THE UNIVERSITY of TENNESSEE KNOXVILLE

Lecture 1. Machine Learning in Full Electron Microscopy Workflow

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Pacific Northwest NATIONAL LABORATORY


2002 - 2022
Since 2022
2022 -2023
SCANNING PROBE MICROSCOPY
Electrical and Electromechanical Phenomena at the Nanoscale
Sergei Kalinin
Alexei Gruverman
EDITORS
VOLUME 1
Springer
Sergei V. Kalinin
Alexei Gruverman
Editors
Scanning Probe Microscopy of Functional Materials
Springer
Volume 7
World Scientific Series in Nanoscience and Nanotechnology
Scanning Probe Microscopy for Energy Research
Dawn A Bonnell • Sergei V Kalinin
Editors
World Scientific
Volume 1
World Scientific Series on Emerging Technologies – Volume 1
HANDBOOK ON BIG DATA AND MACHINE LEARNING IN THE PHYSICAL SCIENCES
In 2 Volumes
Volume 1
Big Data Methods in Experimental Materials Discovery
Editors
Surya R. Kalidindi • Sergei V. Kalinin
Turab Lookman
Editors-in-Chief
Sergei V. Kalinin • Ian Foster
World Scientific

Kevin Roccapriore

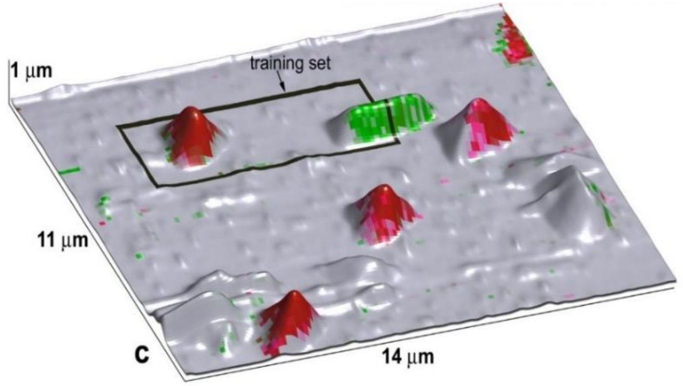
Founder, AtomQ

Build quantum world one atom at a time

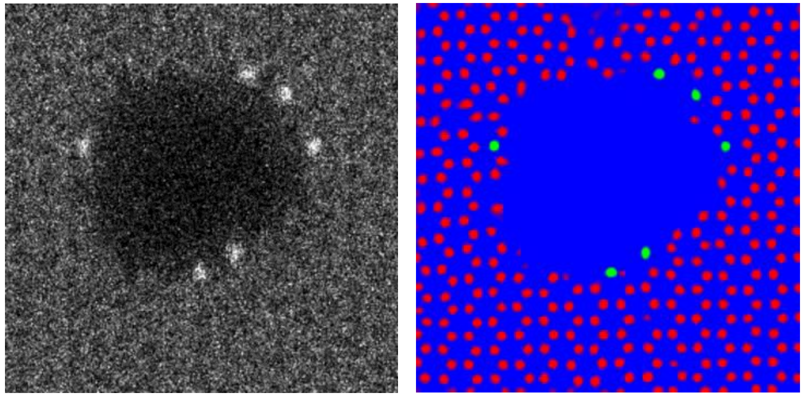
- Staff scientist at Oak Ridge National Laboratory's Center for Nanophase Materials Sciences (CNMS).
- PhD in Physics from the University of North Texas in 2018, which was nanofabrication for nanophotonics applications.
- Post-doc at ORNL (2018-2021), focusing on machine learning in the scanning transmission electron microscope (STEM) toward automated experiments, including STEM-EELS and 4D-STEM.
- Currently, focuses on ML on-the-fly in the STEM for precise atomic fabrication by site-specific atom targeting with the electron beam, automated experiments learning and utilizing structure-property relationships, and automated multidimensional data acquisition and analysis.

Machine Learning in Microscopy

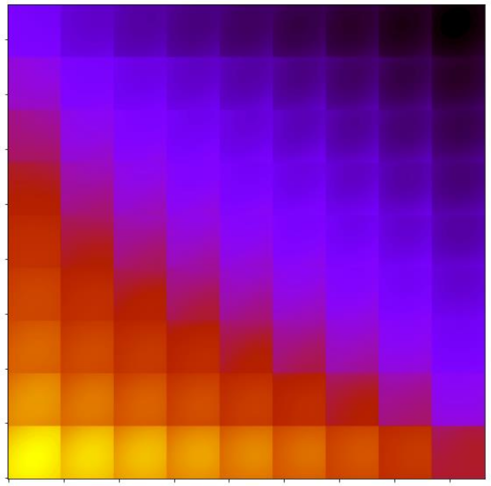
Shallow networks for data analysis



Deep networks for data analysis



Deep networks for learning physics



2006 2008 2010 2012 2014 2016 2018 2020 2022

Principal component analysis

Neural network recognition

NN-based theory-experiment marching

Physics-based ML

Deep learning

- ELIT ML
- Invariant autoencoders

M.P. Nikiforov, A.A. Vertegel, V.V. Reukov, G.L. Thompson, S.V. Kalinin, and S. Jesse, *Functional recognition imaging using artificial neural networks: Applications to rapid cellular identification by broadband electromechanical response*, Nanotechnology **20**, 405708 (2009).

O.S. Ovchinnikov, S. Jesse, P. Bintacchit, S. Trolier-McKinstry, and S.V. Kalinin, *Disorder identification in hysteresis data: recognition analysis of random-bond random-field Ising model*, Phys. Rev. Lett. **103**, 157203 (2009).

Microscopy AND Microanalysis

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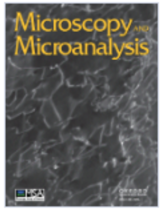

Volume 16, Issue S2
1 July 2010

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JOURNAL ARTICLE

Towards the Thinking Microscope

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OS Ovchinnikov, S Jesse, SV Kalinin, HJ Chang, SJ Pennycook, AY Borisevich

Microscopy and Microanalysis, Volume 16, Issue S2, 1 July 2010, Pages 160–161,
https://doi.org/10.1017/S1431927610062720
Published: 01 August 2010

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Abstract

Extended abstract of a paper presented at Microscopy and Microanalysis 2010 in Portland, Oregon, USA, August 1 – August 5, 2010.

Issue Section: Instrumentation and Techniques Symposia > Aberration-Corrected Electron Microscopy: Exploring Materials Through New Eyes

ACS Nano > Vol 15/Issue 8 > Article

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REVIEW | July 16, 2021

Automated and Autonomous Experiments in Electron and Scanning Probe Microscopy

Sergei V. Kalinin*, Maxim Ziatdinov, Jacob Hinkle, Stephen Jesse, Ayana Ghosh, Kyle P. Kelley, Andrew R. Lupini, Bobby G. Sumpter, and Rama K. Vasudevan

nature > nature materials > progress article > article

Progress Article | Published: 23 September 2015

Big–deep–smart data in imaging for guiding materials design

Sergei V. Kalinin, Bobby G. Sumpter & Richard K. Archibald

Nature Materials **14**, 973–980 (2015) Cite this article

Abstract

Harnessing big data, deep data, and smart data from state-of-the-art imaging might accelerate the design and realization of advanced functional materials. Here we discuss new opportunities in materials design enabled by the availability of big data in imaging and data analytics approaches, including their limitations, in material systems of practical interest. We specifically focus on how these tools might help realize new discoveries in a timely manner. Such methodologies are particularly appropriate to explore in light of continued improvements in atomistic imaging, modelling and data analytics methods.

nature > npj computational materials > perspectives > article

Perspective | Open access | Published: 20 December 2023

Machine learning for automated experimentation in scanning transmission electron microscopy

Sergei V. Kalinin, Debangshu Mukherjee, Kevin Roccapriore, Benjamin J. Blaiszik, Ayana Ghosh, Maxim A. Ziatdinov, Anees Al-Najjar, Christina Doty, Sarah Akers, Nageswara S. Rao, Joshua C. Agar & Steven R. Spurgeon

npj Computational Materials **9**, Article number: 227 (2023) Cite this article

Image atomic columns

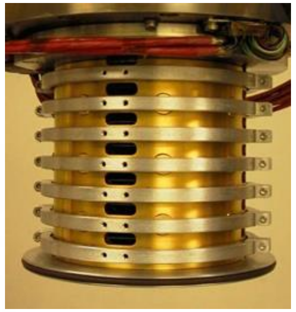
Spectra from single atoms

Image light atoms and sensitive materials

Diffraction from subatomic volumes

Beams with orbital momentum

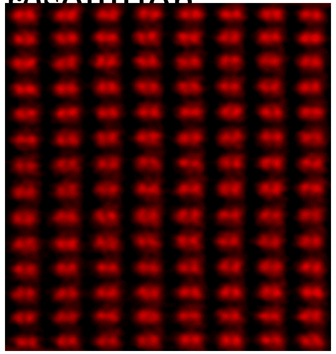
Beam manipulation

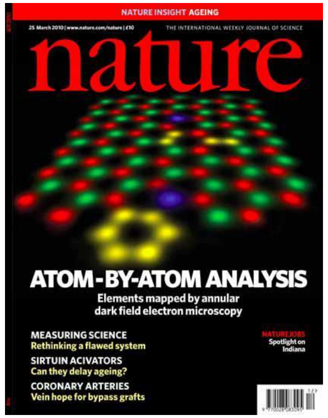


First AC (Nion)

1997

0.61 A resolution





2002 Prototype correctors

2006 TEAM project

2012 Broad adoption of AC STEM

2012 Segmented detectors

2014 4D STEM

2016 Vortex beams

2018

Vortex beams

High-res EELS

In-situ holders

4D STEM

Physics extraction

Atomic manipulation

Source

Sample

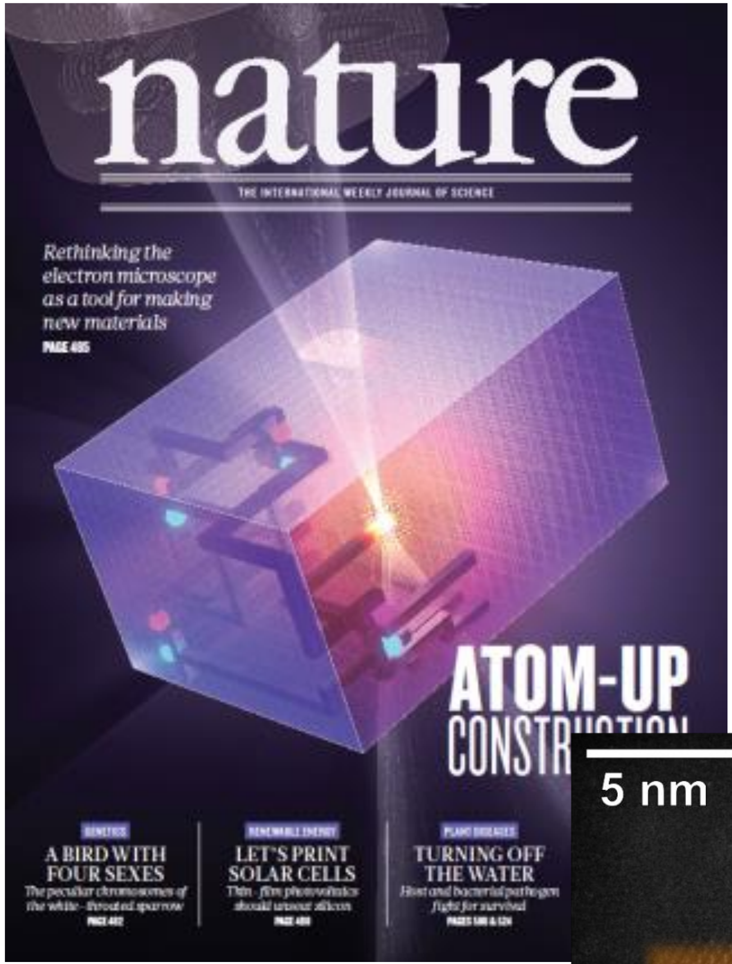
Detector

Data

Feedback

Images from A. Lupini

Electron Microscopy yields large volumes of spatially-resolved data on atomic coordinates (bond length and bond angles), low energy and core-level excitations, and scattering from sub-atomic volumes. In addition, electron beams can be used to manipulate matter atom by atom.



Volume 75, Issue 6
June 2022

< Previous Article   Next Article >

A quantum lab in a beam FREE

Advances in electron microscopy have revolutionized atomic-scale imaging, characterization, and manipulation of materials.

Sergei V. Kalinin; Stephen Jesse; Andrew R. Lupini

Check for updates

Physics Today **75** (6), 30–36 (2022);
https://doi.org/10.1063/PT.3.5018

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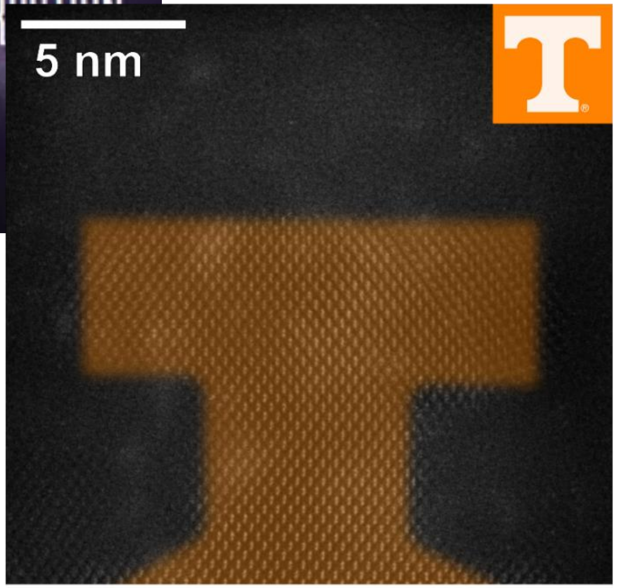
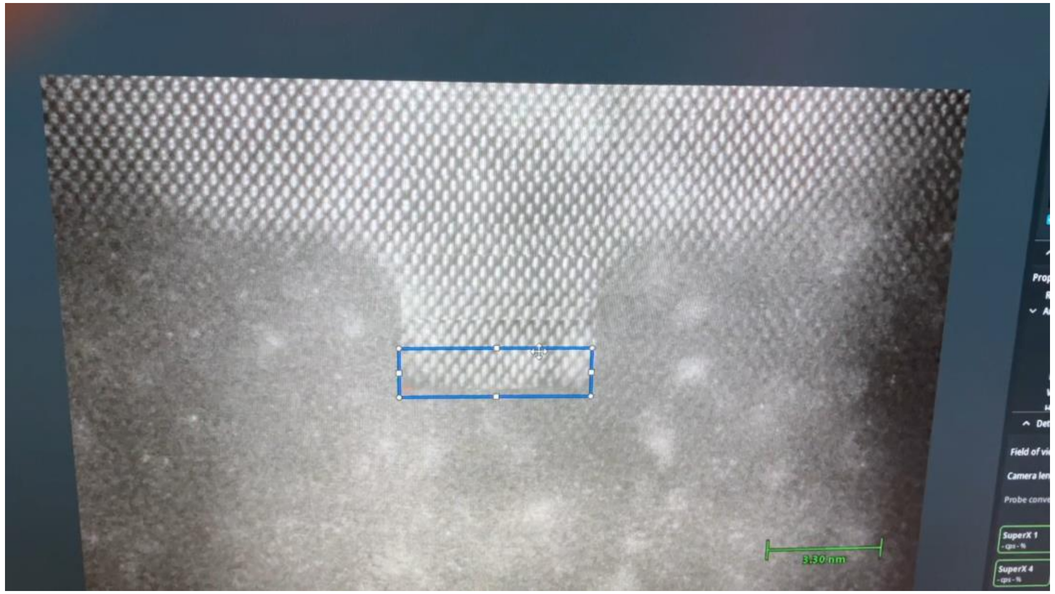
Topics

Electron spectroscopy, Scanning electron microscopy, Scanning tunneling microscopy, Transmission electron microscopy, Quantum structures, Quantum mechanical systems and processes

The history of science is filled with questions about the nature of matter, its constituent elements, how properties emerge from the elements' arrangements, and how the arrangements can guide or be guided by energy flows. The answers to those questions have progressed from philosophical proposals of atomic theory to practical demonstrations of atoms' existence to modern quantum theory. And, importantly, the answers have been based on experimental measurements.



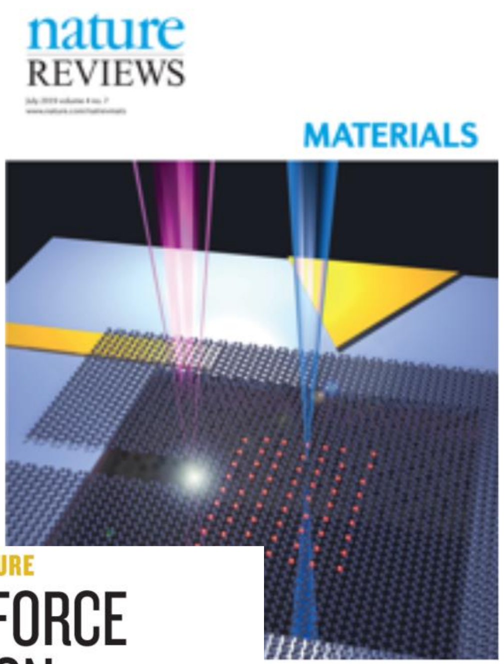


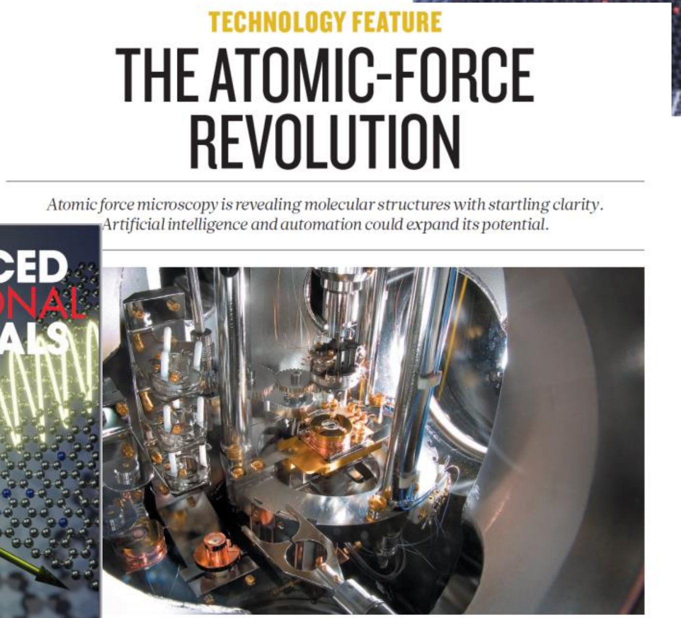
ML for Automated Microscopy?

Microscopy today:
- Primary component of research in materials, physics and biology
- 1000s of high-end (S)TEM platforms, ~10,000 overall
- 1000s of high-end UHV SPMs, >50,000 ambient
- Chemical and mass-spectrometric imaging

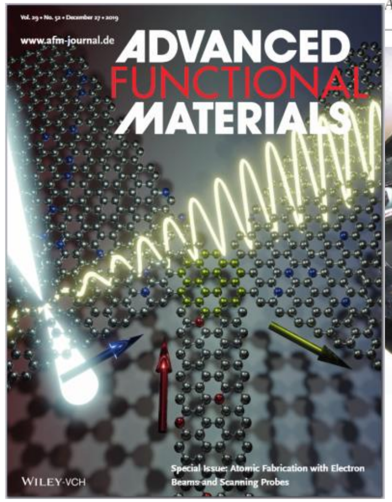
What do microscopists do?
- Most of the time - sit alone in the dark room and turn knobs 😌
- Limited amount of collected data
- Case for automation: CryoEM

Unsurprisingly, inspired by autonomous cars, etc. – multiple proposals to make automated microscopes!



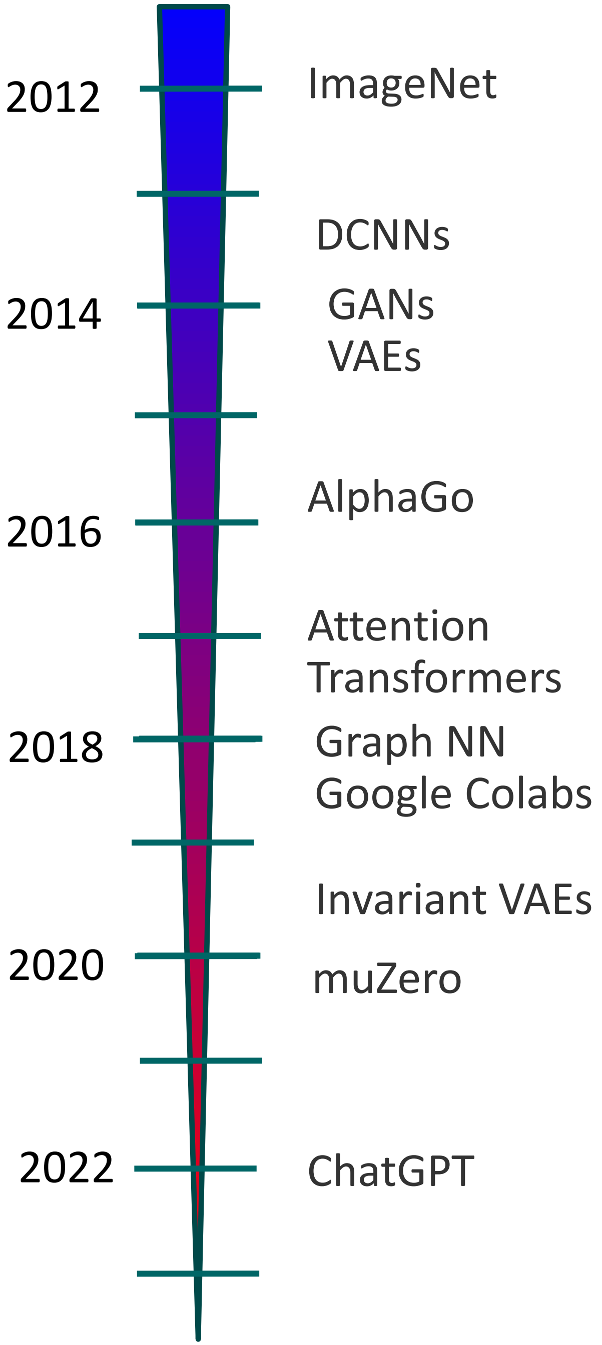


July 2019



Why Machine Learning?

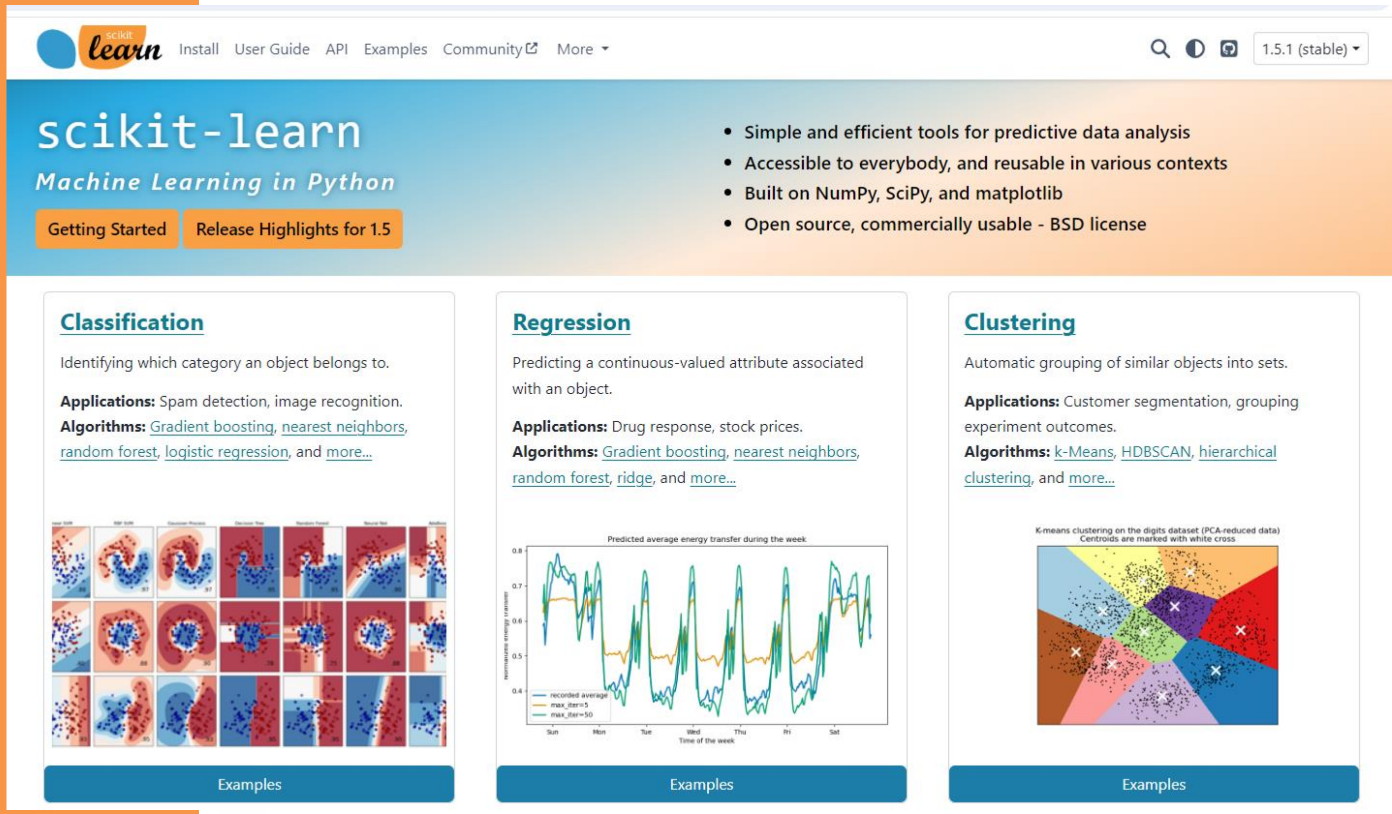
- Last decade has experienced an explosive growth of machine learning and artificial intelligence applications
- These developments have spanned areas from computer vision to medicine to autonomous systems and games
- However, the progress and impact as applied to experimental physical sciences has been minimal....

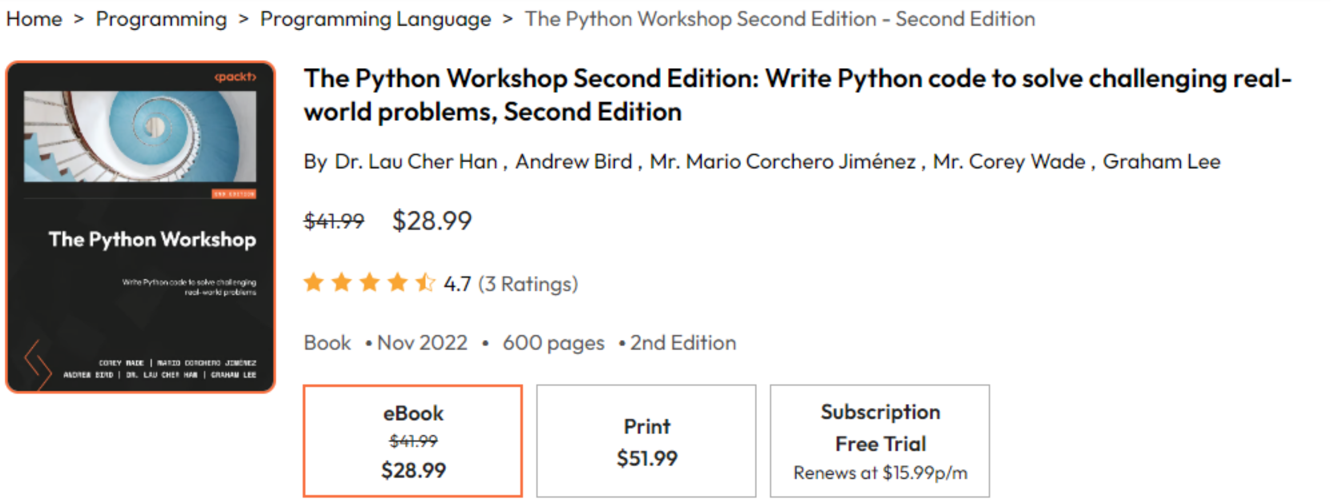


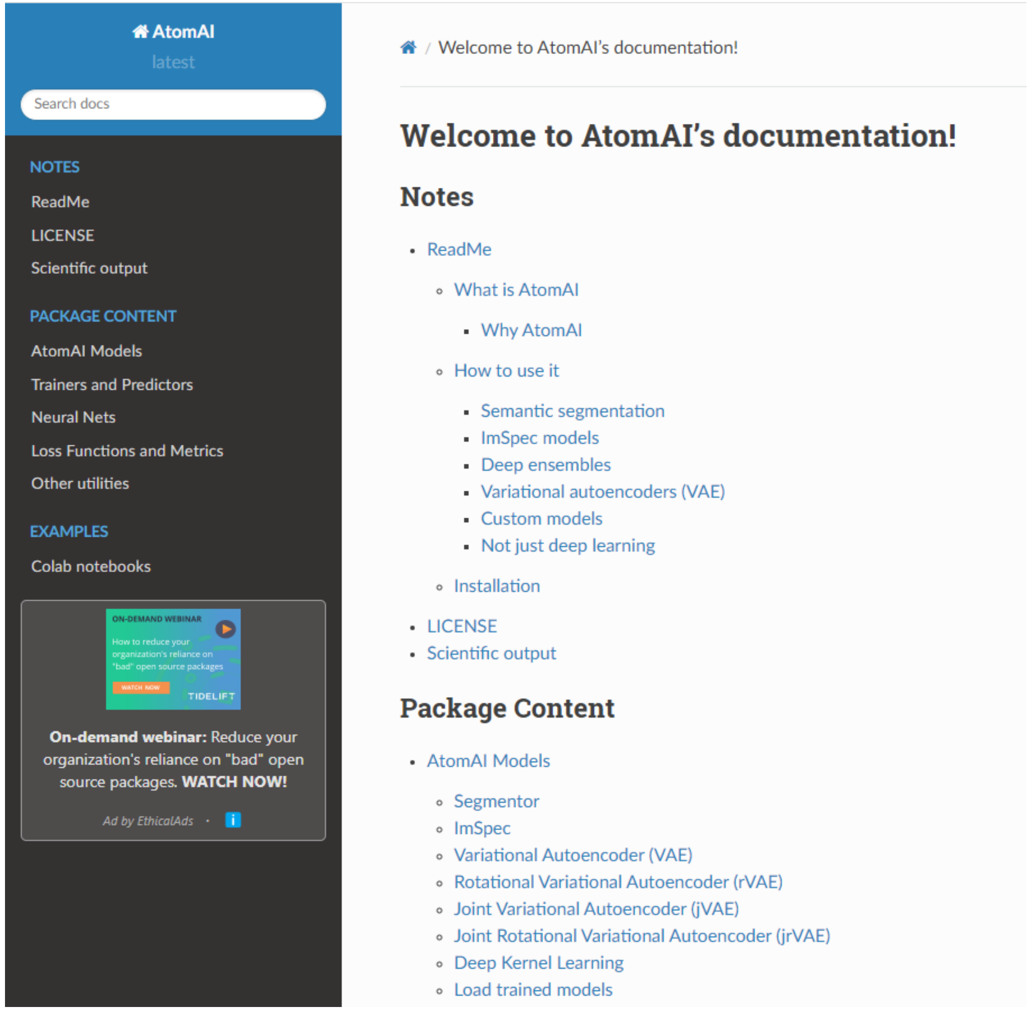
Why is it difficult?

- Requires domain expertise and domain-specific goals
- Deeply causal and hypothesis drive nature of domain sciences
- No single answer: culture, not a method
- Infrastructure, open code, open data
- **Most important:** active nature of scientific process

- Packtpub.com
- Manning.com
- GitHub

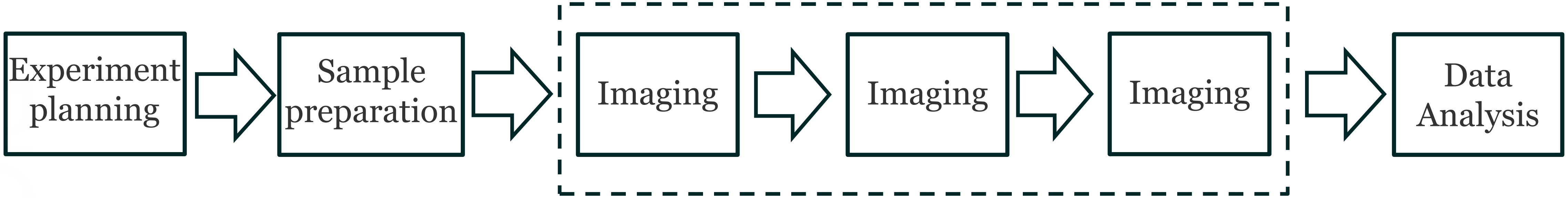






What is your goal?

Level 5: Downstream Use of Microscopy Data: Incorporates microscopy data into theory analysi pipelines, closing the synthesis-characterization-discovery loop.



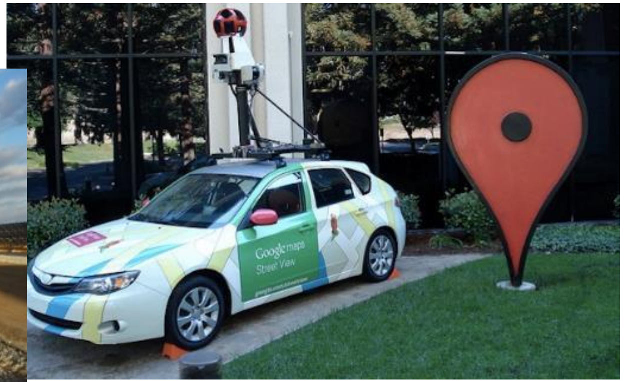
Level 4: Upstream Task Planning:
ML is used for planning experiments, including sample selection and integrating microscopy with materials synthesis.

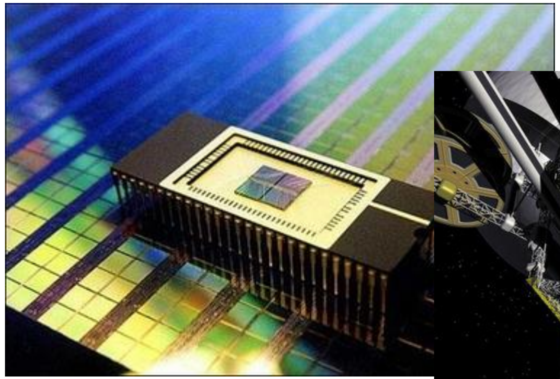
Level 2: Real-Time Analytics
- ML helps represent data in a form that is more understandable to humans.
- Decisions are still made and orchestrated by humans.

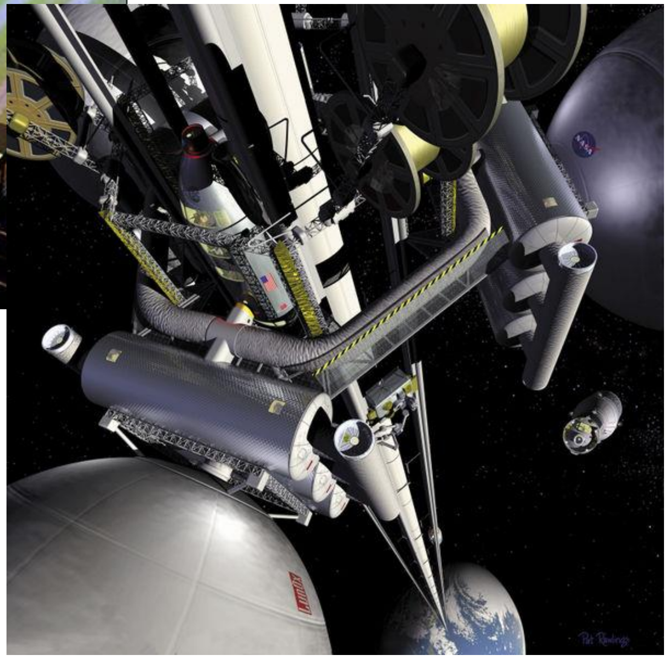
Level 1: Post-Acquisition Data Analysis

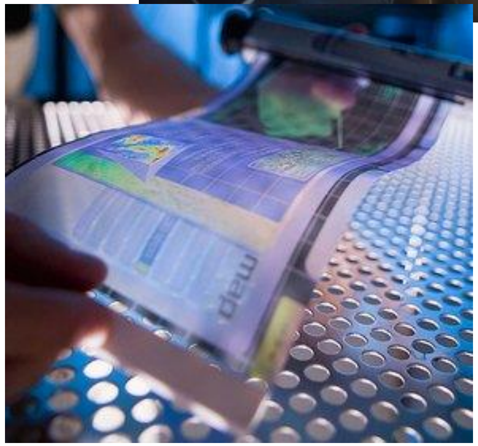
Level 3: ML Agent Introducing Decisions: automated microscopy

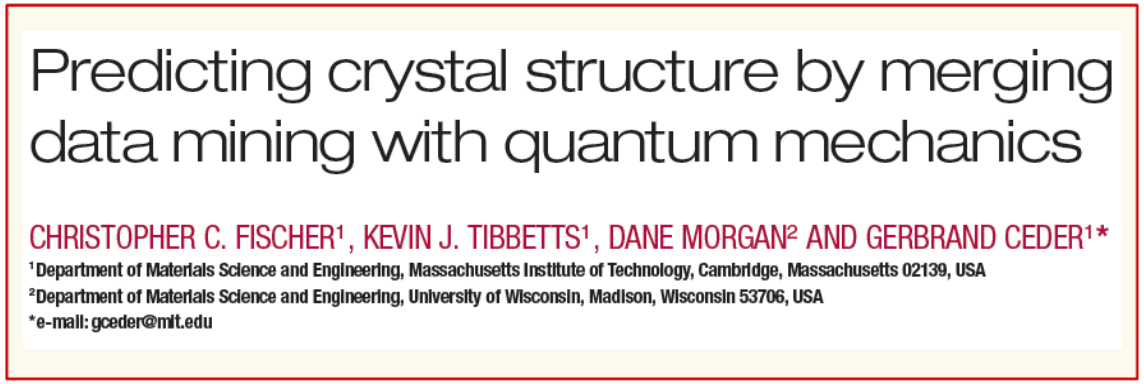
The World is a Material Opportunity











Predicting crystal structure by merging data mining with quantum mechanics

CHRISTOPHER C. FISCHER[1], KEVIN J. TIBBETTS[1], DANE MORGAN[2] AND GERBRAND CEDER[1]*

[1]Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

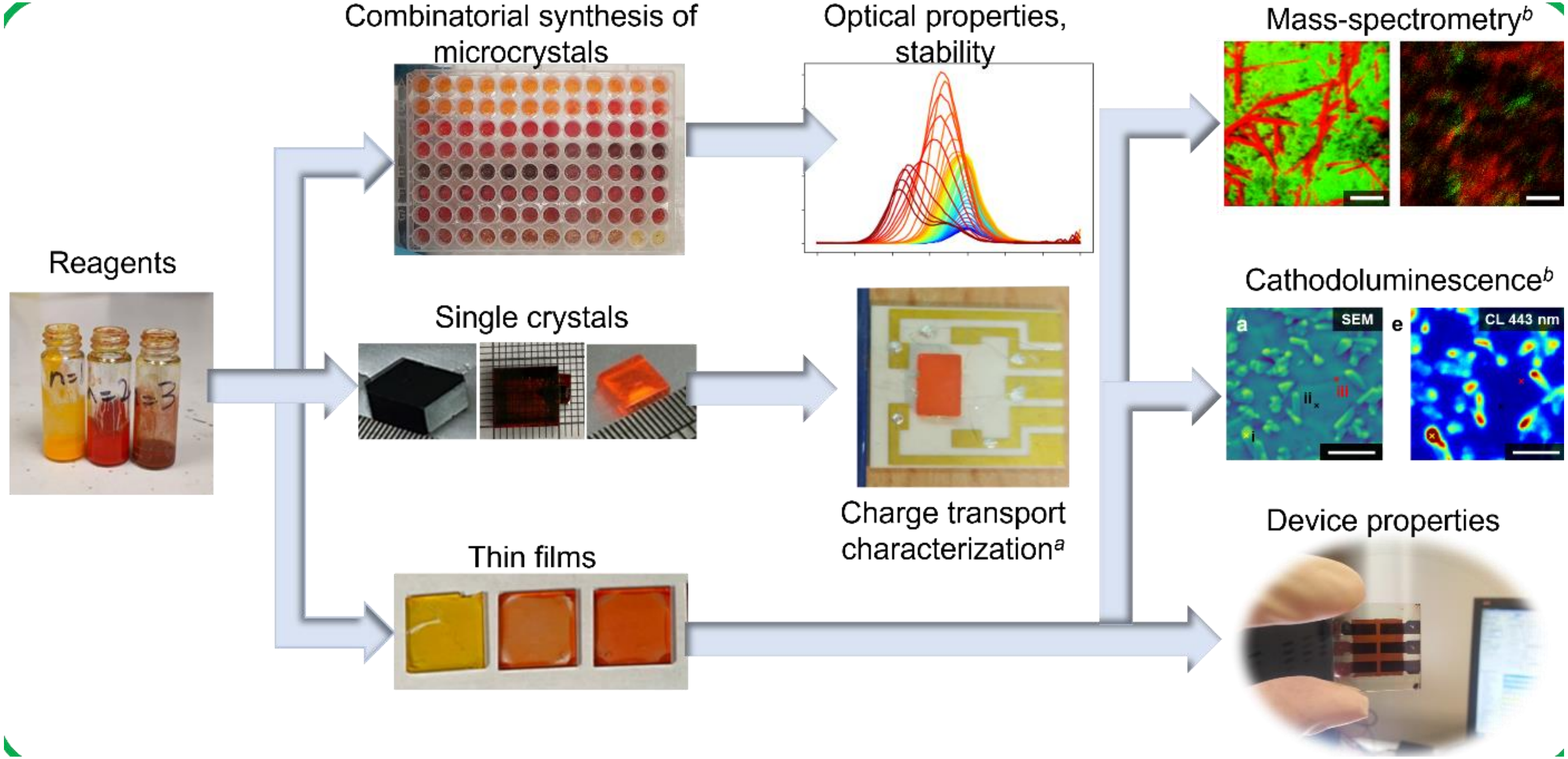
[2]Department of Materials Science and Engineering, University of Wisconsin, Madison, Wisconsin 53706, USA

*e-mail: gceder@mit.edu

- **"Improve":** Renewable energy, structural materials
- **"Discover":** RT superconductivity, high mechanical stress materials
- **"Engineer":** Quantum computing, single-atom catalysts, biomolecules

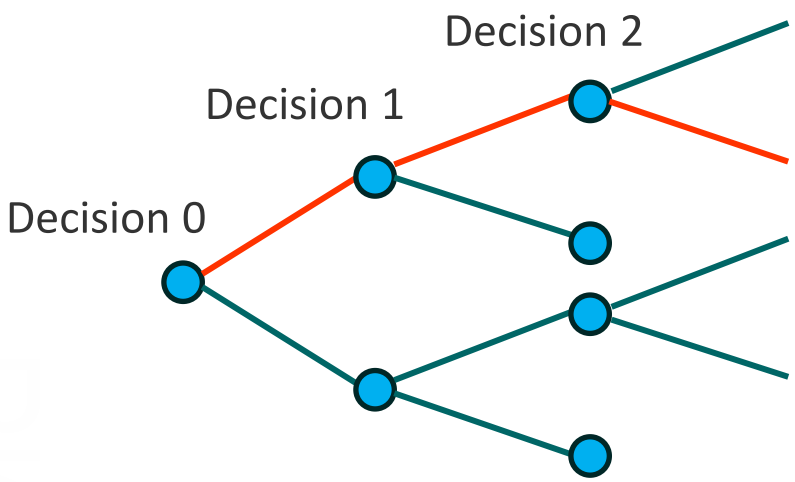
Functionality, manufacturability, cost

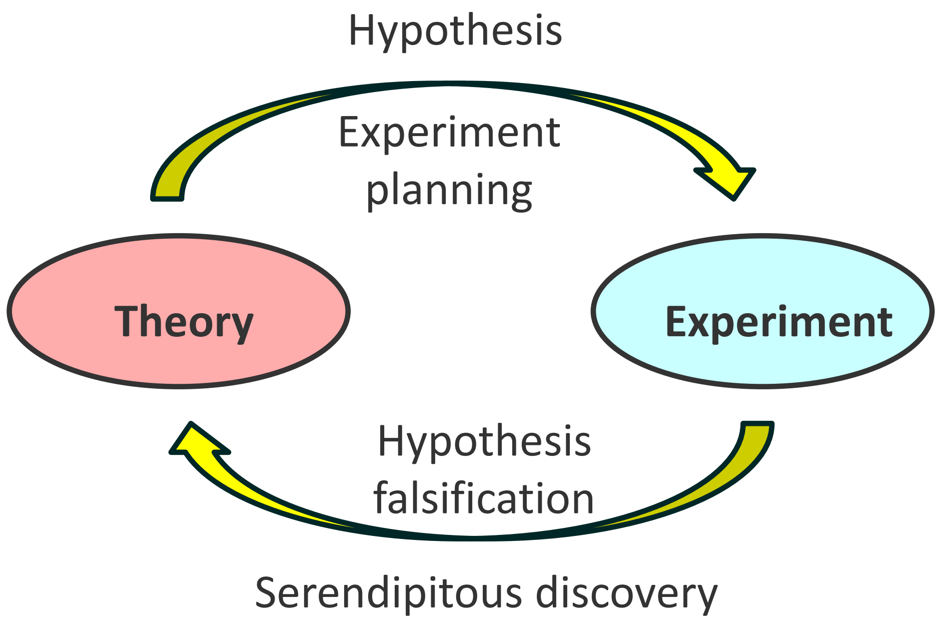
Decision science of workflows



- Multiple levels of decision making based on perceived gain, latencies, and costs
- Iterative cycles between low-cost and expensive measurements
- Learning **basic science/models** as a strategy to minimize cost and answer interventional and counterfactual questions

Decision science of workflows

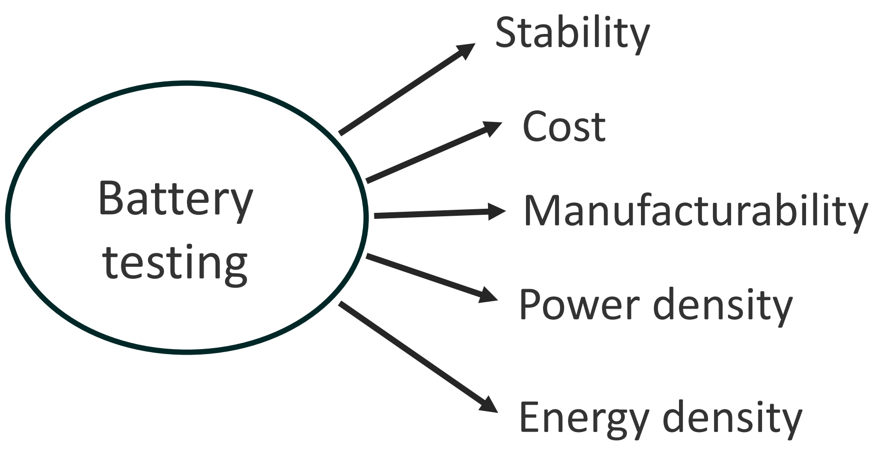


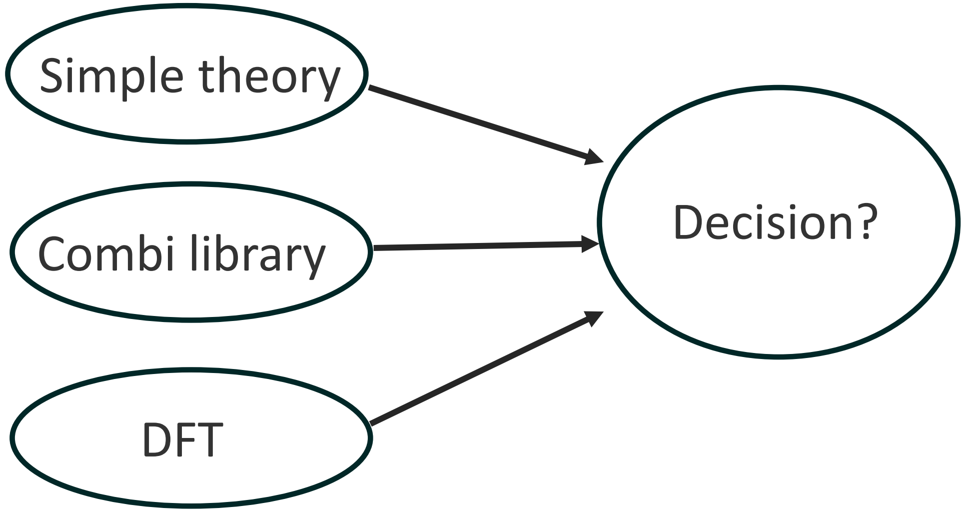


- **Experiment is a combinatorial space of opportunities:**
  → Investing only in scaling of throughput is only a linear improvement
- **Knowledge of physics often allows to reduce complexity: combinatorial to linear**:
  → Basic science pays off (with time)!
- **Science is a cycle between theory-driven hypothesis generation and experiment**:
  → We need to accelerate all elements of the cycle
- **Experimental and computational tool development:**
  → Currently constrained by human paradigm

If the part of a workflow is automated, our autonomous decision-making ability should match the level of autonomy!

Decision science of workflows

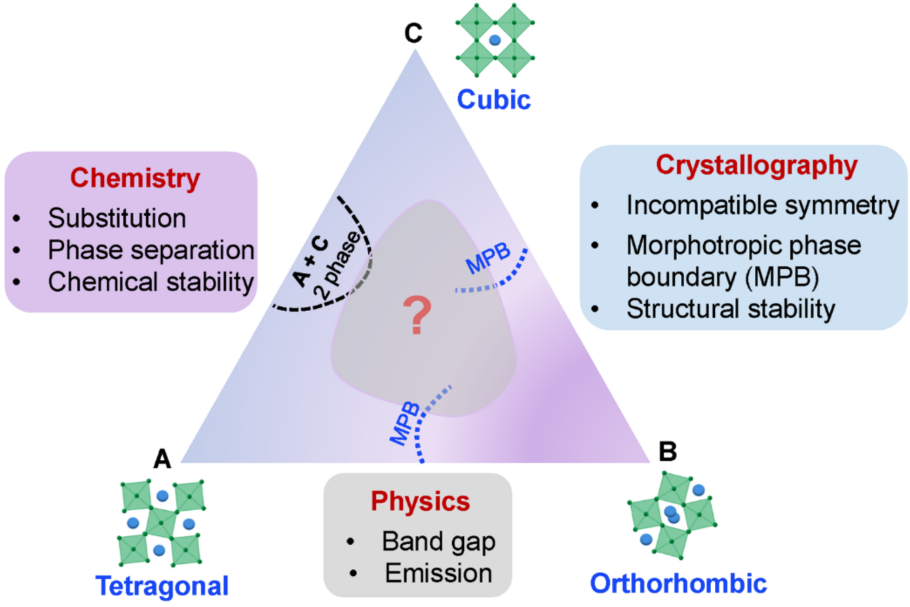




1. We need to balance multiple functionalities
2. Integrate multiple sources of data
3. Make decisions considering costs, latencies, physical inferential biases, and beliefs

Key consideration: **reward function**

1. Pure physical discovery (symbolic laws)
2. Data-driven exploration
3. Materials optimization
4. …

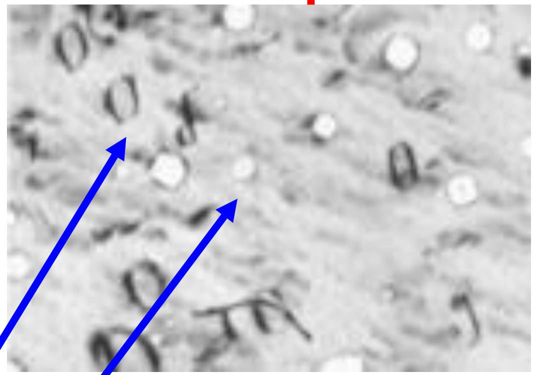


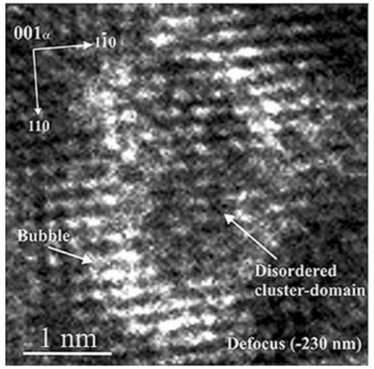
Workflows for Nuclear Materials Design

Traditional experiment:

1. Always based on workflows
2. Ideated, orchestrated, and implemented by humans
3. The “gain of value” during the workflow implementation is uncertain

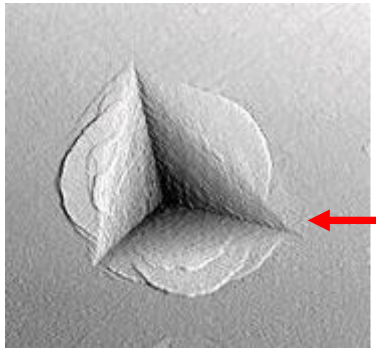
Materials selection





Prior knowledge

Operator curiosity

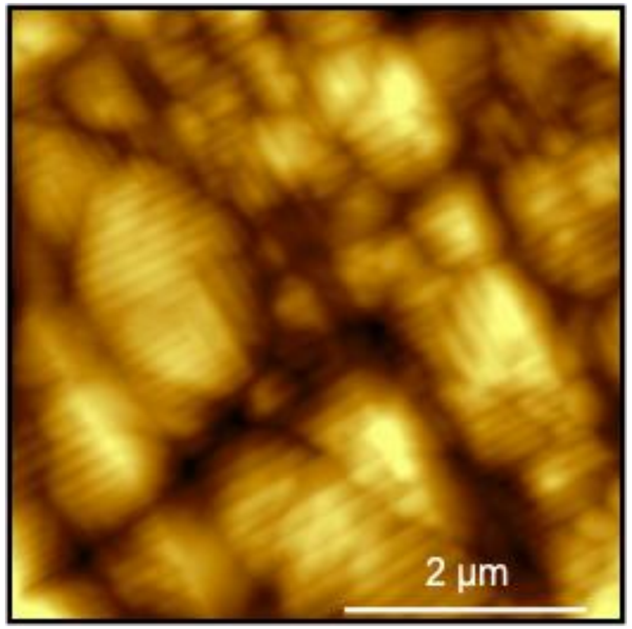


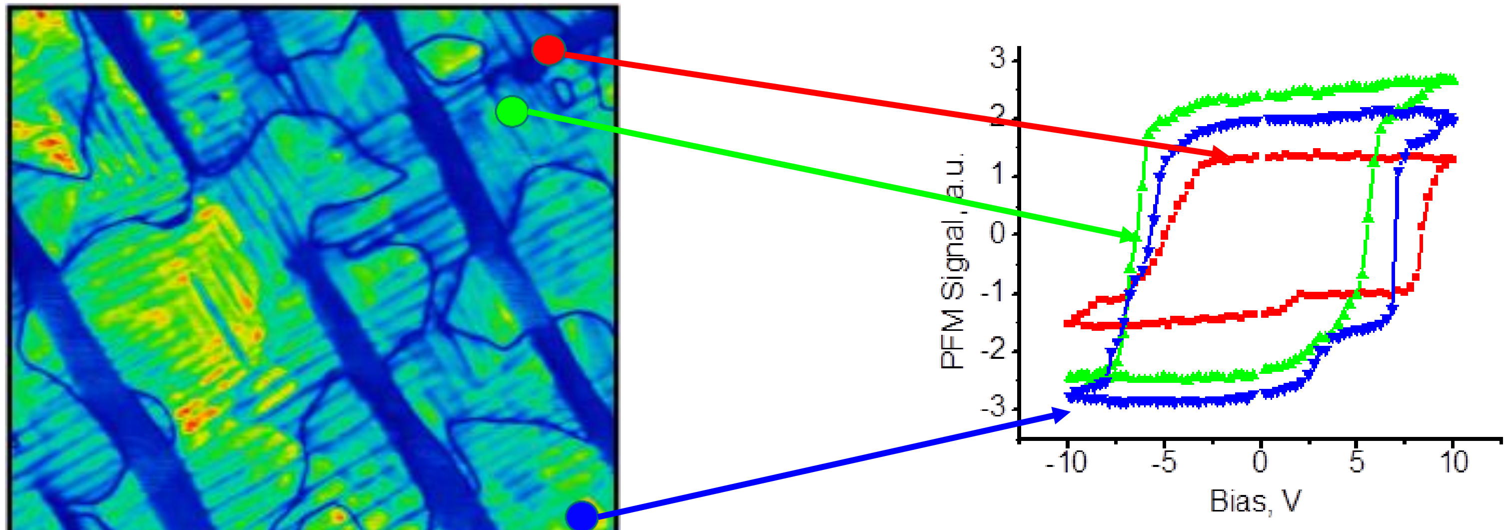
Value

Value of the step is key element:

- Either based on prior knowledge
- Or defined in a sense of the reinforcement learning Q-function

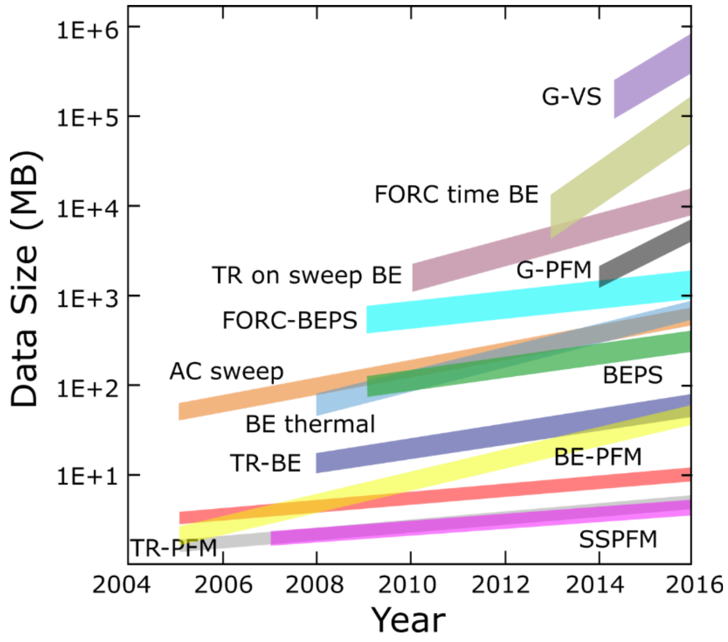
Decision Making in SPM



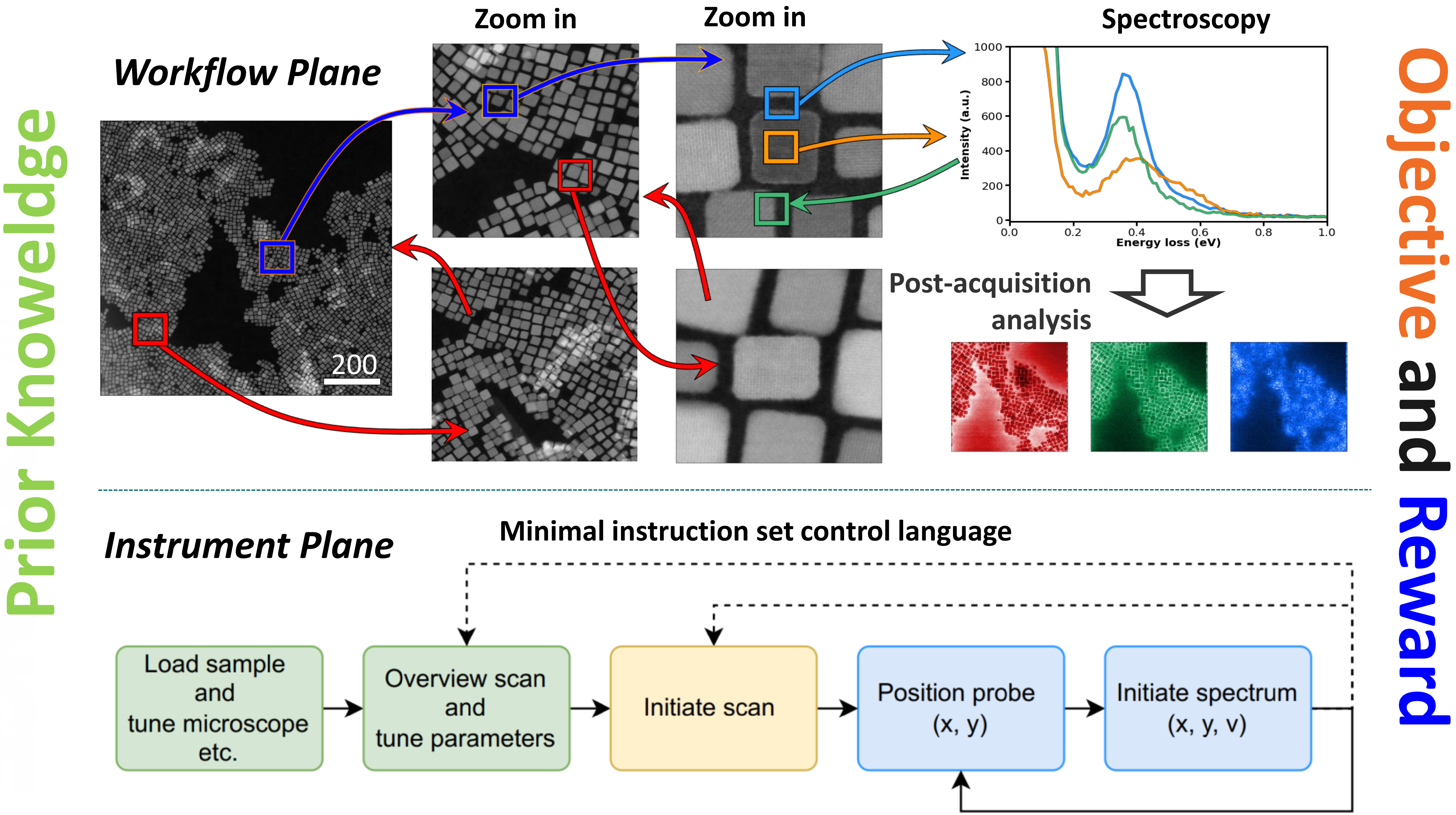


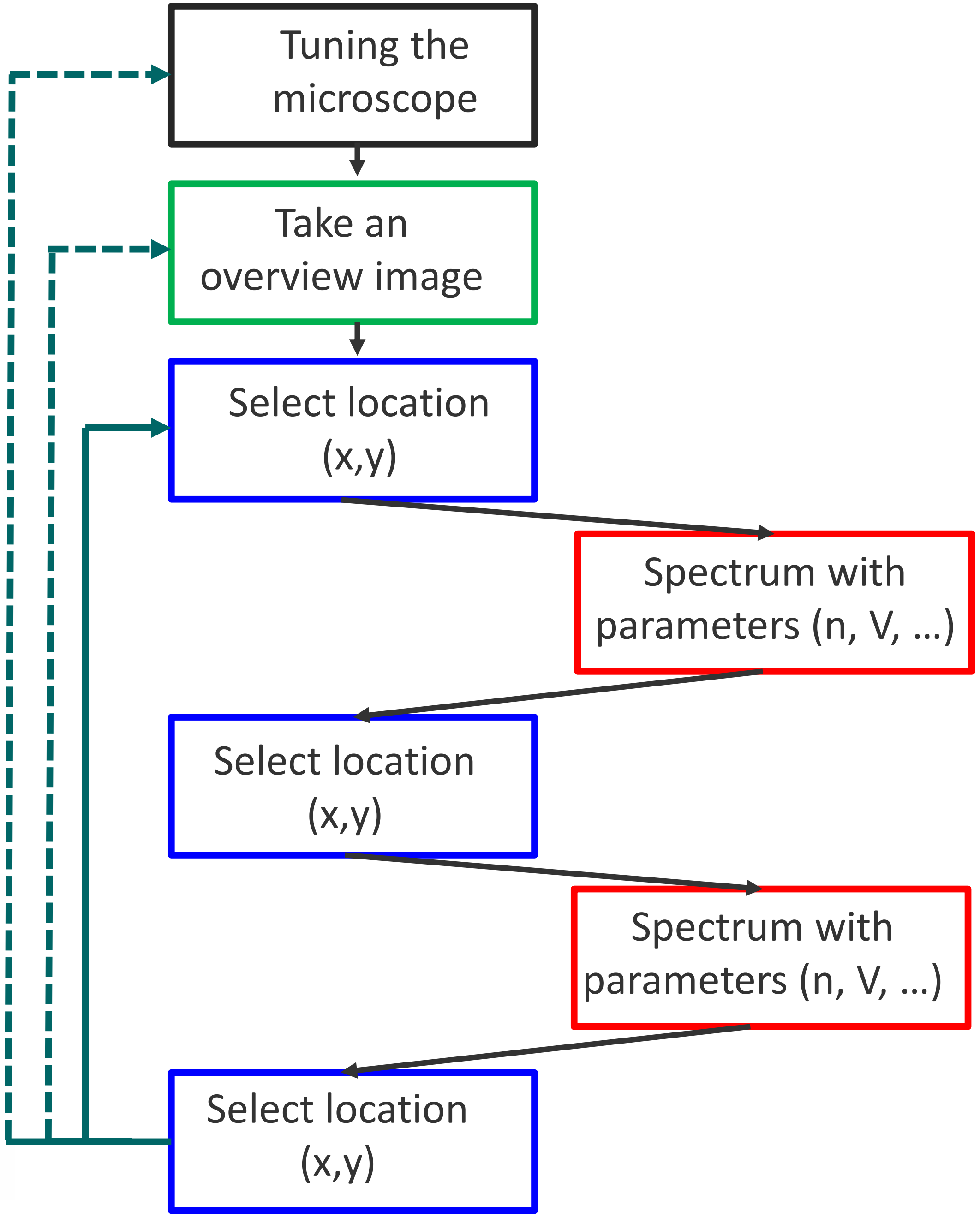
- Interesting functionalities are expected at the certain elements of domain structure
- We can guess some; we have to discover others

- **Experimental objectives →ML Rewards**
  - Microscope optimization
  - Properties of a priori known regions of interest
  - Discovery of regions with interesting properties
  - Physical theory falsification



Workflows in STEM





To implement the ML workflows, we start from emulating the human operations:

- Well defined and explainable commands
- Extensive domain expertise
- Potentially available data from experiments

Development of ML workflows can give rise to more complex imaging modalities

- Data volumes and dimensionalities above human level
- More complex modes of sampling
- “Guardian angel” modules

However, we always have to think about

- Reward function(s) for imaging problem
- Reward functions for materials problem
- Overall objective

Challenges for the Automated Experiment

Elements of realistic workflow design

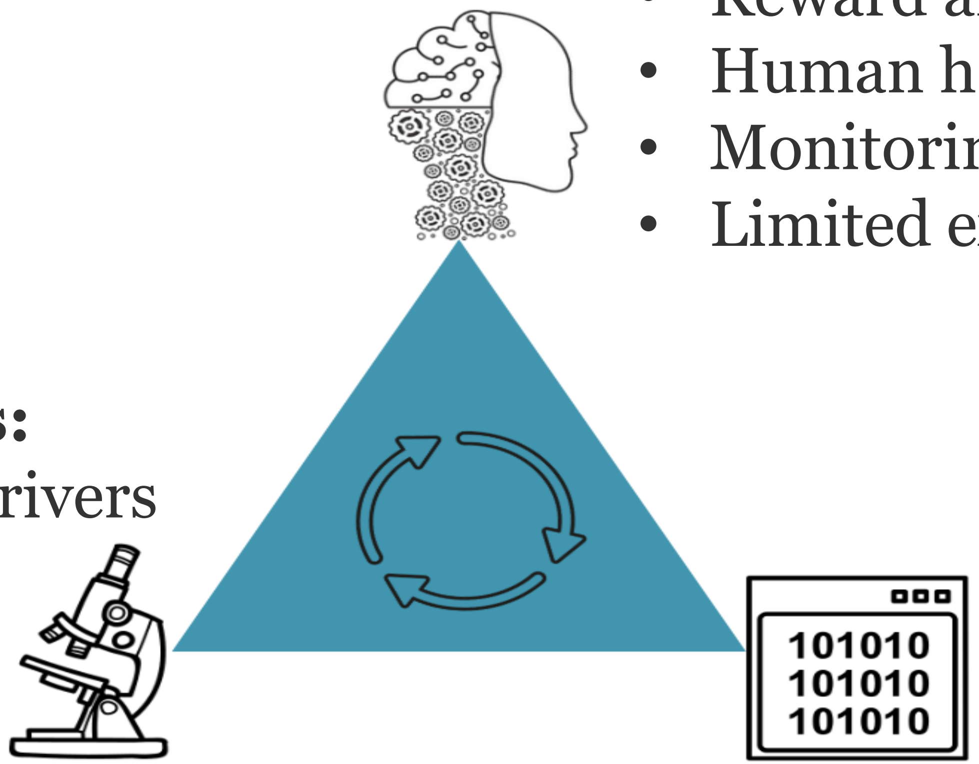
- Co-orchestration of multiple measurement modalities
- Building theory in the loop
- Integration between multiple domains

Workflow design:

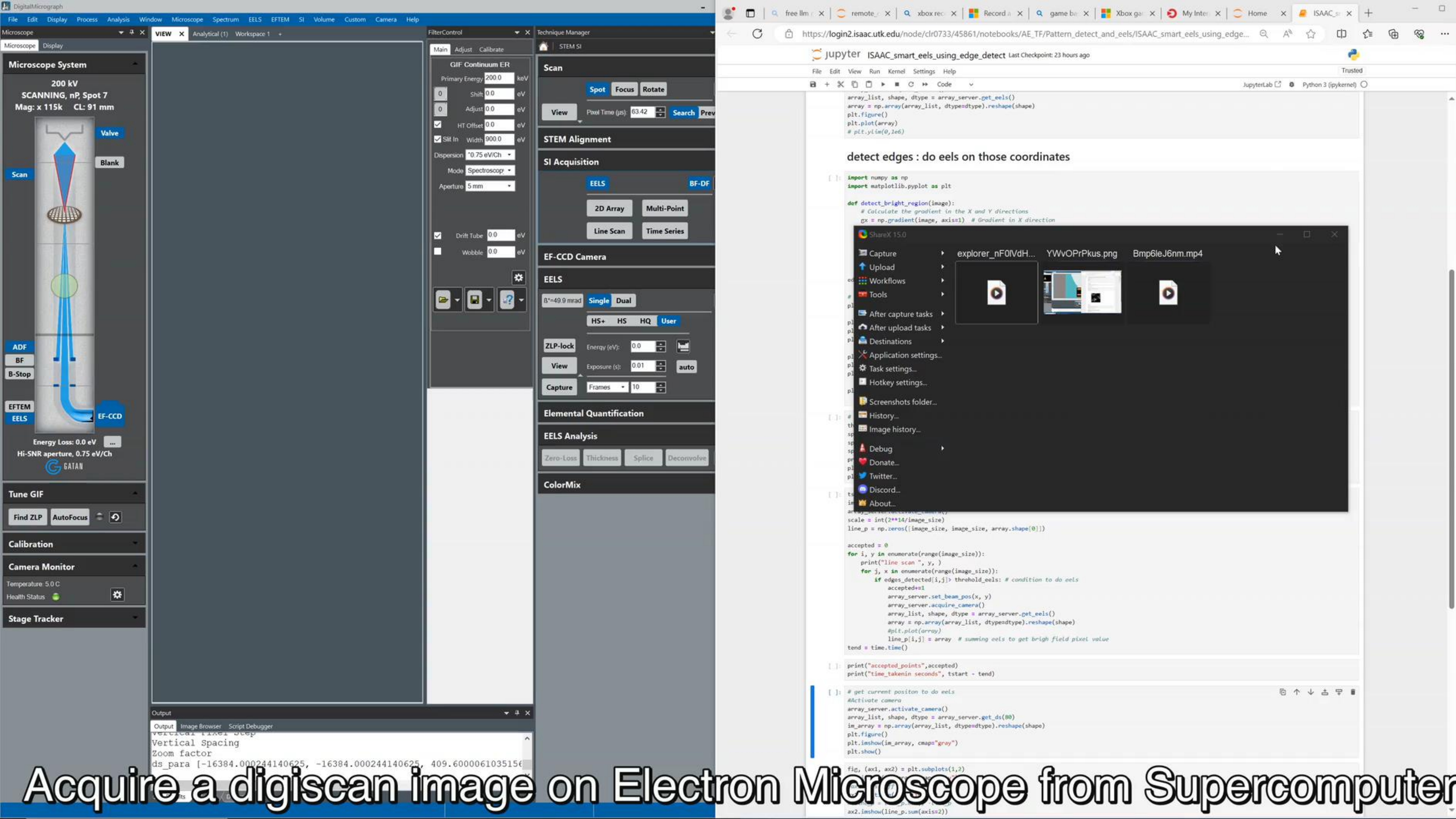
- Reward and value functions
- Human heuristics
- Monitoring
- Limited experimental budget

Engineering controls:

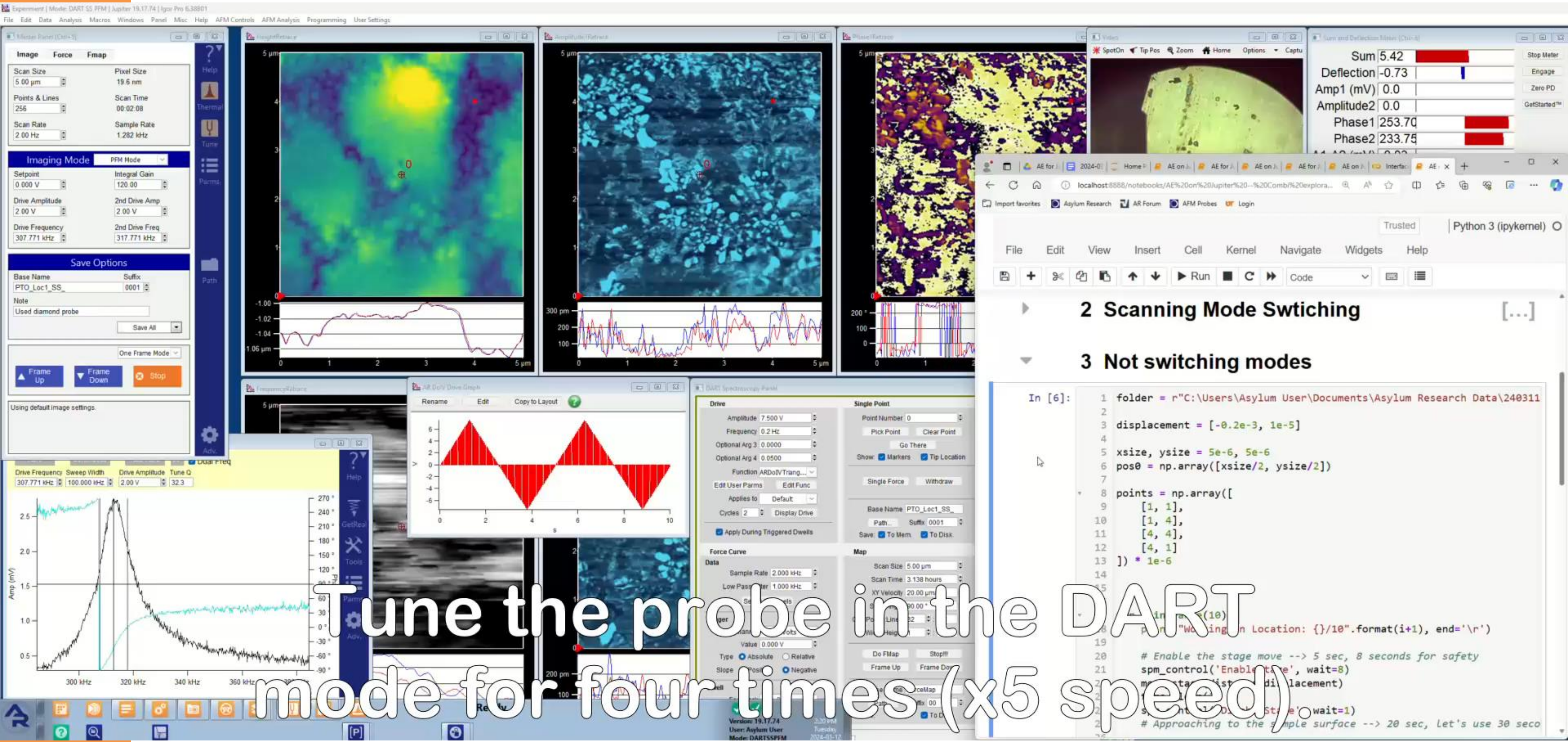
- Instrument-specific drivers
- Hyperlanguage
- Python APIs



ML/AI



Acquire a digiscan image on Electron Microscope from Supercomputer



Tune the probe in the DART mode for four times (x5 speed)

File Edit View Insert Cell Kernel Widgets Help

Trusted | gpuenv

Run | Code

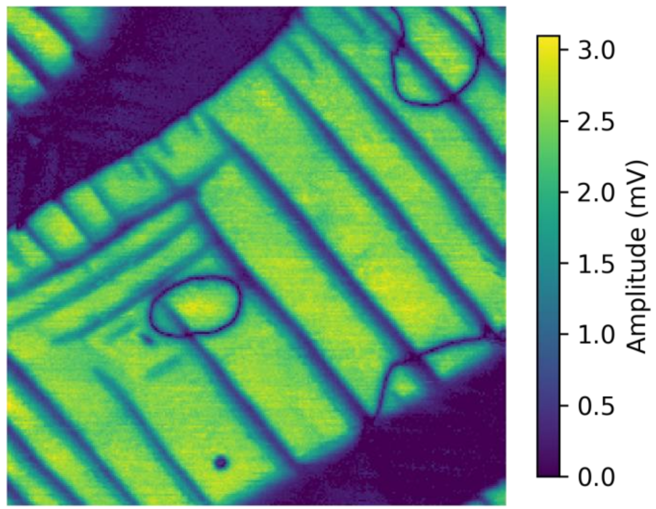
```
move_(-volt*2-(offsetvx), 0, 0-offsetvy, 0, move_speed)
```

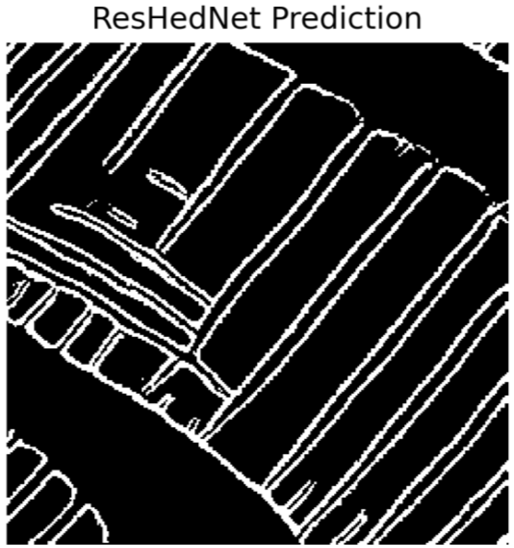

```
scanning line #56
```

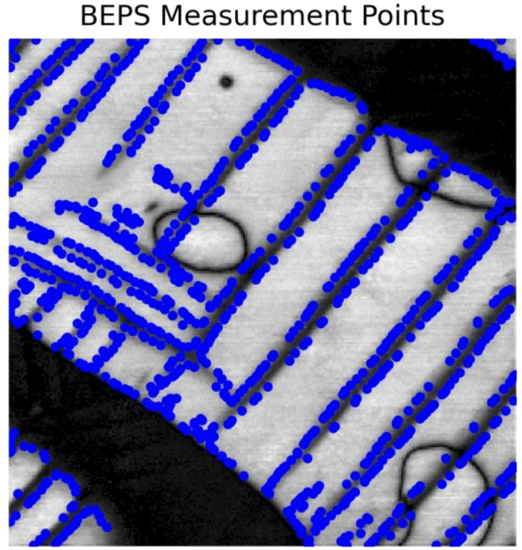
In [ ]:

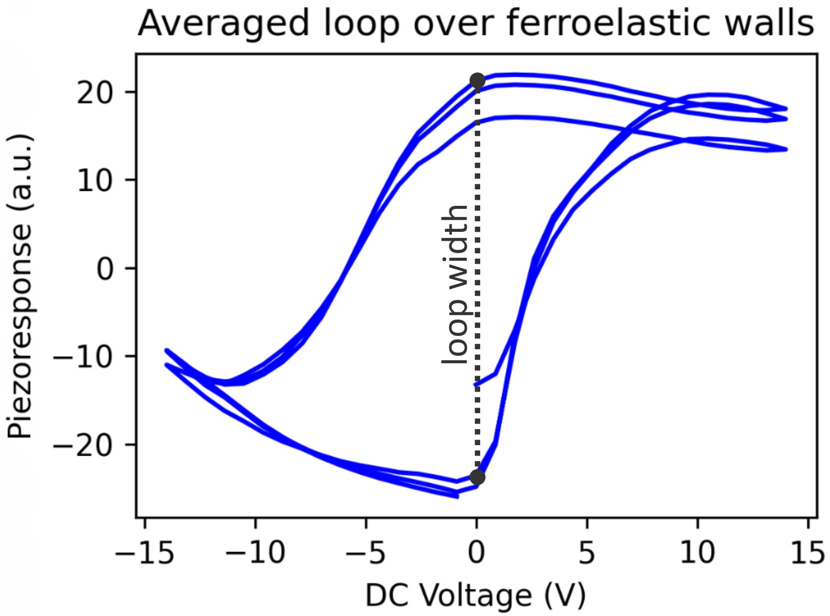
In [ ]:

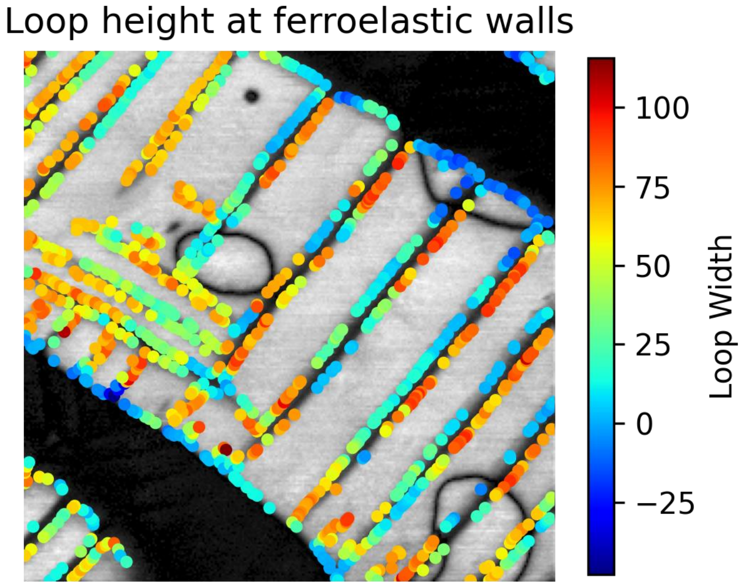
Mapping Activity of Domain Walls

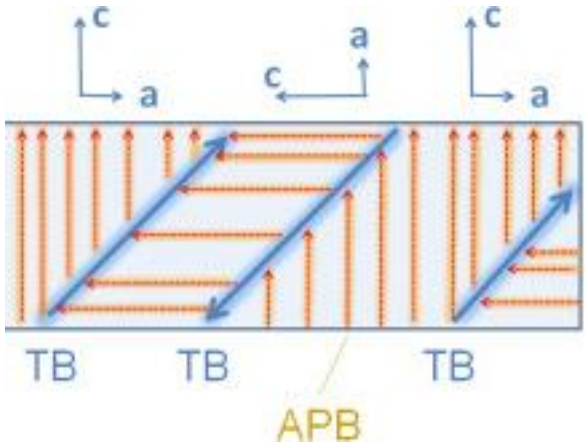






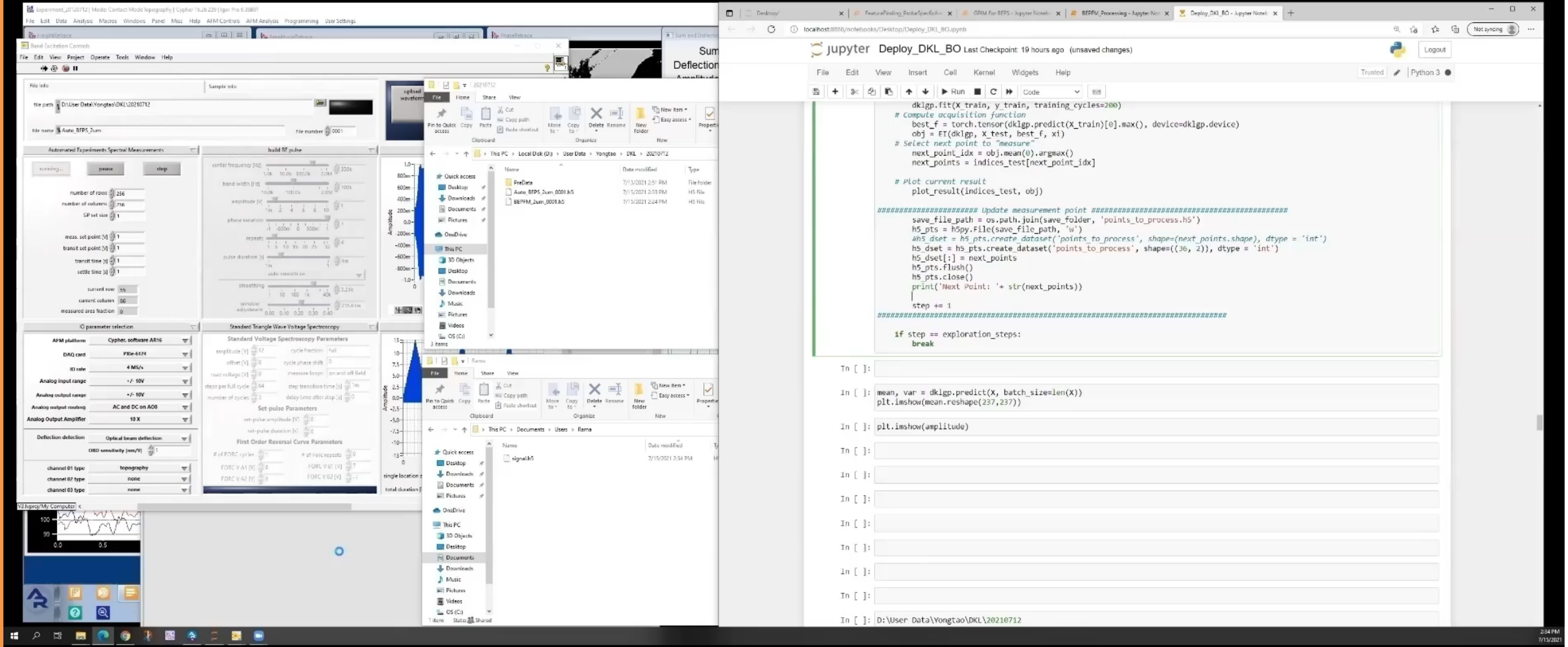






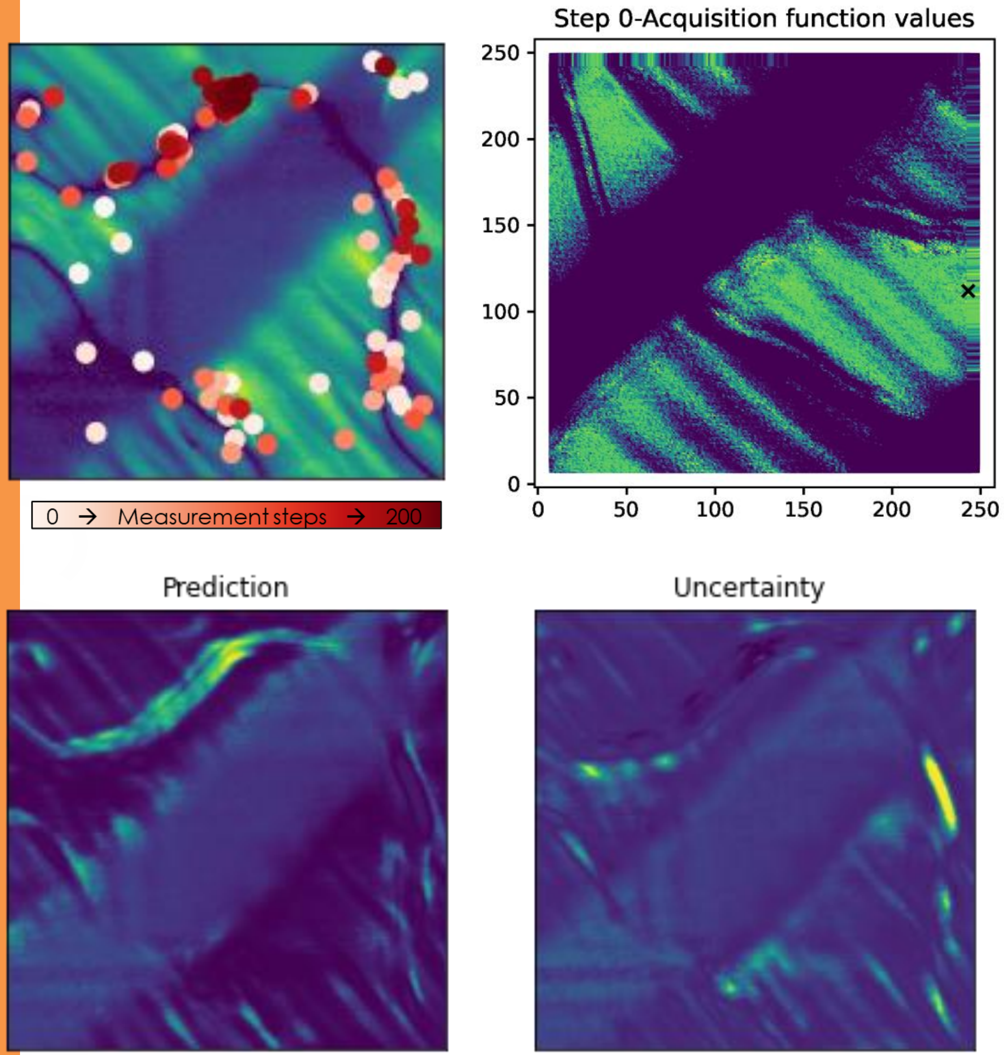
Liu, Y., Kelley, K.P., Funakubo, H., Kalinin, S.V., Ziatdinov, M., Arxiv, submitted

Deep Kernel Learning AE

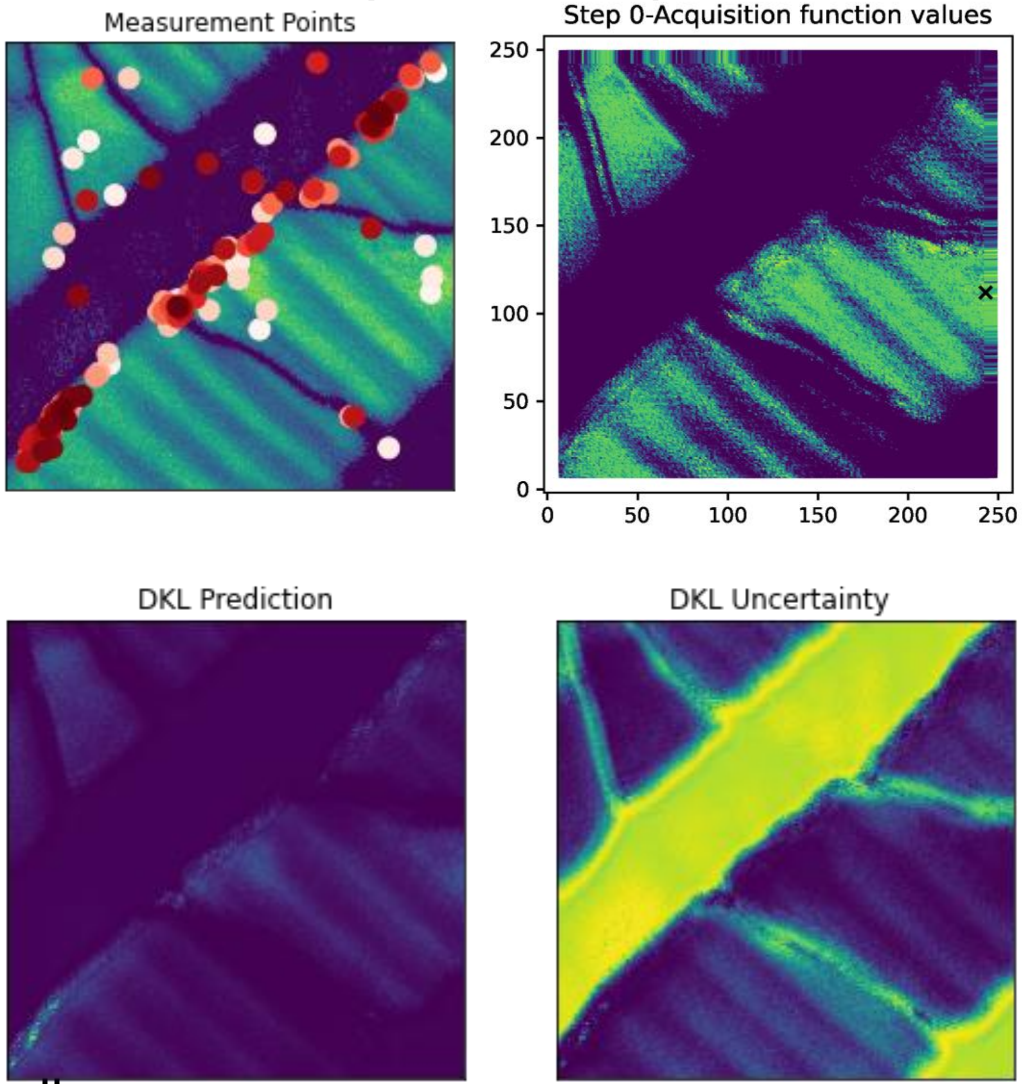


Deep Kernel Learning SPM

Guided by: On field loop area



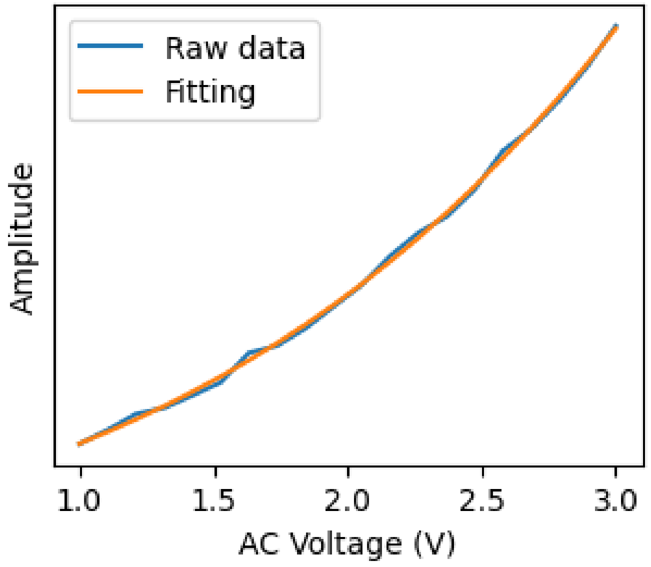
Guided by: Off field loop area



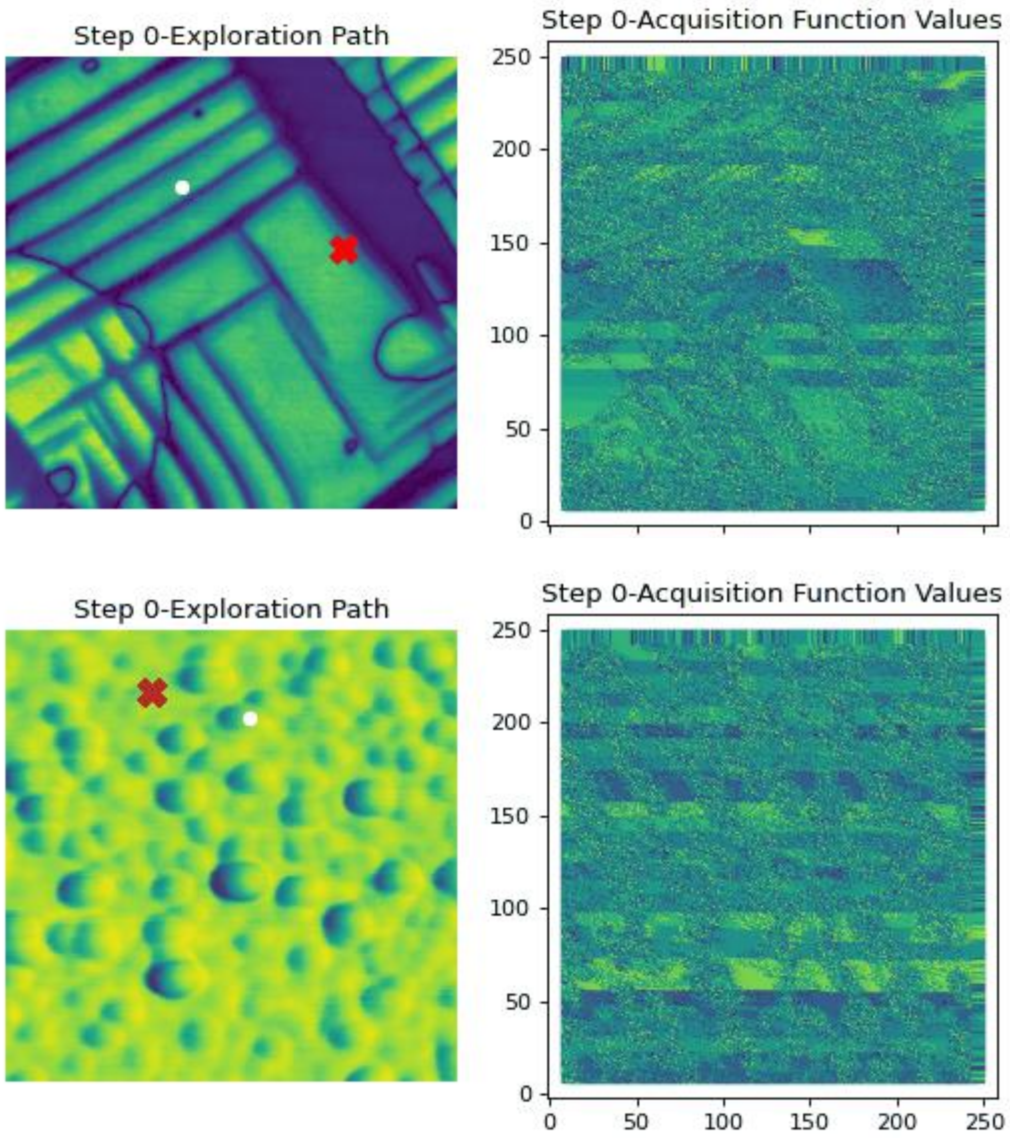
- ➢ Large loop opening corresponding 180º domain walls
- ➢ This behavior can be attributed to the large polarization mobility of 180º walls

Liu, Yongtao, et al, Nature Machine Intelligence 4, 4 (2022): 341-350.

Why human in the loop?



- 200-step automated experiment
- PFM amplitude was used as structure ima
- V_{AC} sweep curve at each location was fitte
  $y = Ax^3 + Bx^2 + Cx$
- A, B, C, and A/B were used as the target function to guide DKL-V_{AC} measurement.



The methodologies of classical ML (hyperparameter optimization, cross-validation) are rarely applicable for active learning!

The dance of policies and rewards

Rewards and objectives:

- What is our (hierarchical) objective?
- Can we define reward(s)?

Inferential biases:

- What do we know before the experiment?
- What do we (hope to) learn after the experiment?

Experiment planning – policies and values

- How do we plan experiment in advance (policies or values based on rewards)?
- Can we ascribe value to certain steps?
- Do we change our policies during experiment?