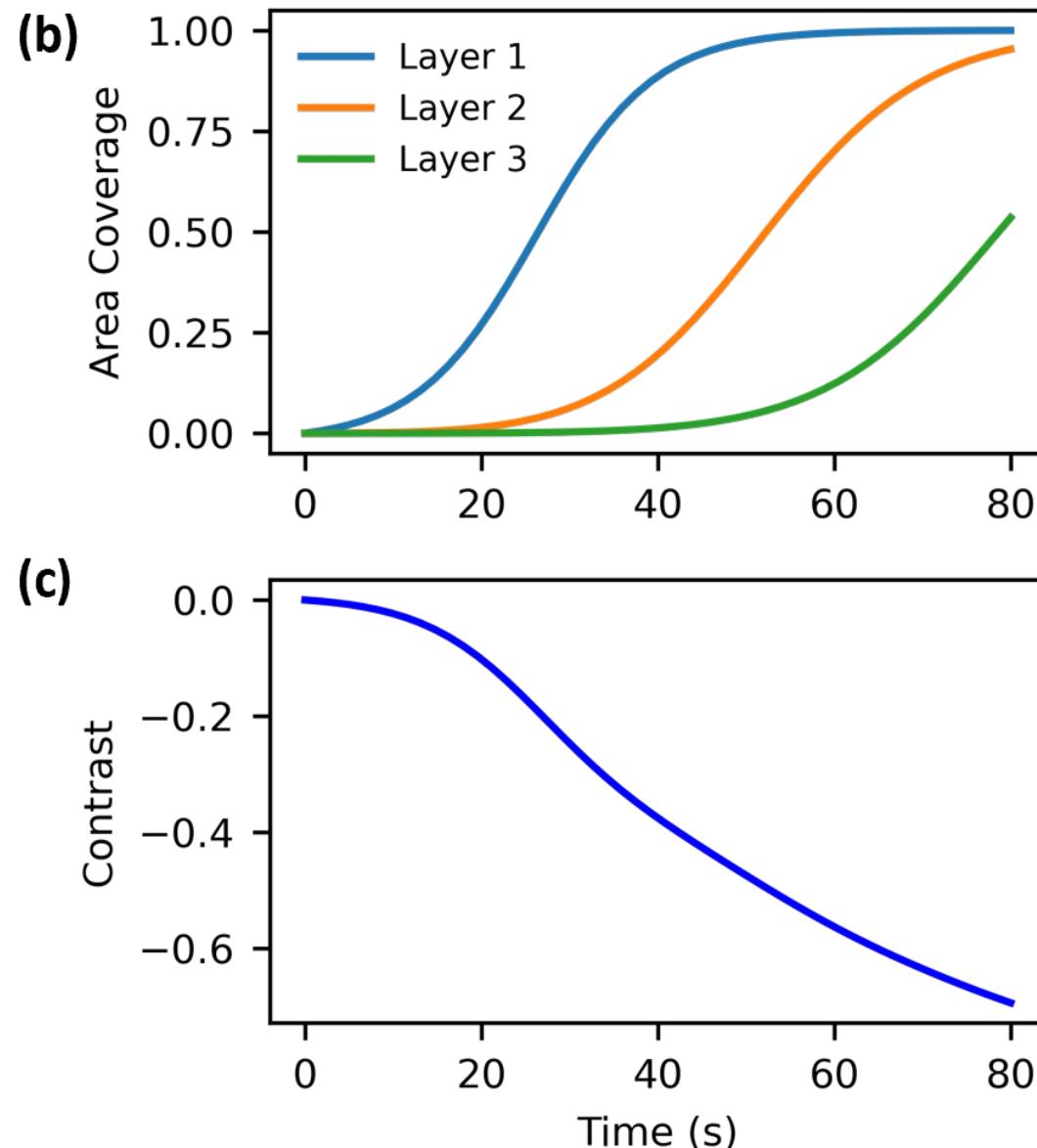
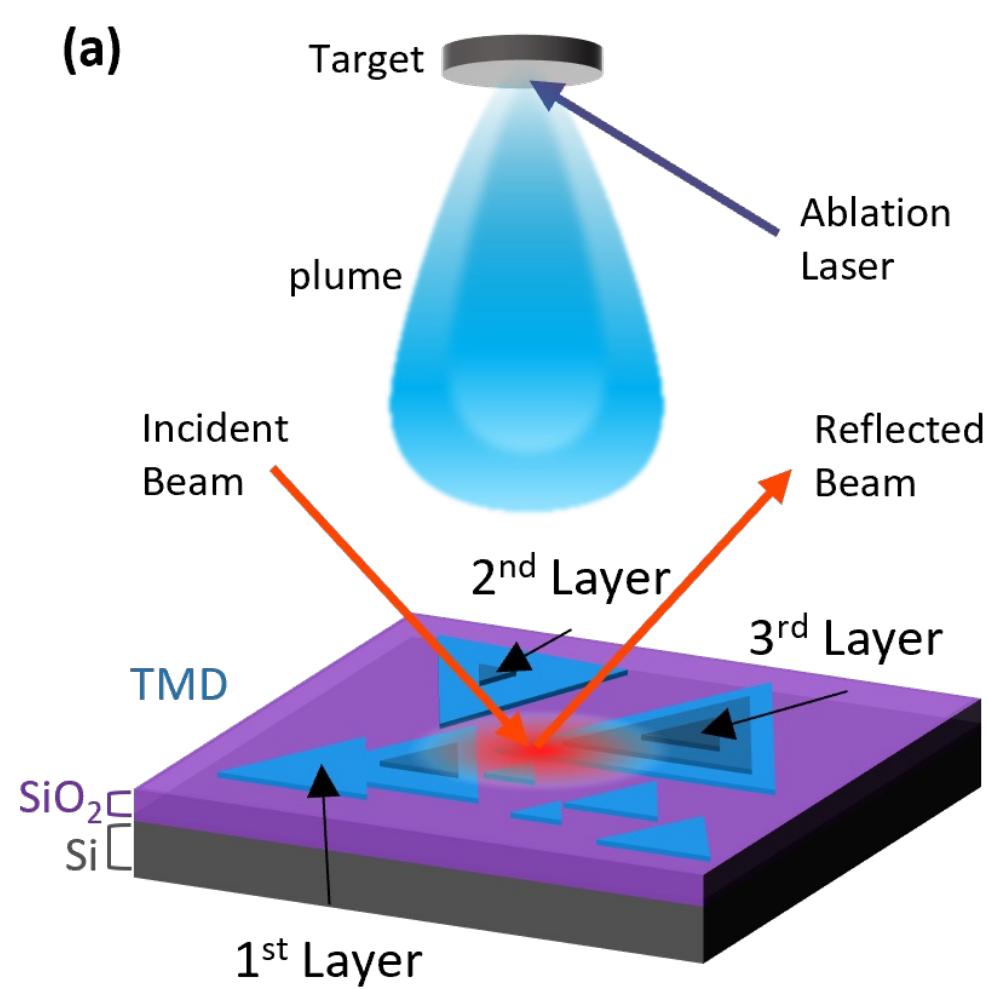
Contrast (observations)

$$c_r(t) = \sum_{i=1}^N (c_i - c_{i-1})f_i(t)$$

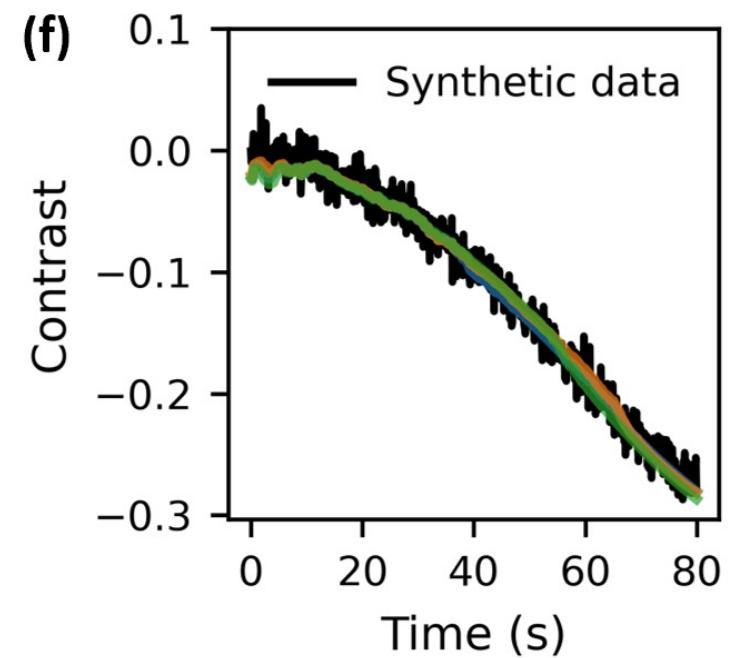
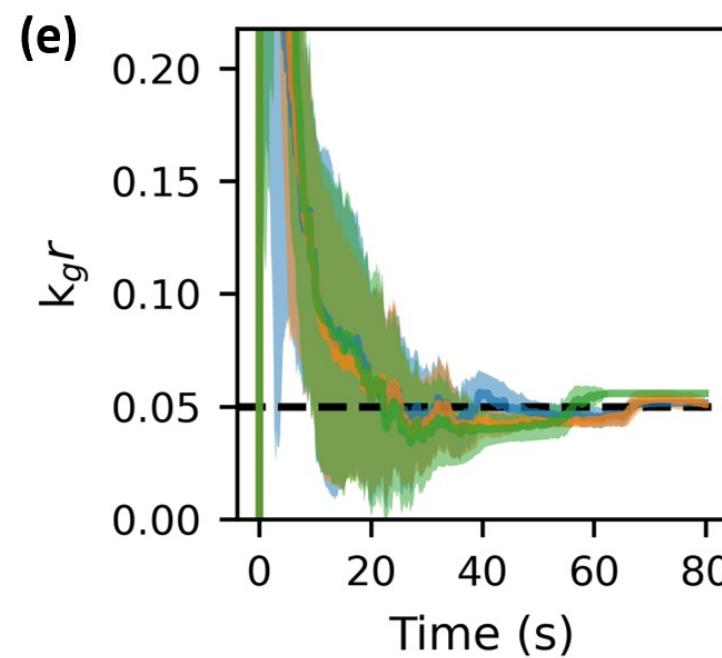
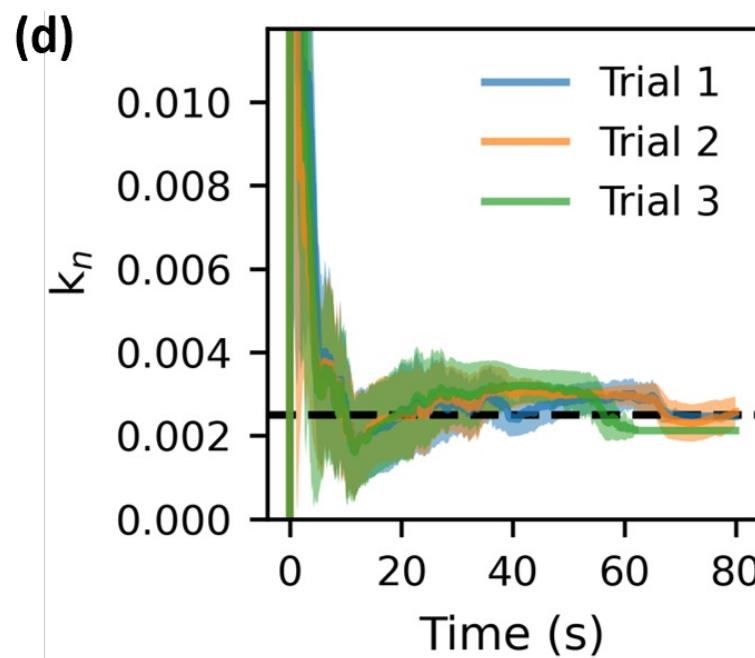
Growth of 2D material with PLD can be represented by a simple ordinary differential equation, with two unknown parameters (nucleation and growth coefficients)



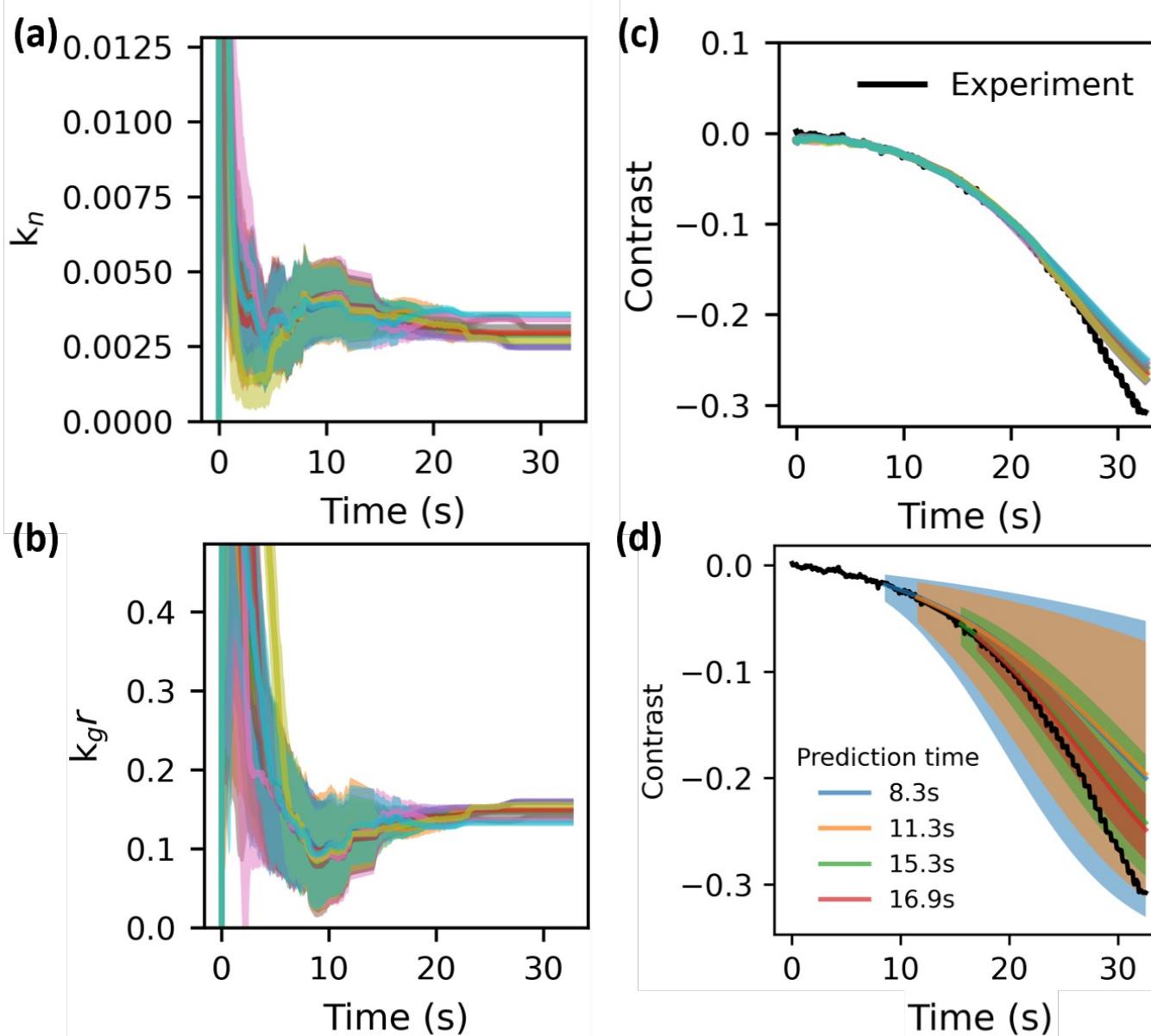
Simple two-step kinetic model for growth of thin film



Simulated Data



Real Data, Offline Testing



Next-step: use this for real-time control in an online setting



Future Directions

LLMs as scientific user assistants

GPT converts human demonstration to Python script

ChatGPT4 Response:

```
import os
import win32com.client
import numpy as np
import time
import h5py
import sidpy
import pyNSID
import matplotlib.pyplot as plt
from tqdm import tqdm

# Import Acquisition from Acroscopy
from Acroscopy import Acquisition

# Initialize the Acroscopy Acquisition
newexp = Acquisition(exe_path = r'C:\Path\To\BEPyAE.exe') # Replace with your actual path

# Initialize BEPyAE with AR18 (set offline_development=True for offline development)
newexp.init_BEPyAE(offline_development=True)

# Set BE parameters
# Set the center frequency to 400 kHz (400000 Hz)
newexp.define_be_params(be_params_dict = {"center_frequency_Hz_00": 400000, "band_width_Hz_01": 100, # bandwidth is an example
                                         "amplitude_V_02": 1, "phase_variation_03": 1,
                                         "repeats_04": 4, "req_pulse_duration_s_05": 4,
                                         "auto_smooth_ring_06": 1},
                        do_create_be_waveform = True, feedback=False)

# Conduct a BE line scan
# The line scan will go from [-1, -1] to [1, 1]
qk_fit, com_spec, chn1, chn2, chn3 = newexp.do_line_scan(line_scan_params_dict = {"num_BE_pulses_01": 32,
                                         "start_x_pos_00": -1, "start_y_pos_01": -1,
                                         "stop_x_pos_02": 1, "stop_y_pos_03": 1},
                                         upload_to_daq = True, do_sine_scan = True)
```

ChatGPT

To conduct a Band Excitation (BE) line scan measurement using the AEcroscopy API, as per your request, we need to set up both the line scan parameters and the BE parameters. For your specific case, we'll set the line scan to go from point `[-1, -1]` to `[1, 1]` and set the center frequency of the BE pulse to `400 kHz`.

Here's the Python code to achieve this:

Prompt (human demonstration of SPM experiment):

Provide Python code for conducting a BE line scan measurement from point [-1, -1] to [1, 1] with a center frequency of 400kHz

Instructions:

1. **Path to BEPyAE.exe:** Update the `exe_path` with the actual path to your `BEPyAE.exe` file.
2. **BE Parameters:** The `define_be_params` function is set with a center frequency of 400 kHz. You may need to adjust other parameters like `band_width_Hz_01`, `amplitude_V_02`, etc., based on your specific requirements.
3. **Line Scan Parameters:** The `do_line_scan` function's parameters are set for a scan from `[-1, -1]` to `[1, 1]`. The `num_BE_pulses_01` parameter (number of BE pulses) can be adjusted based on your requirements.

1. Scientific AI Assistants: for users, an ability to query assistants to help to plan experiments, guide data analysis, and refer to similar works

2. Workflow Planning and Execution: AI agents capable of digesting literature along with prior experiments performed at the Center, to propose new experiments and write and execute computational and experimental workflows

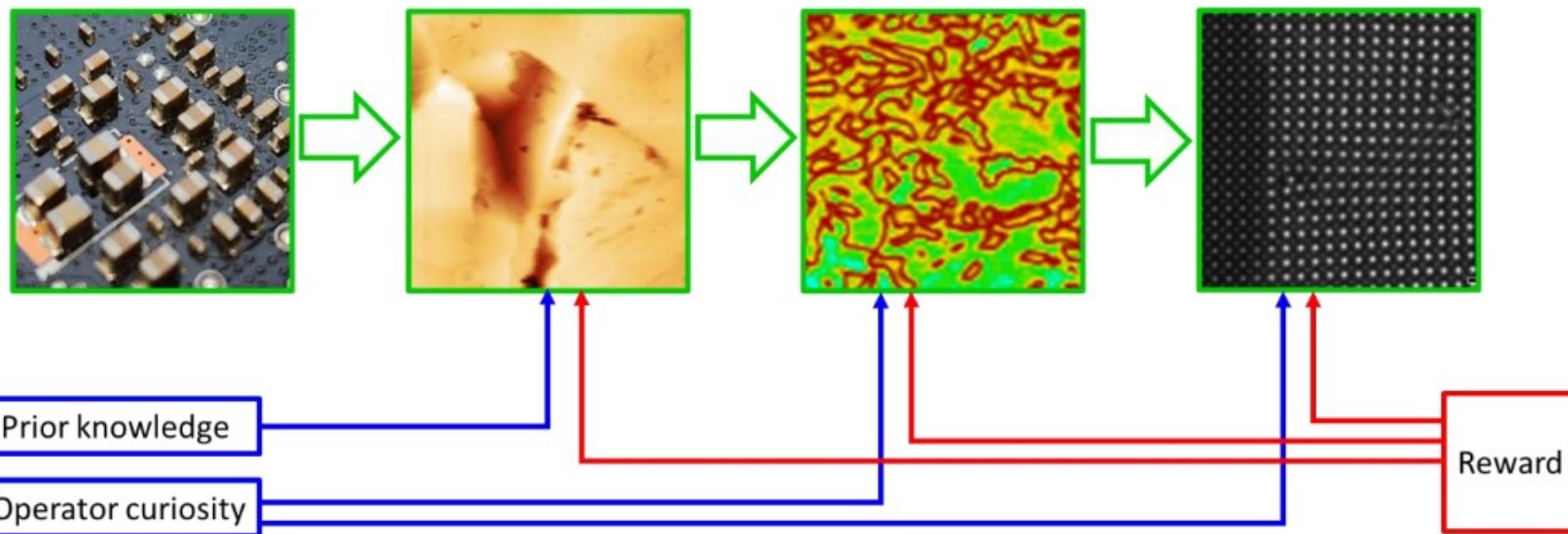
3. More context: Existing LLMs lack scientific context. Simulations can provide 'tuning' to ensure physically relevant proposals.

Liu, Checa, Vasudevan MLST (accepted) (2024)

Large Language Models (LLMs) can be used to help plan, write, and execute workflows on scientific characterization and synthesis tools

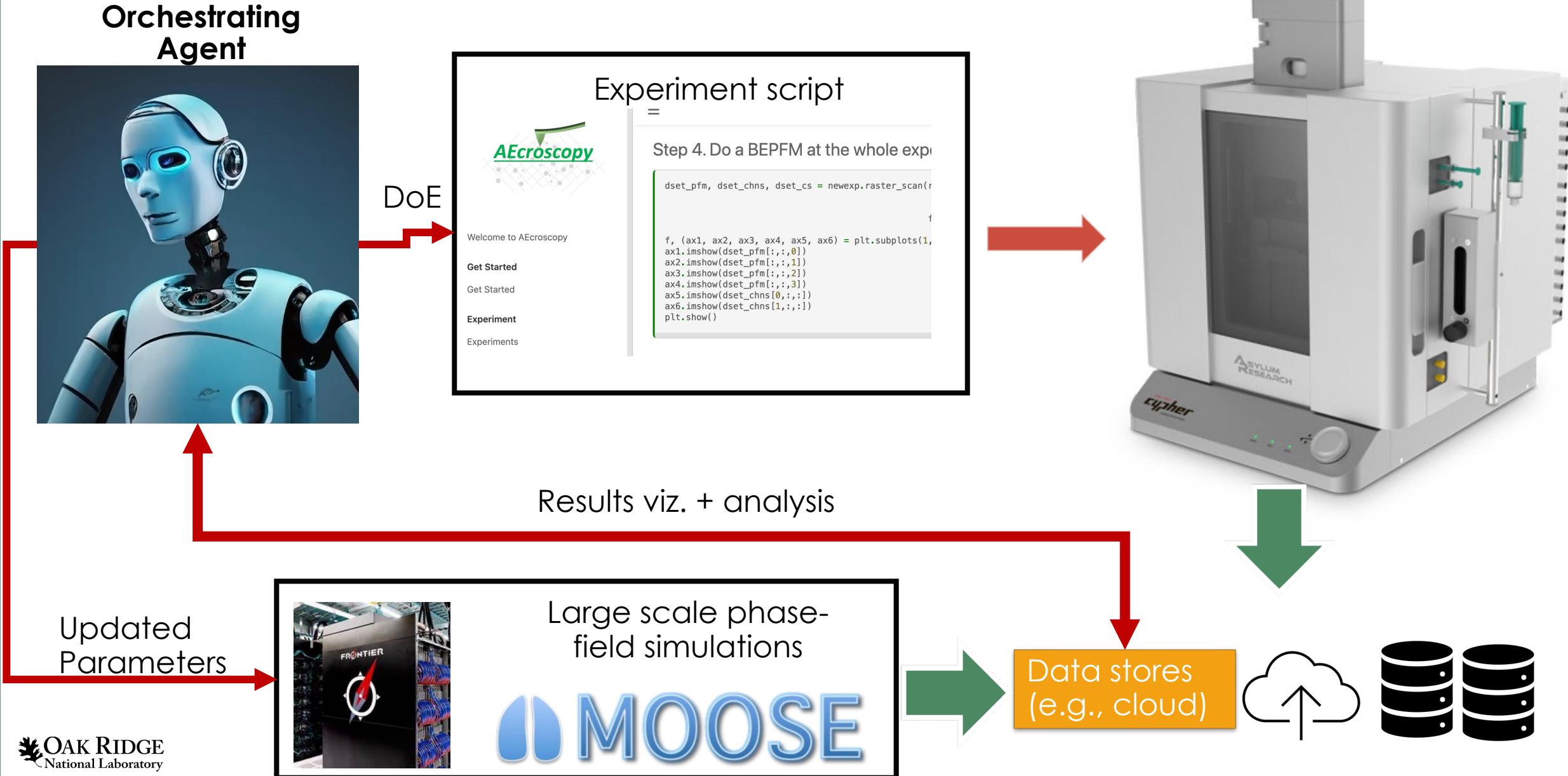
Multi-step workflow optimization

- Science is about multiple tools working in concert. We need tools to know what experiment is likely to produce the most information based on prior knowledge, past literature, etc.
- This presents algorithmic challenges.



Kalinin, Ziatdinov, Ahmadi, Ghosh, Roccapirore, Liu, Vasudevan, *Appl. Phys. Rev.* **11**, 011314 (2024)

Autonomous theory-experiment workflow



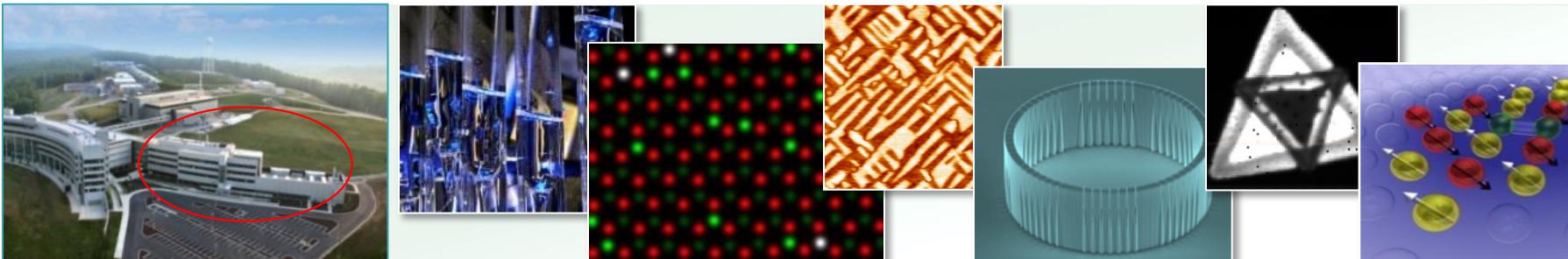
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- **Free access to laboratories, equipment and expertise if you agree to publish**
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- **Joint proposals with neutron sources (SNS, HFIR)**

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- **Nanofabrication** – direct-write, microfluidics, cleanroom
- **Advanced Microscopy** – AFM, STM, aberration-corrected TEM/STEM, atom-probe tomography
- **Functional Characterization** – laser spectroscopy, transport, magnetism, electromechanics
- **Theory and Modelling** – including gateway to leadership-class high performance computing



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Thank you

