THE UNIVERSITY of TENNESSEE KNOXVILLE

Day 5: Rewards and Decision Making for Automated Microscopy

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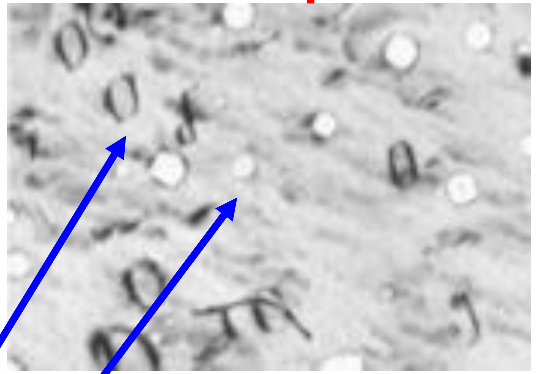
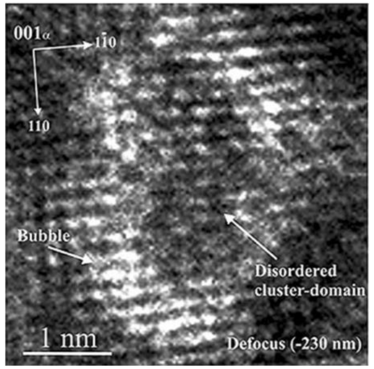
Workflows for Nuclear Materials Design

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

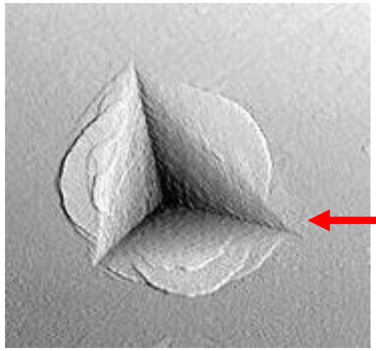
Materials selection





Prior knowledge

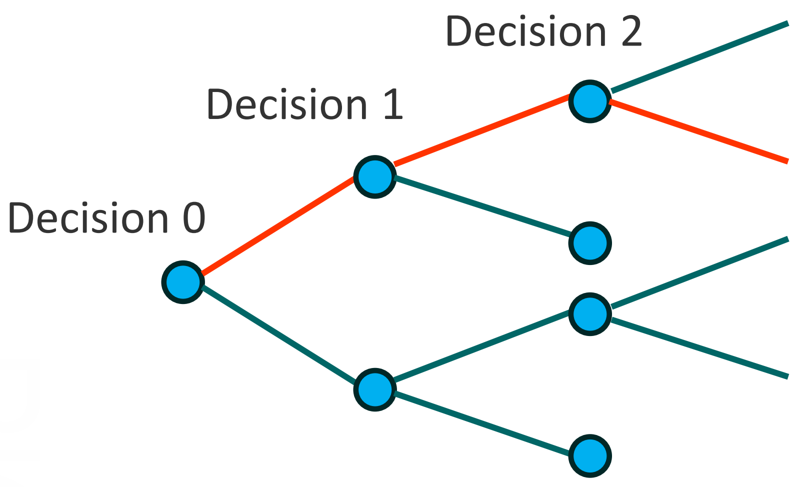
Operator curiosity

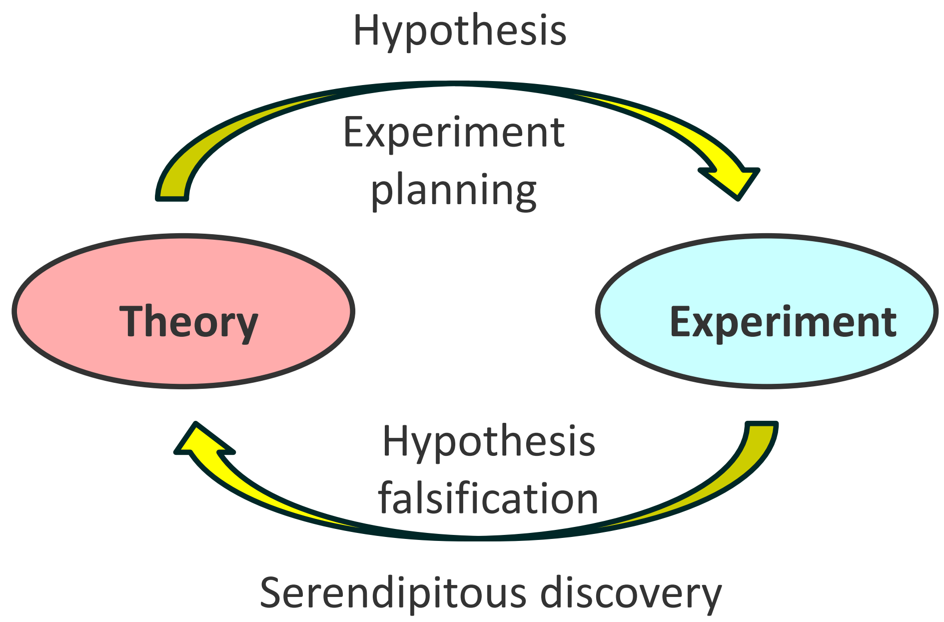


Value

Value of the step is key element:
- Either based on prior knowledge
- Or defined in a sense of the reinforcement learning Q-function

Decision science of workflows

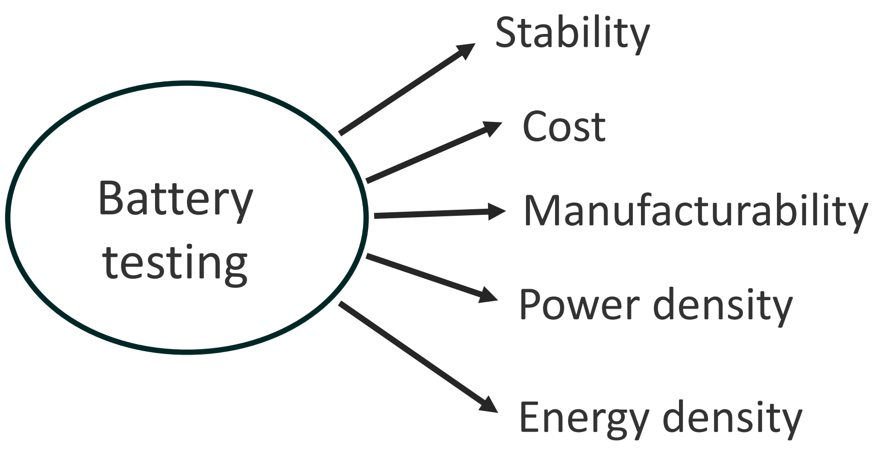


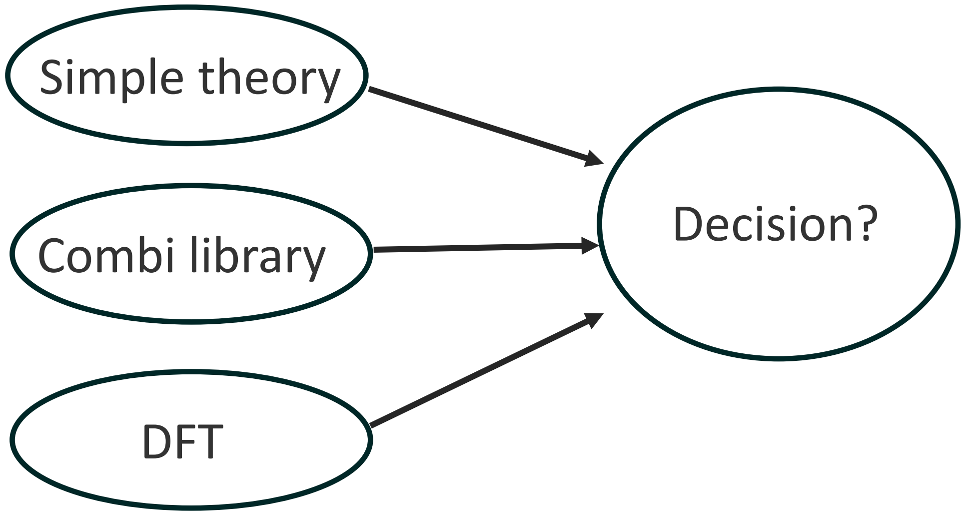


- **Experiment is a combinatorial space of opportunities:**
  → Investing only in scaling of throughput is only a linear improvement
- **Knowledge of physics often allows to reduce complexity: combinatorial to linear**:
  → Basic science pays off (with time)!
- **Science is a cycle between theory-driven 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

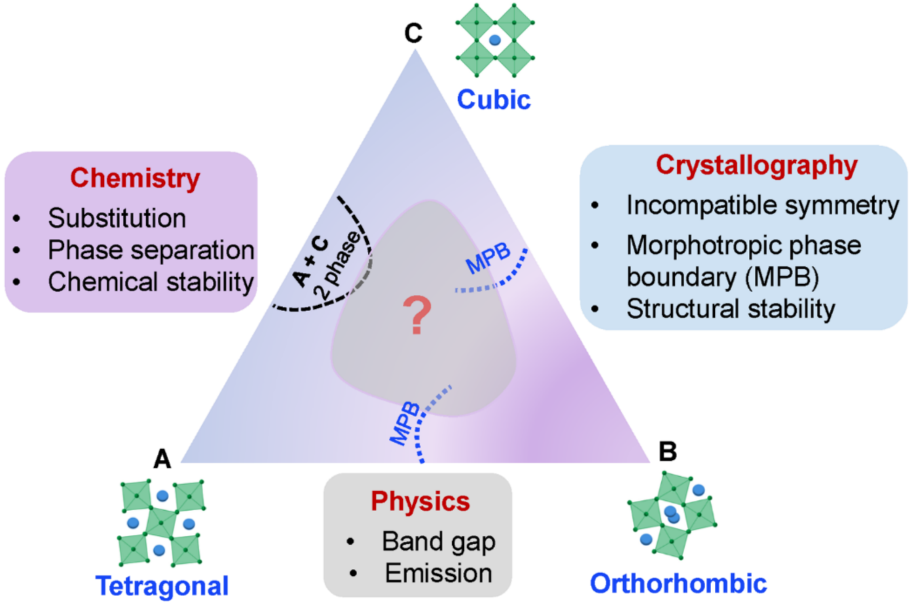




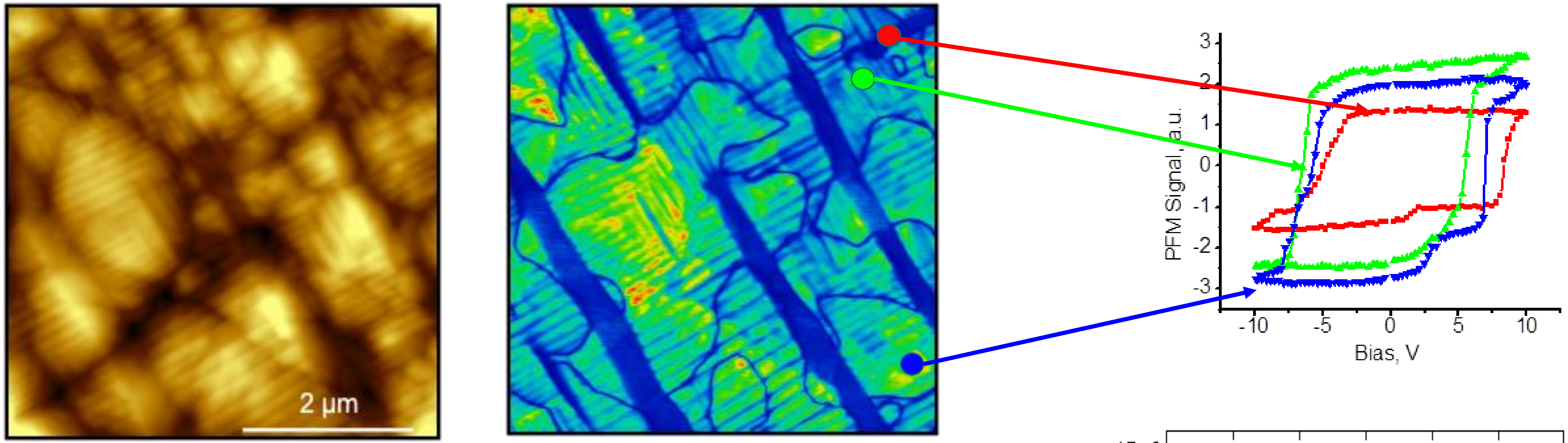
1. We need to balance multiple functionalities
2. Integrate multiple sources of data
3. Make decisions considering costs, latencies, physical inferential biases, and beliefs

Key consideration: reward function

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

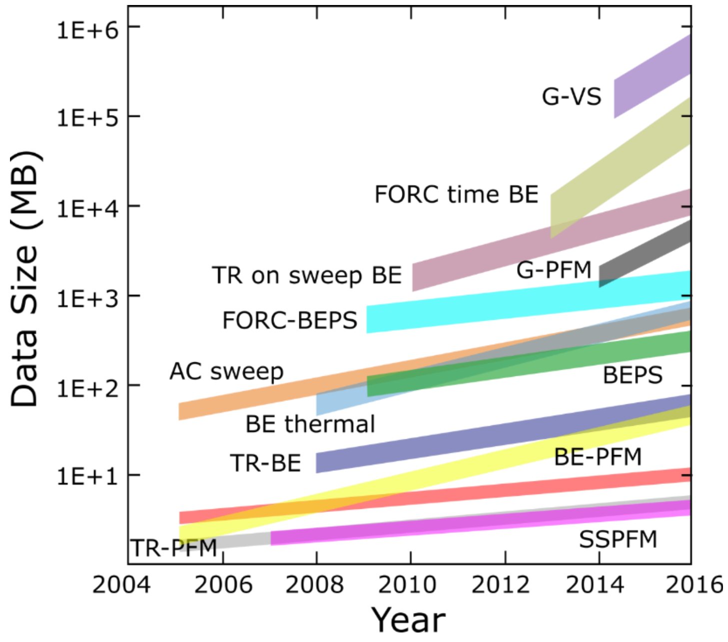


Decision Making in SPM

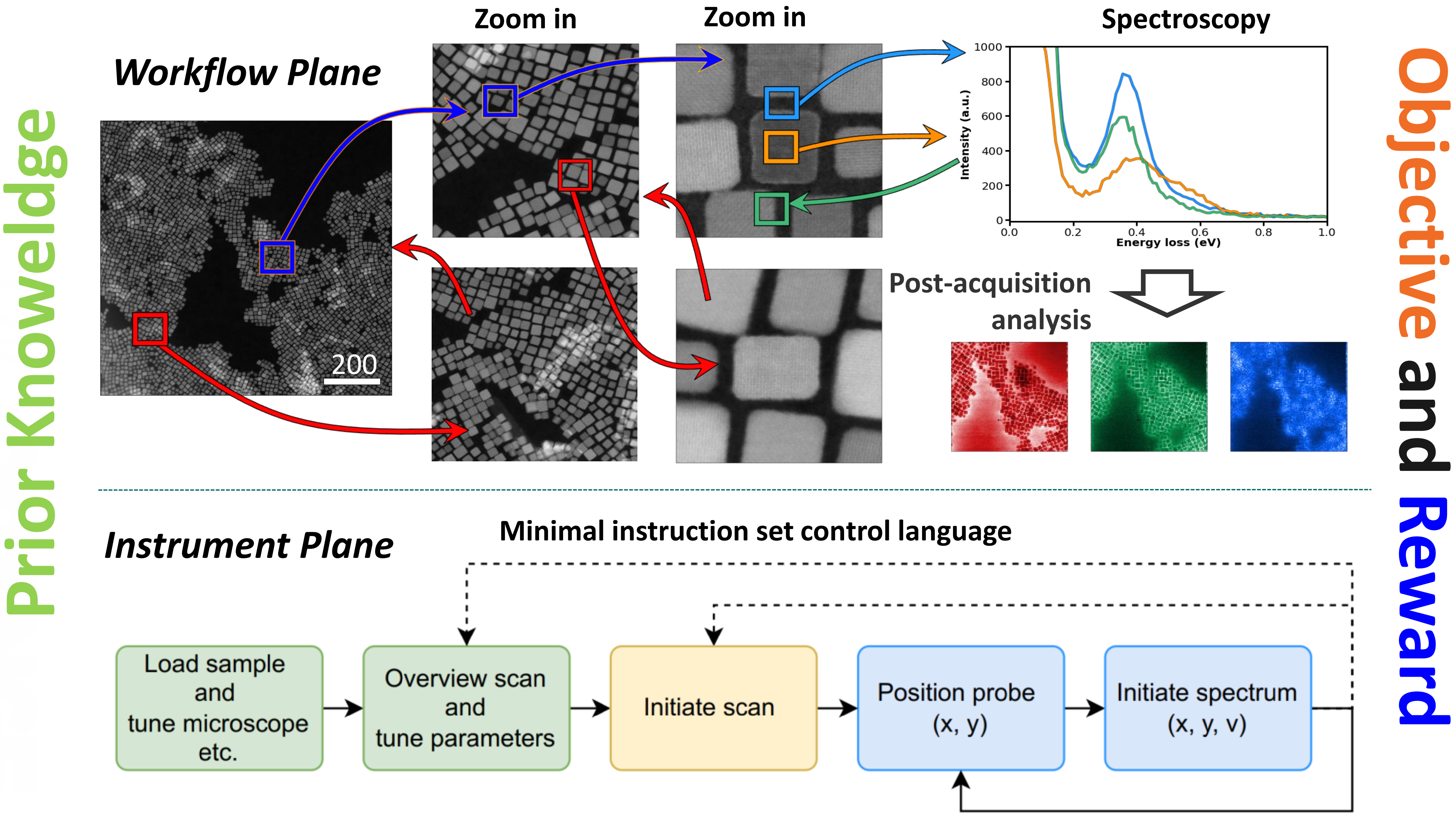


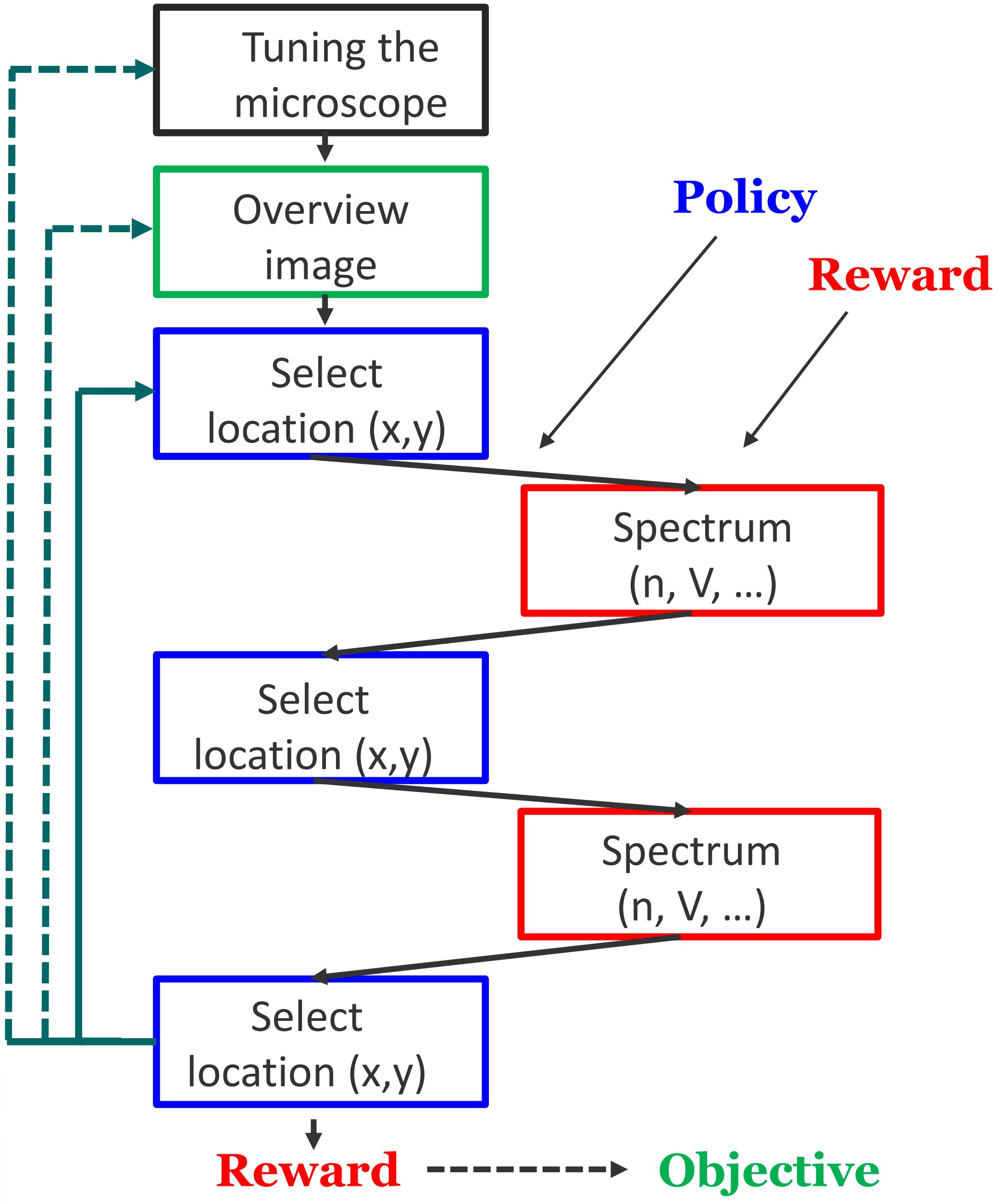
- Interesting functionalities are expected at the certain elements of domain structure
- We can guess some; we have to discover others

- **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

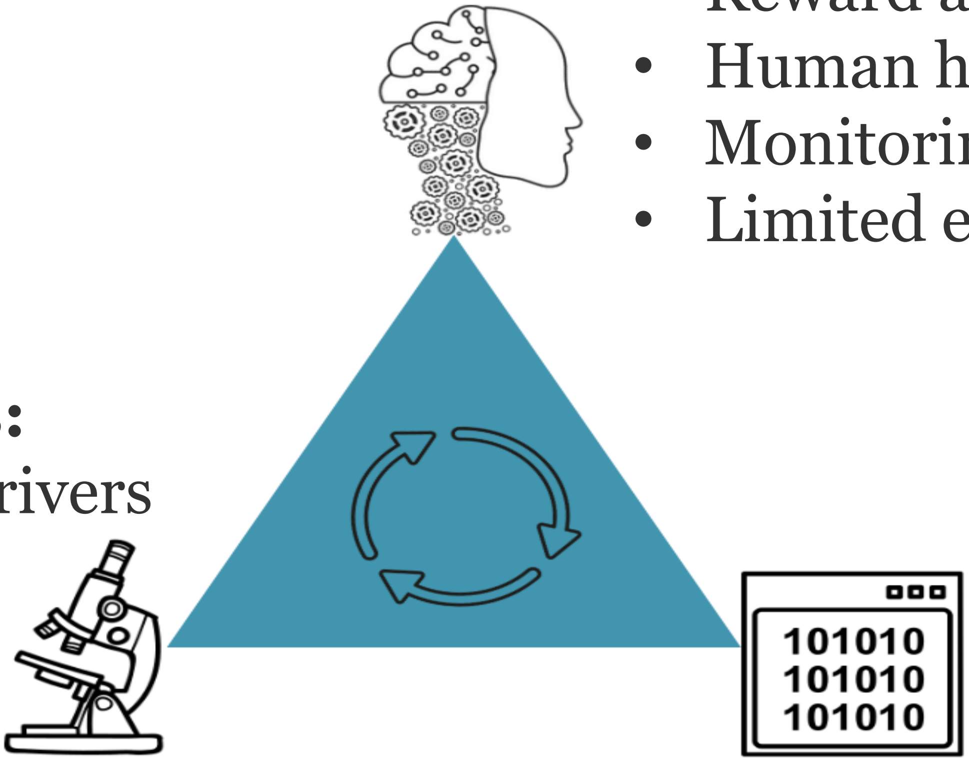
- Co-orchestration of multiple measurement modalities
- Building theory in the loop
- Integration between multiple domains

Workflow design:

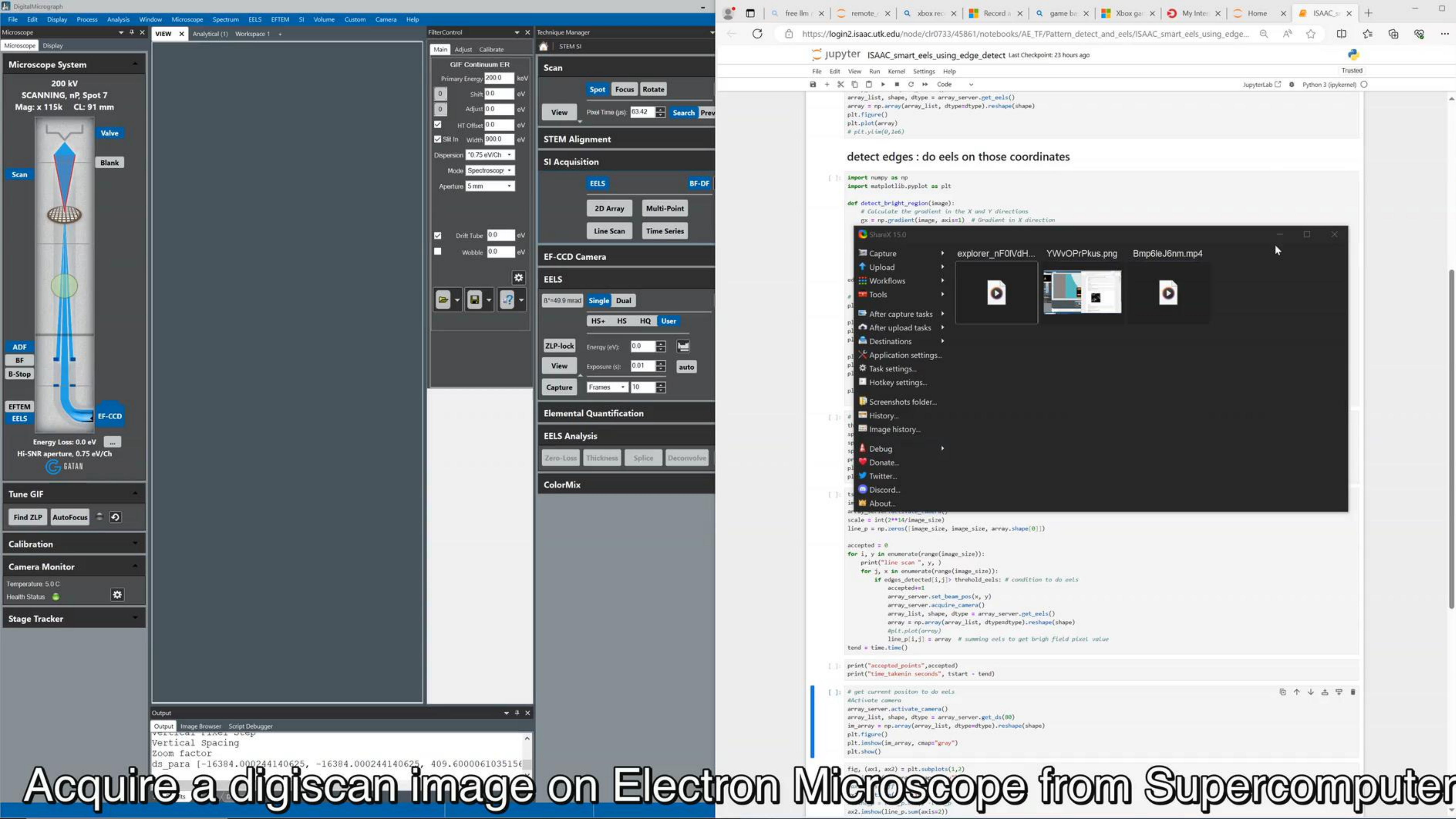
- Reward and value functions
- Human heuristics
- Monitoring
- Limited experimental budget

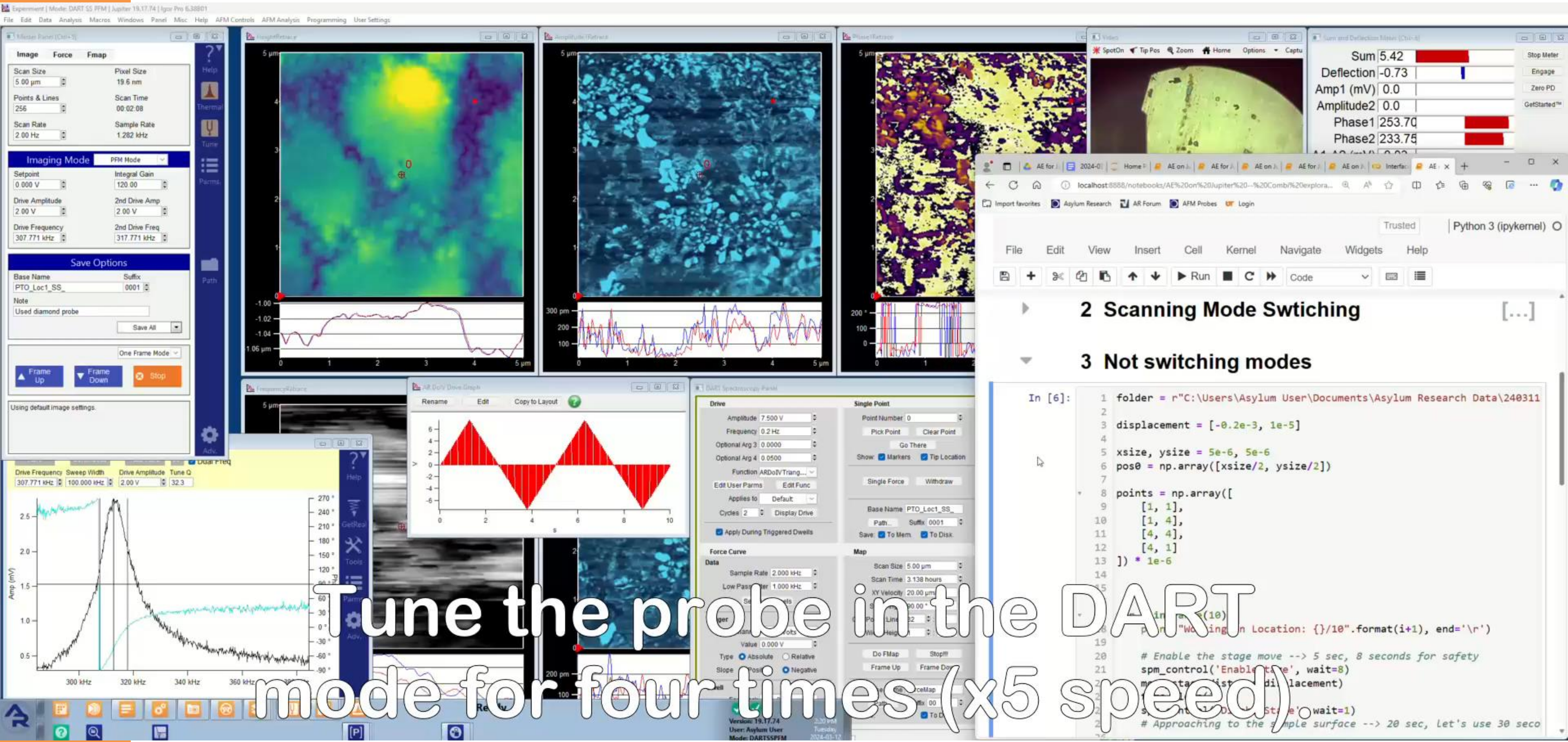
Engineering controls:

- Instrument-specific drivers
- Hyperlanguage
- Python APIs



ML/AI


DigitalMicrograph
File Edit Display Process Analysis Window Microscope Spectrum EELS EFTEM SI Volume Custom Camera Help
Microscope System
200 kV
SCANNING, nP, Spot 7
Mag: x 115k CL: 91 mm
Energy Loss: 0.0 eV
Hi-SNR aperture, 0.75 eV/Ch
GATAN
Tune GIF
Find ZLP AutoFocus
Calibration
Camera Monitor
Temperature: 5.0 C
Health Status
Stage Tracker
FilterControl
GIF Continuum ER
Primary Energy 200.0 keV
Technique Manager
STEM SI
Scan
Spot Focus Rotate
STEM Alignment
SI Acquisition
2D Array Multi-Point
Line Scan Time Series
EF-CCD Camera
EELS
Elemental Quantification
EELS Analysis
ColorMix
Output
Vertical Spacing
Zoom factor
ds_para [-16384.000244140625, -16384.000244140625, 409.600006103515
jupyter ISAAC_smart_eels_using_edge_detect Last Checkpoint: 23 hours ago
detect edges : do eels on those coordinates
import numpy as np
import matplotlib.pyplot as plt
def detect_bright_region(image):
# Calculate the gradient in the X and Y directions
gx = np.gradient(image, axis=1) # Gradient in X direction
ShareX 15.0
Capture
Upload
Workflows
Tools
After capture tasks
After upload tasks
Destinations
Application settings...
Task settings...
Hotkey settings...
Screenshots folder...
History...
Image history...
Debug
Donate...
Twitter...
Discord...
About...
print("accepted_points",accepted)
print("time_takenin seconds", tstart - tend)
# get current positon to do eels
#Activate camera
array_server.activate_camera()
array_list, shape, dtype = array_server.get_ds(80)
im_array = np.array(array_list, dtype=dtype).reshape(shape)
plt.figure()
plt.imshow(im_array, cmap="gray")
plt.show()
Acquire a digiscan image on Electron Microscope from Supercomputer


Imaging Mode
Save Options
2 Scanning Mode Swtiching
3 Not switching modes
folder = r"C:\Users\Asylum User\Documents\Asylum Research Data\240311
displacement = [-0.2e-3, 1e-5]
xsize, ysize = 5e-6, 5e-6
pos0 = np.array([xsize/2, ysize/2])
points = np.array([
[1, 1],
[1, 4],
[4, 4],
[4, 1]
]) * 1e-6
Tune the probe in the DART
mode for four times (x5 speed)

File Edit View Insert Cell Kernel Widgets Help

Trusted gpuenv

Run Code

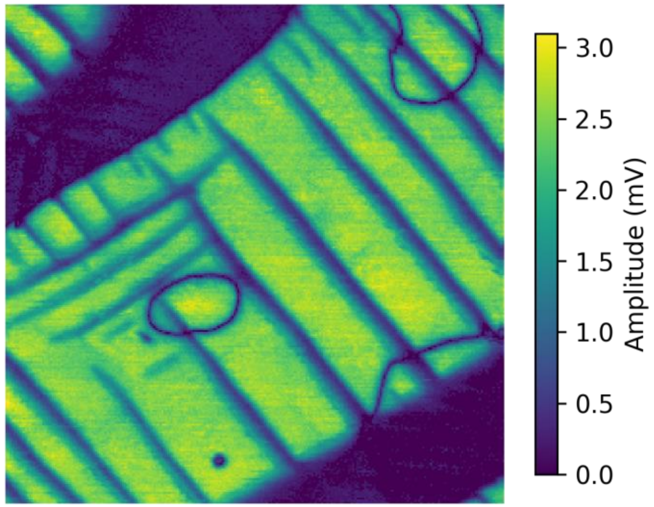
```
move_(-volt*2-(offsetvx), 0, 0-offsetvy, 0, move_speed)
```

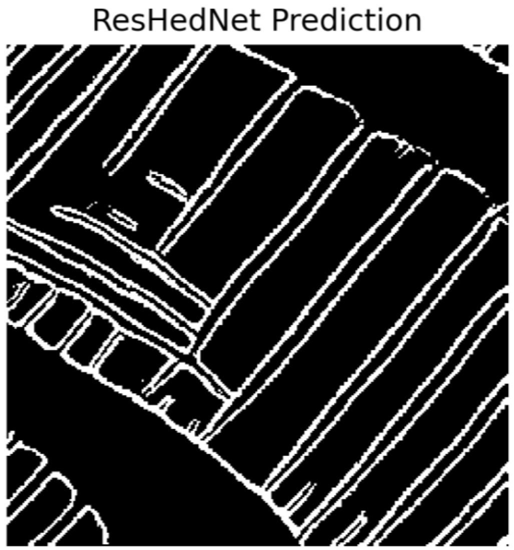

```
scanning line #56
```

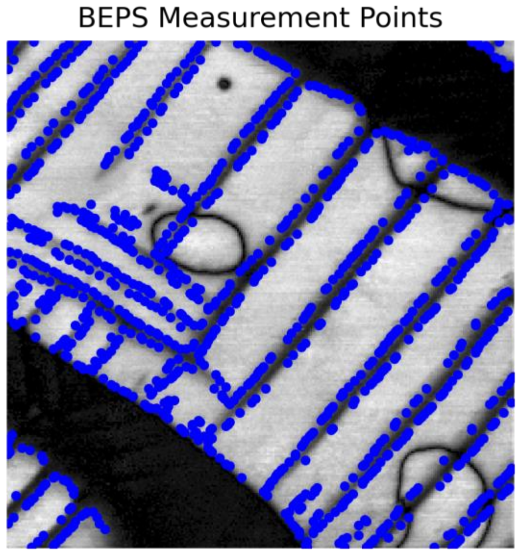
In [ ]:

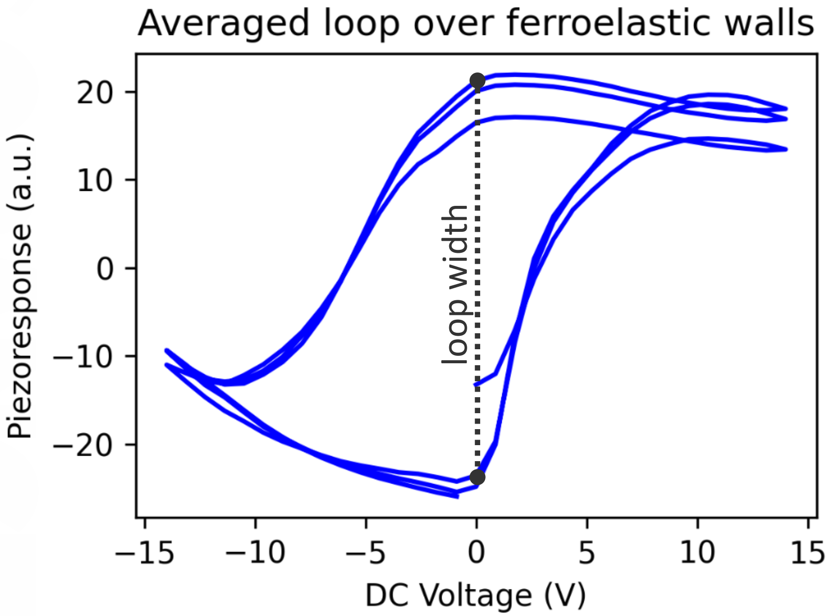
In [ ]:

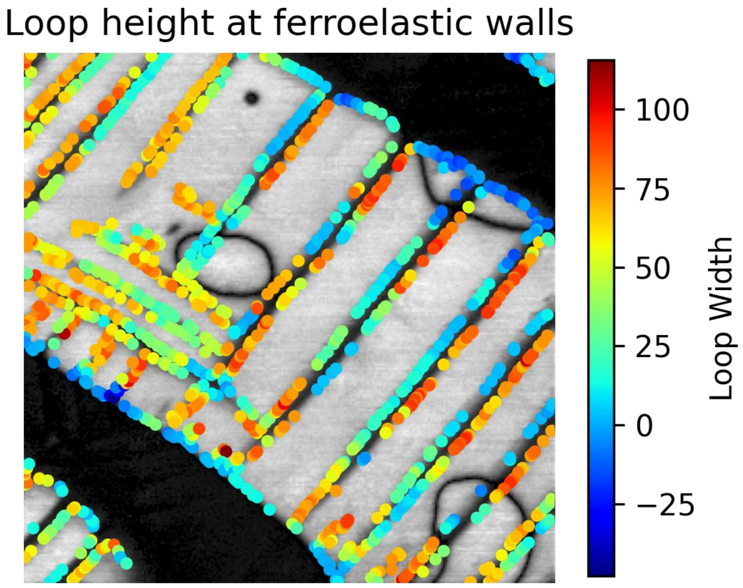
Mapping Activity of Domain Walls

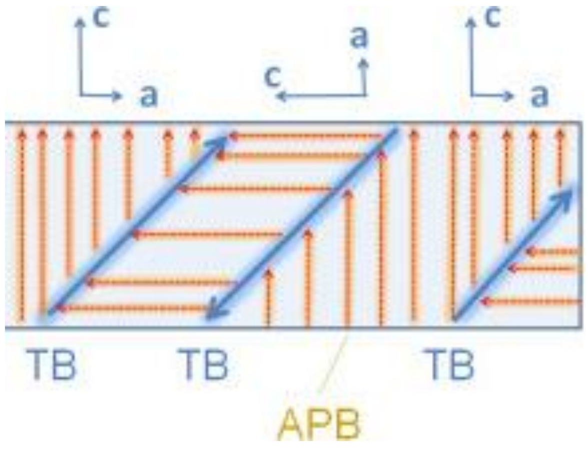








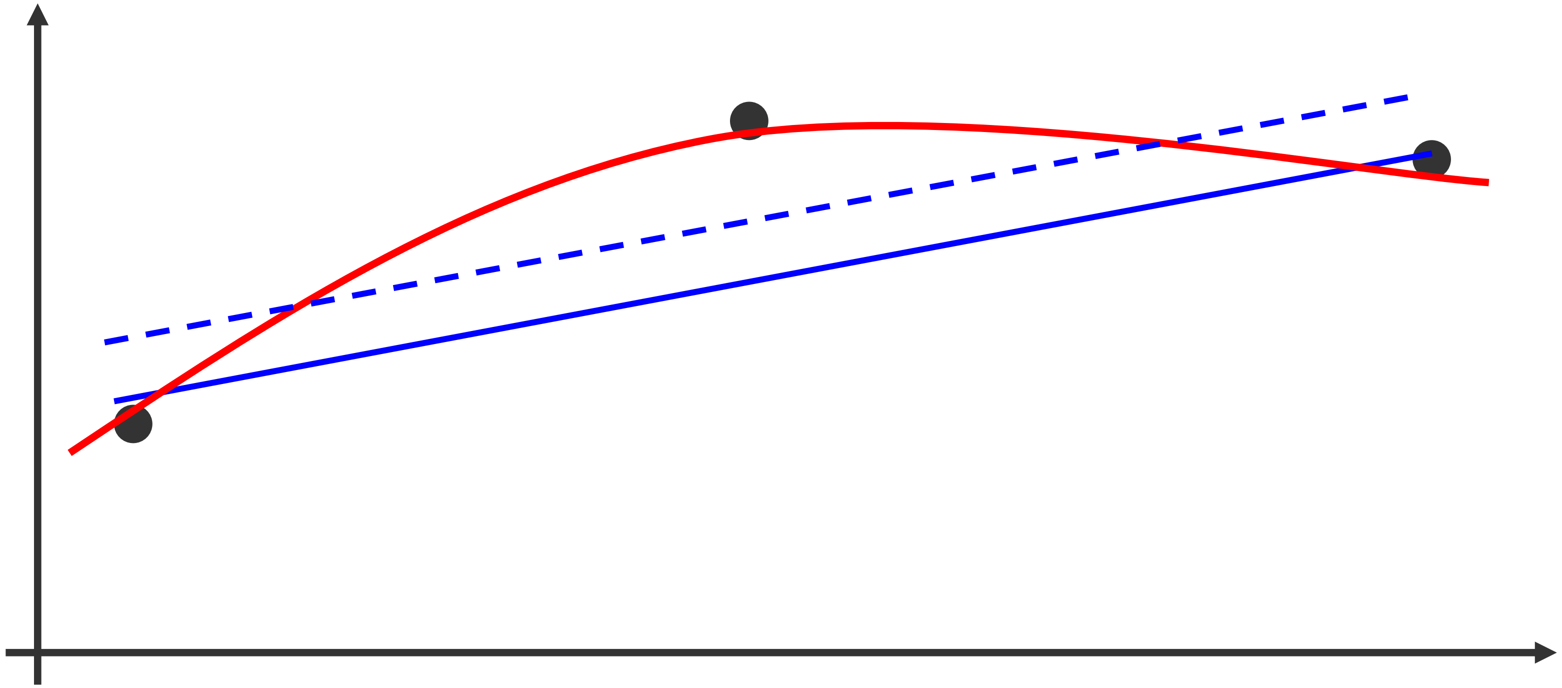




Liu, Y., Kelley, K.P., Funakubo, H., Kalinin, S.V., Ziatdinov, M., Arxiv, submitted

Gaussian Processes

What do we know if we do not know anything?



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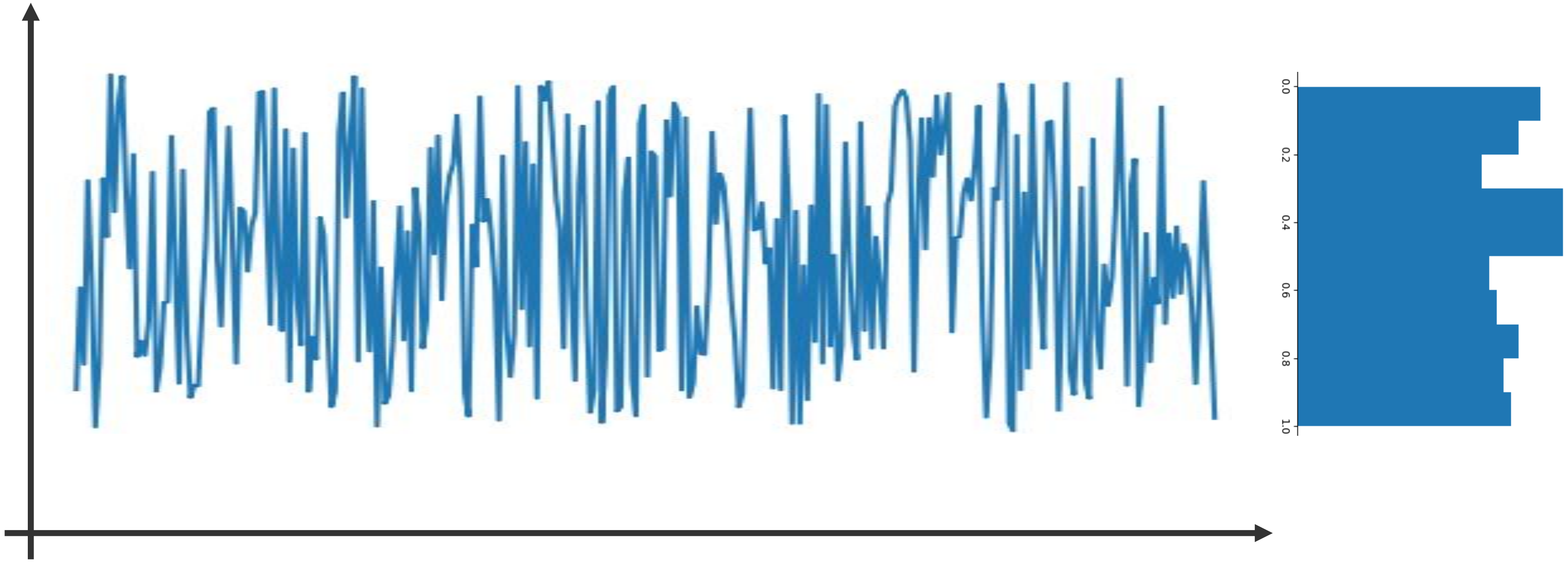
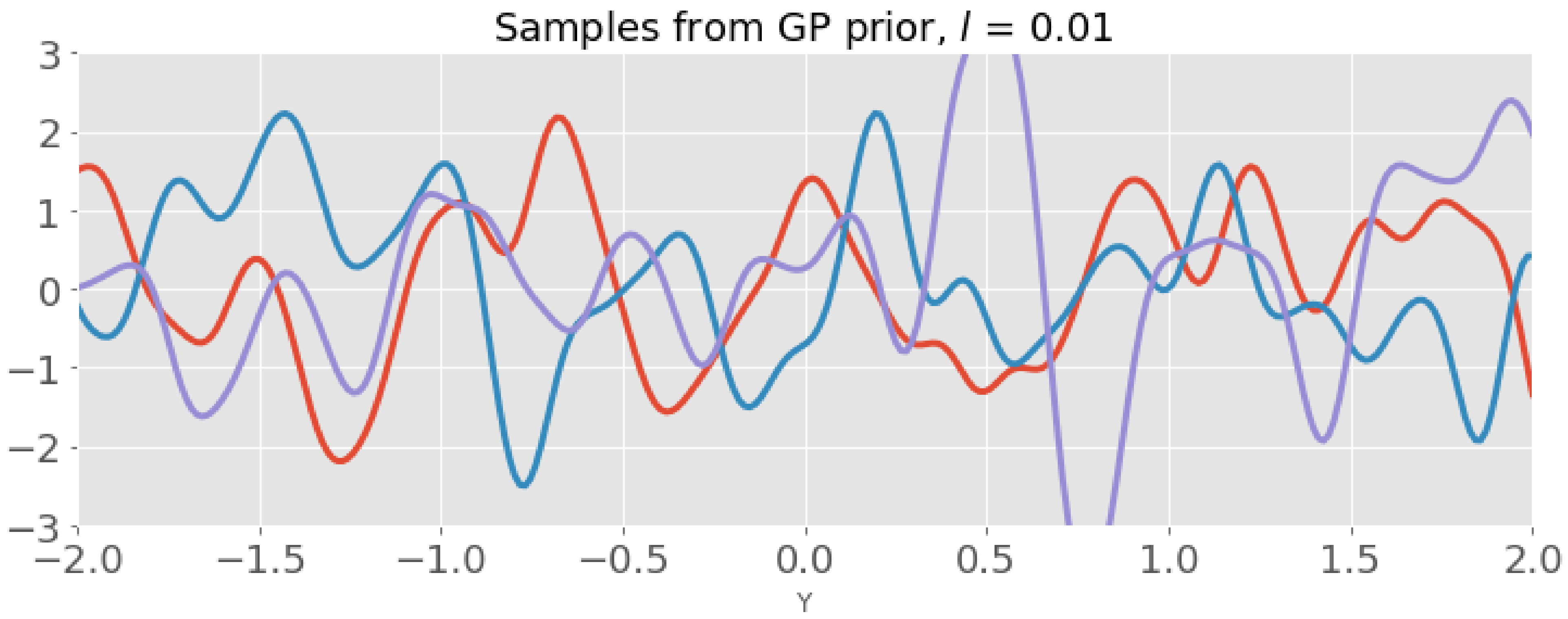


Gaussian Process Regression

- Covariance matrix determines what type of functions we will allow.

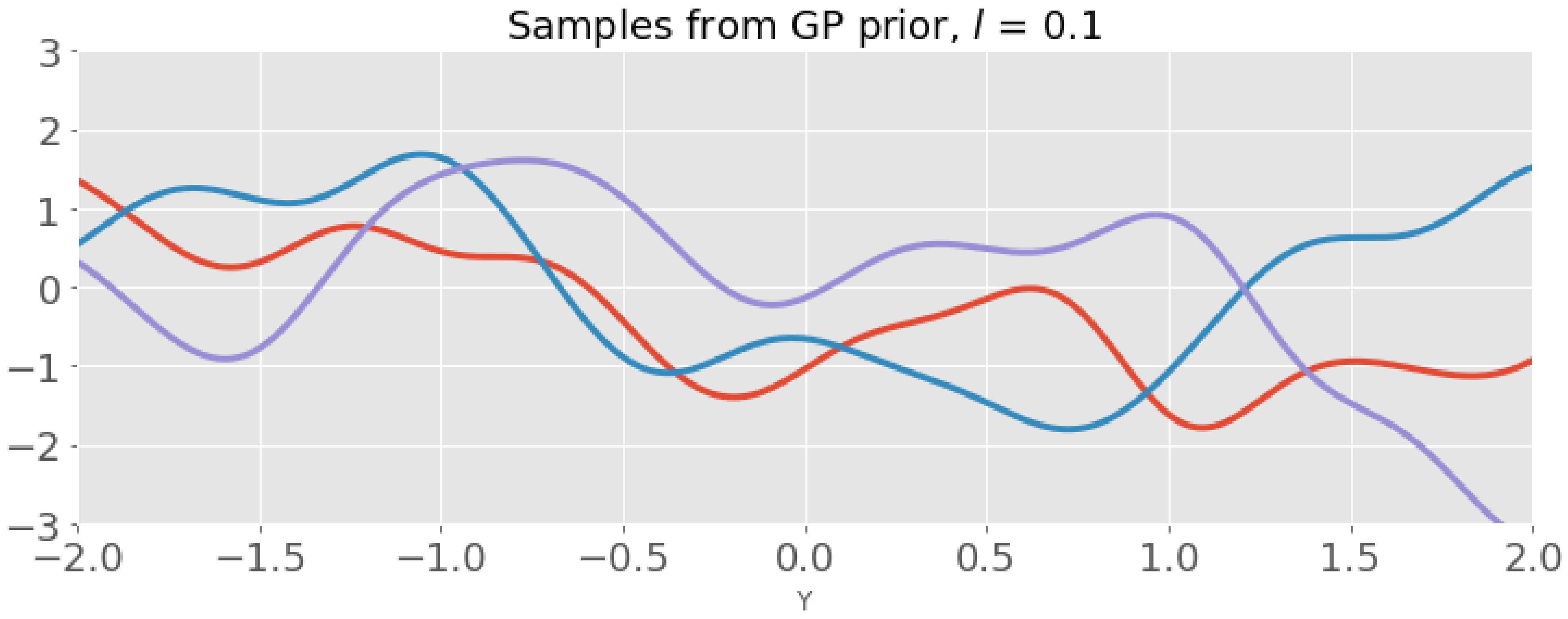
$$\mathrm{k}(\mathrm{x},\mathrm{x}') = \exp\left(-\frac{1}{2l}(x-x')^2\right)$$



Gaussian Process Regression

- Covariance matrix determines what type of functions we will allow.

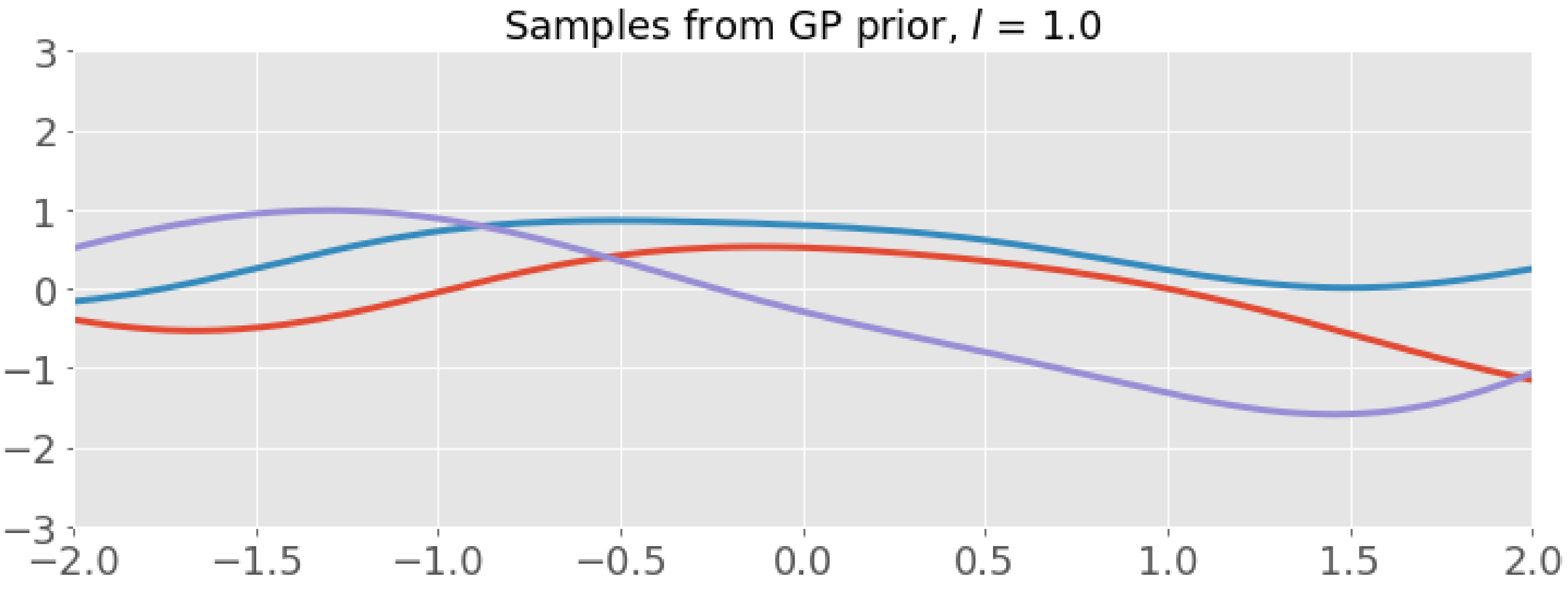
$$k(x, x') = \exp\left(-\frac{1}{2l}(x - x')^2\right)$$



Gaussian Process Regression

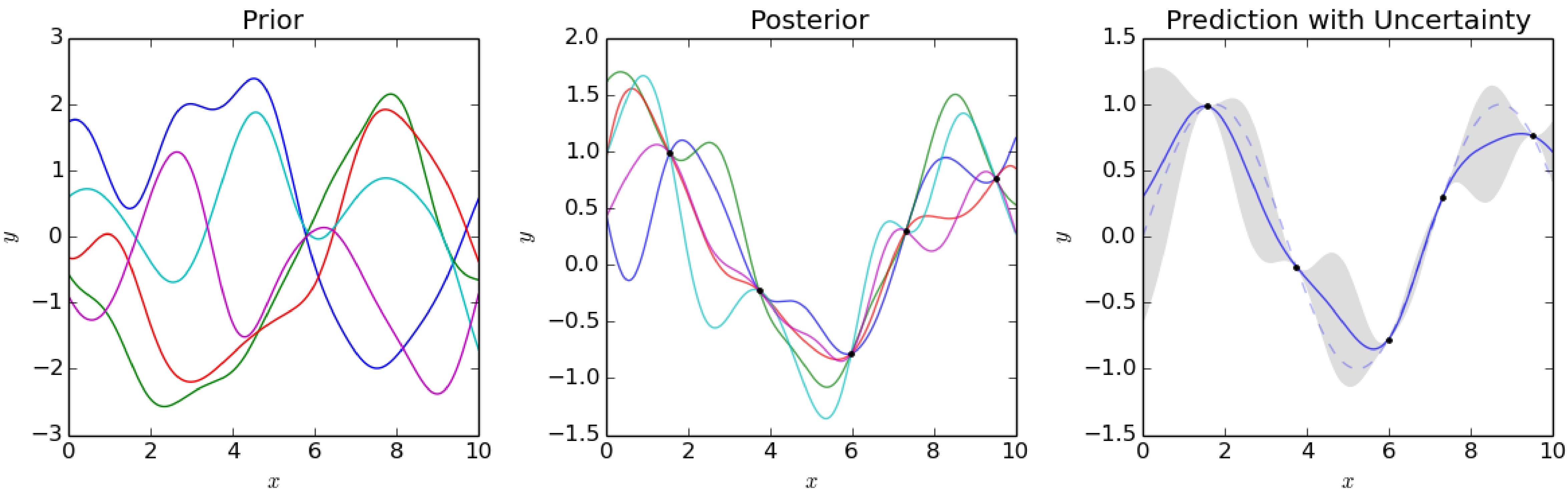
- Covariance matrix (kernel) determines what type of functions we will allow.

$$k(x, x') = \exp\left(-\frac{1}{2l}(x - x')^2\right)$$



L controls the length scale – sort of how far points should be to make them independent of each other.

Gaussian Process Regression

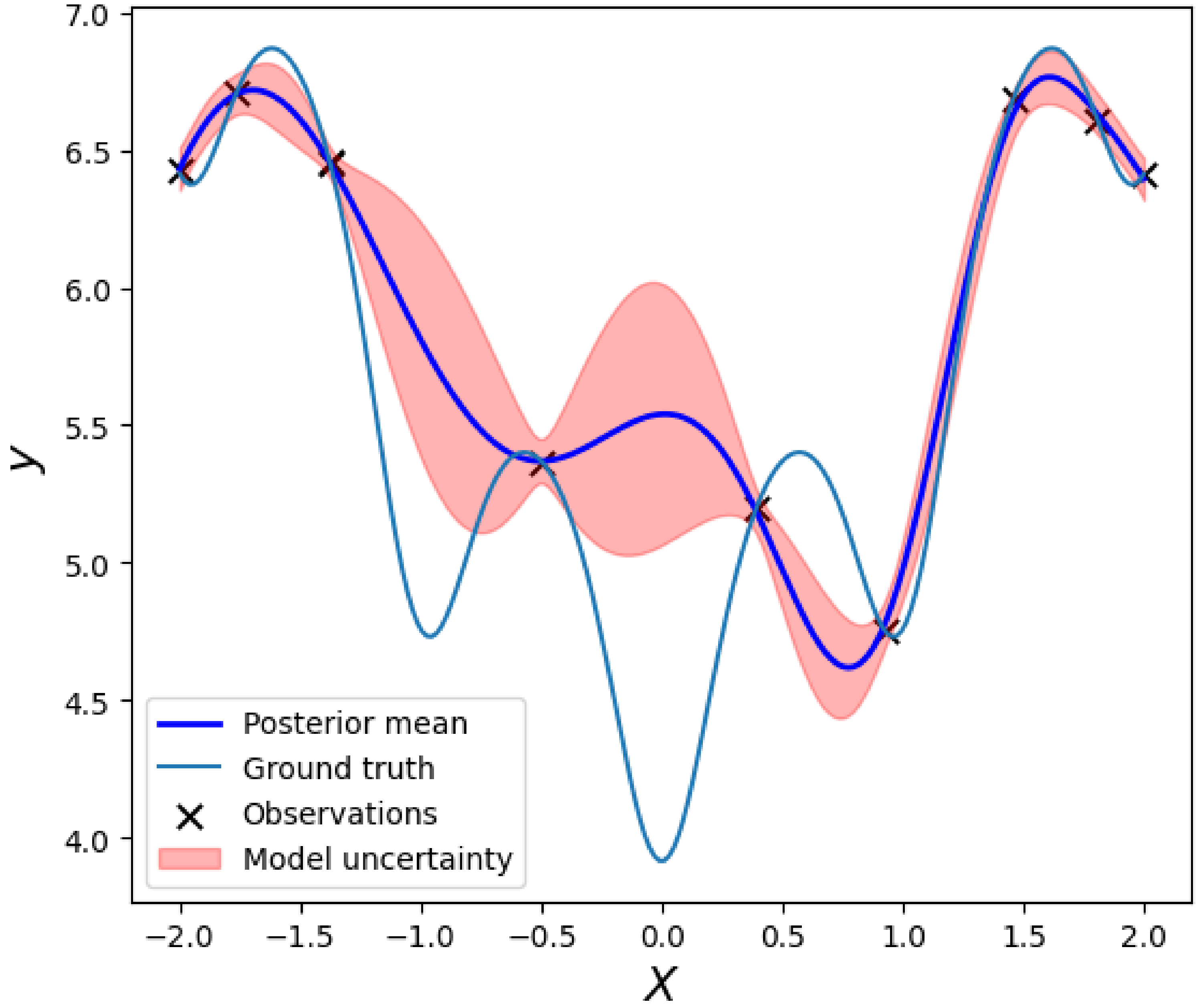


Prior:	What can the function be before the measurement
Data:	Measurement
Posterior:	What can the function be after measurement
Policy:	How do we balance exploration and exploitation (acquisition function)

GP Vocabulary

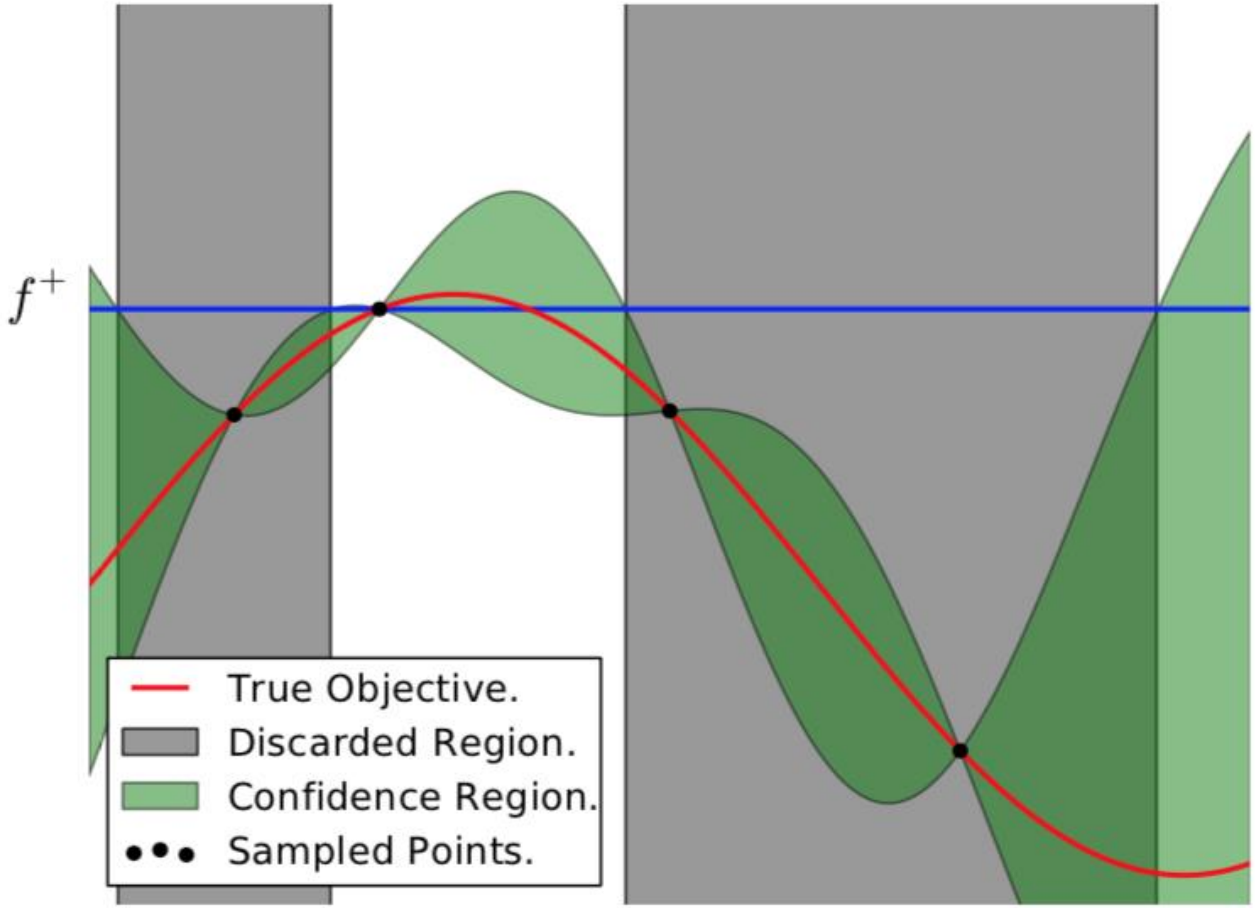
- Gaussian Process
- Kernel and kernel parameters
- Kernel Priors
- Noise Priors
- Posteriors

Colab



7.0
6.5
6.0
5.5
5.0
4.5
4.0
y
−2.0
−1.5
−1.0
−0.5
0.0
0.5
1.0
1.5
2.0
X
Posterior mean
Ground truth
Observations
Model uncertainty

Bayesian Optimization

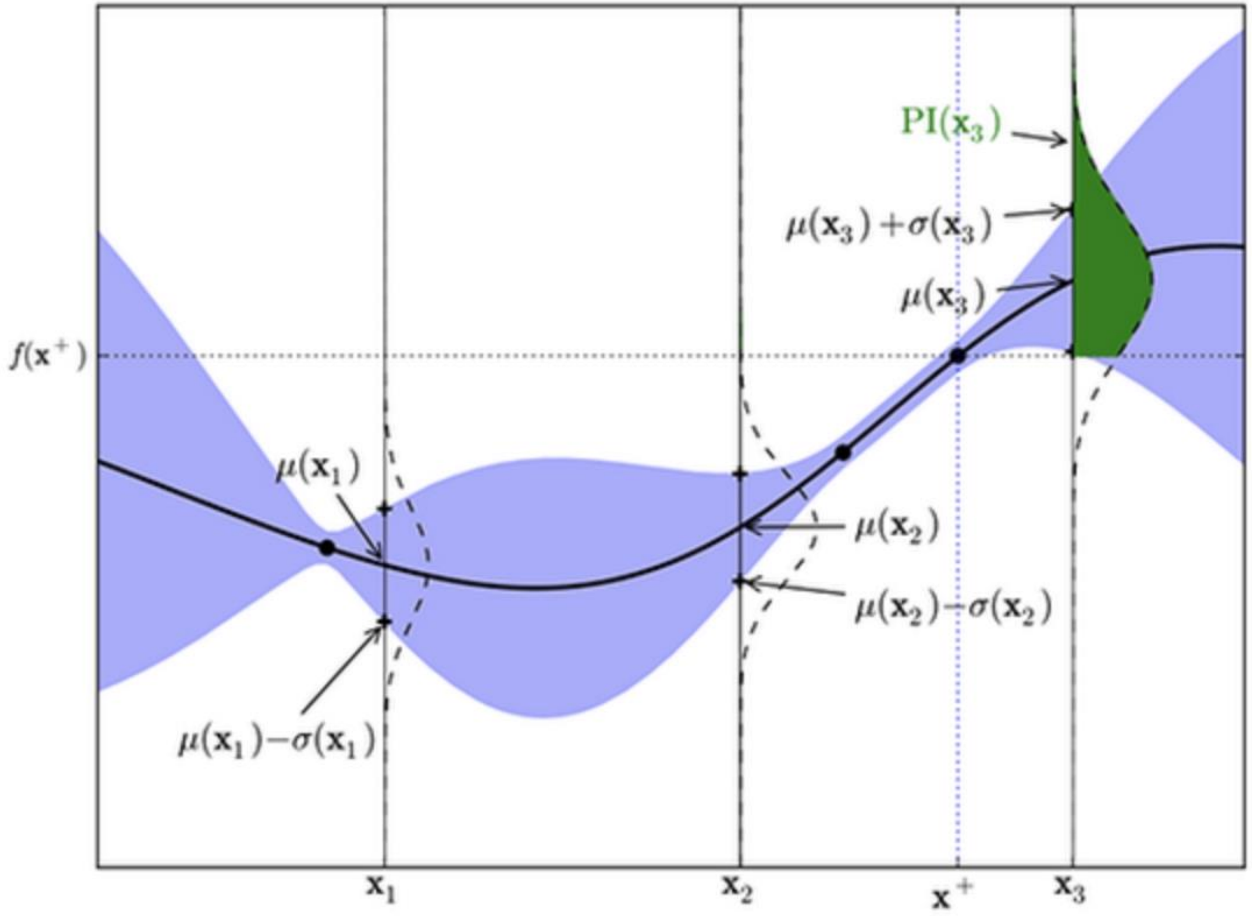


- We have some measurements in space X, and we want to maximize some property f(X).
- How can we decide what point to measure next to best maximize f?
- We need to balance the exploration of the space with exploitation of regions near we have already know

N. de Freitas et al., Taking the Human Out of the Loop: A Review of Bayesian Optimization , *Proceedings of the IEEE* **104**, 148 (2015)

Acquisition Functions

Probability of Improvement
Acquisition Function



1. **Upper confidence bound:** simplest possible - just take the upper confidence bound from the prediction

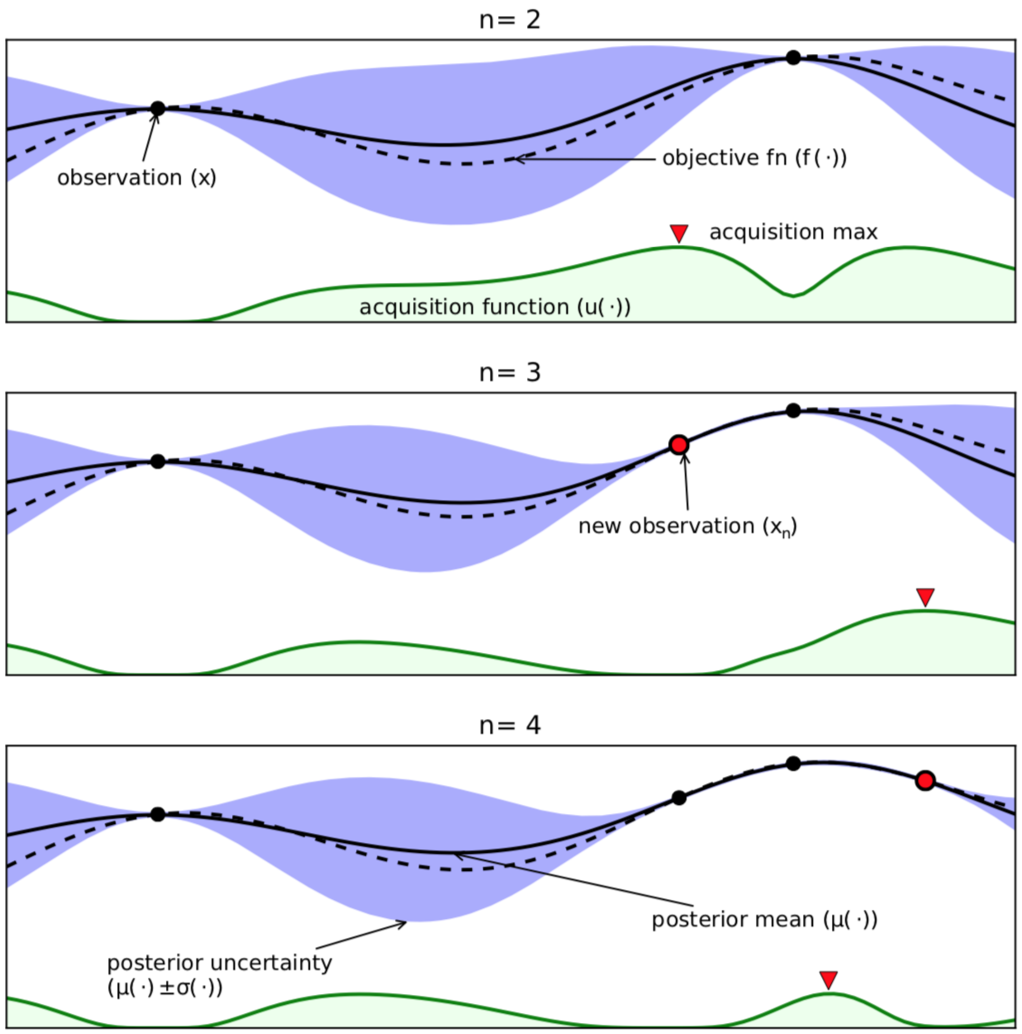
2. **Probability of Improvement:** Integral from current functional maximum to upper limit of distribution as test point

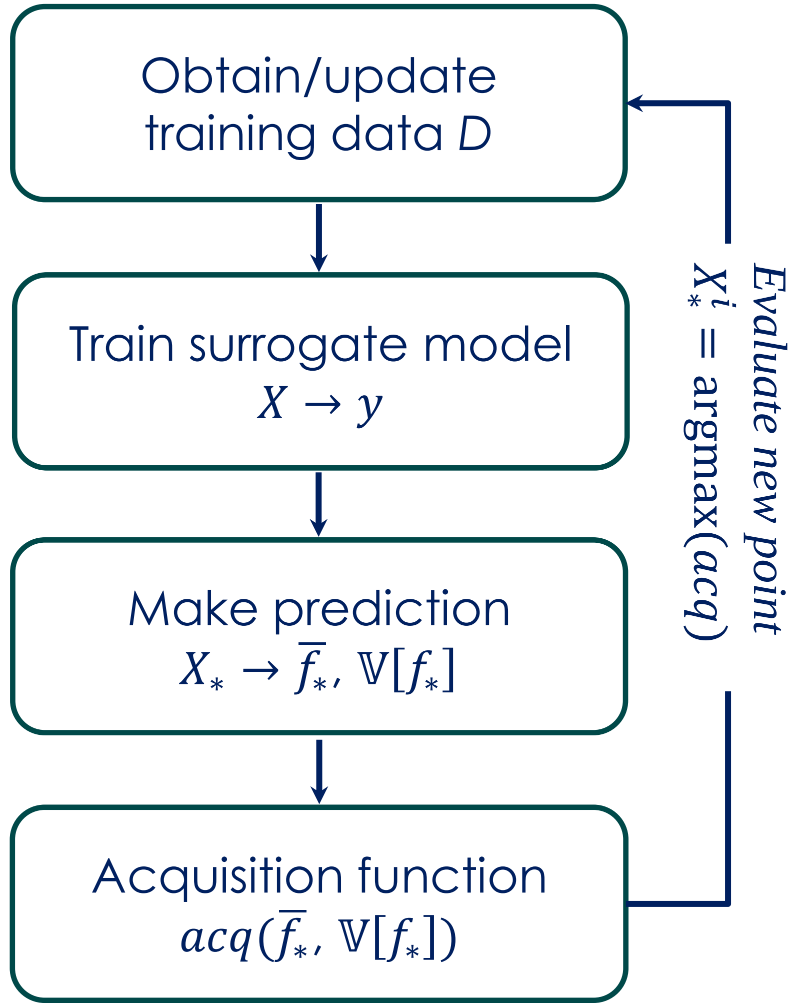
3. **Expected Improvement:** Instead of probability of improvement, we want to maximize the expected increase in the function value

4. **There are (always) more…**

The basics: Bayesian Optimization

X, y: (sparse) Training data
X_*: New (not yet evaluated) points

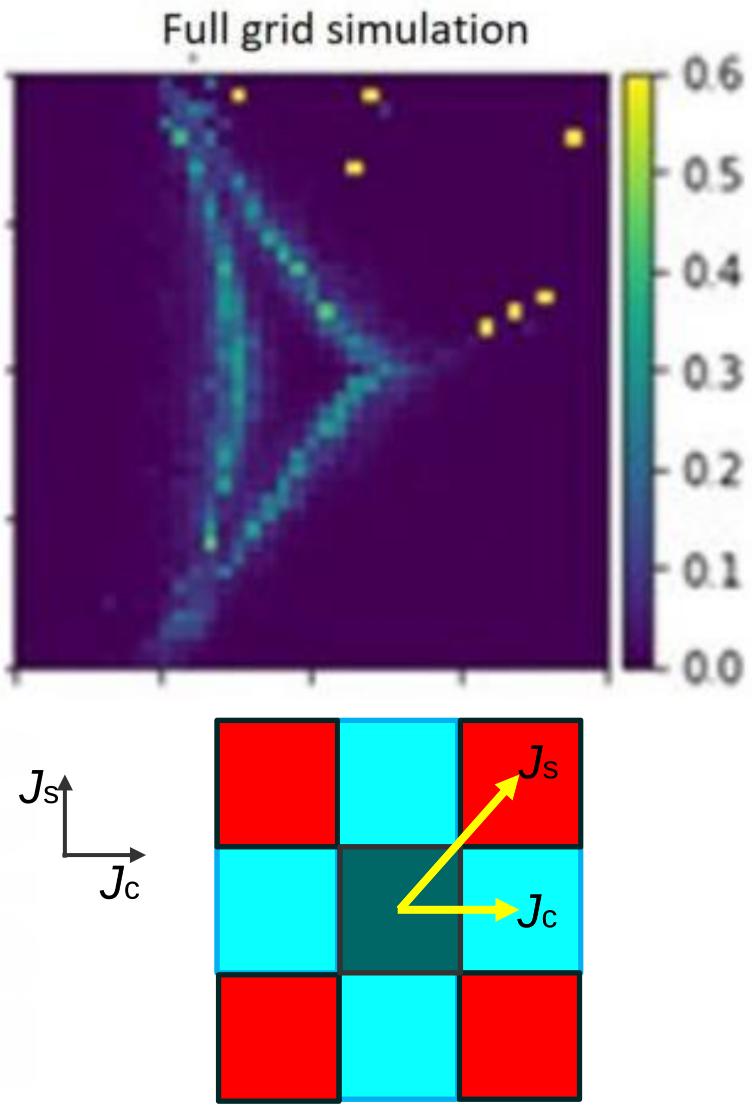


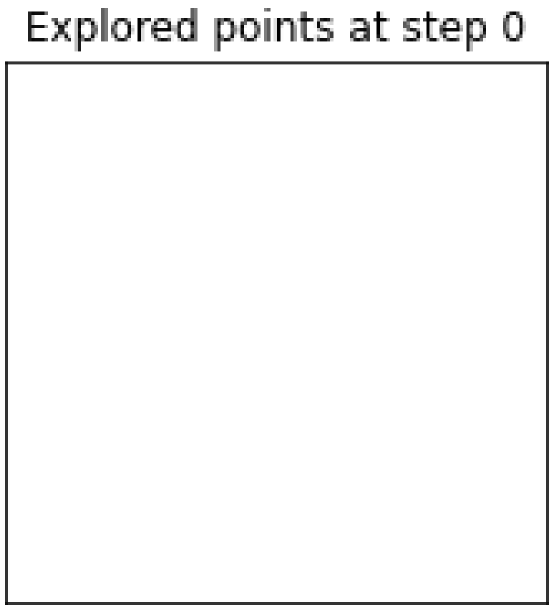


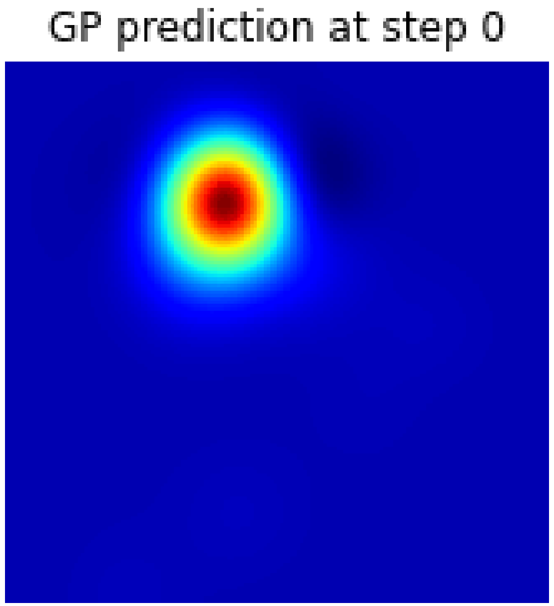
N. de Freitas et al., Taking the Human Out of the Loop: A Review of Bayesian Optimization , *Proceedings of the IEEE* **104**, 148 (2015)

Bayesian Optimization for physical discovery

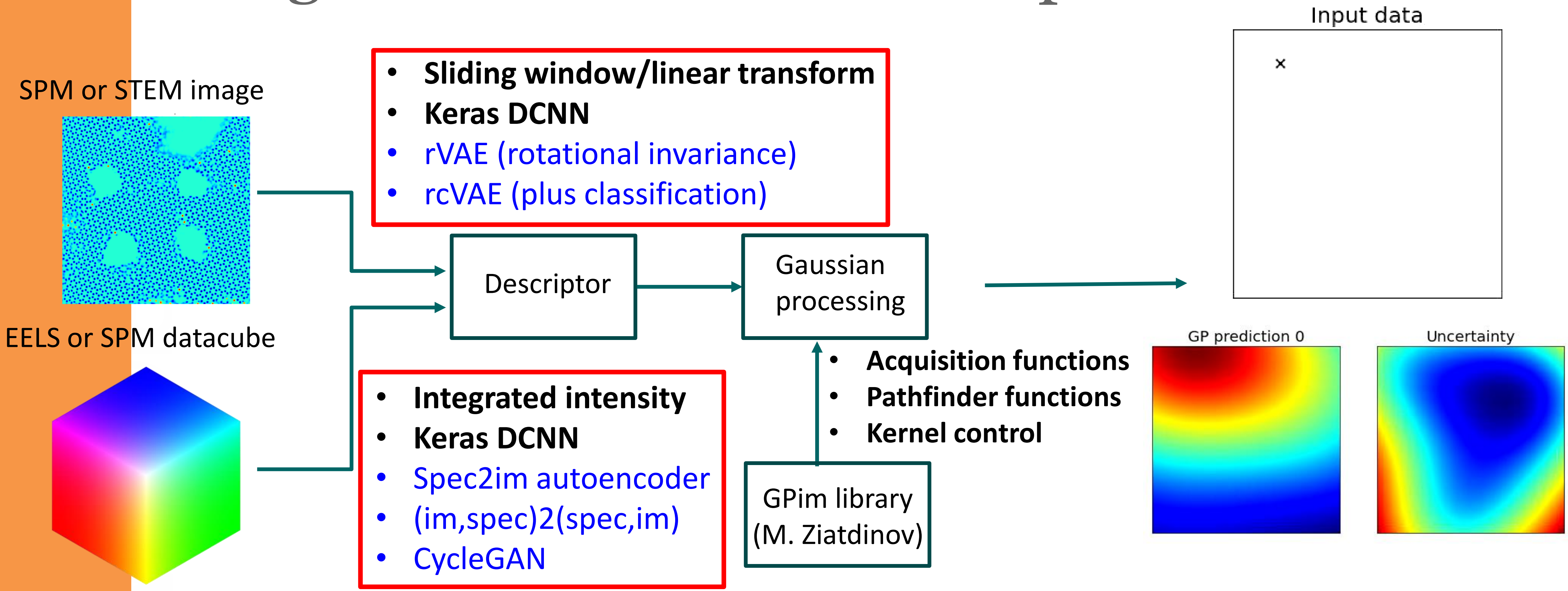
Discovering regions where heat capacity is maximized in NNN Ising model





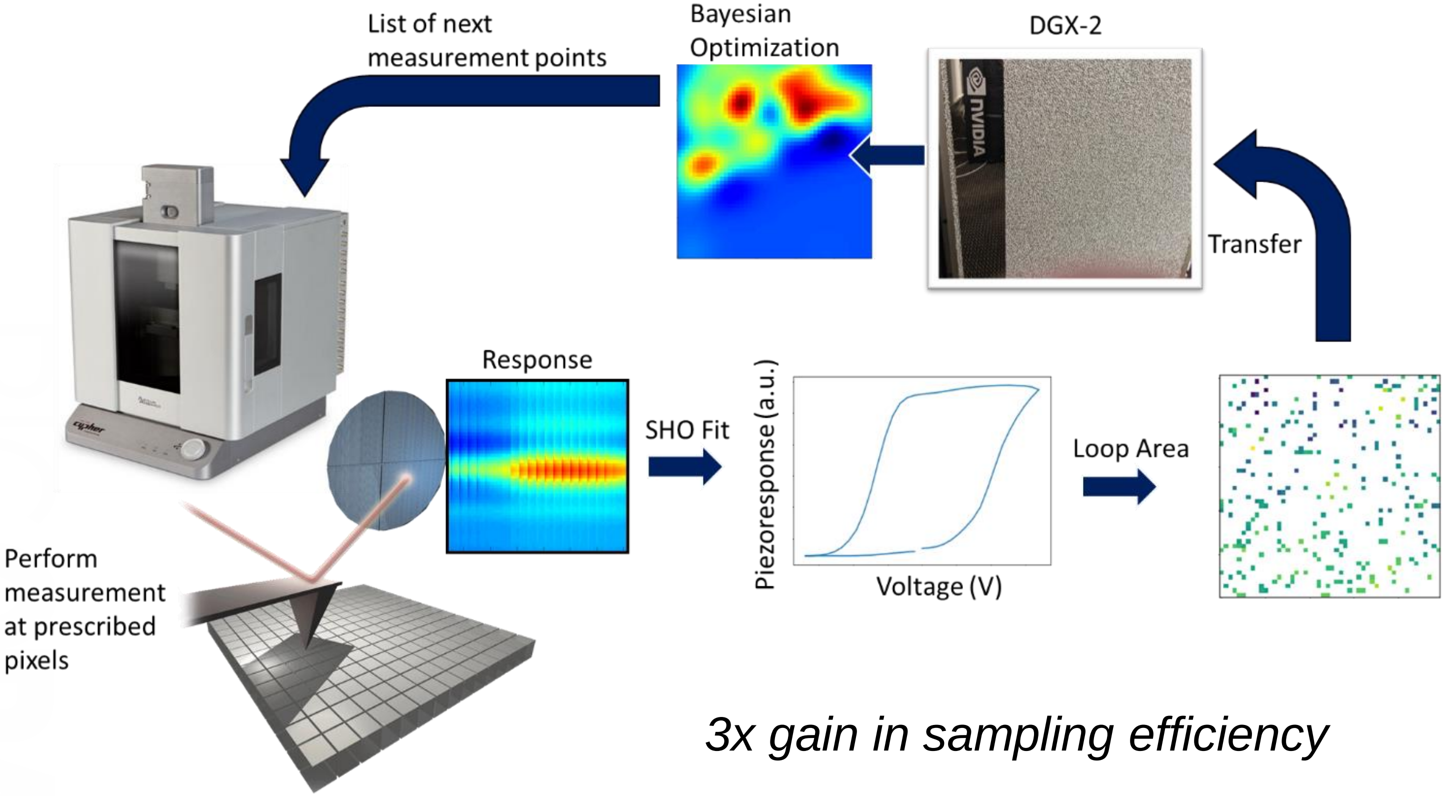


Going real time: automated experiment



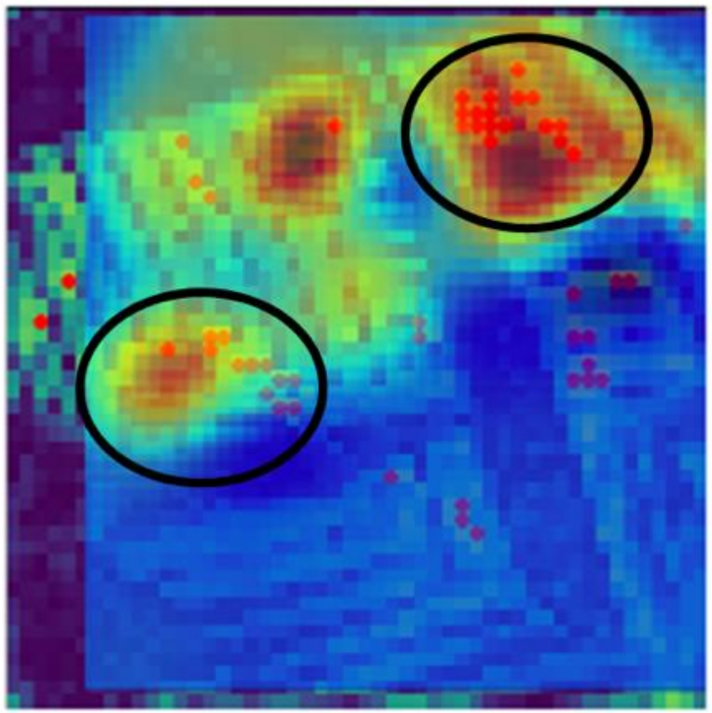
- AE based on structural analysis for STEM data
- AE based on spectral data in PFM
- AE based on DL for EELS data
- Feature of interest finding for mesoscopic images

BO for Self-Driving Microscopy



3x gain in sampling efficiency

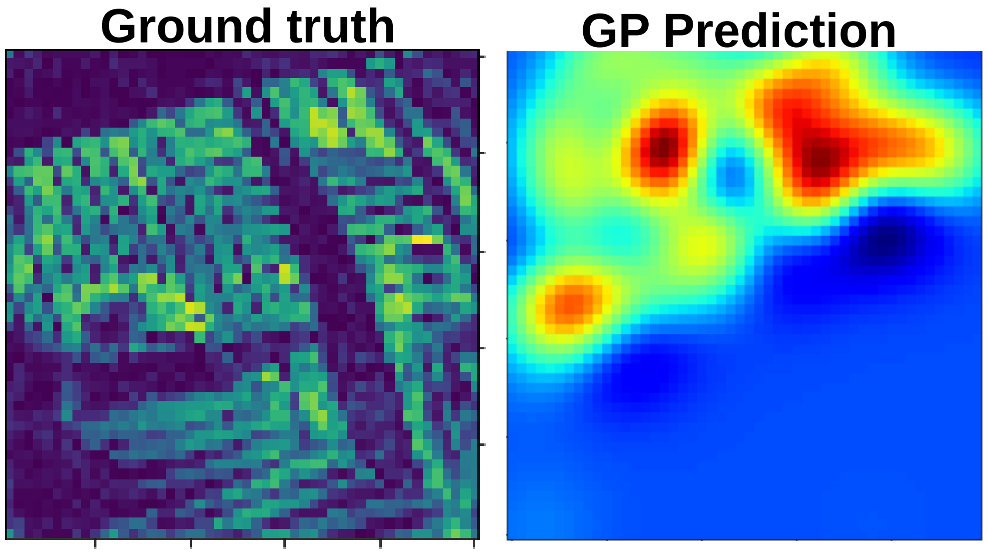
Comparison with "ground truth"



R. K. Vasudevan, K. Kelley, H. Funakubo, S. Jesse, S. V. Kalinin, M. Ziatdinov, ***ACS Nano*** (2021) *https://doi.org/10.1021/acsnano.0c10239*

Classical GP based BO

- Purely data-driven: limited advantage in high dimensional spaces
- Predicts scalar functions
- Typically used assuming equal cost of measurements
- And targeting fully automated process



Vasudevan et al, ACS Nano 2021

These assumptions rarely comport to real world scenarios

- We typically have ample (but partial) physical knowledge
- Multiple proxy signals
- Our observed data is very often high-dimensional (spectra, images)
- Cost and latencies of measurements is determined by physical equipment
- We can co-orchestrate measurements
- Humans are a part of the process (if process is slow)

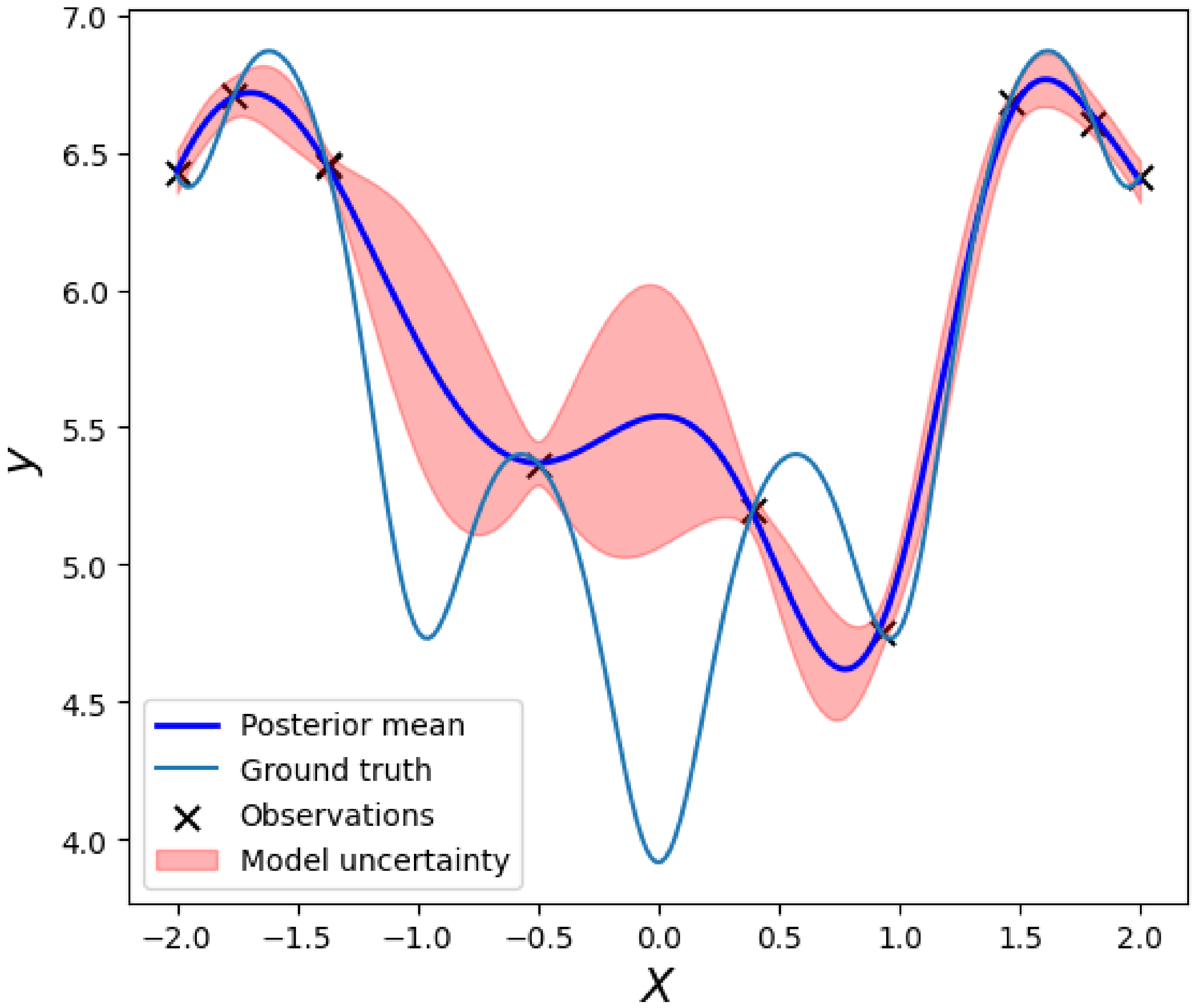
Bayesian Inference

What have we learned from lecture 2

- Gaussian Process
- Kernel and kernel parameters
- Kernel Priors
- Noise Priors
- Posteriors

- Bayesian Optimization
- Bayesian Optimization based on Gaussian Process

- Acquisition Functions
- Cost-award BO

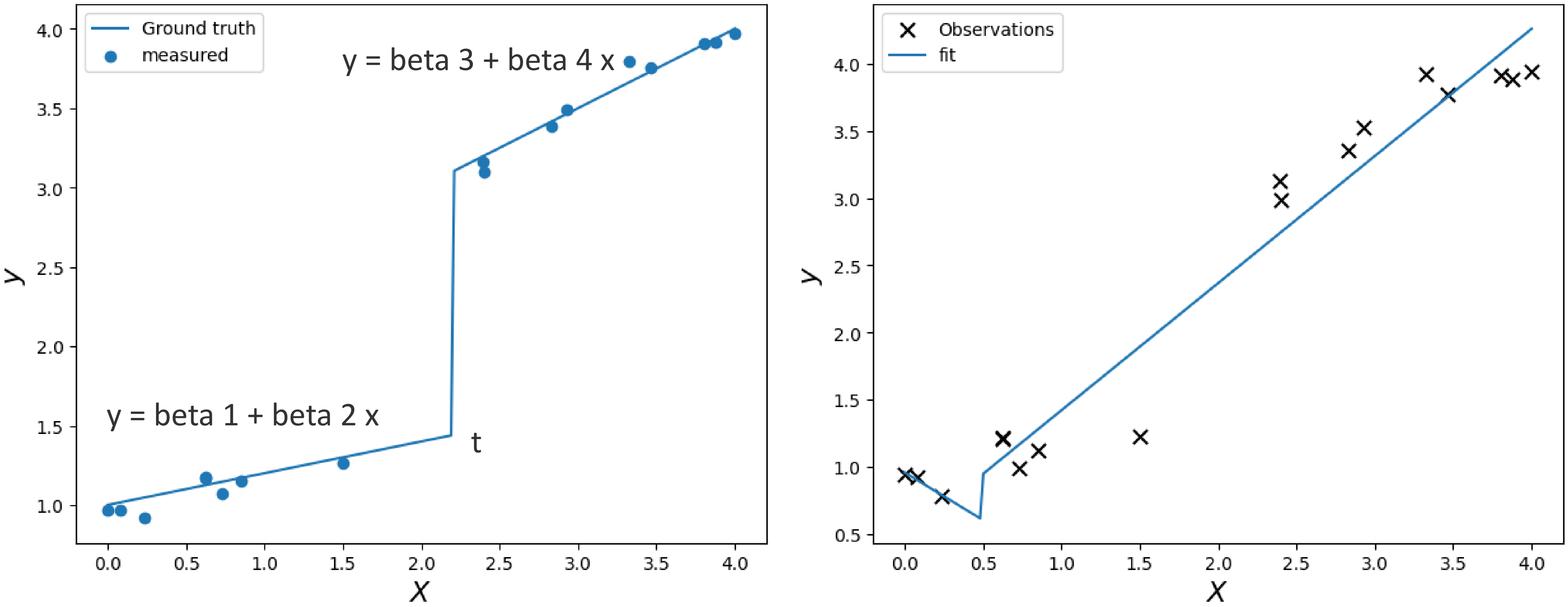


We can introduce noise in GP models

- Heteroscedastic GP: learn function and noise
- Measured Noise GP: measure function and noise
  – Needs model for noise prediction at unmeasured locations
- Noise in position in parameter space

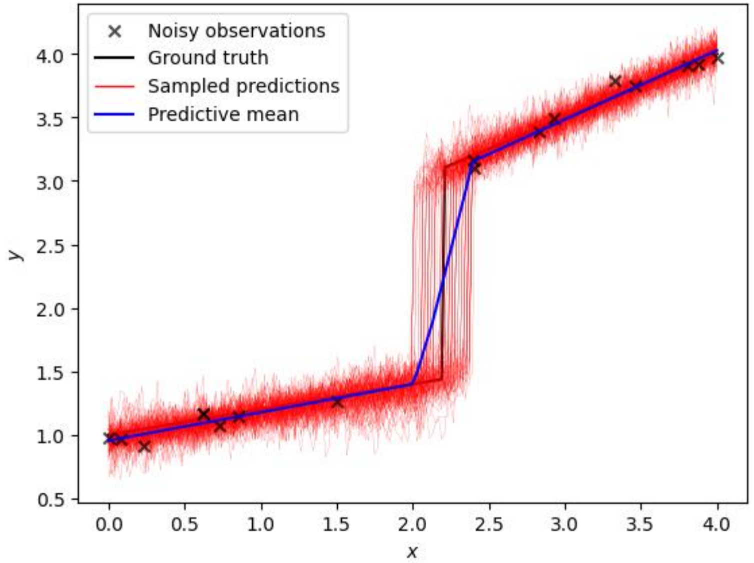
Need to balance priors based on extant knowledge;
do **not** treat these as optimization parameters

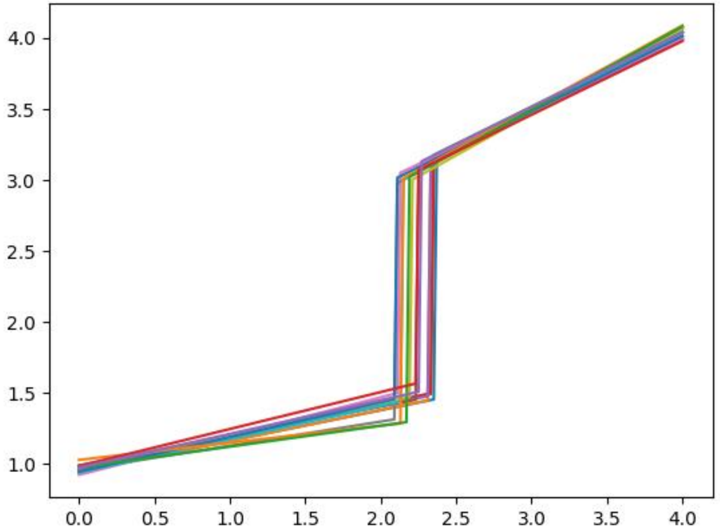
Least Square Fits can be a problem

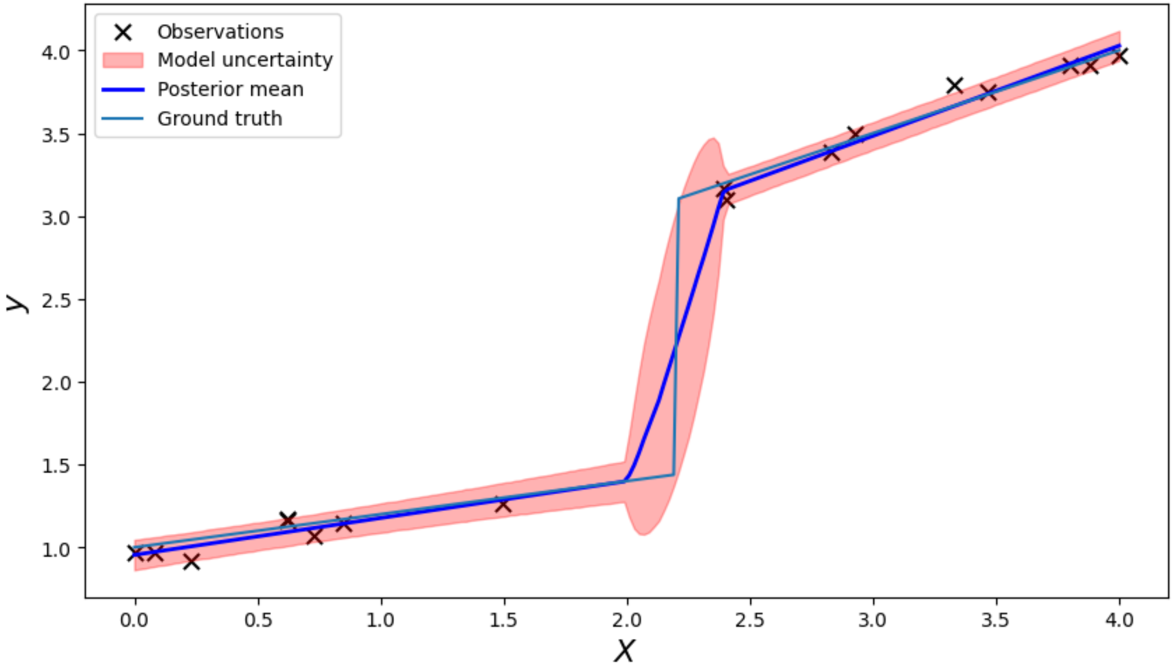


- No way to incorporate prior knowledge
- Convergence to metastable minima
- No feedback when gradient descent gets stuck

Bayesian Inference



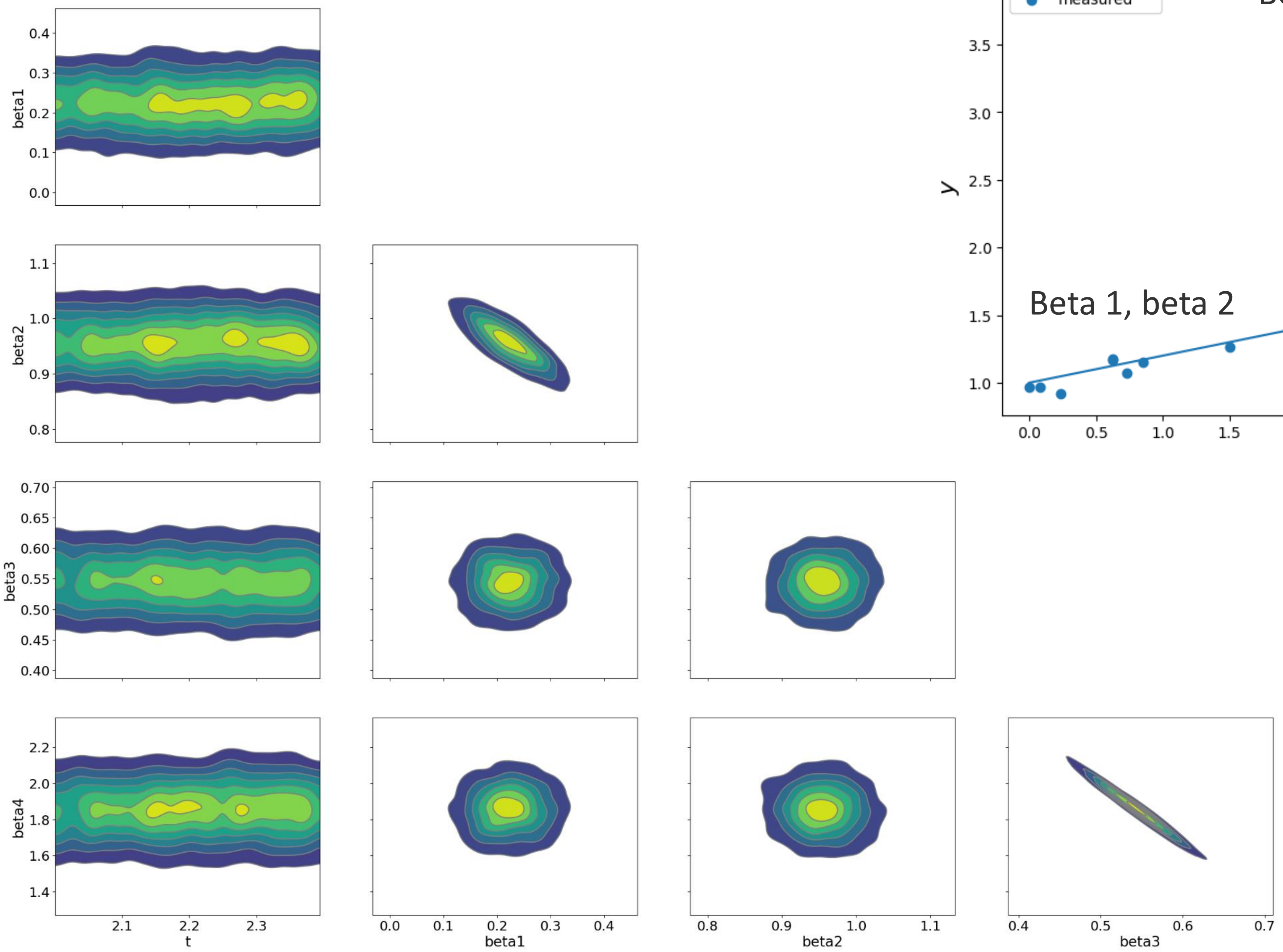


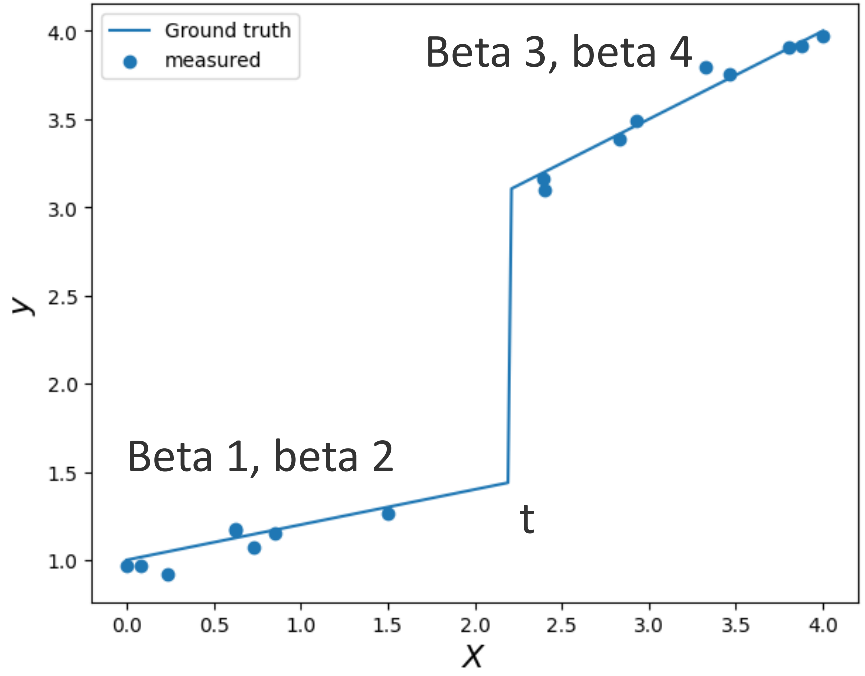


Bayesian inference:

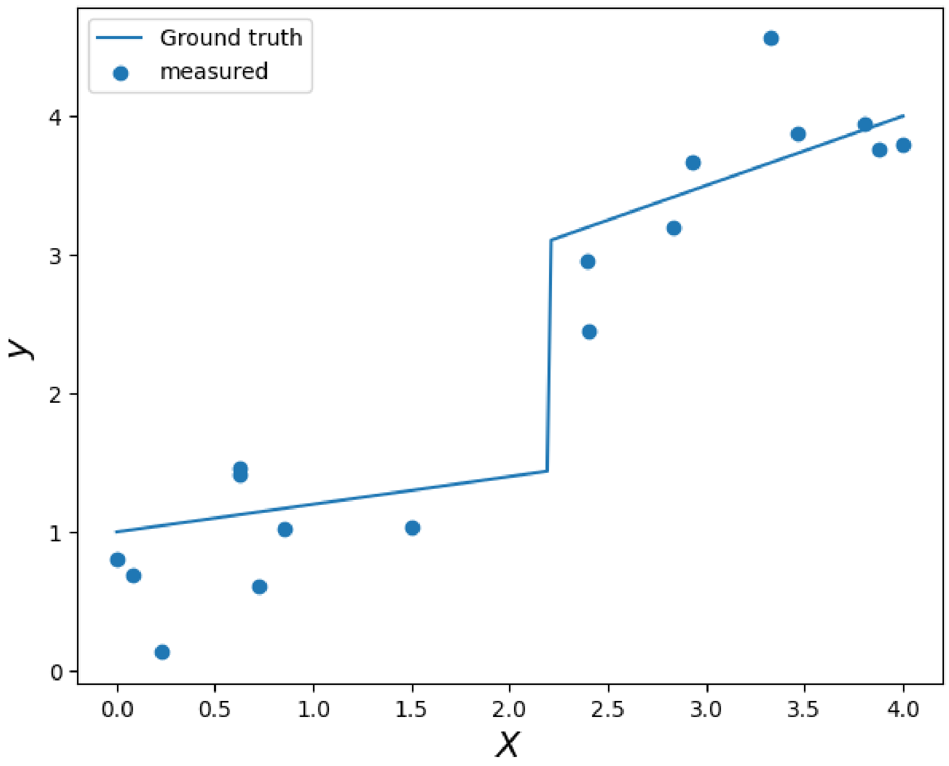
- Priors on parameter values
- Sampling values or mean functions
- Predictive mean and uncertainty
- Posteriors on parameter values

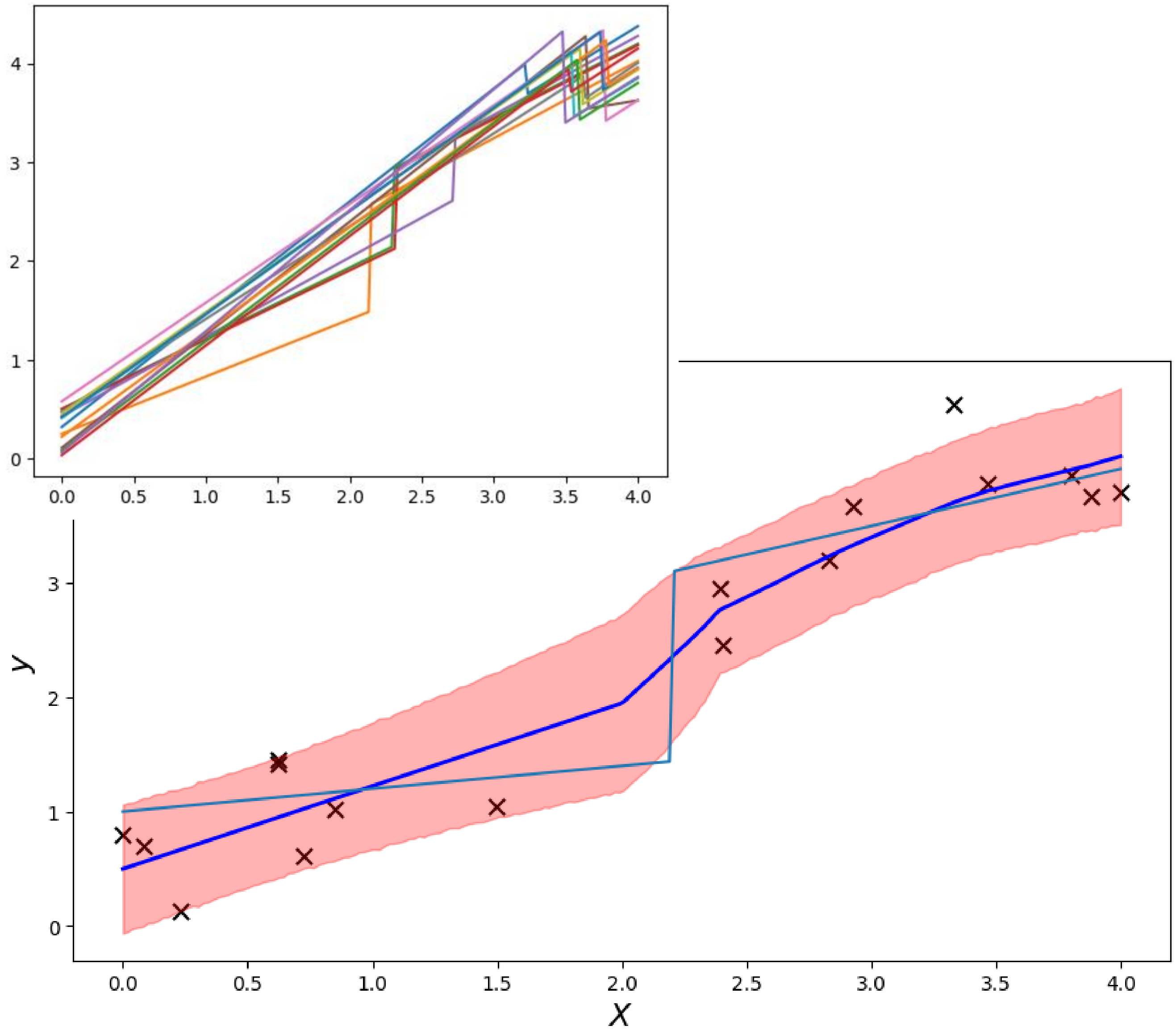
Bayesian Inference



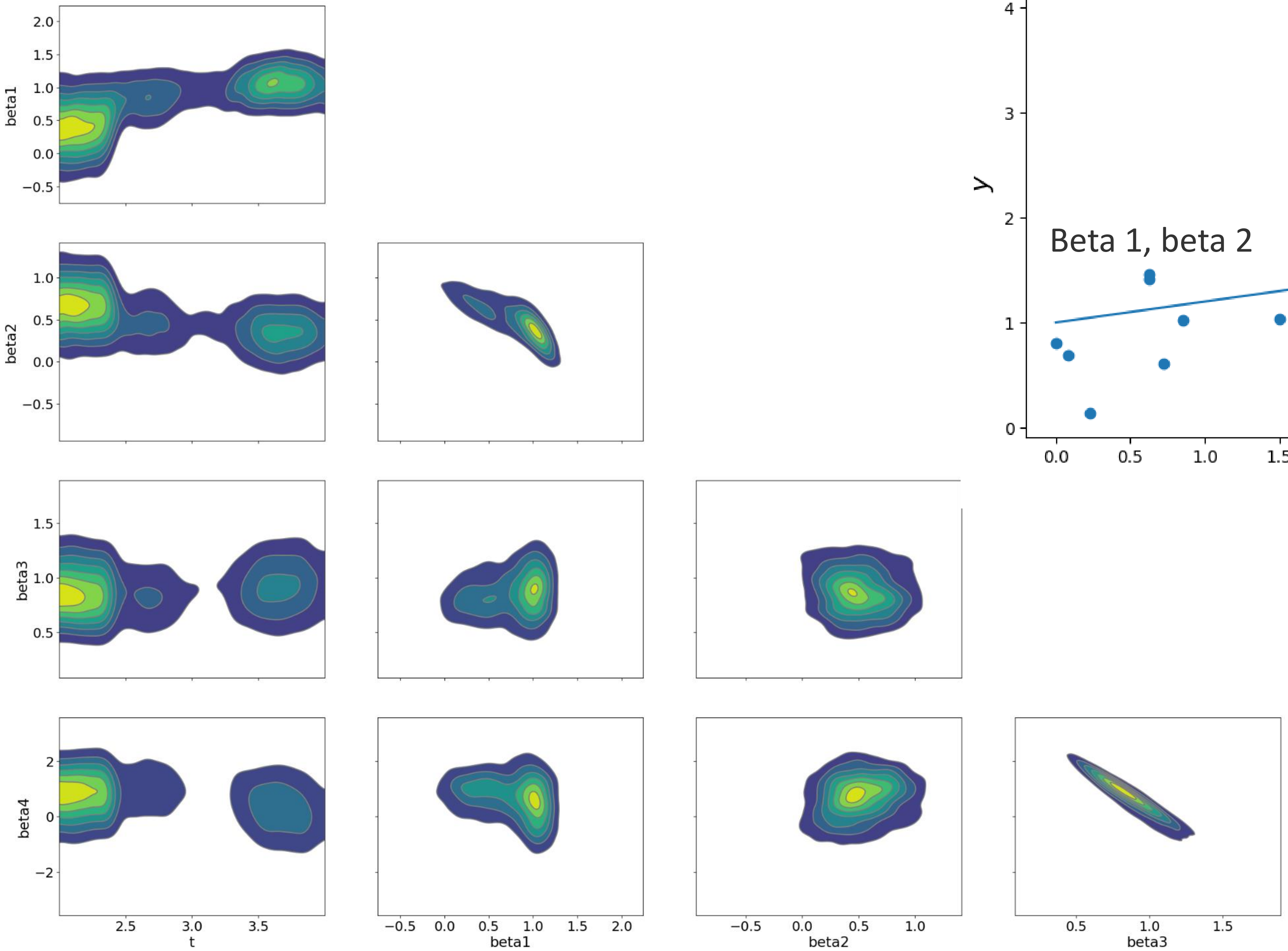


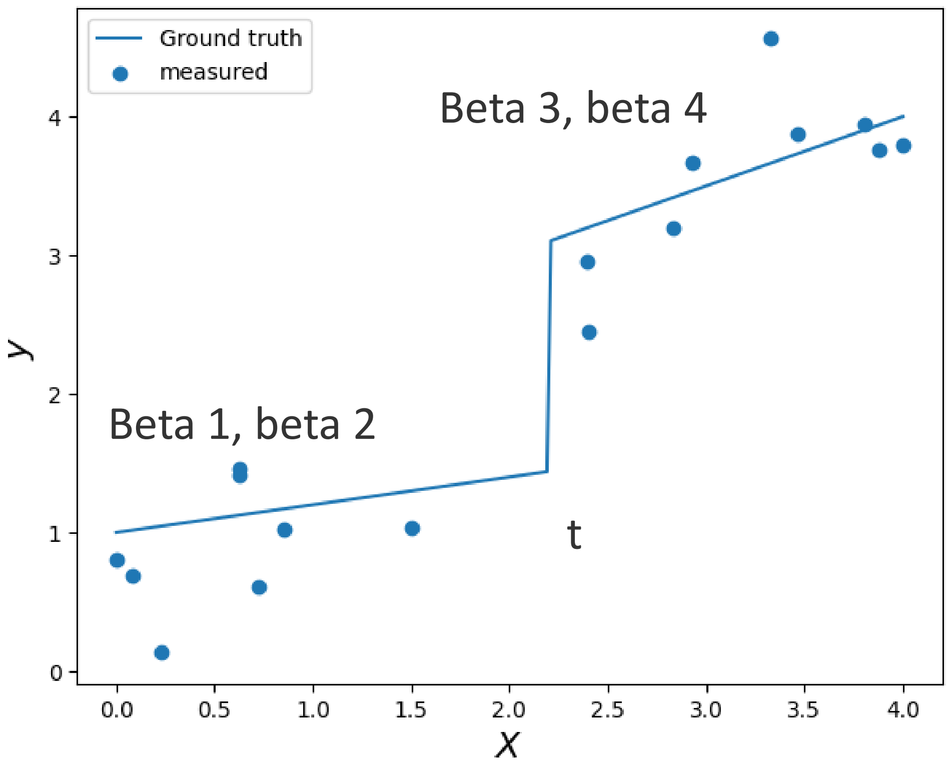
Bayesian Inference



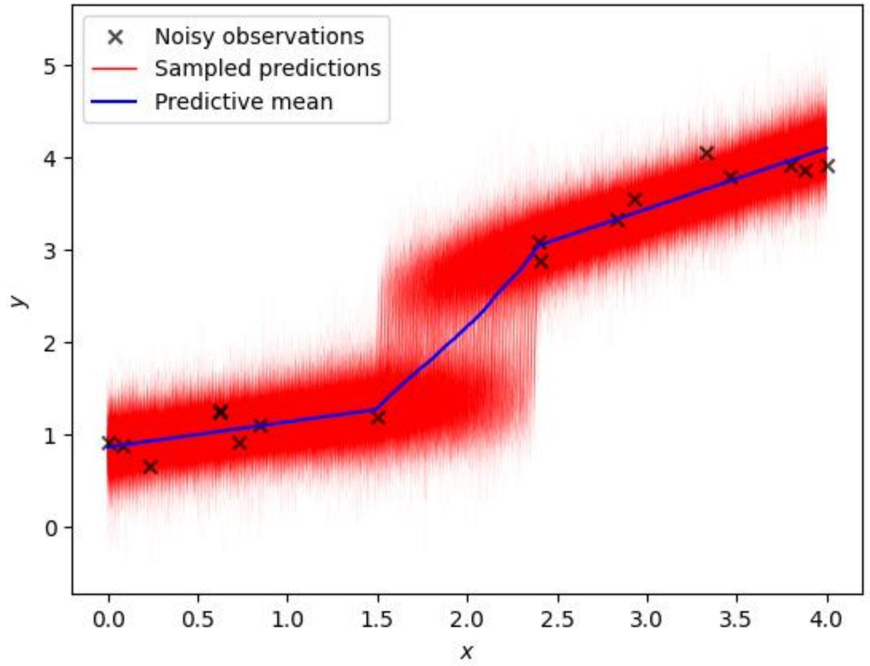


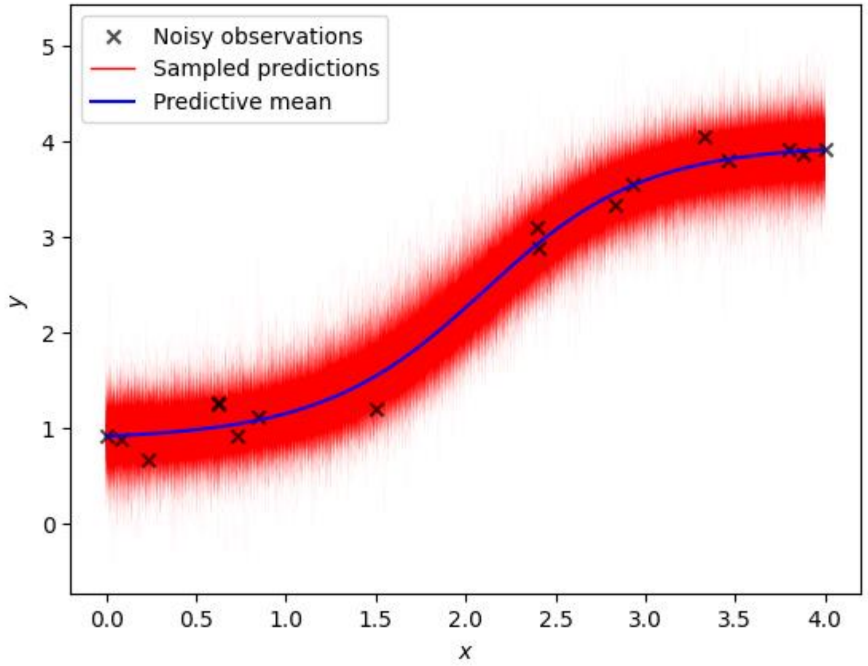
Bayesian Inference





Comparison of models: WAIC






```
1 az.compare(mcmc_all, ic="waic")
```

	rank	elpd_waic	p_waic	elpd_diff	weight	se	dse	warning	scale
piecewise	0	1.305878	3.207341	0.000000	5.027393e-01	2.217737	0.000000	True	log
sigmoidal	1	0.845156	4.073488	0.460722	4.972607e-01	2.829716	2.693397	True	log
linear2	2	-6.792783	2.312571	8.098661	0.000000e+00	2.842782	3.265961	True	log
power_law2	3	-7.820722	3.031942	9.126600	0.000000e+00	2.667691	3.113230	True	log
exponential1	4	-9.072673	2.661915	10.378551	0.000000e+00	2.251935	2.767500	True	log
linear1	5	-9.886713	1.539532	11.192591	9.836576e-14	3.207155	3.532880	True	log

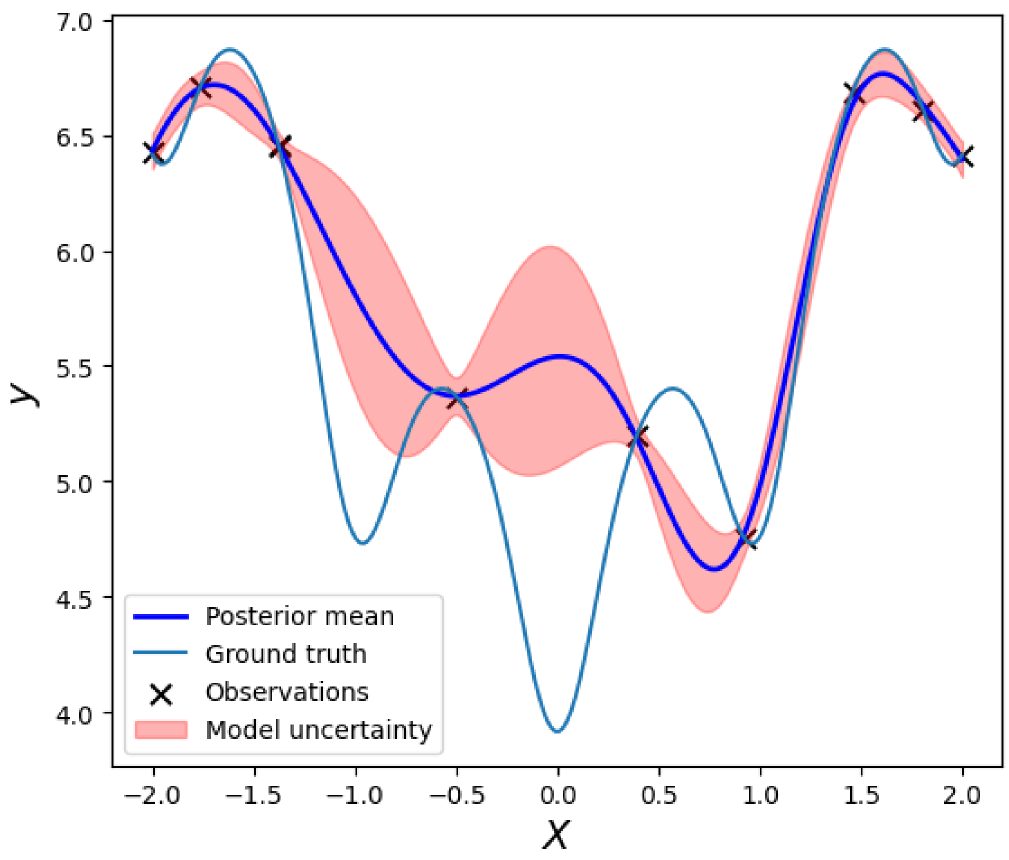
Automated Experiment:

… as a scientist…

Bayesian optimization:

1. Works only in low-dimensional spaces
2. The correlations are defined by the kernel function (very limiting)
3. We do not use any knowledge about physics of the system
4. We do not use cheap information available during the experiment (proxies)

- Classical Bayesian Optimization is useful for microscope tuning and imaging optimization, but almost useless for exploration in image plane
- Limited to low D: we need Deep Kernel Learning for Structure-Property relationship discovery
- No physics priors: we need structured Gaussian Processes to learn physics



GP Augmented with Structural model

Define a probabilistic model:

$$\mathbf{y} \sim MVNormal(\mathbf{m}, \mathbf{K})$$

$$K_{ij} = \sigma^2 \exp(0.5(x_i - x_j)^2 / l^2$$

$$\sigma \sim LogNormal(0, s_1)$$

$$l \sim LogNormal(0, s_2)$$

- We substitute a constant GP prior mean function **m** with a structured probabilistic model of the expected behavior.
- This probabilistic model reflects our prior knowledge about the system, but it does not have to be precise.
- The model parameters are inferred together with the kernel parameters via the Hamiltonian Monte Carlo.
- The fully Bayesian treatment of the model allows additional control over the optimization via the selection of priors for the model parameters.

Prediction on new data X_*:

$$\mathbf{f}_*^i \sim MVNormal\left(\mu_{\boldsymbol{\theta}^i}^{\text{post}}, \Sigma_{\boldsymbol{\theta}^i}^{\text{post}}\right)$$

replaced with

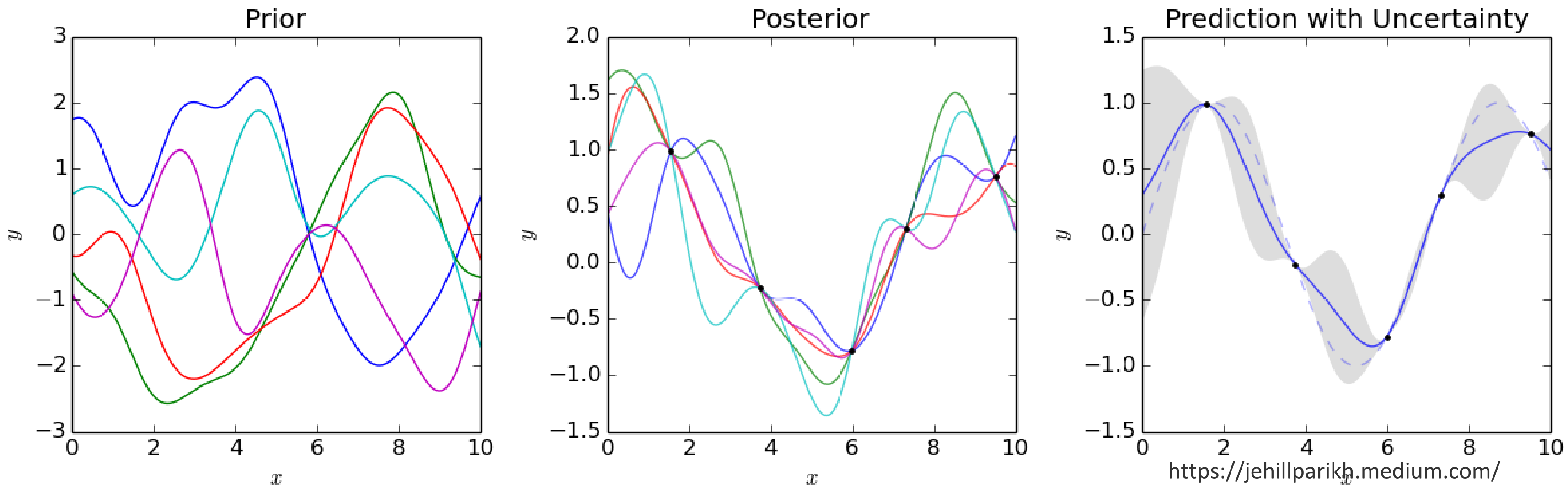
$$\mu_{\boldsymbol{\theta}^i}^{\text{post}} = \mathbf{m}(X_*) + \mathbf{K}(X_*, X|\boldsymbol{\theta}^i)\mathbf{K}(X, X|\boldsymbol{\theta}^i)^{-1}(\mathbf{y} - \mathbf{m}(X)) \rightarrow \mu_{\boldsymbol{\Omega}^i}^{\text{post}} = \mathbf{m}(X_*|\phi^i) + \mathbf{K}(X_*, X|\boldsymbol{\theta}^i)\mathbf{K}(X, X|\boldsymbol{\theta}^i)^{-1}(\mathbf{y} - \mathbf{m}(X|\phi^i))$$

$$\Sigma_{\boldsymbol{\theta}^i}^{\text{post}} = \mathbf{K}(X_*, X_*|\boldsymbol{\theta}^i) - \mathbf{K}(X_*, X|\boldsymbol{\theta}^i)\mathbf{K}(X, X|\boldsymbol{\theta}^i)^{-1}\mathbf{K}(X, X_*|\boldsymbol{\theta}^i)$$

$\boldsymbol{\Omega}^i = \{\phi^i, \boldsymbol{\theta}^i\}$ is a single HMC posterior sample with the kernel and prob model parameters

GP Augmented with Structural Model

Standard Gaussian process aims to discover function based on learned correlations (kernel)



Probabilistic model

$$m = y_0 - \sum_{n=1}^{N} L_n \quad (N{=}2)$$

$$y_0 \sim Uniform(-10, 10)$$

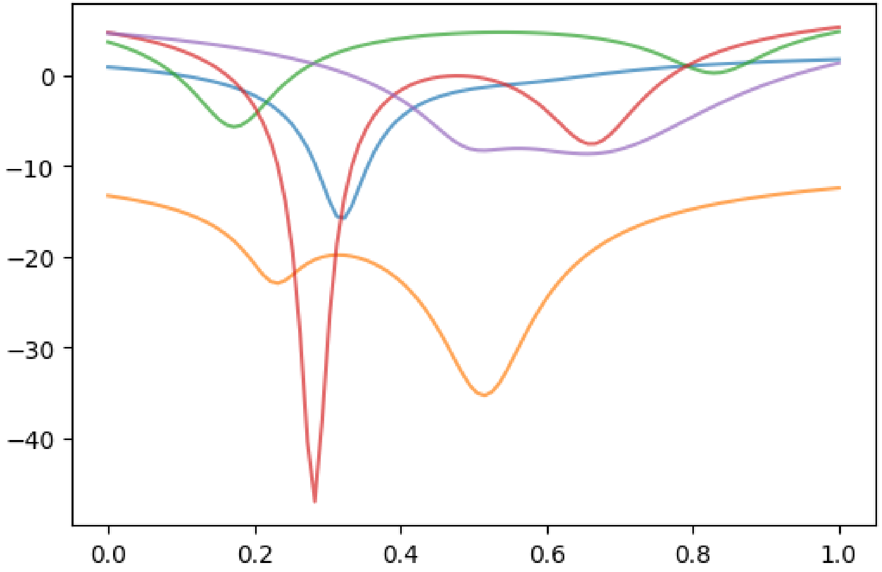
$$L_n \sim \frac{A_n}{\sqrt{(x - x_n^0)^2 + w_n^2}}$$

$$A_n \sim LogNormal(0, 1)$$

$$w_n \sim HalfNormal(.1)$$

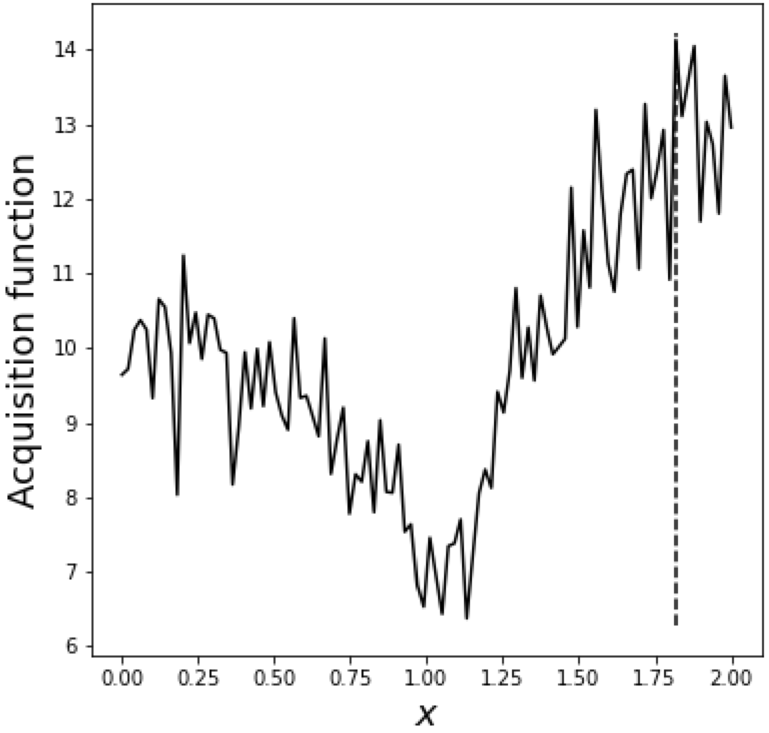
$$x_n^0 \sim Uniform(0, 1)$$

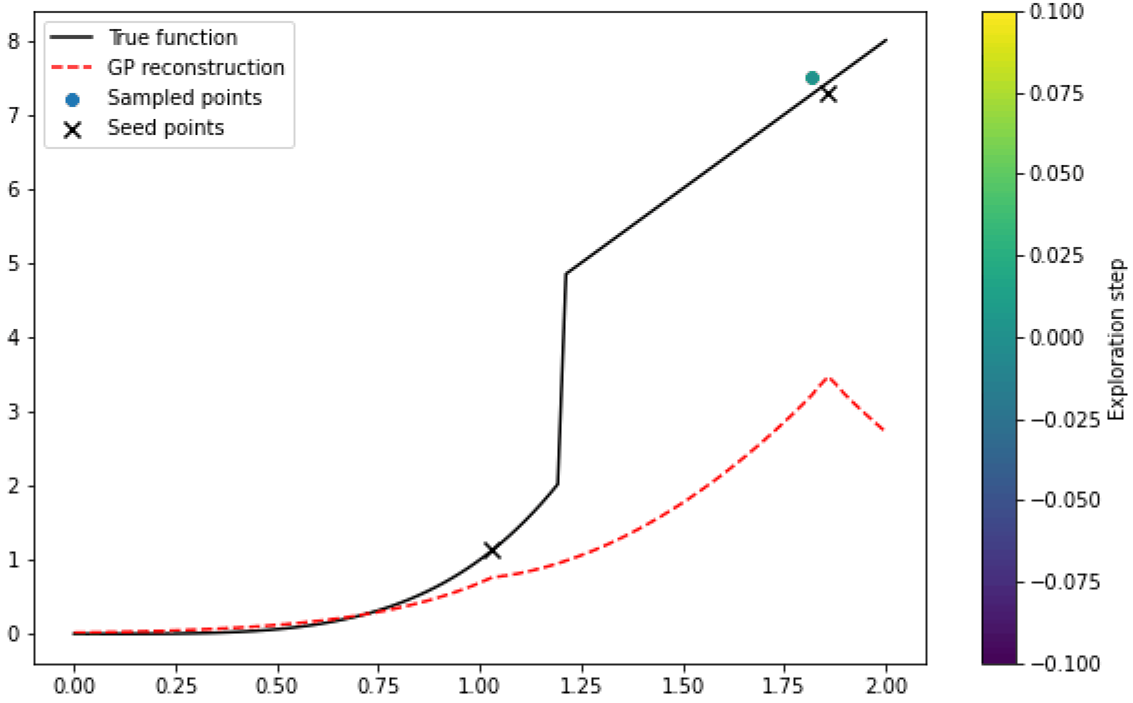
Prior predictive distribution



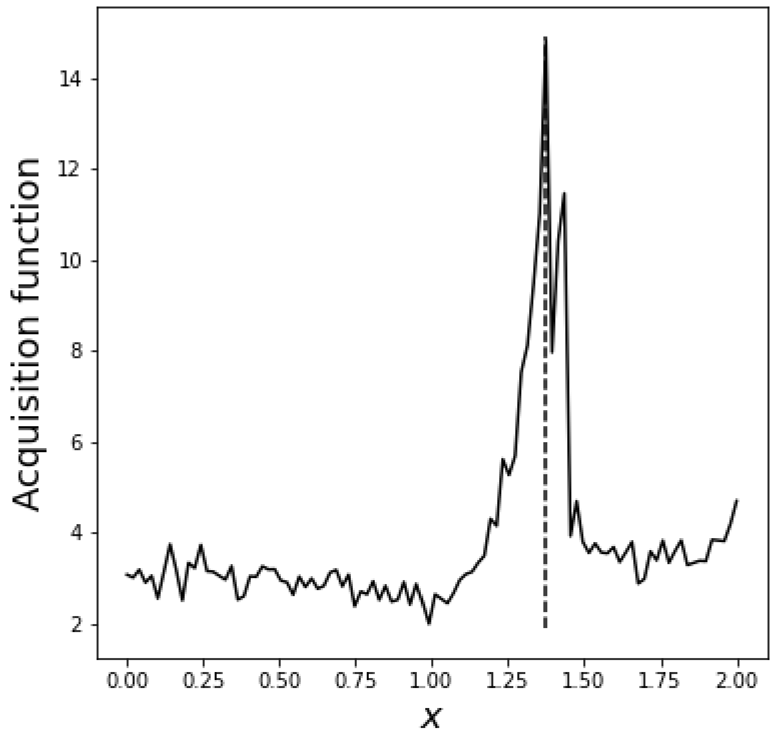
This model simply tells us that there are two minima in our data but does not assume to have any prior knowledge about their relative depth, width, or distance

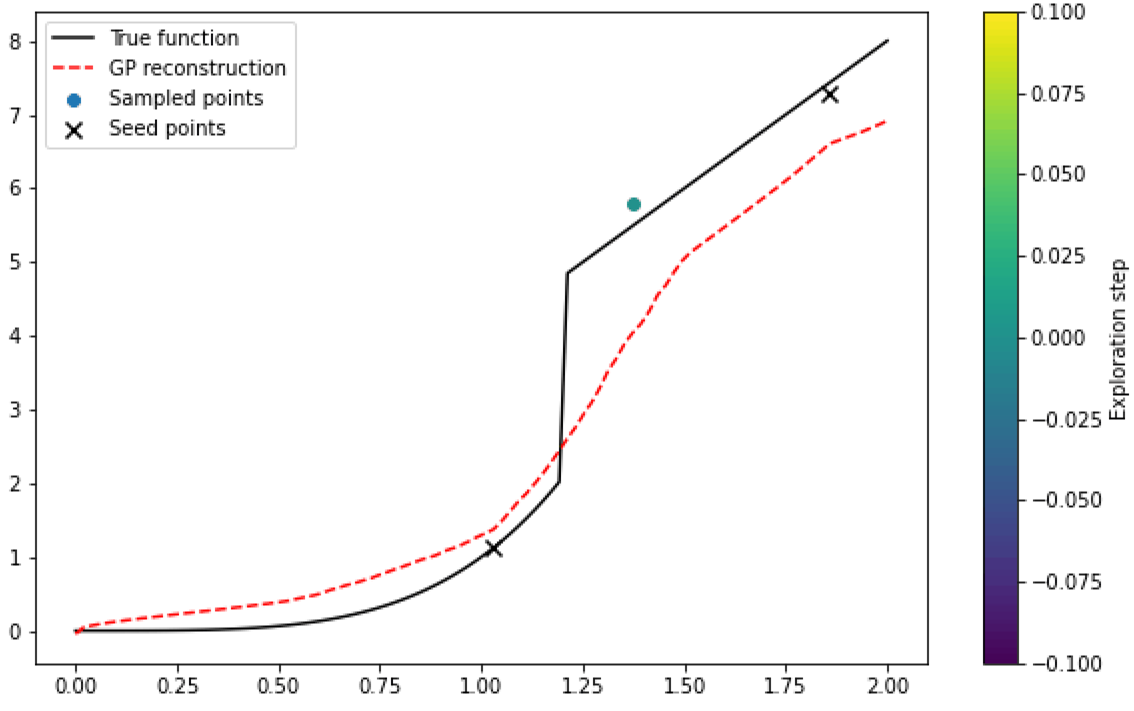
Simple GP search





Structured GP search



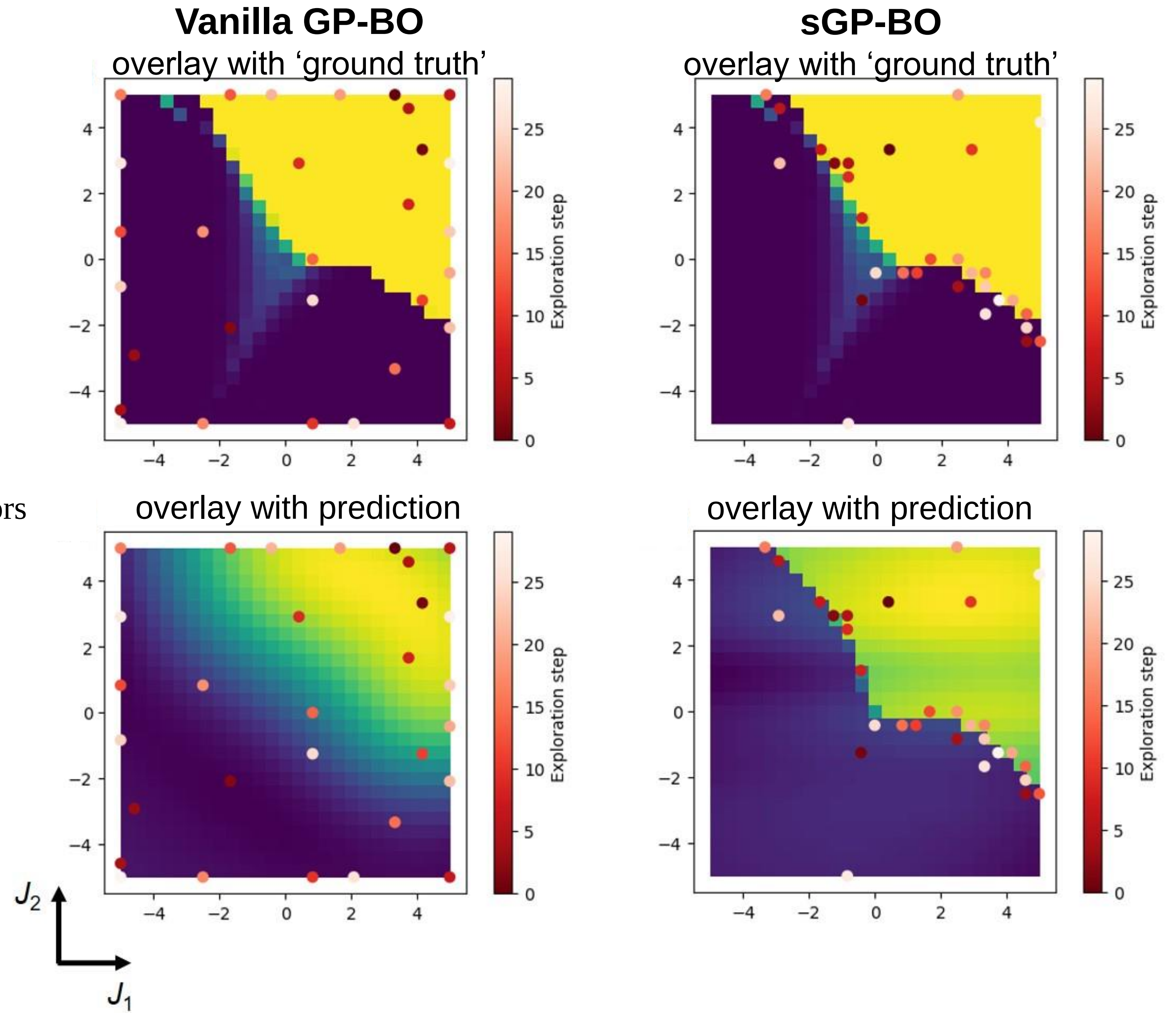


Application to Ising model

Probabilistic model

$A/\tanh(\frac{f(J_1)+f(J_2)}{w})$

where $f(J)$ is a third-degree polynomial with normal priors on its parameters



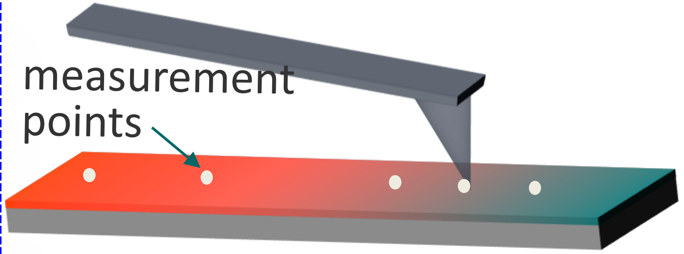
Colab

Hypothesis Active Learning

Co-navigation of experimental and hypothesis spaces

Goal: Learn (1) physical property distribution and (2) a correct model of system's behavior

Synthesis
1. Chemical synthesis
2. Film growth
3. ….



composition gradient sample

Physical measurement

Vector

Scalar

Statistical descriptor (e.g., latent variable)

Physical descriptor (e.g., conductivity, polarization, etc)

New point

*s*GP

Sample model

Physical hypotheses
1. Model 1, priors p_1, reward r_1
2. Model 2, priors p_2, reward r_2

n. Model *n*, priors p_n, reward r_n

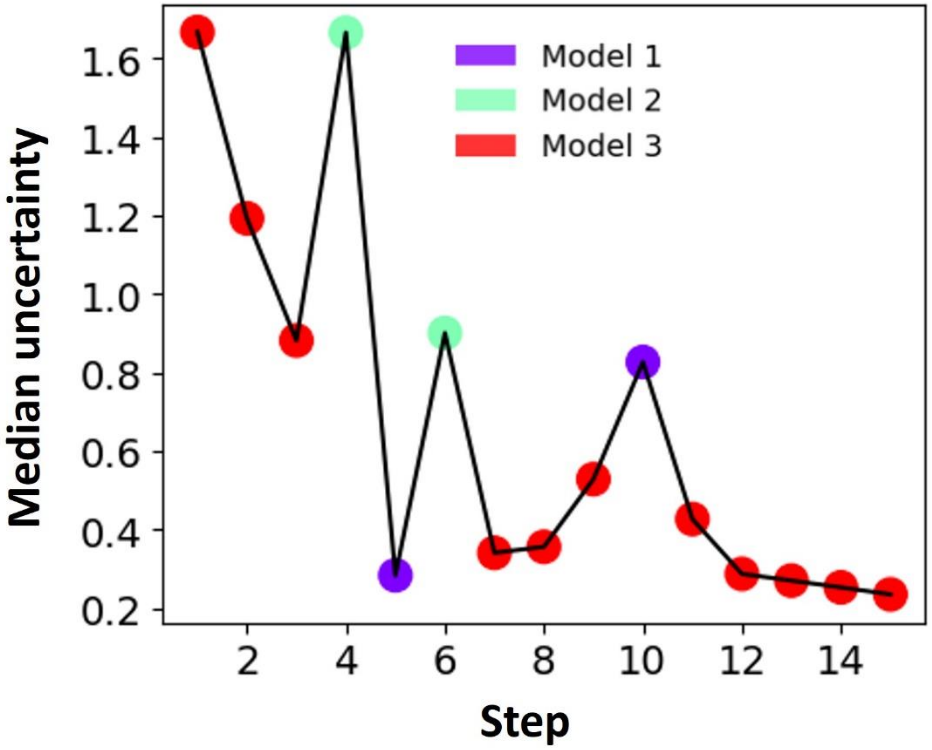
Model's reward

M. Ziatdinov et al. *Advanced Materials* 2201345 (2022)
Y. Liu et al., *Patterns* 4, 100704 (2023)

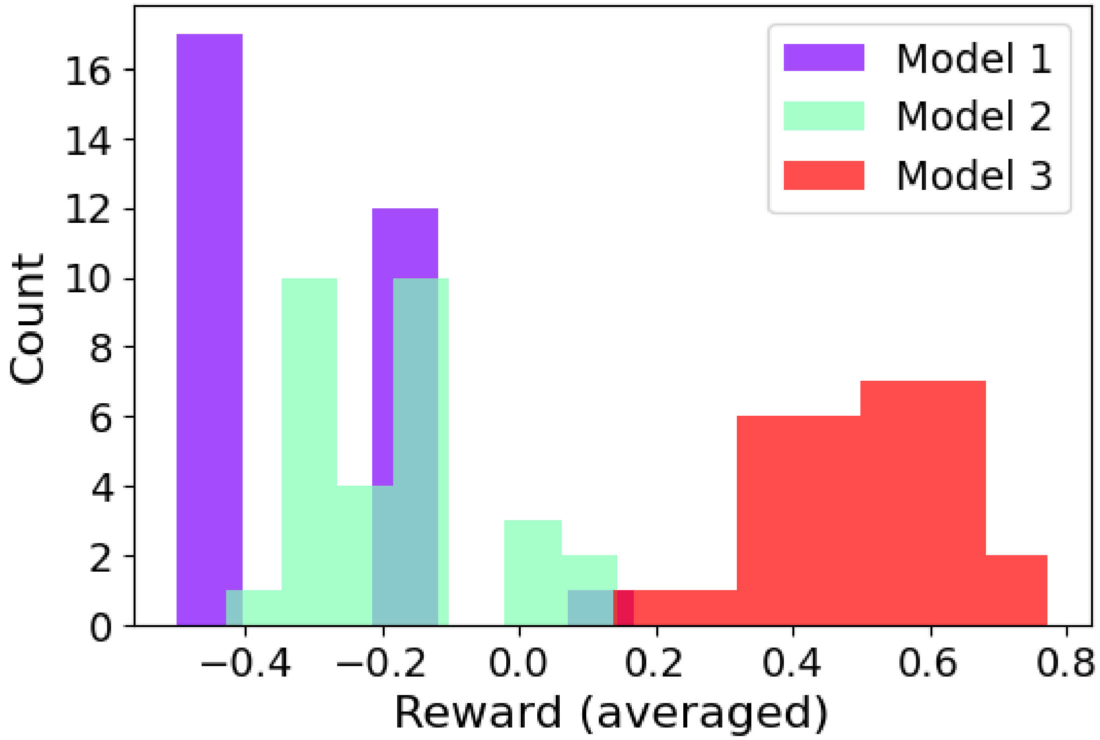
Hypothesis Learning: Synthetic data

Synthetic data represents a 1D discontinuous phase transition

Evolution of uncertainty for a single seed

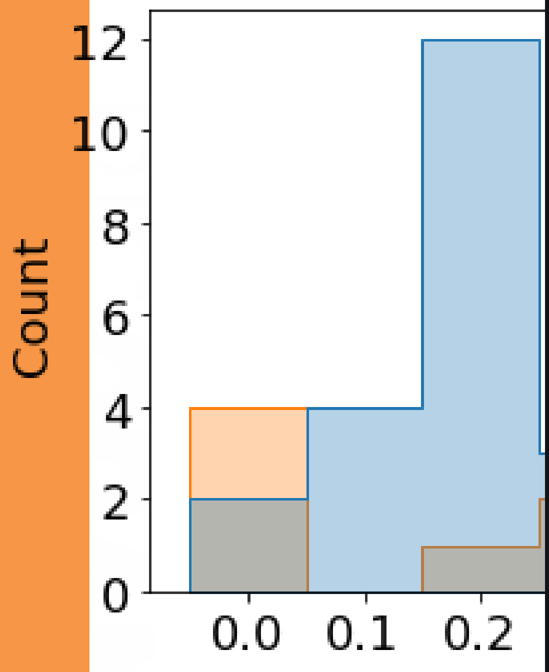


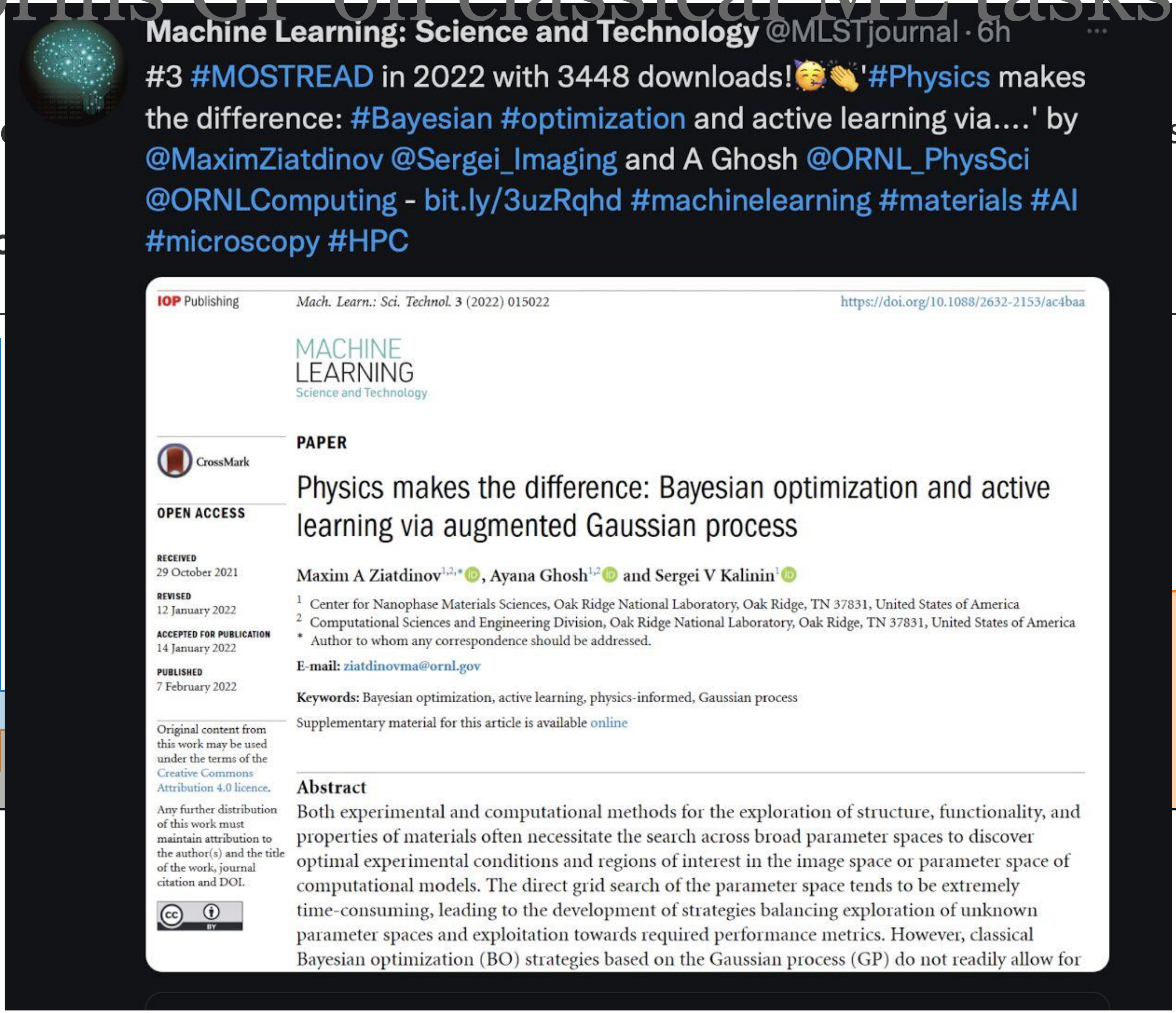
Results for 30 random seeds



The hypothesis learning learns a correct data distribution with a small number of sparse measurements while also identifying a correct model that describes the system's behavior

Outperforms GP on classical ML tasks

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PAPER

Physics makes the difference: Bayesian optimization and active learning via augmented Gaussian process

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Keywords: Bayesian optimization, active learning, physics-informed, Gaussian process

Supplementary material for this article is available online

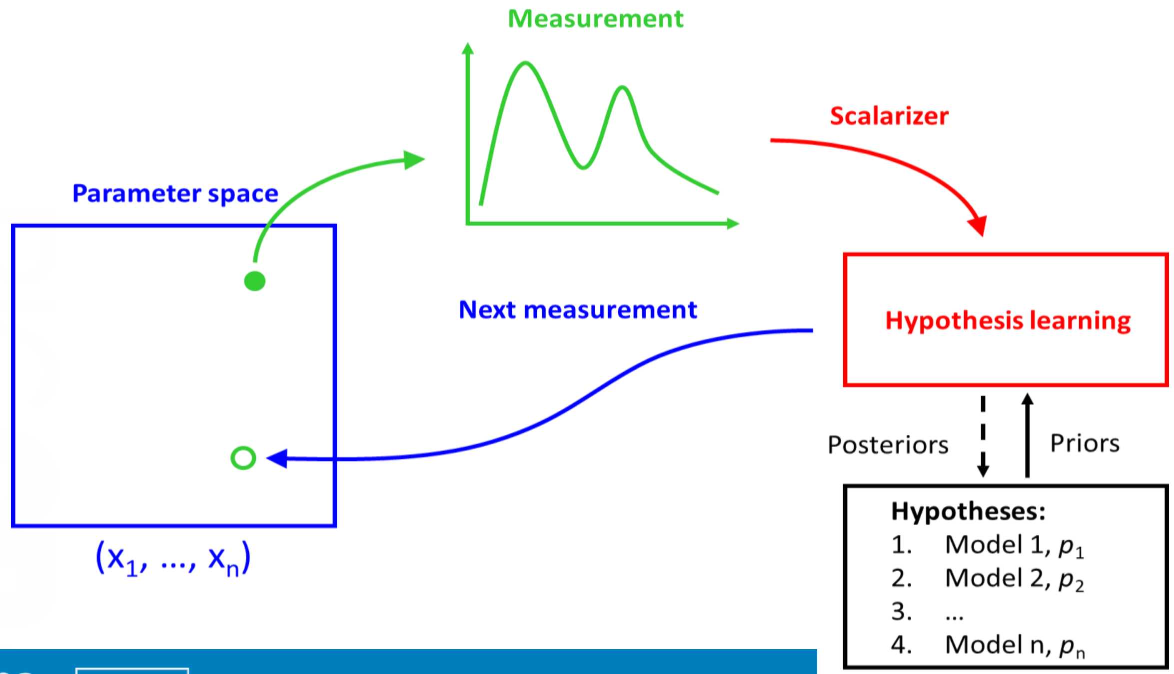
Abstract

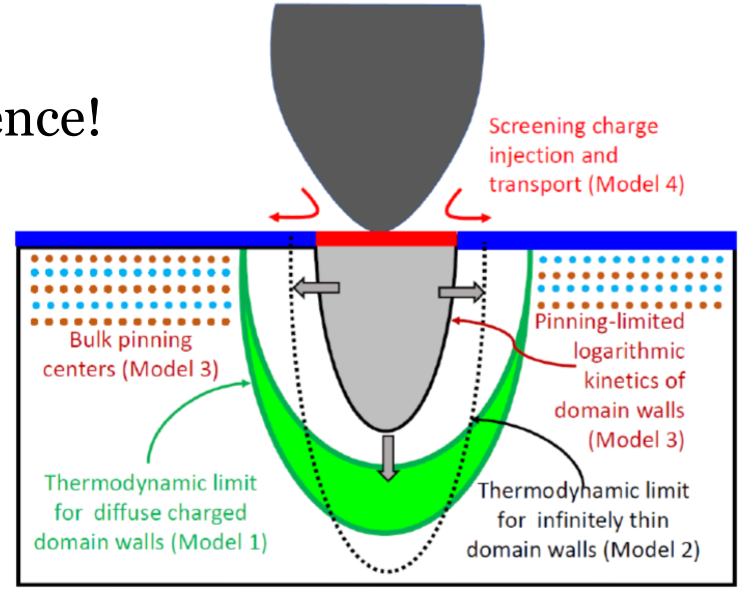
Both experimental and computational methods for the exploration of structure, functionality, and properties of materials often necessitate the search across broad parameter spaces to discover optimal experimental conditions and regions of interest in the image space or parameter space of computational models. The direct grid search of the parameter space tends to be extremely time-consuming, leading to the development of strategies balancing exploration of unknown parameter spaces and exploitation towards required performance metrics. However, classical Bayesian optimization (BO) strategies based on the Gaussian process (GP) do not readily allow for

M. Ziatdinov et al., *Machine Learning: Science and Technology* 3, 015022 (2022)

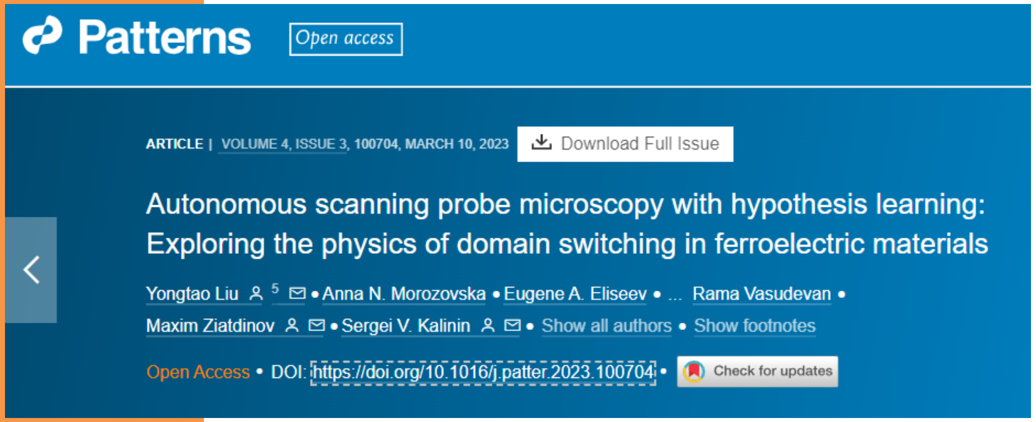
Hypothesis Learning

- Can ML algorithm think like a scientist?
- Yes – automated experiment can pursue hypothesis-driven science!





		Model Equation
Thermodynamic 1	Model I	$r(V) = r_{cr} + r_0\sqrt{\left(\frac{V}{V_c}\right)^{2/3} - 1}$
Thermodynamic 2	Model II	$r(V) = r_{cr} + r_0\sqrt[3]{\left(\frac{V}{V_c}\right)^{2} - 1}$
Wall pinning	Model III	$r(V,t) = V^{\alpha}\log\tau$
Charge injection	Model IV	$r(V,t) = V^{\alpha}\tau^{\beta}$

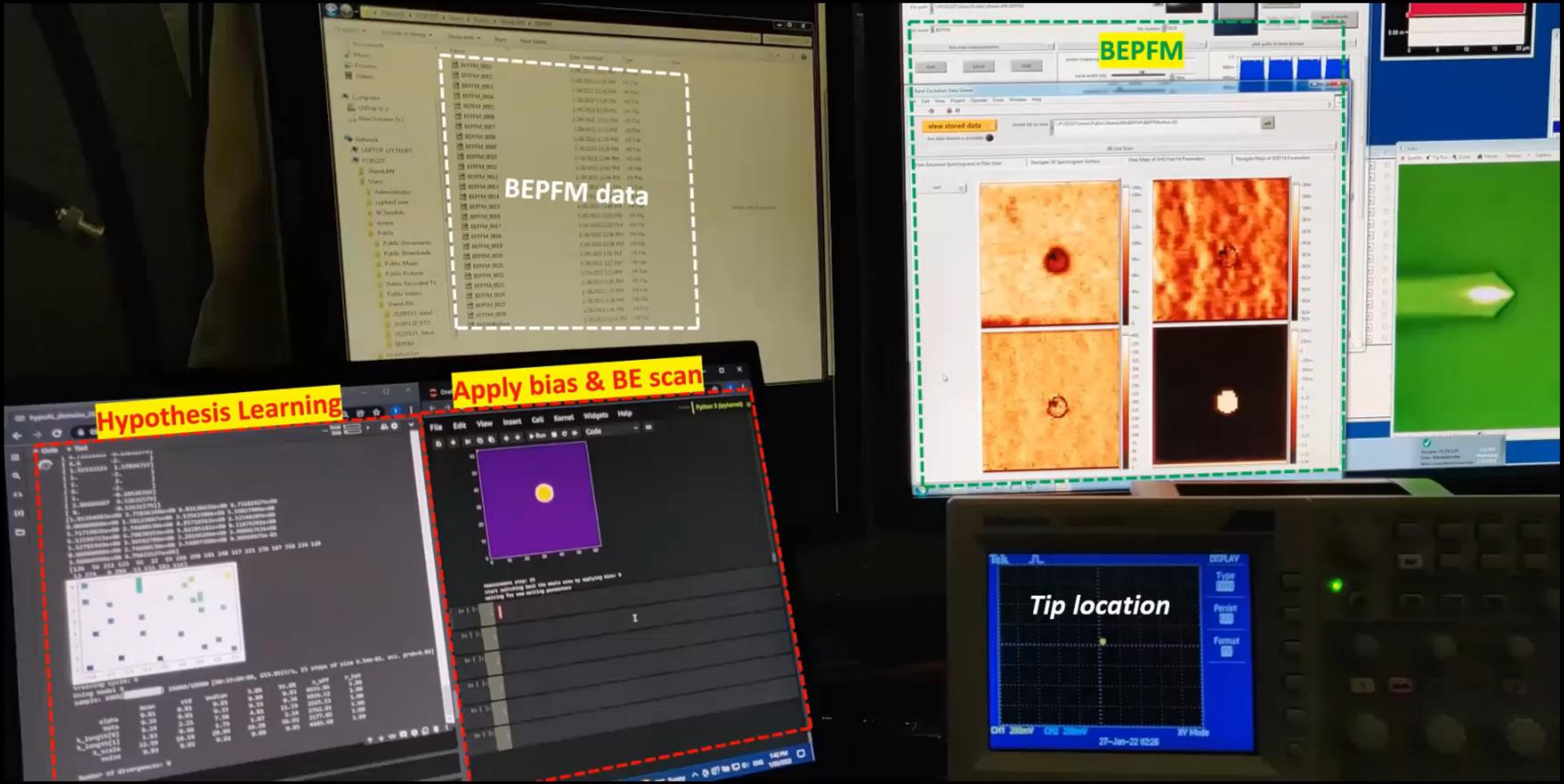

Patterns Open access

ARTICLE | VOLUME 4, ISSUE 3, 100704, MARCH 10, 2023 Download Full Issue

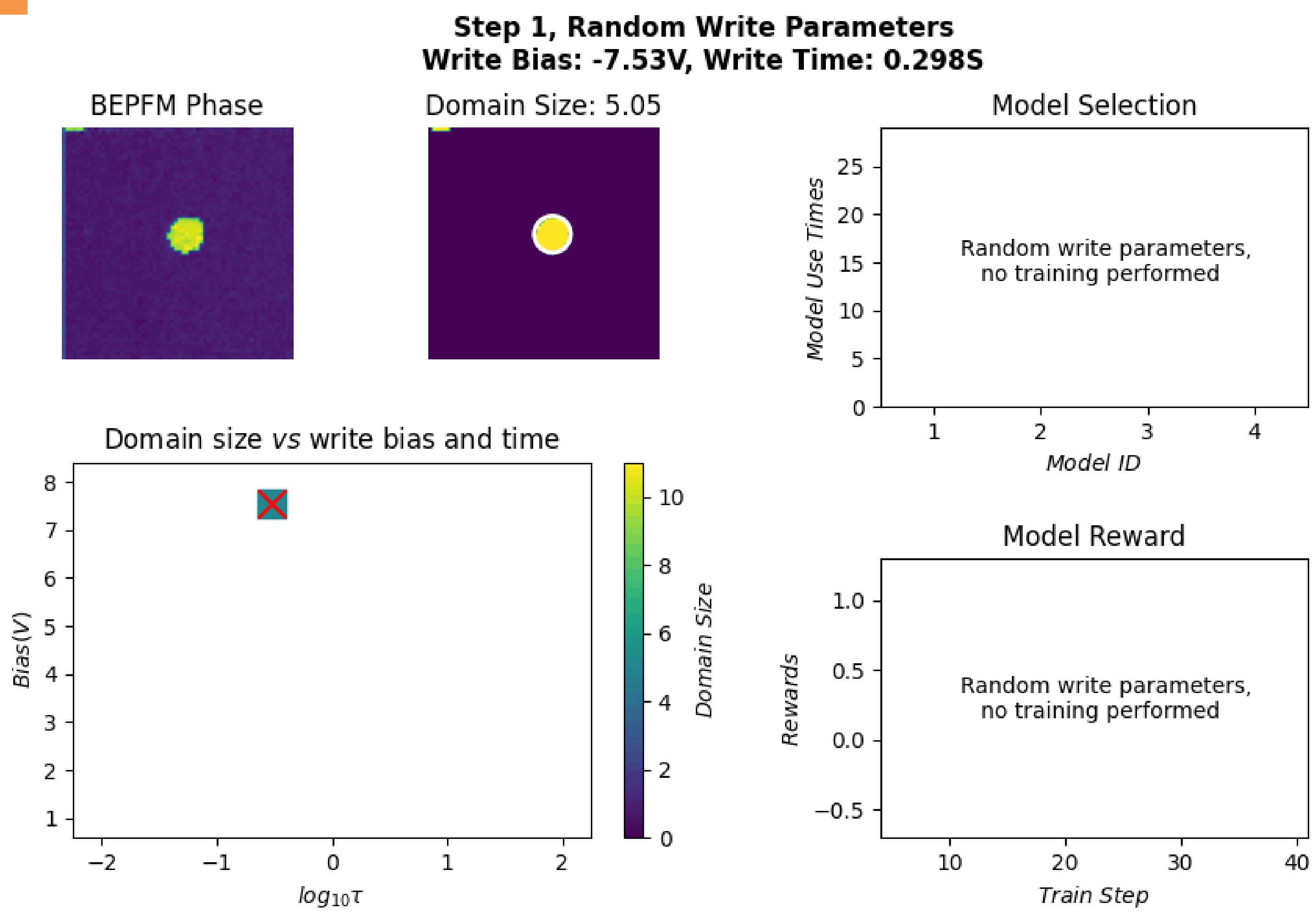
Autonomous scanning probe microscopy with hypothesis learning: Exploring the physics of domain switching in ferroelectric materials

Yongtao Liu [5] • Anna N. Morozovska • Eugene A. Eliseev • … Rama Vasudevan • Maxim Ziatdinov • Sergei V. Kalinin • Show all authors • Show footnotes

Open Access • DOI: https://doi.org/10.1016/j.patter.2023.100704 • Check for updates


BEPFM data
BEPFM
Hypothesis Learning
Apply bias & BE scan
Tip location

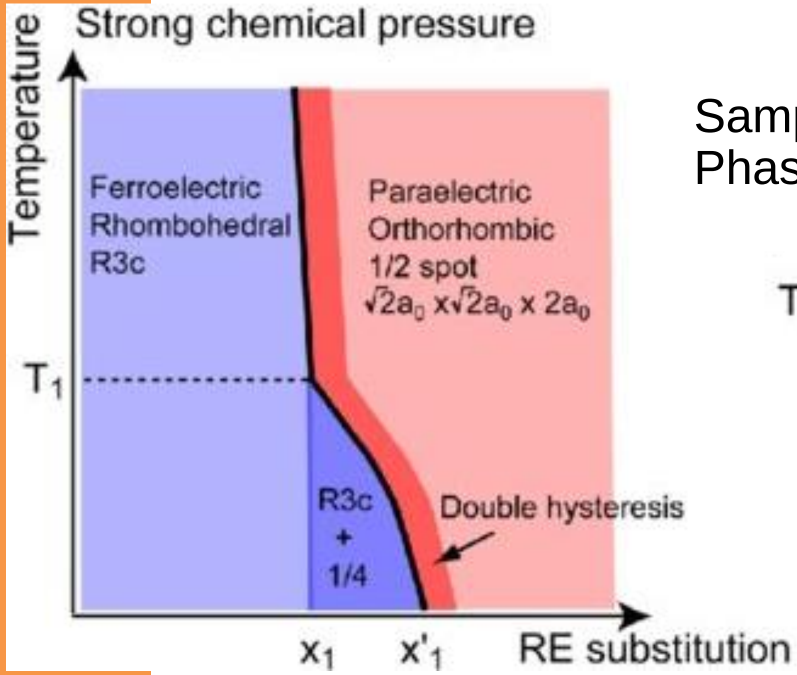
Hypothesis learning in action



Y. Liu, arxiv 2202.01089
Y. Liu, arxiv 2112.06649

- ML algorithm has 4 competing hypothesis on domain switching mechanisms
- These hypothesis represent full set of possibilities for this system
- The microscope chooses experimental parameters in such a way as to establish which hypothesis is correct fastest
- Important: the same approach can be implemented in synthesis and electrical characterization
- Machine learning meets hypothesis-driven scientific discovery!

Combinatorial Synthesis



Sample by I. Takeuchi, UMD
Phase diagram by N. Valanoor et al.

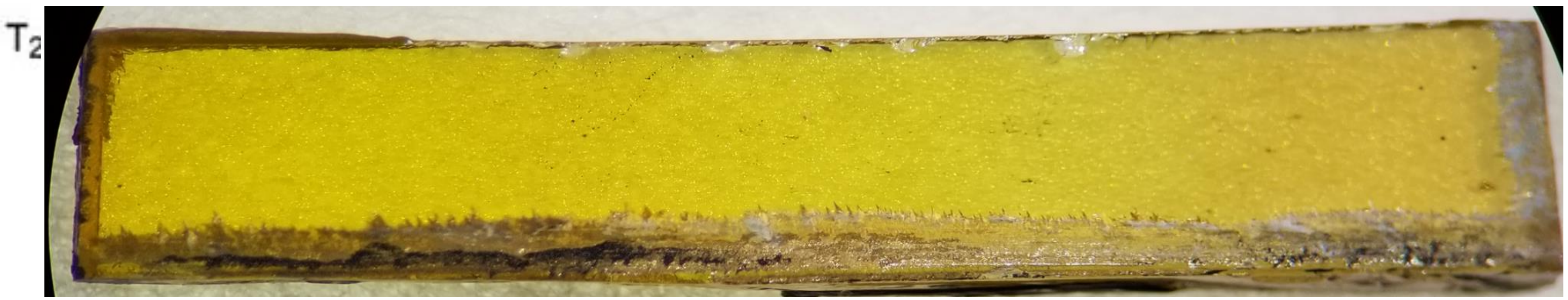
ADVANCED MATERIALS

Research Article

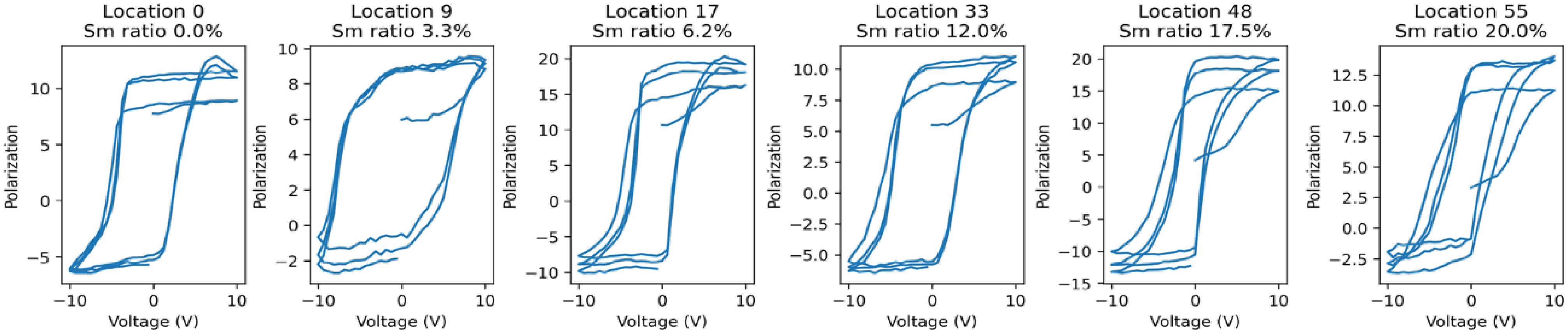
Hypothesis Learning in Automated Experiment: Application to Combinatorial Materials Libraries

Maxim A. Ziatdinov, Yongtao Liu, Anna N. Morozovska, Eugene A. Eliseev, Xiaohang Zhang, Ichiro Takeuchi, Sergei V. Kalinin

First published: 12 March 2022 | https://doi.org/10.1002/adma.202201345 | Citations: 17



BiFeC Linear est. 7%Sm $BiFeO_3$ 20%Sm $BiFeO_3$



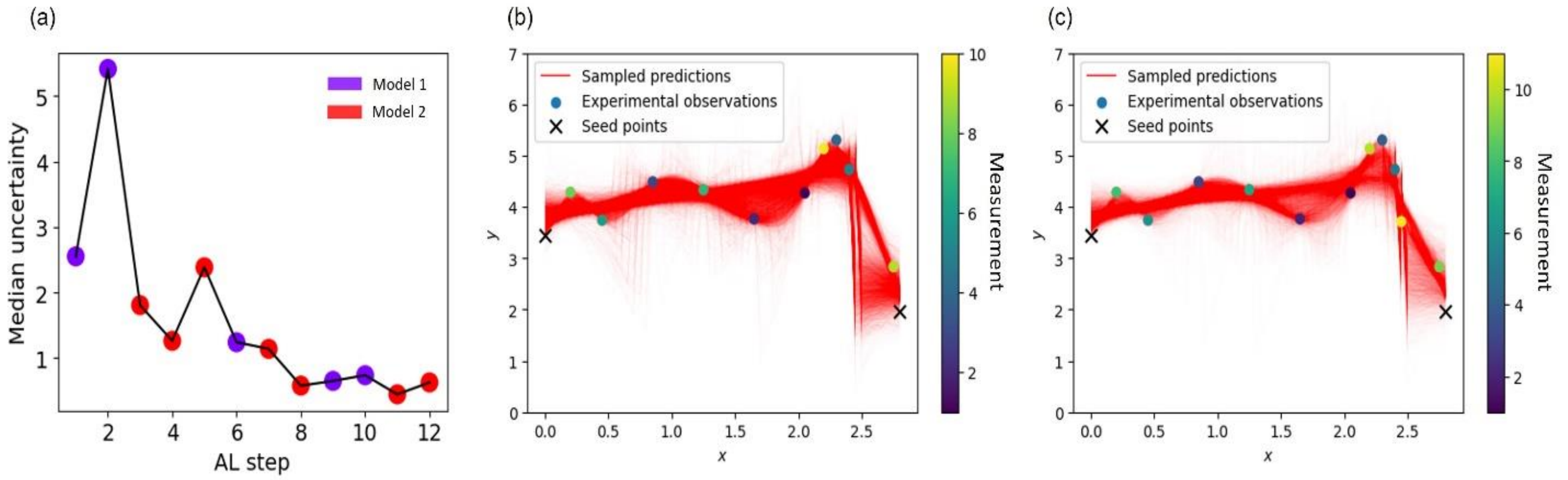
Hypothesis Selection for Ferroelectric

Model 1 (second order phase transition):

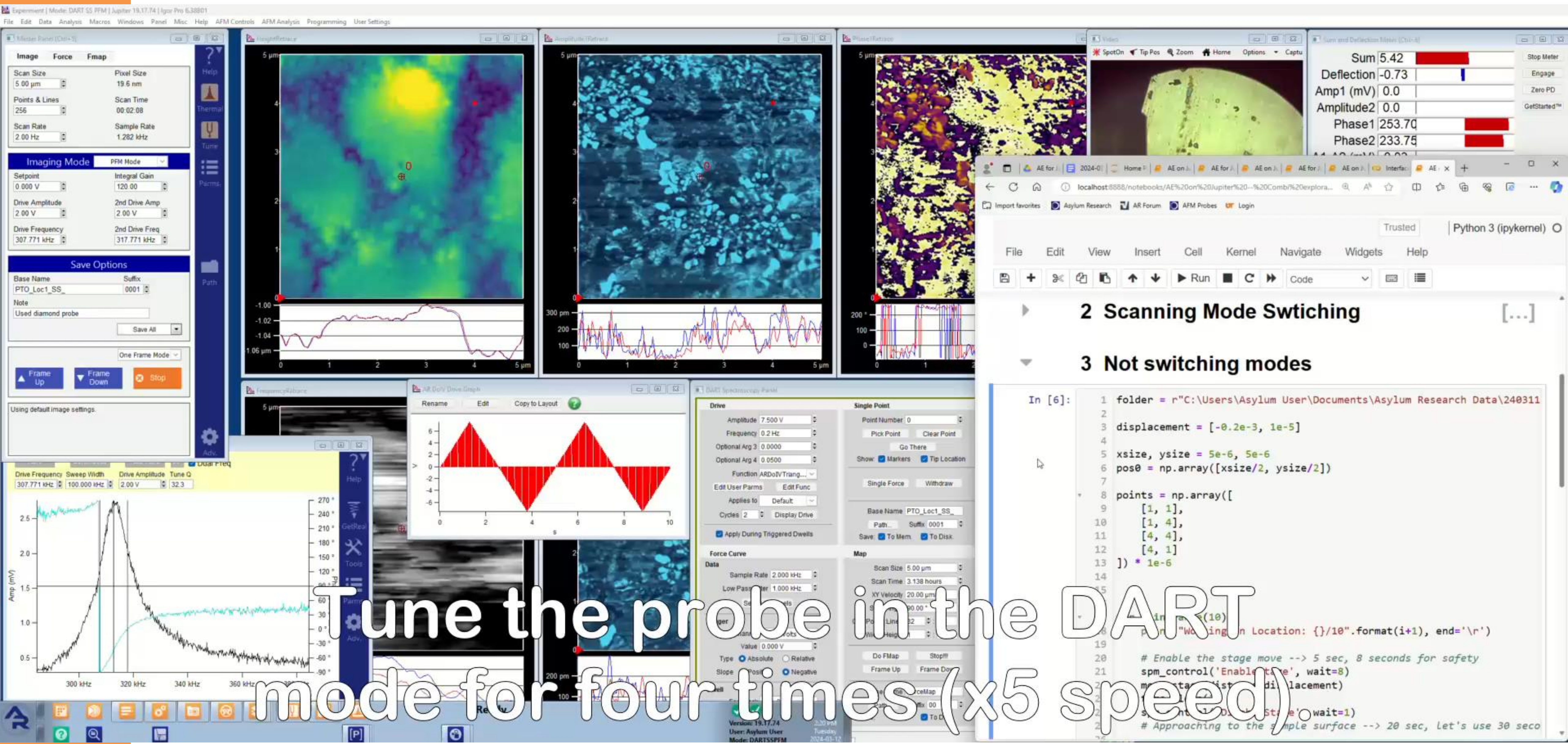
$$S = \begin{cases} S_0\left(1 - \dfrac{x}{x_0}\right)^2 + C, & x \le x_c, \\ C, & x > x_c \end{cases}$$

Model 2 (first order phase transition):

$$S = \begin{cases} S_0\left(1 - \dfrac{x}{x_0}\right)^{\frac{5}{4}} + C_0, & x \le x_c, \\ C_1, & x > x_c \end{cases}$$



THE UNIVERSITY of TENNESSEE KNOXVILLE



Colab 2

Deep Kernel Learning

Two modes of operations

Structural imaging (Cheap)

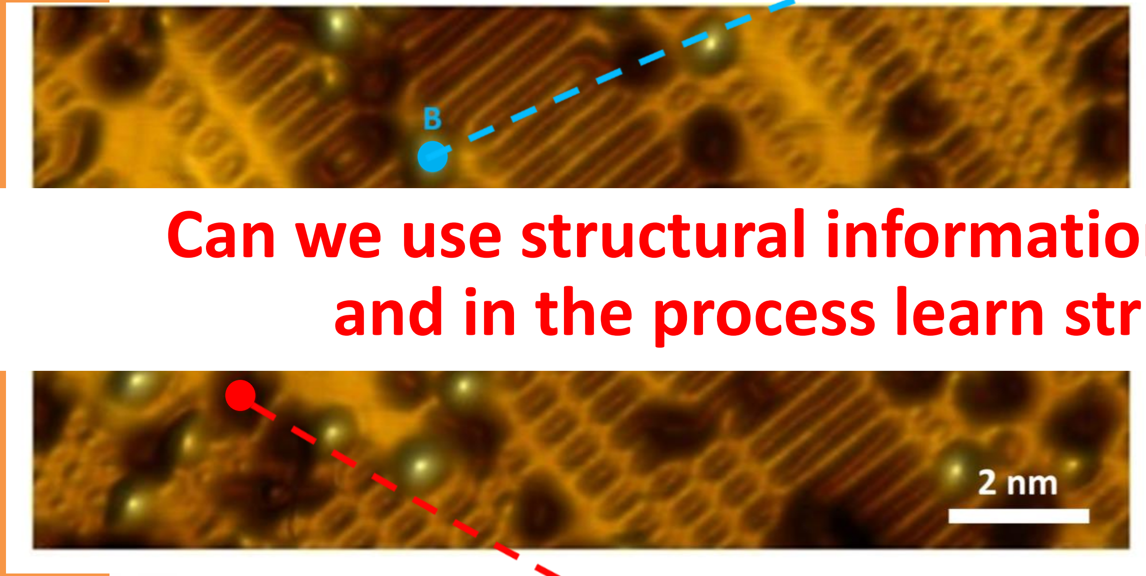
Topography in STM, amplitude/phase in SPM, (HA)ADF-image in STEM, etc.

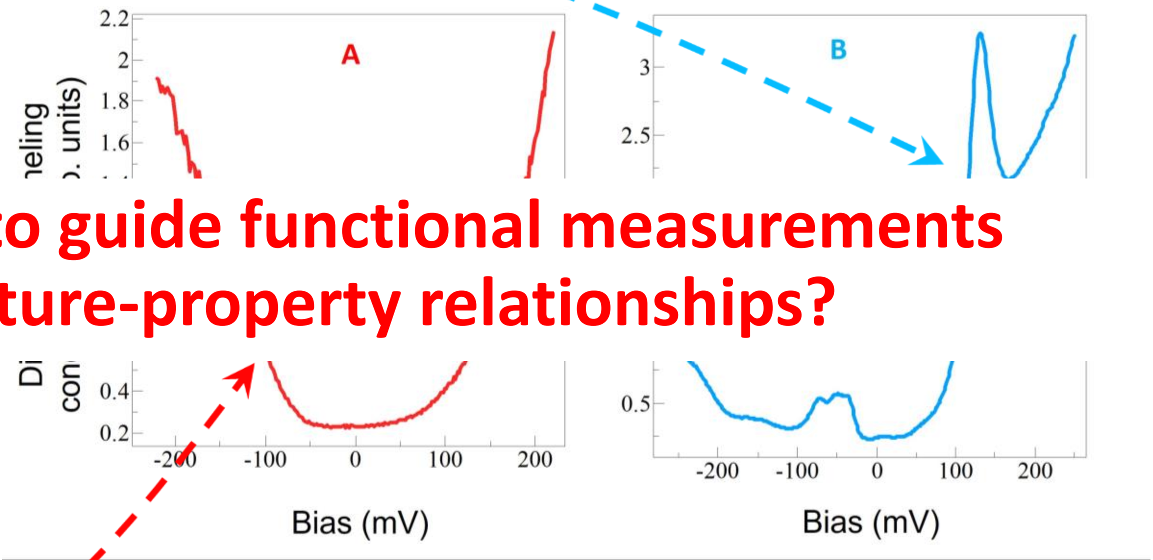
These are FAST measurements (from seconds to minutes)

Functional imaging (Costly)

Scanning tunneling spectroscopy (STS), polarization loops in SPM, EELS in STEM, etc.

These are SLOW and/or DESTRUCTIVE measurements (from minutes to days)



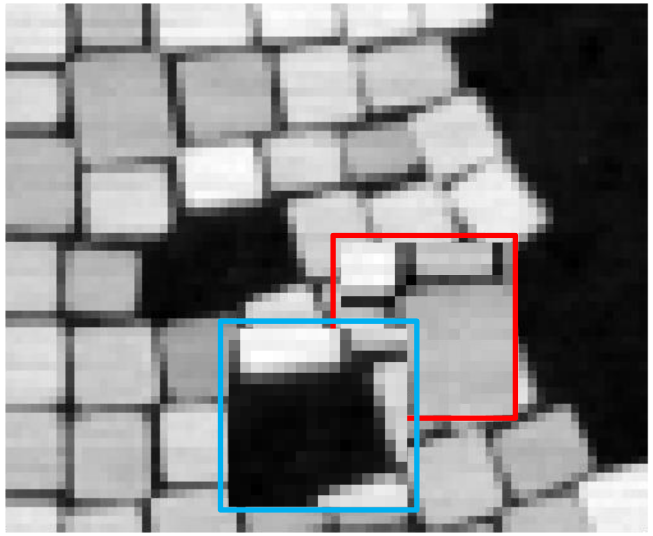


Can we use structural information to guide functional measurements and in the process learn structure-property relationships?

Physics discovery in active experiments

- Suppose we know what physical behavior/property we are interested in (superconductivity, ferroelectric switching, plasmonic modes, etc.)
- This behavior is encoded in spectra that we can measure everywhere in the sample (size of superconducting gap, polarization loop area, peak intensity, etc.)
- We want to identify (local) structural features where this behavior is maximized/minimized
- We want to achieve this with as few measurements as possible (**< 5% of the entire grid**)

Structure



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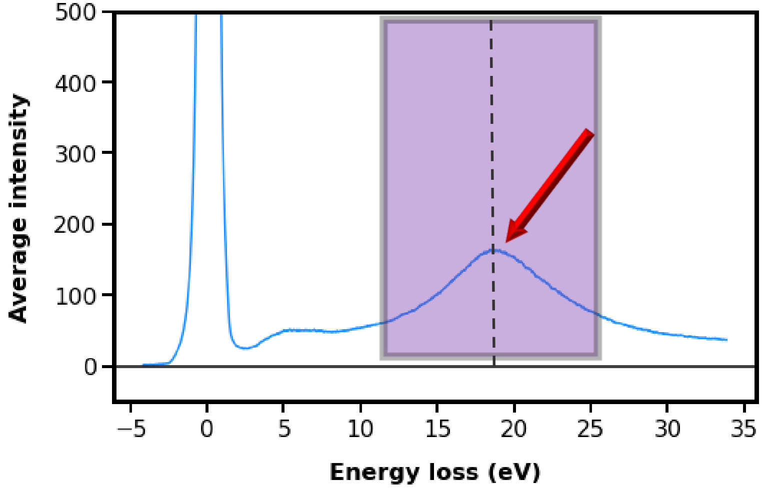
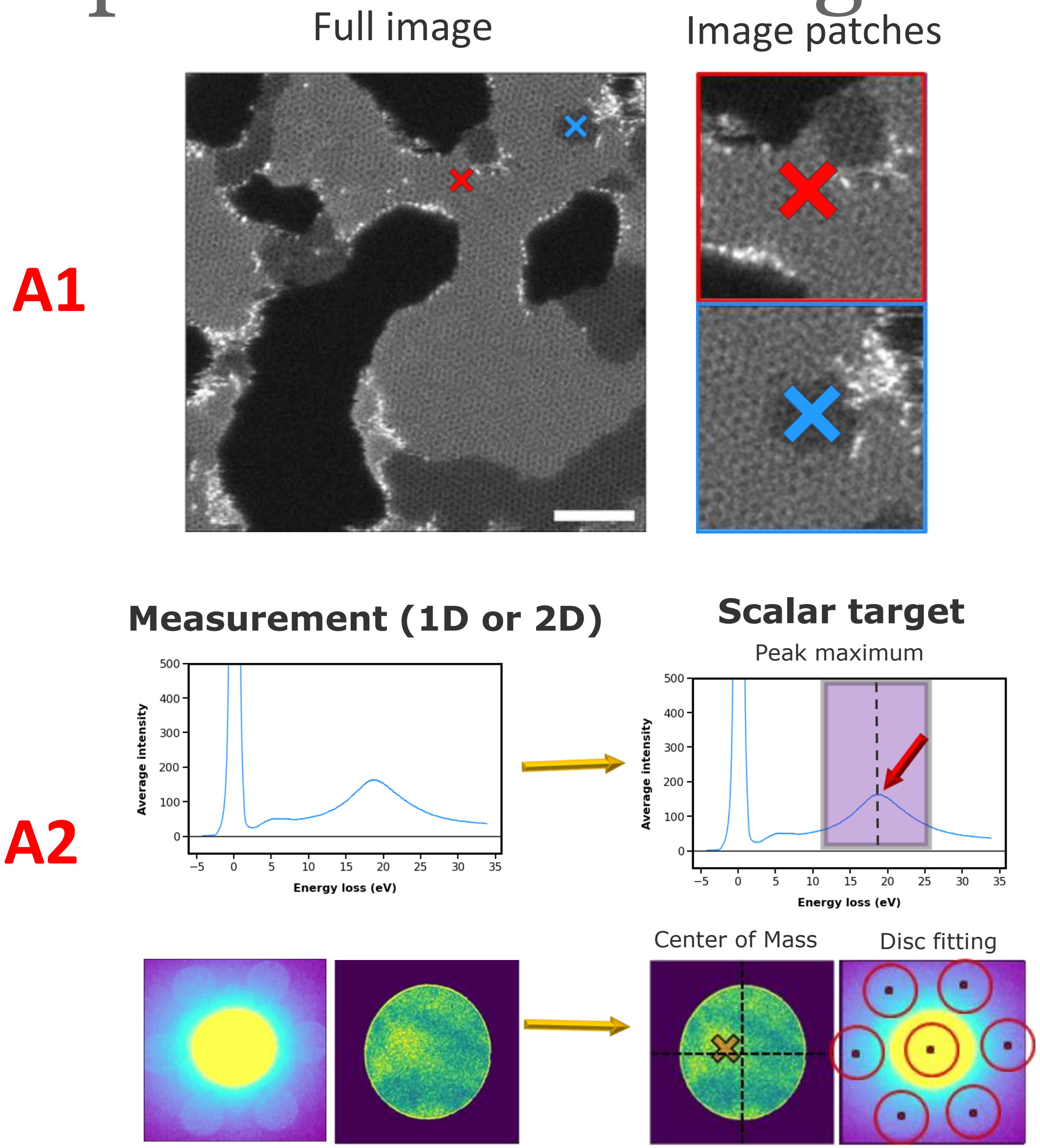
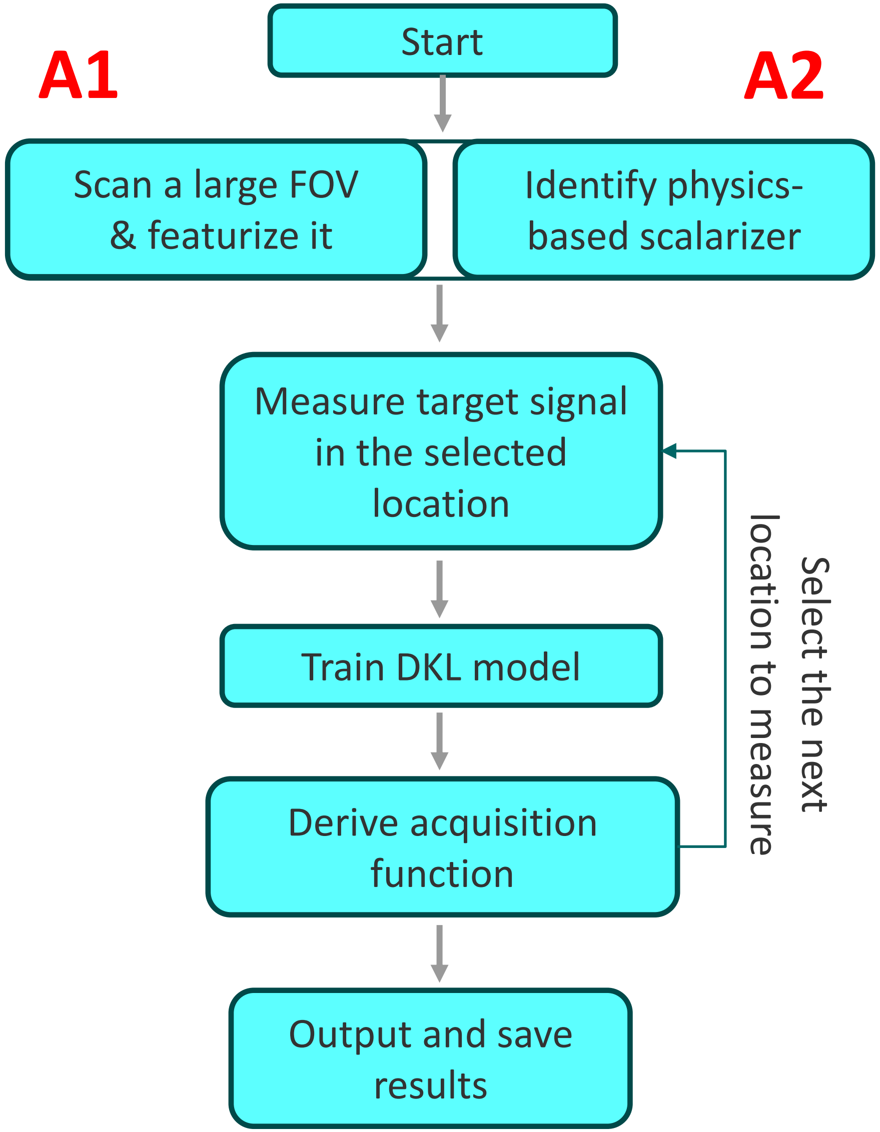
Physical behavior of interest

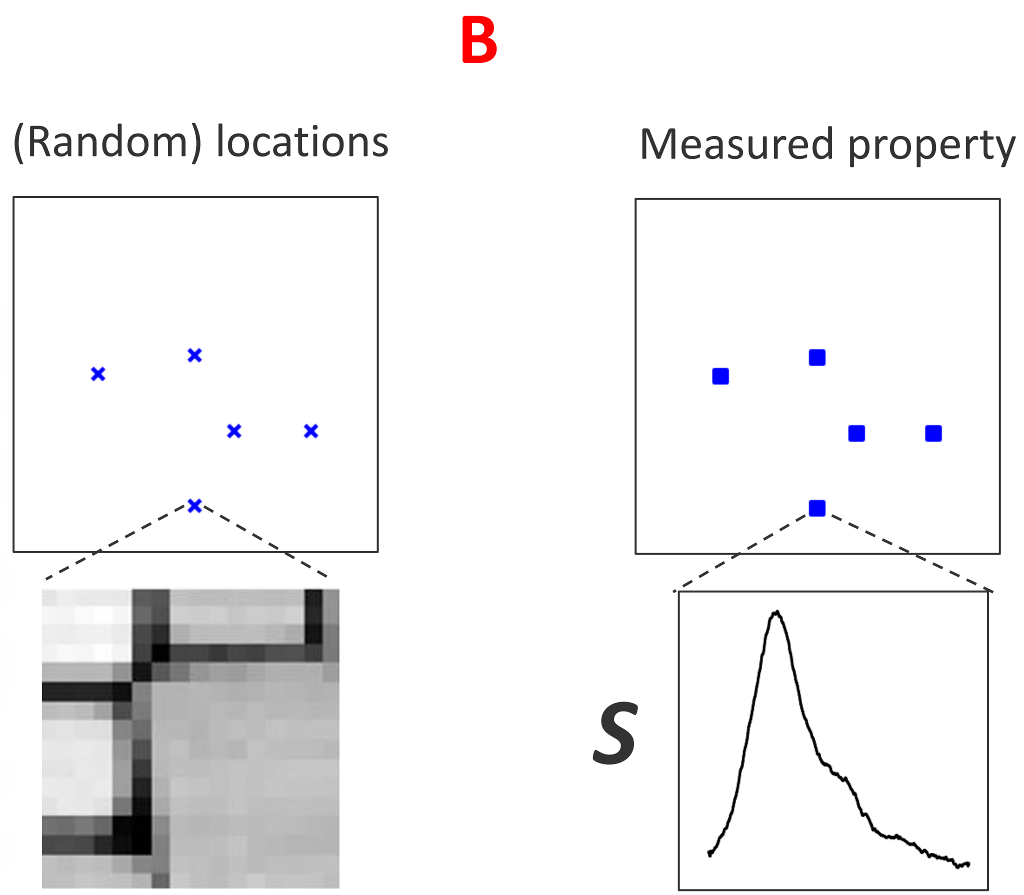


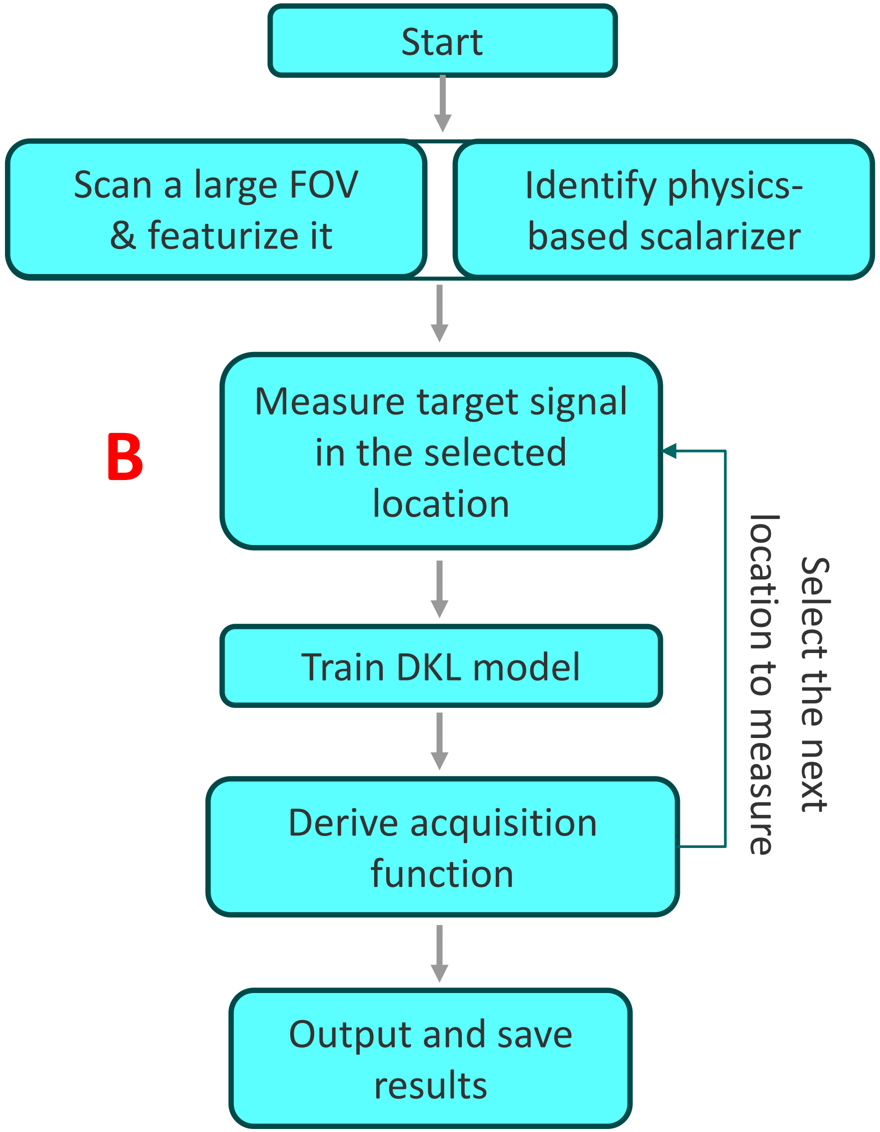
Deep Kernel Learning: Step 1



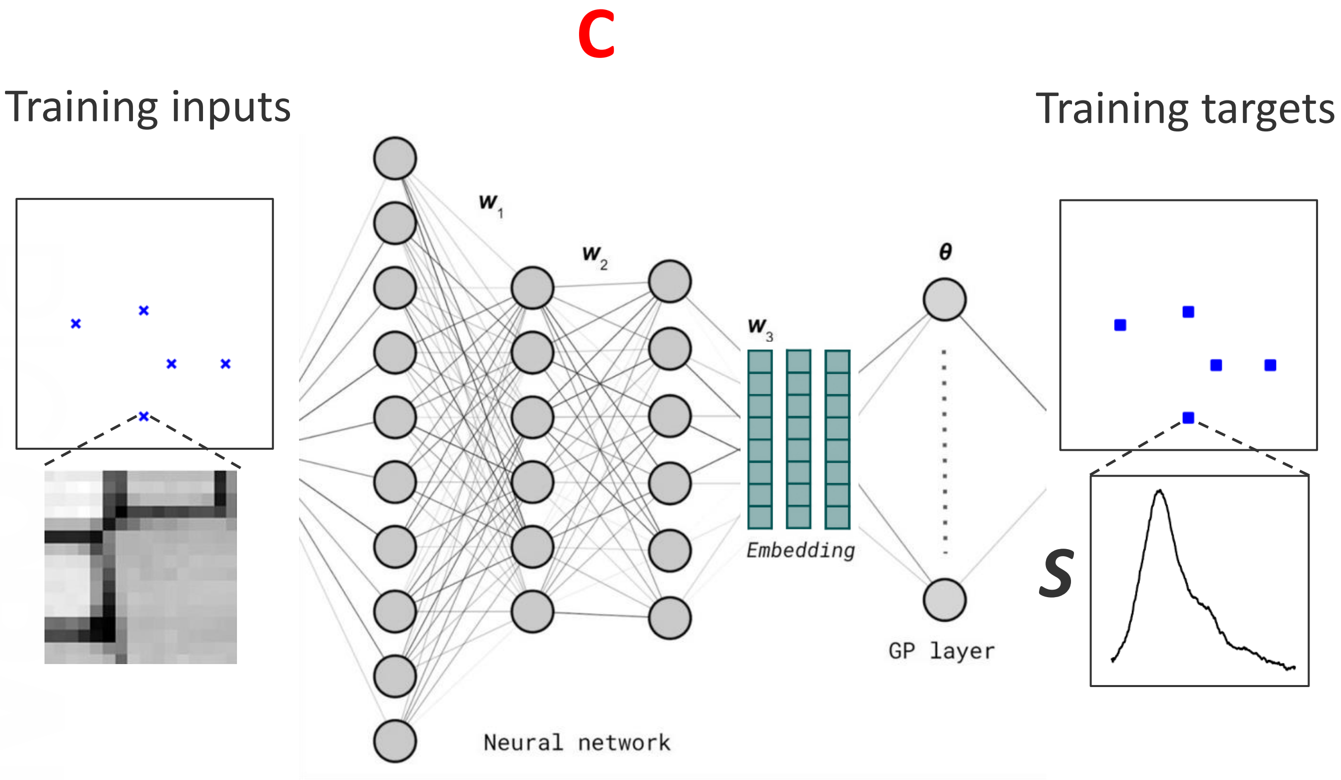


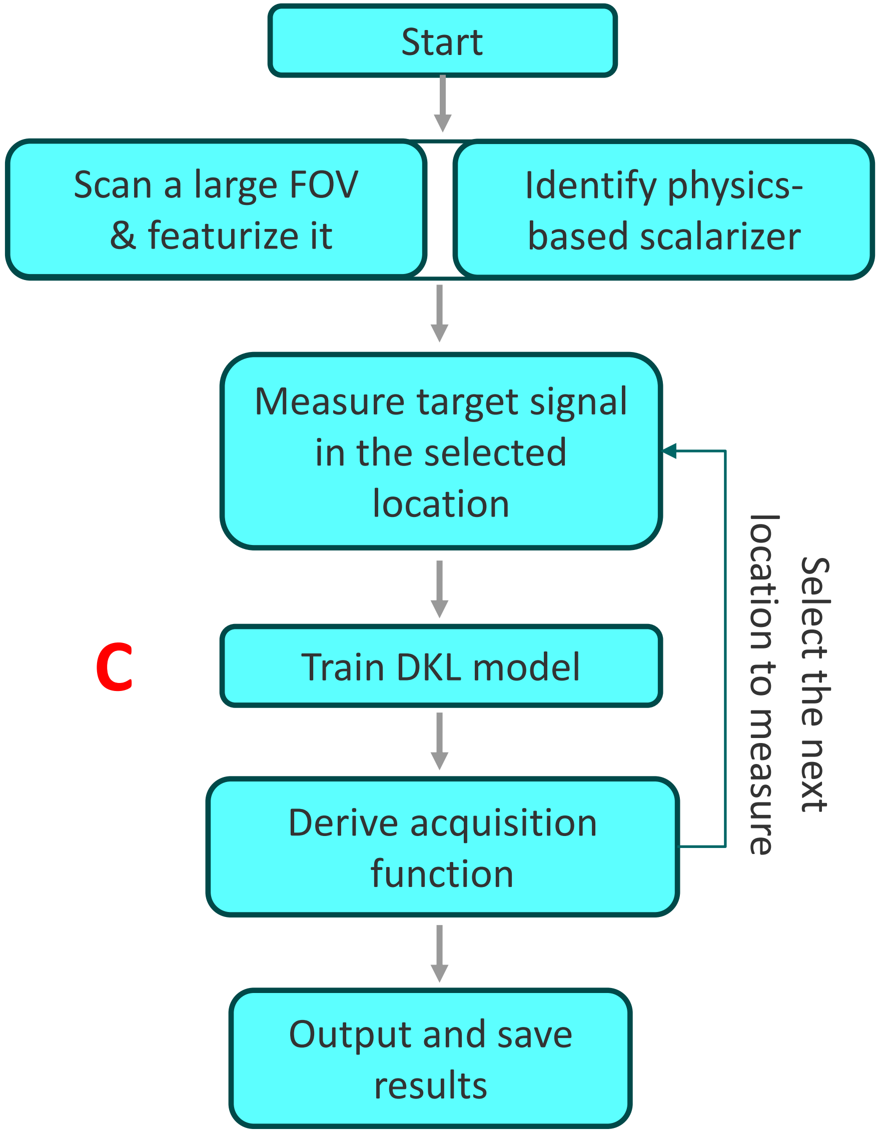
Deep Kernel Learning: Step 2





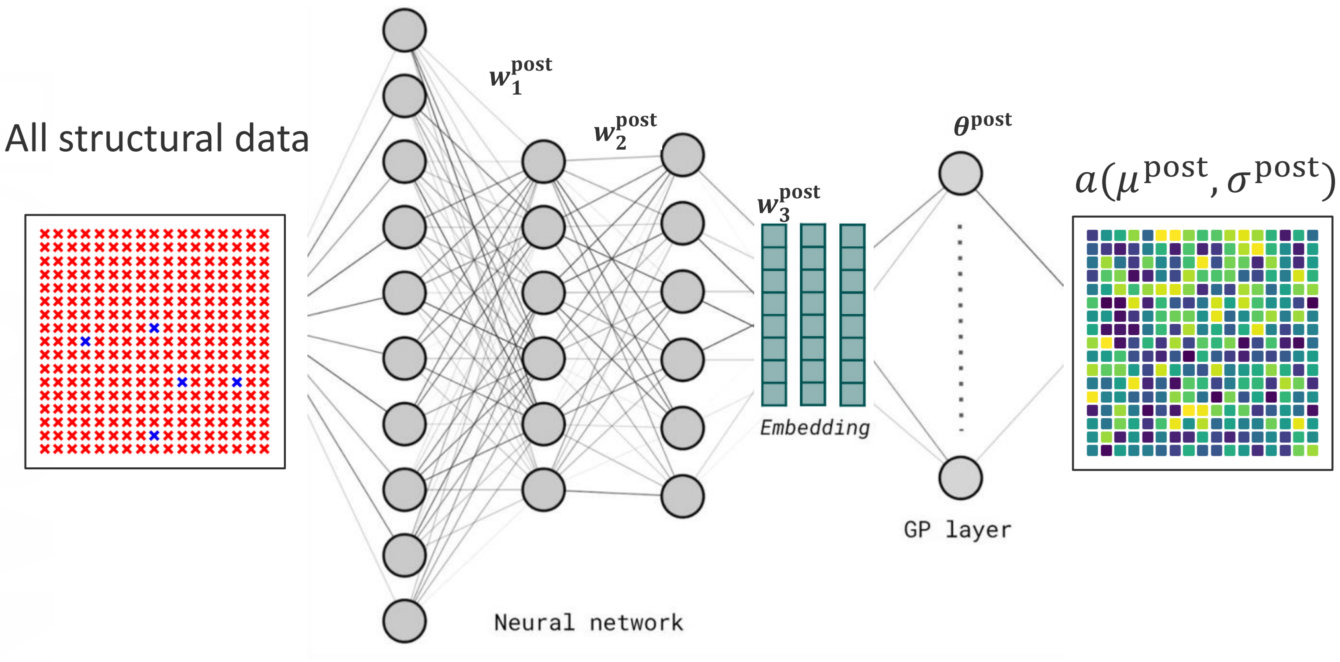
Deep Kernel Learning: Step 3

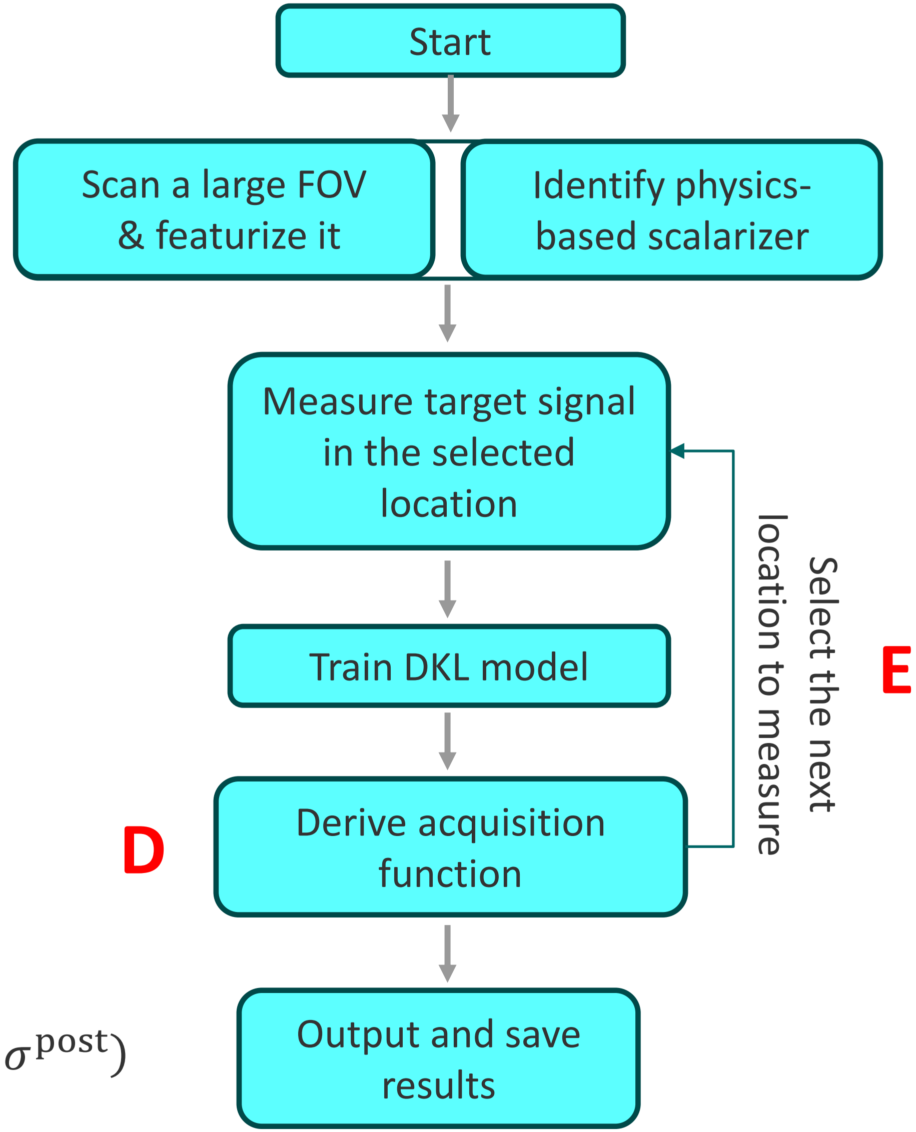




Deep Kernel Learning: Going Active

D





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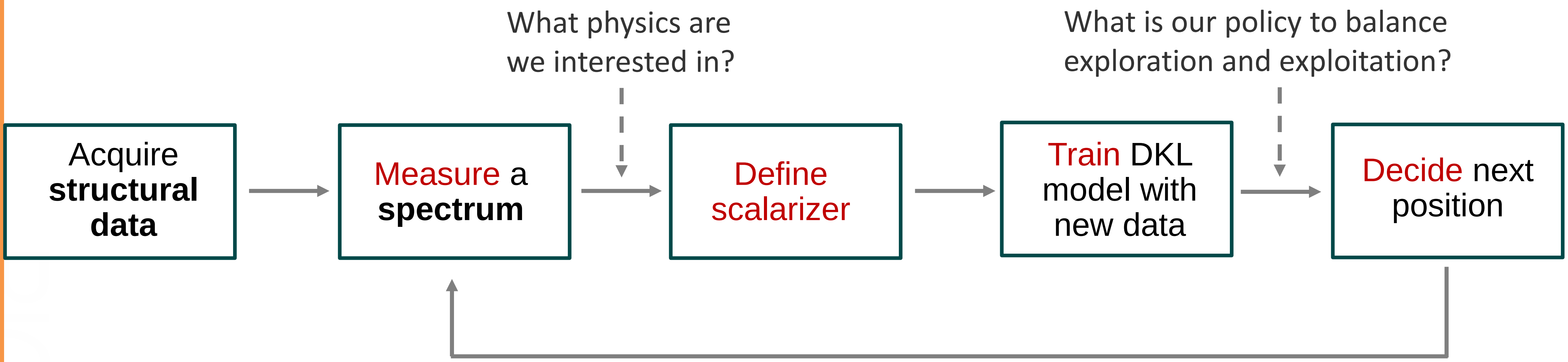
Next point to measure:

$$x_{next} = \arg\max_{\mathbf{x}} a(\mu^{post}, \sigma^{post})$$

Deep Kernel Learning based BO

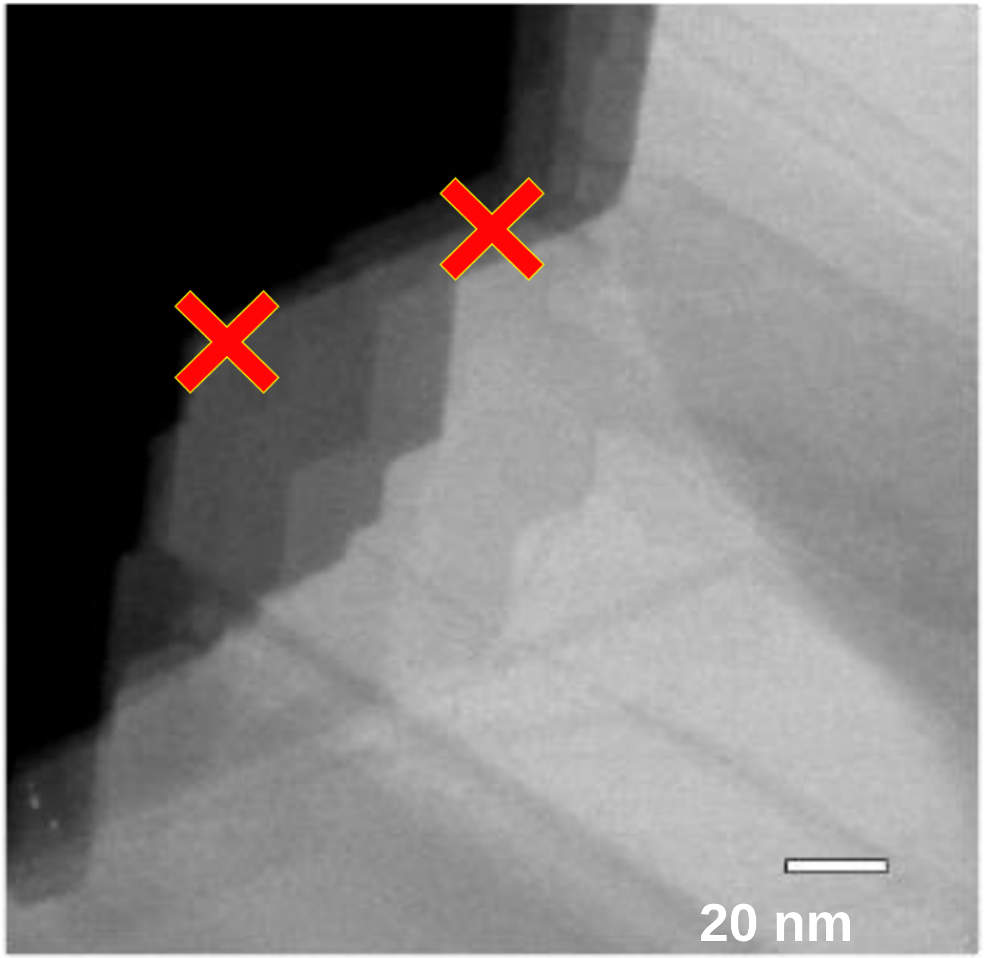


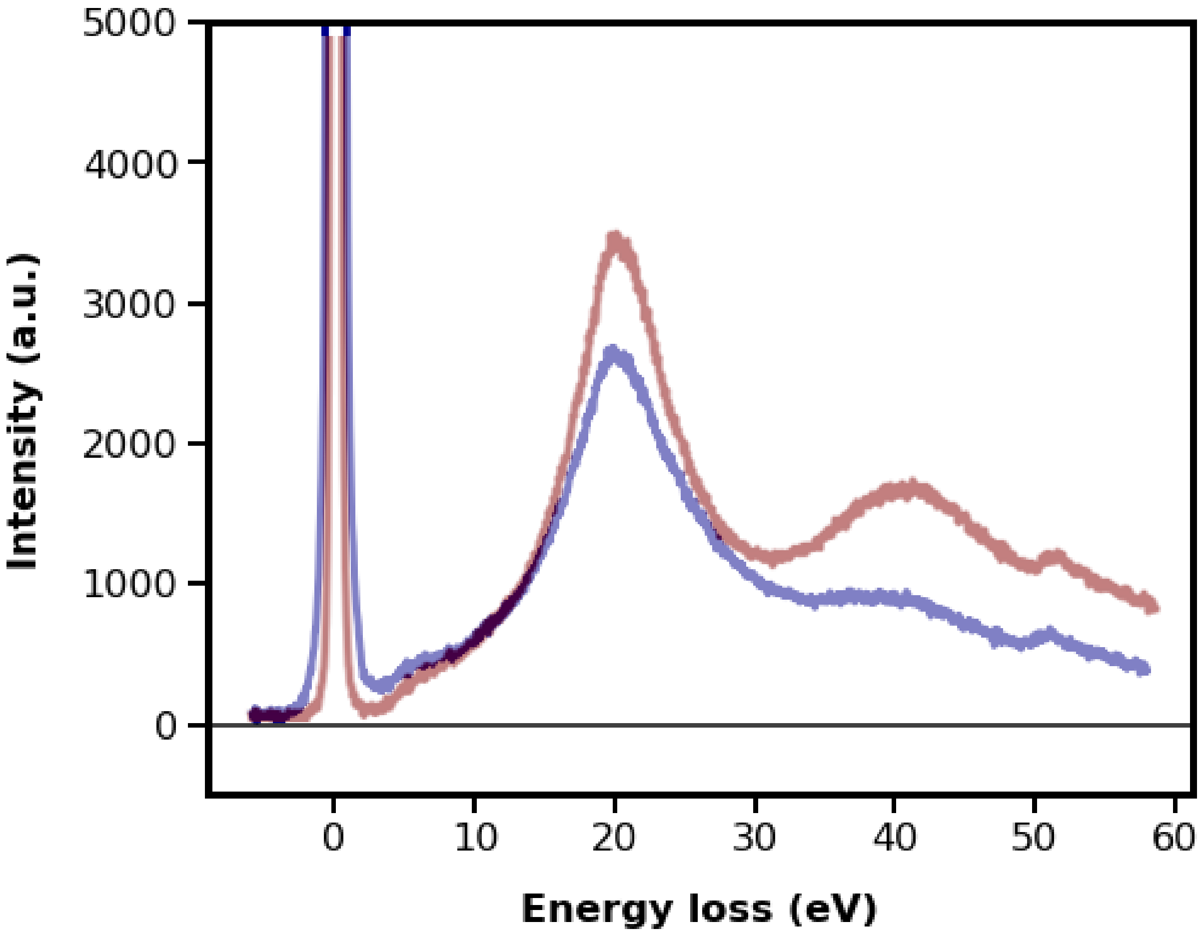
Key concepts:

- **Scalarizer:** (any) function that transforms spectrum into measure of interest. Can be integration over interval, parameters of a peak fit, ration of peaks, or more complex analysis

- **Experimental trace:** collection of image patches and associated spectra acquired during experiment. Note that we collect spectra, not only scalarizers

From Static to Active Learning





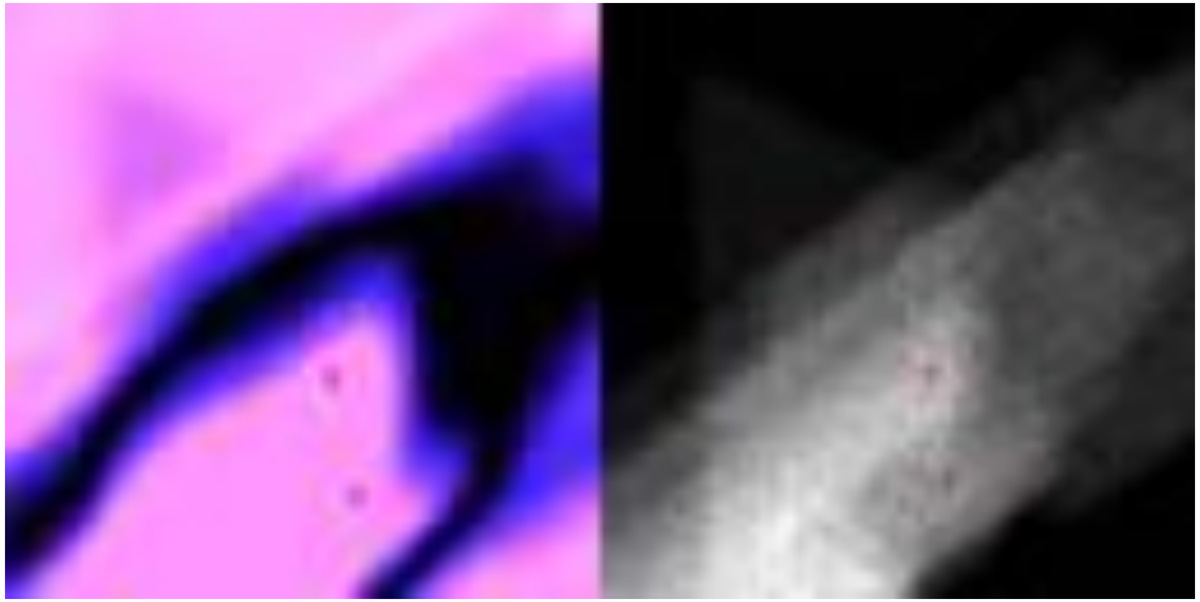
1. What if we have full access to structural information
2. And want to choose locations for (EELS, 4D STEM, CL, EDX) measurements
3. So as to **learn** relationship between structure and spectrum fastest
4. Or **discover** which microstructural elements give rise to specific **desired** spectral features?

Discovering Regions with Interesting Physics

- **Discovering physics in a "new" material $MnPS_3$**
- **Curve fitting** to help enforce physical processes

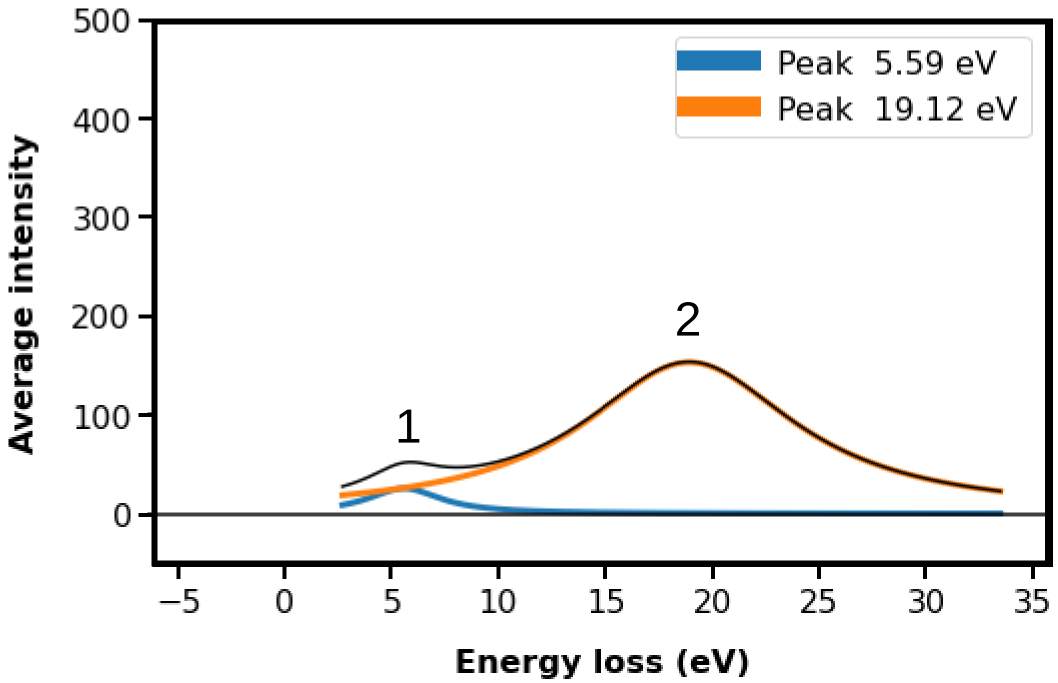
Acquisition function

HAADF-STEM

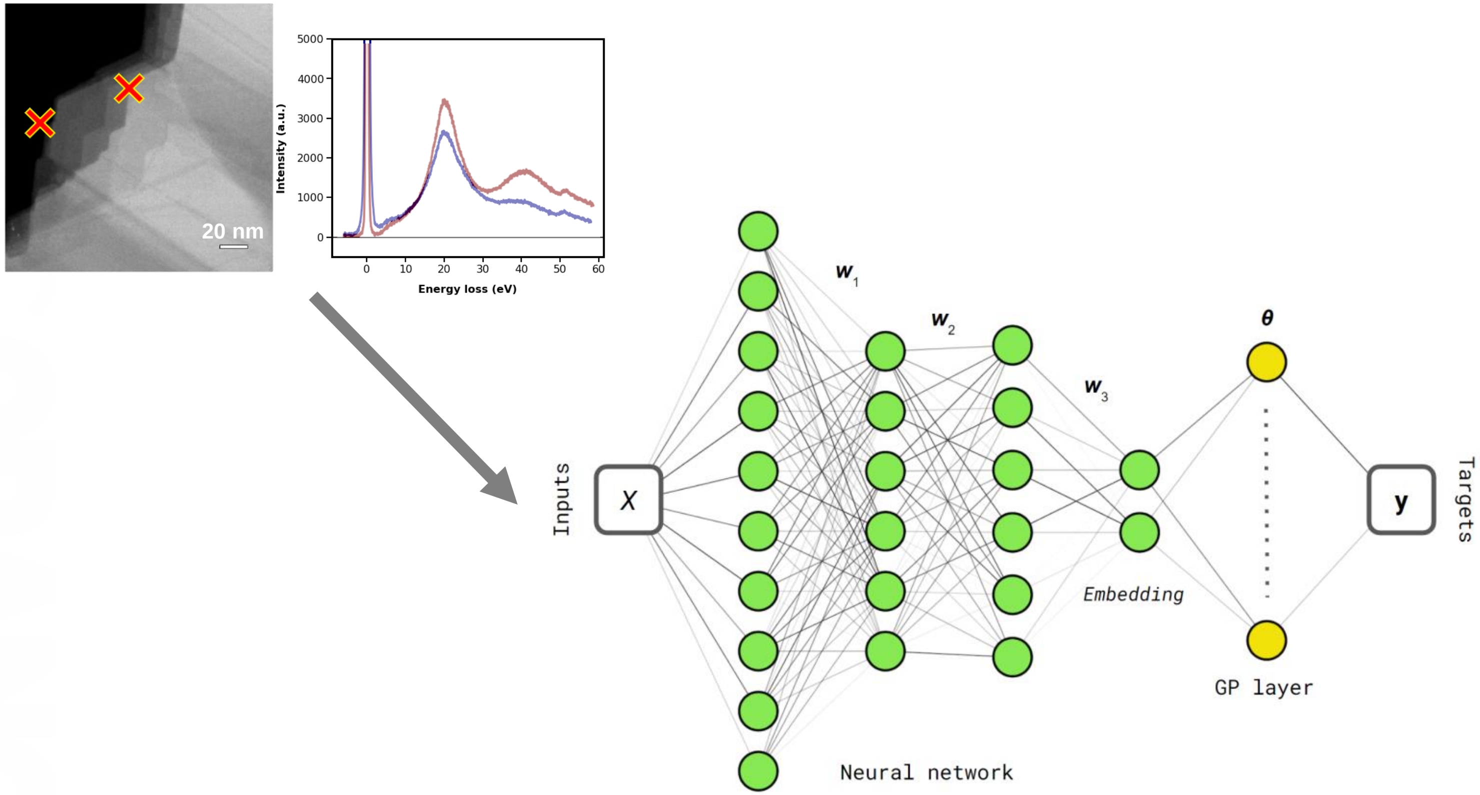


Physics search criteria:

$$\boldsymbol{Ratio} = \boldsymbol{Peak}\,\mathbf{1} \,/\, \boldsymbol{peak}\,\mathbf{2}$$



Deep Kernel Learning



<u>Specify physics criteria</u>

Acquire
structural data

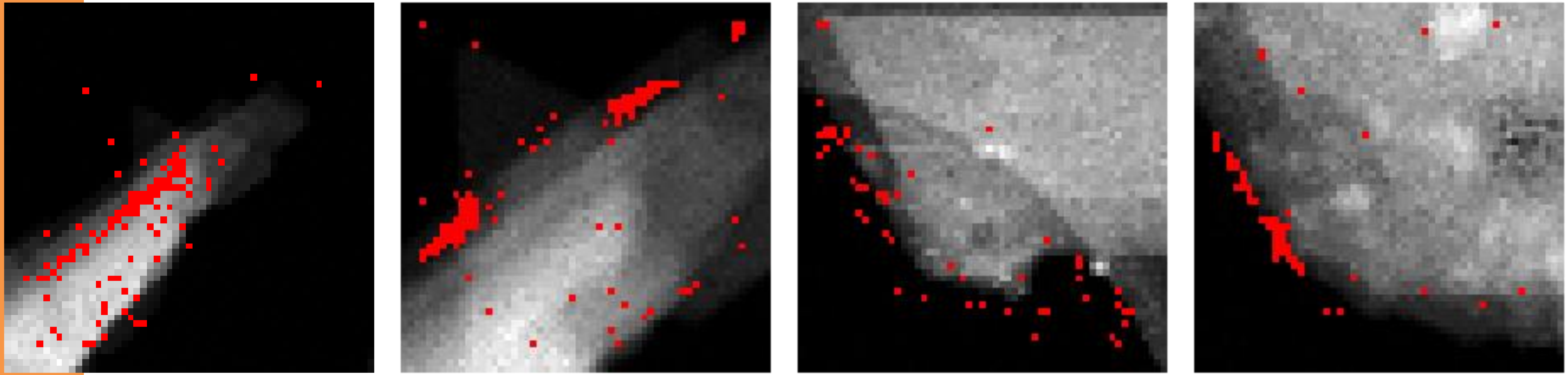
Measure a
spectrum

Train DKL
model with new
data

Decide next
position (optimize
physics criteria)

Allows navigation of the system to search for physics

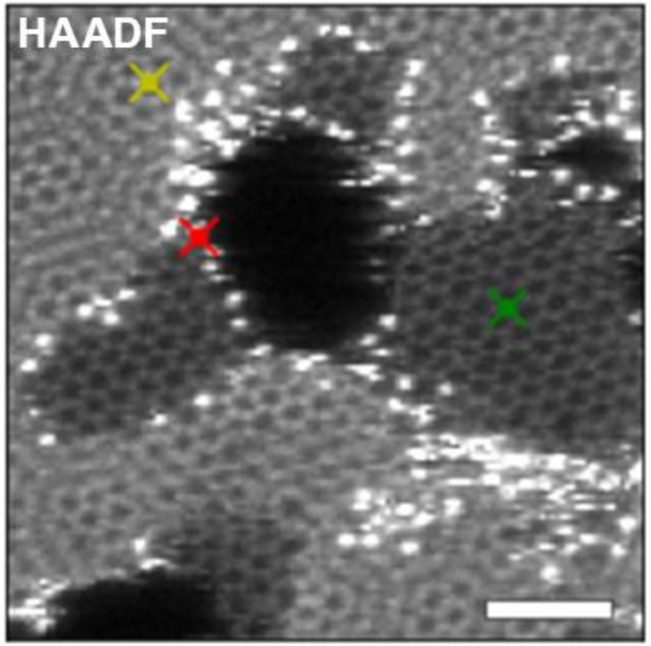
More Examples of Physics Discovery

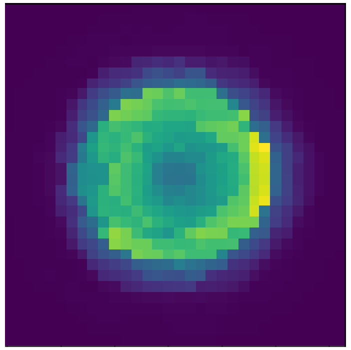


Discovery pathway depends on the reward structure (scalarizer that defines signature of physics we want to discover)!

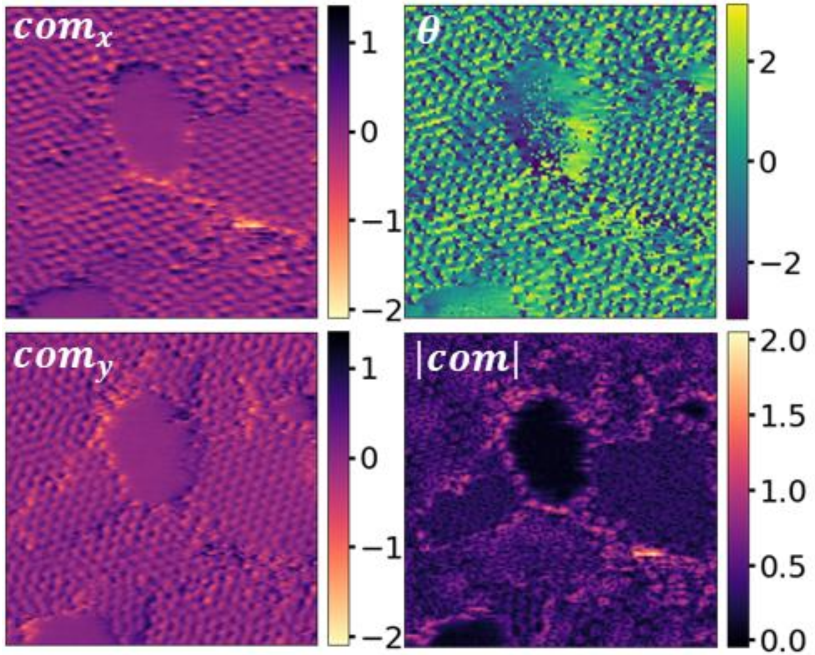
- Currently, we run 4D STEM measurements on a grid.
- What if we want to explore smarter workflows – where microscope chooses where to take 4D STEM measurements?
- **Direct:** We can do it for a priori known objects of interest
- **Inverse:** Or we can aim to discover objects which have predefined signatures of interest in 4D STEM data

4D STEM: Grid, Direct, and Inverse





Quantities to explore
- Electric field
- Potential
- Charge density
- Strain

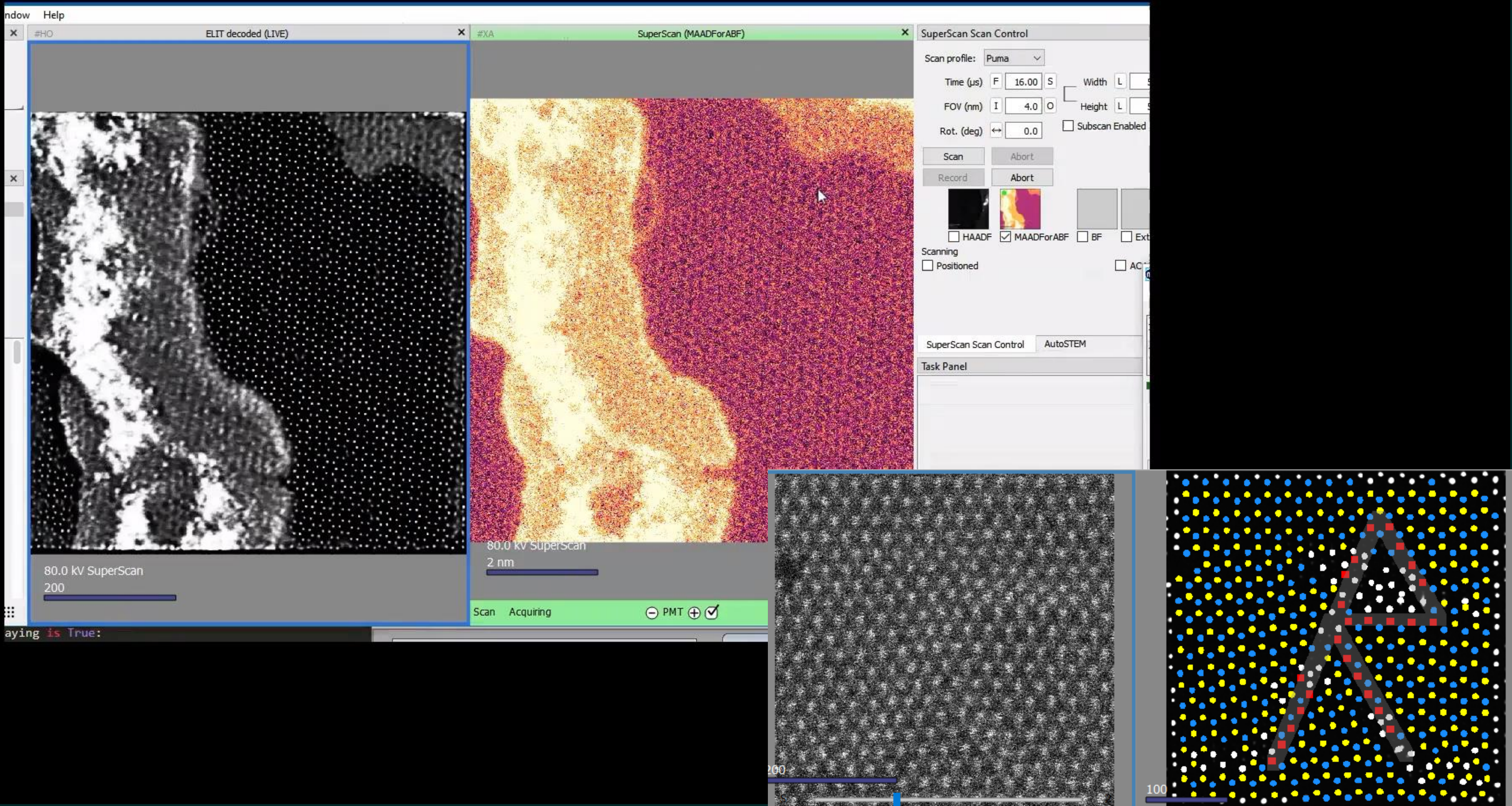


- What can we say about structure?
- Interesting functionalities are expected at the certain structural elements
- We can guess some; we have to discover others
- Multiple goals while running experiment

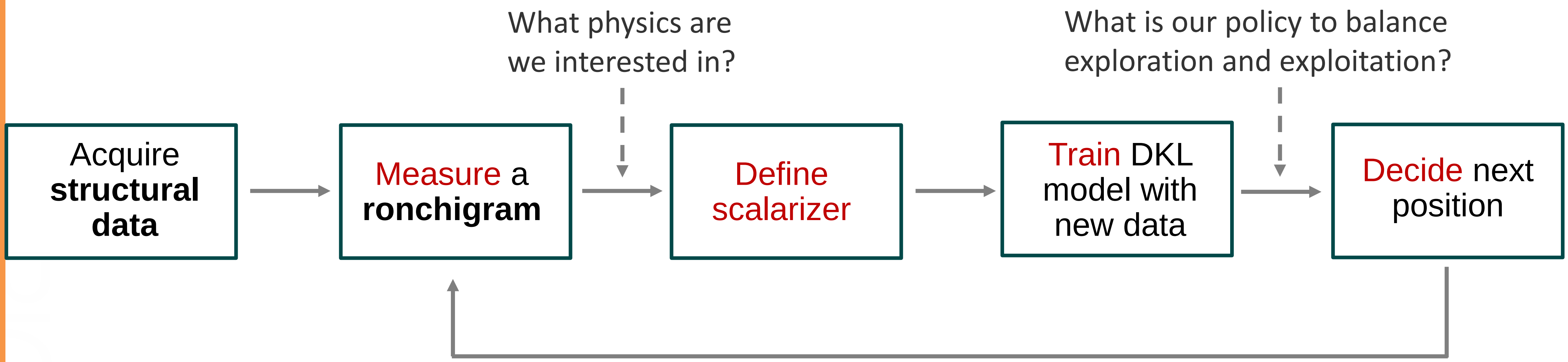
- **Policy:** **what do we do depending on observation**
- **Reward:** **what do we hope to achieve**
- **Value:** **anticipated reward**

Direct experiment: ELIT (2021)

Implementation: Kevin Roccapriore, Ayana Ghosh, Sergei V. Kalinin & Maxim Ziatdinov

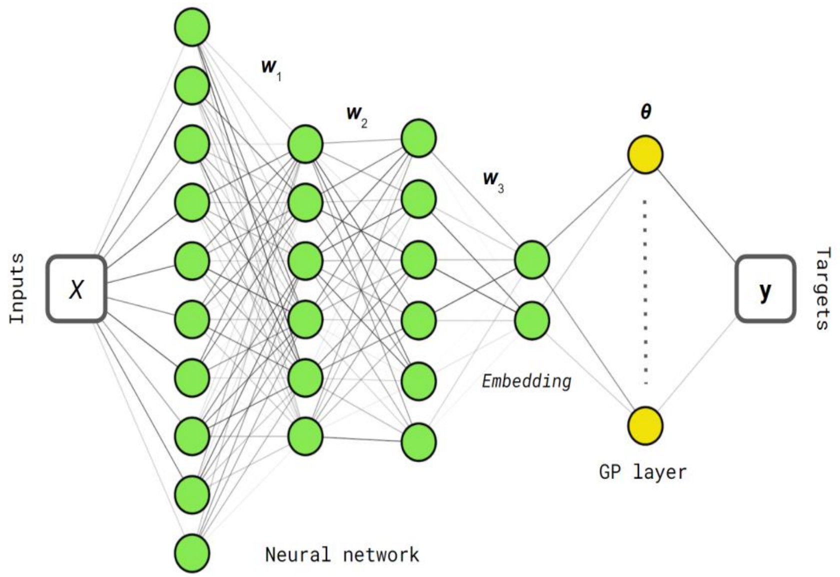


Inverse: Deep Kernel Learning based BO

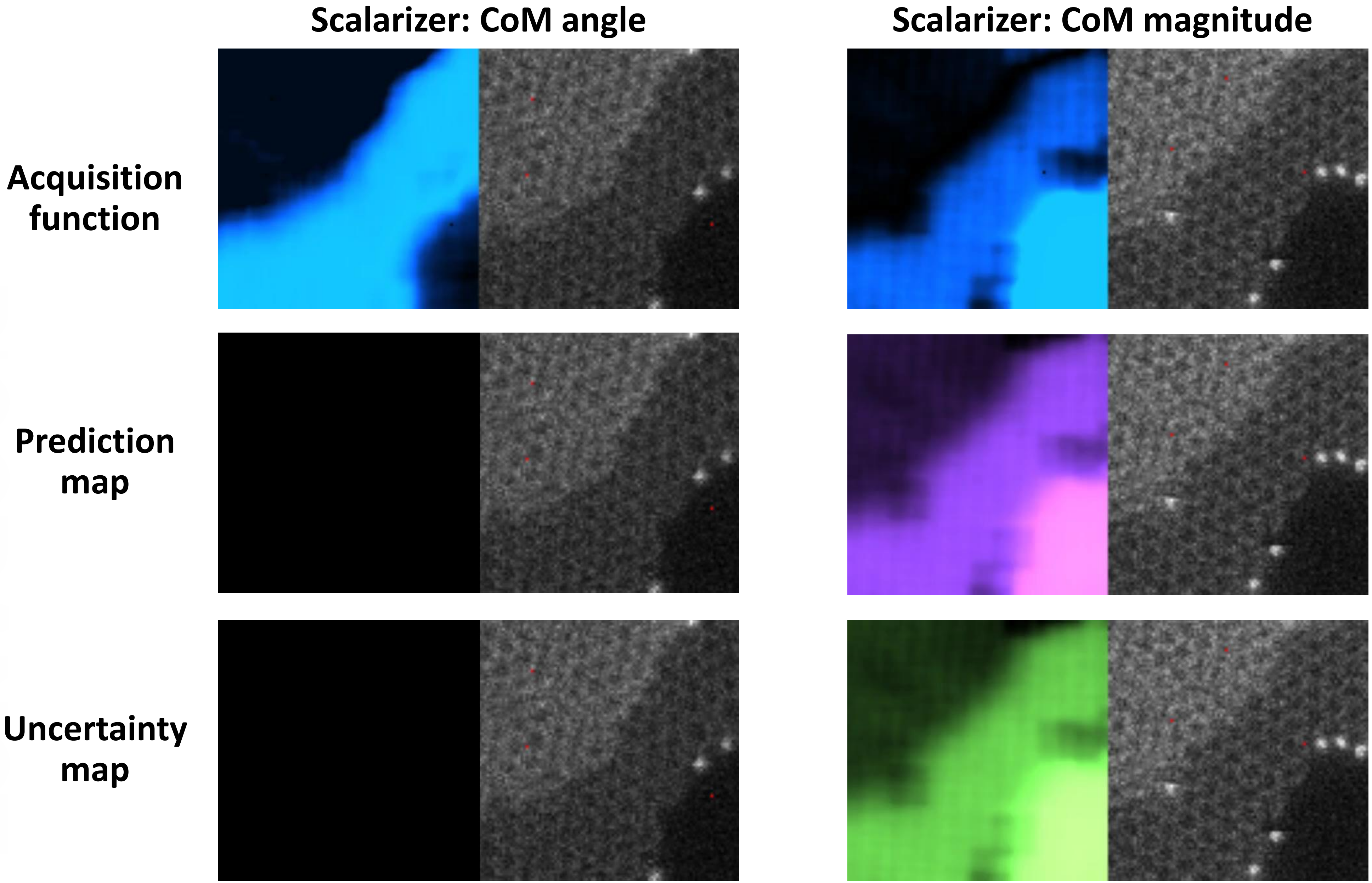


Key concepts:

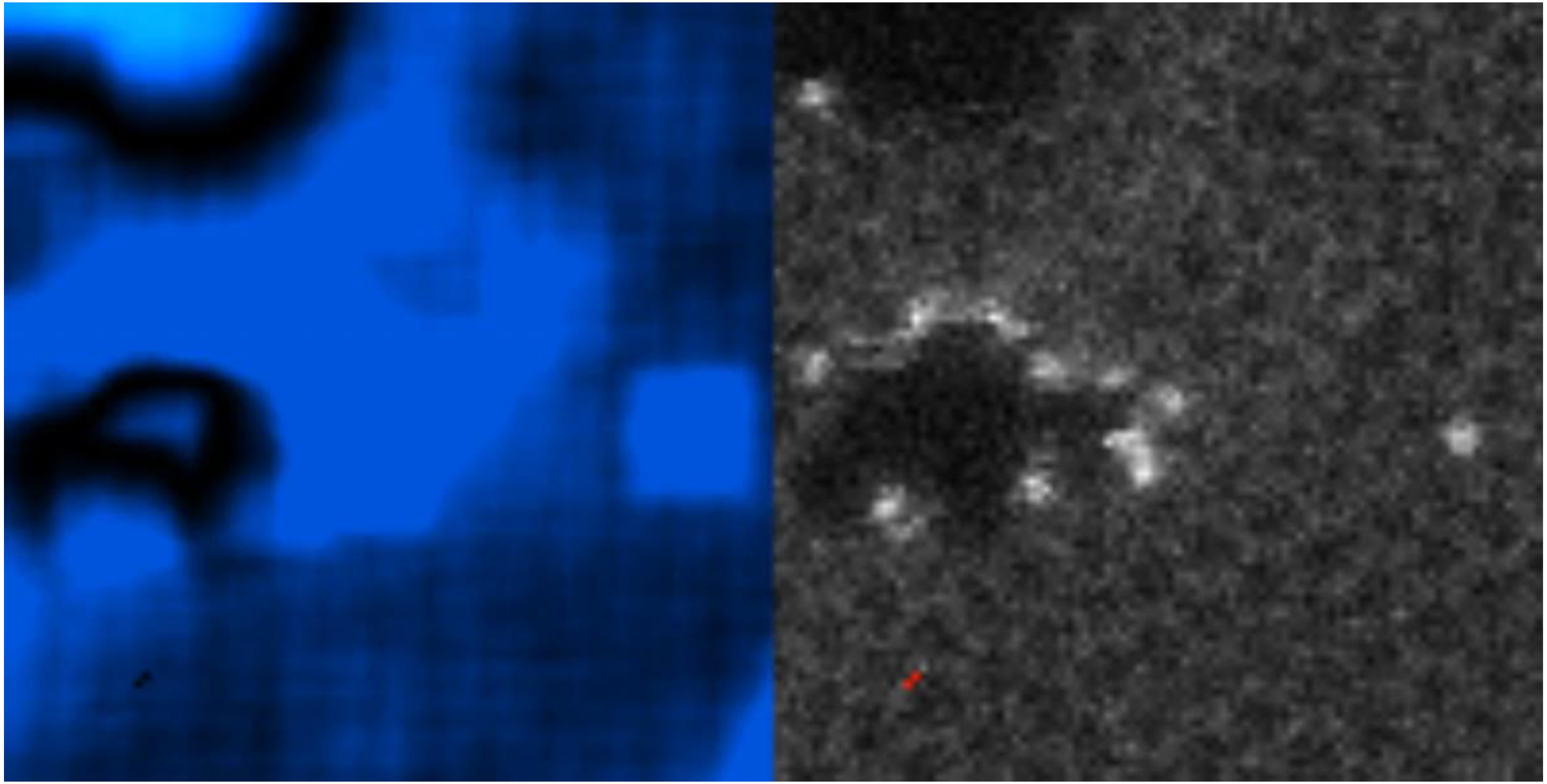
- **Scalarizer:** (any) function that transforms spectrum into measure of interest. Can be integration over interval, parameters of a peak fit, ration of peaks, or more complex analysis
- **Experimental trace:** collection of image patches and associated spectra acquired during experiment. Note that we collect spectra, not only scalarizers



DKL on pre-acquired data



DKL on Active Microscope

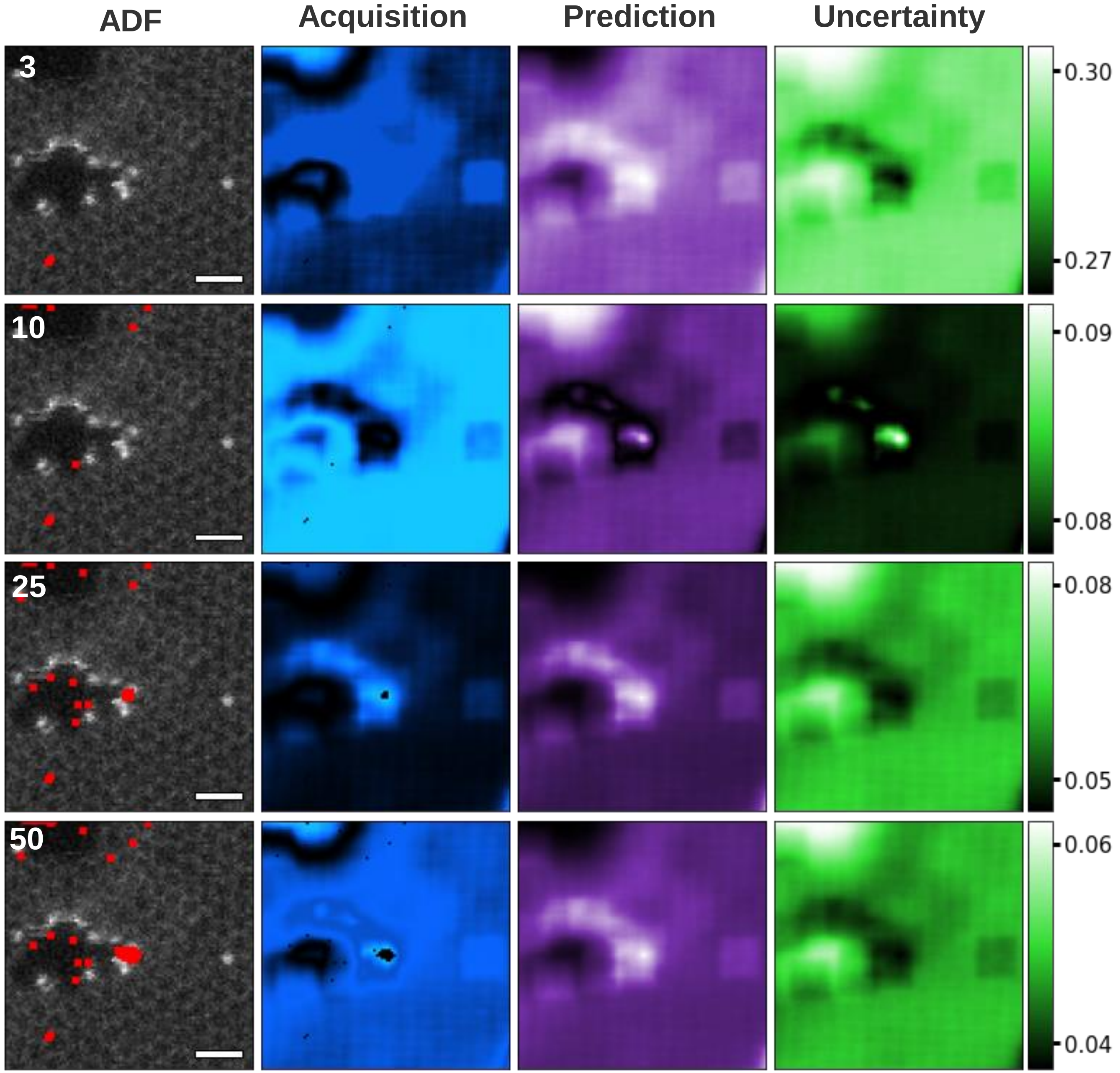


- Different **acquisition functions** can be used:
  - Expected Improvement (**EI**) (usually what was used)
  - Upper Confidence Bound (**UCB**), etc
- Usually based on some combination of **prediction** and **uncertainty**.

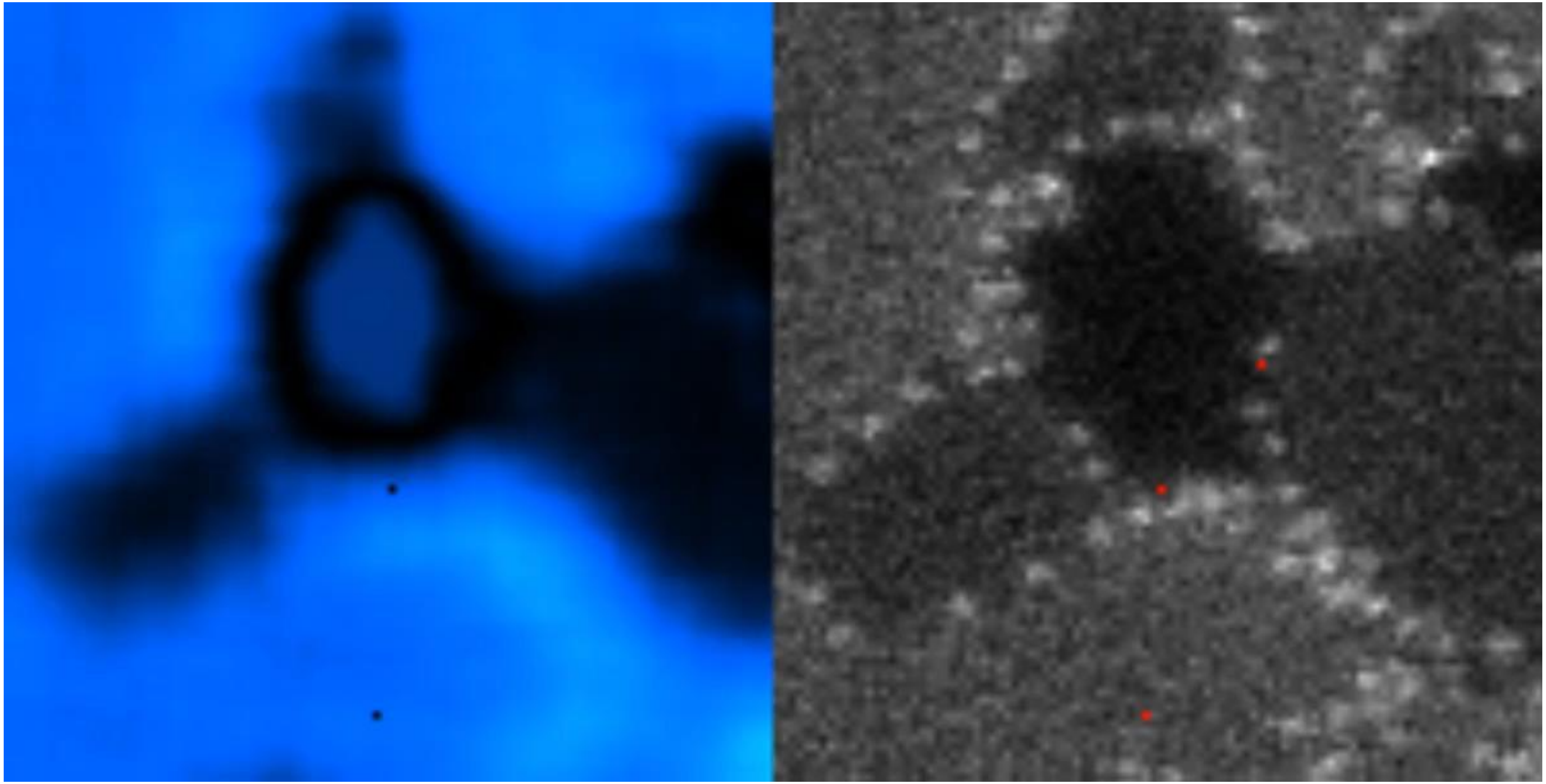
A closer look

Scalarizer: *CoM* magnitude

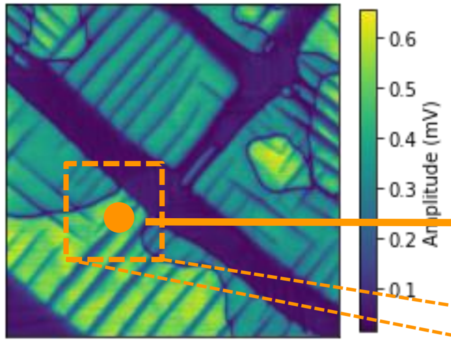
- High uncertainty @ start, but fairly quickly reduces
- Prediction actually doesn't drastically change throughout experiment
  - Structure-property relationship here is fairly rapidly learned
- Note the training can be halted after some criterion is met, making remainder of experiment go much quicker

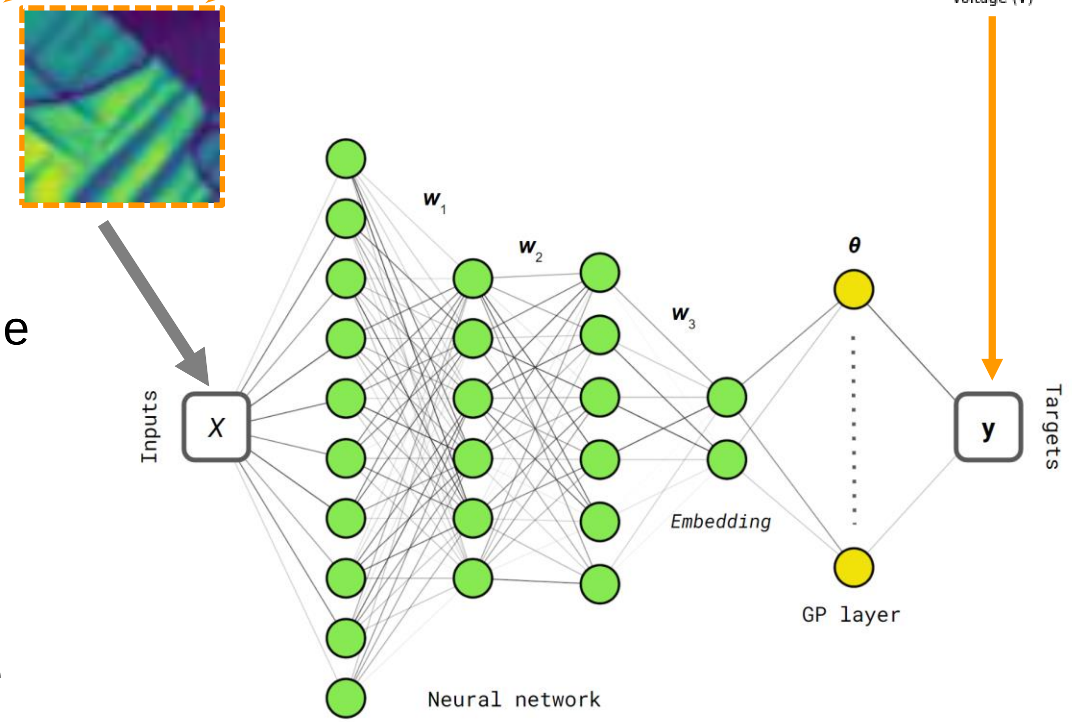


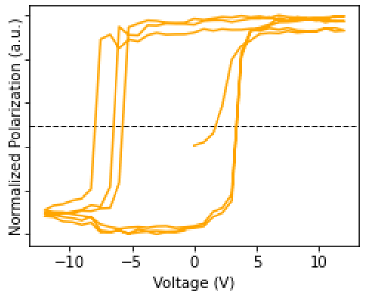
Does it always work?



Deep Kernel Learning







Specify physics criteria

Acquire **structural data**

Measure a **spectrum**

Train DKL model with new data

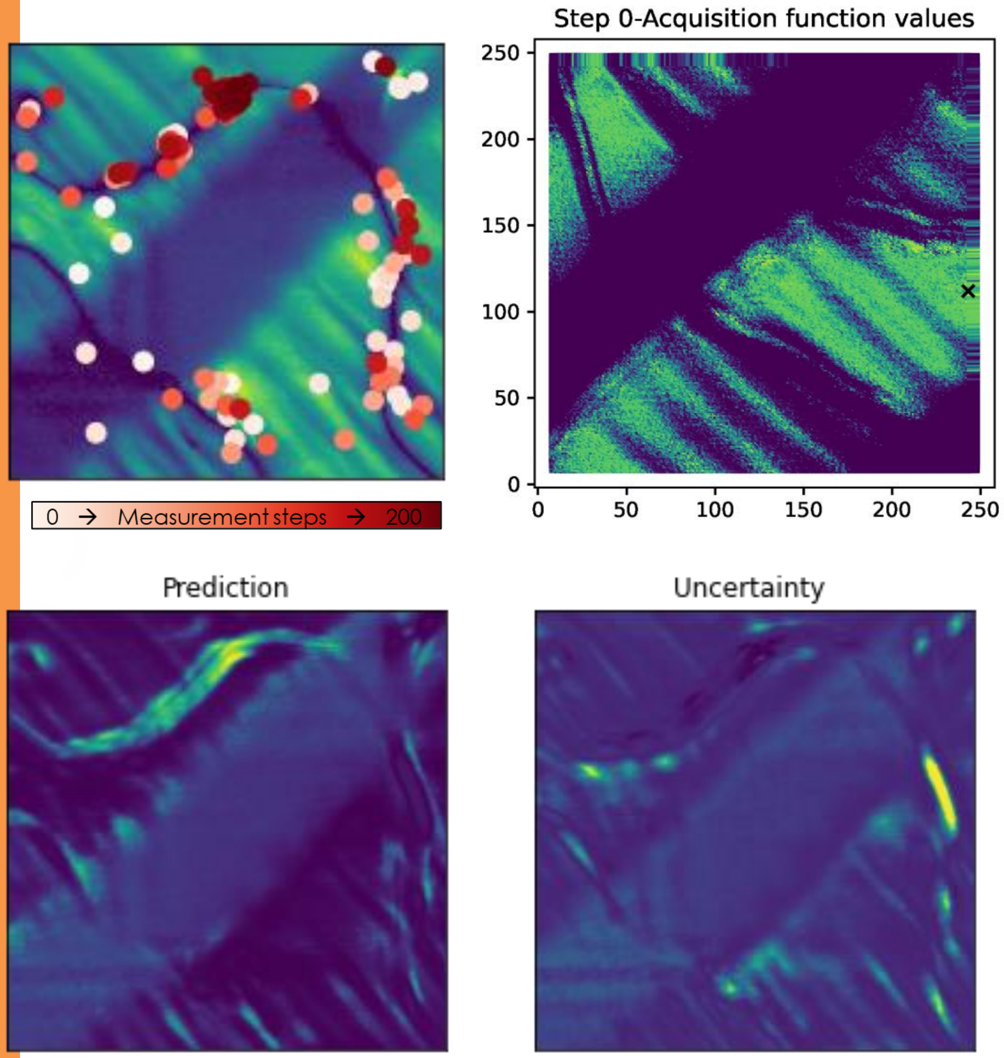
Decide next position (optimize physics criteria)

- All patches are available in the beginning
- Spectra are made available sequentially
- We define what feature in spectrum are we interested in

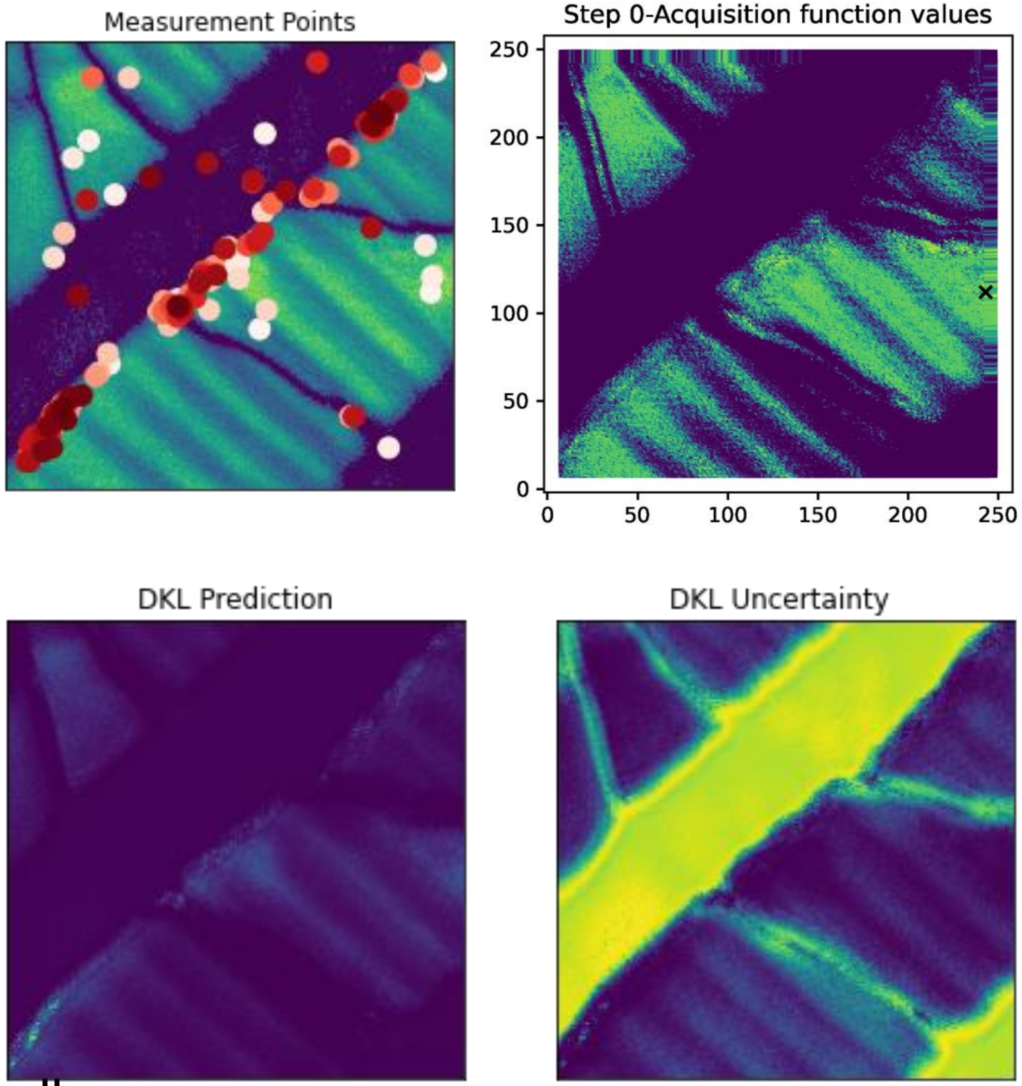
Allows navigation of the system to search for physics

Deep Kernel Learning SPM

Guided by: On field loop area



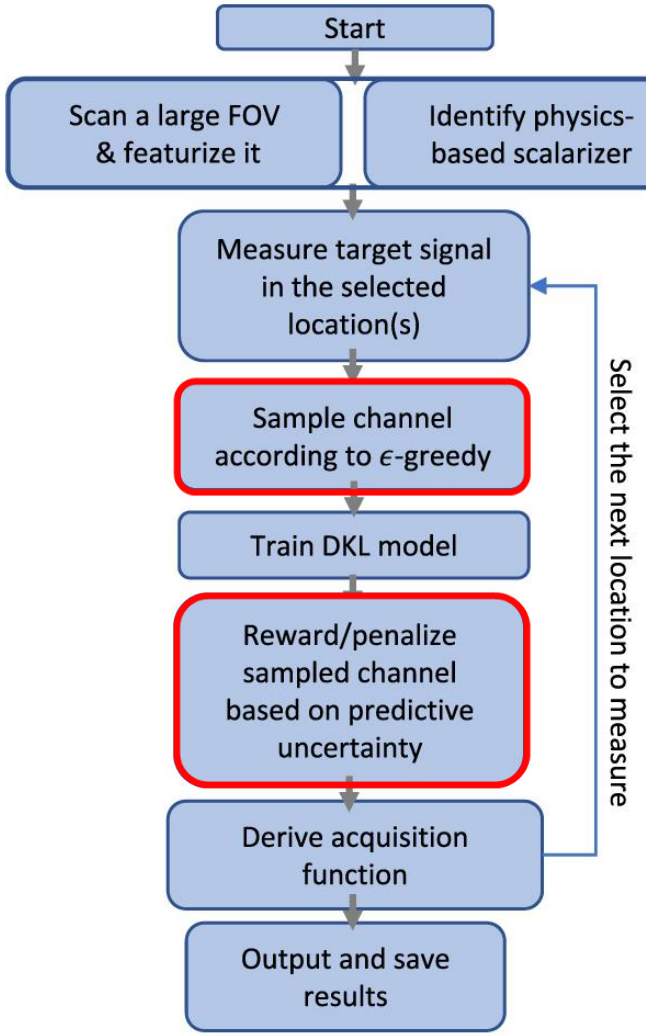
Guided by: Off field loop area

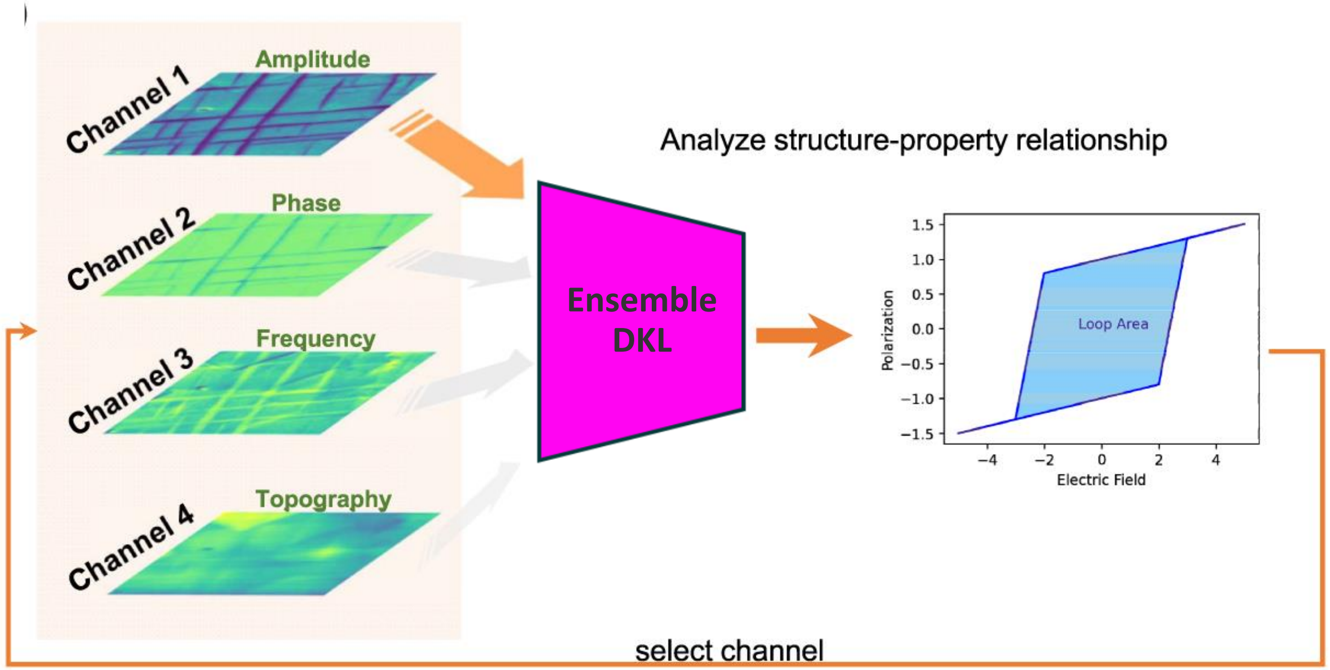


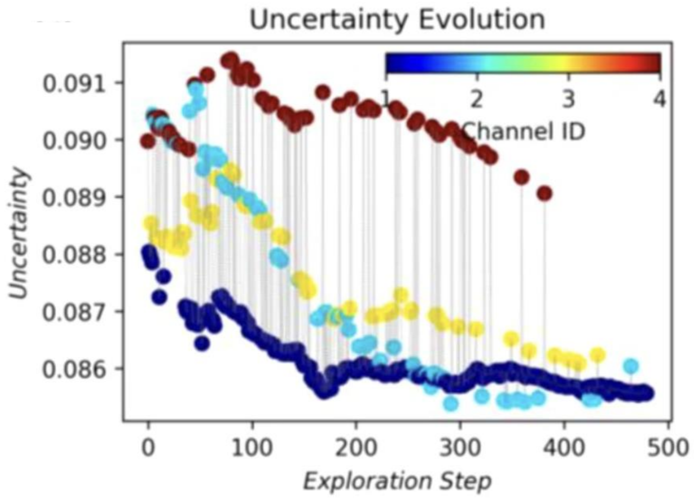
- ➢ Large loop opening corresponding 180º domain walls
- ➢ This behavior can be attributed to the large polarization mobility of 180º walls

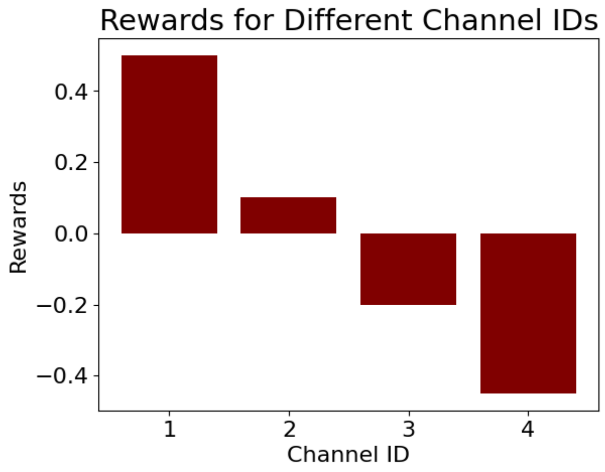
Liu, Yongtao, et al, Nature Machine Intelligence 4, 4 (2022): 341-350.

DKL: Learning the best channel



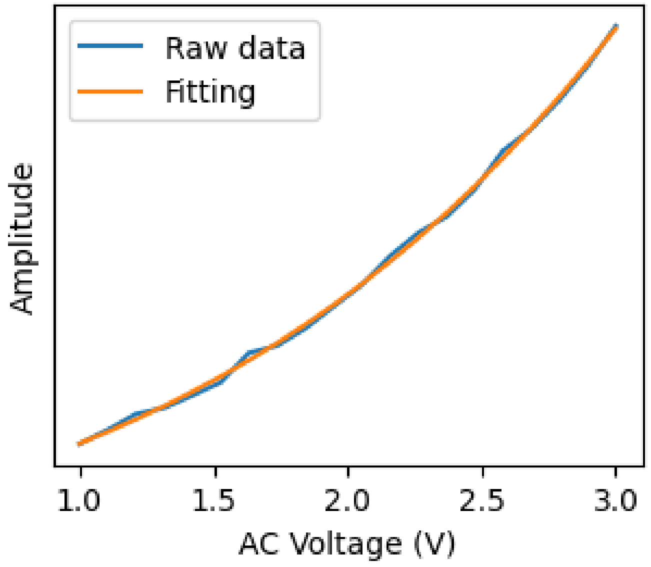




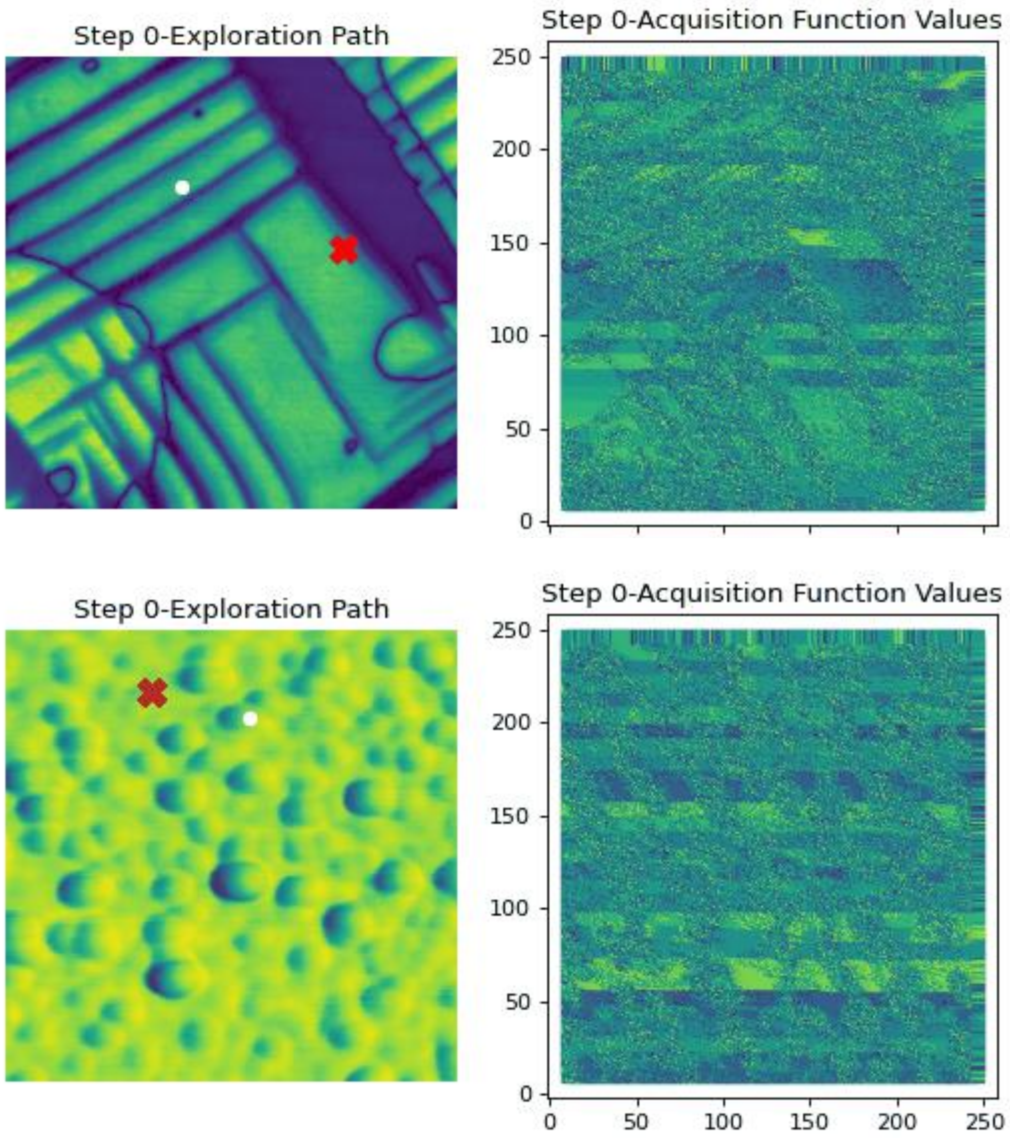


Y. Liu et al. npj Comput Mater **9**, 34 (2023).

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!

- In conventional microscopy experiment, human runs everything directly – defines scan, positions the probe, defines measurement parameters.

- In AE SPM, the policies are defined before the experiment and do not change. Sometimes it works – but not always.

- How would we:
  - (a) explain the AE progression after the experiment and
  - (b) control it during the experiment ?

NESSEE KNOXVILLE

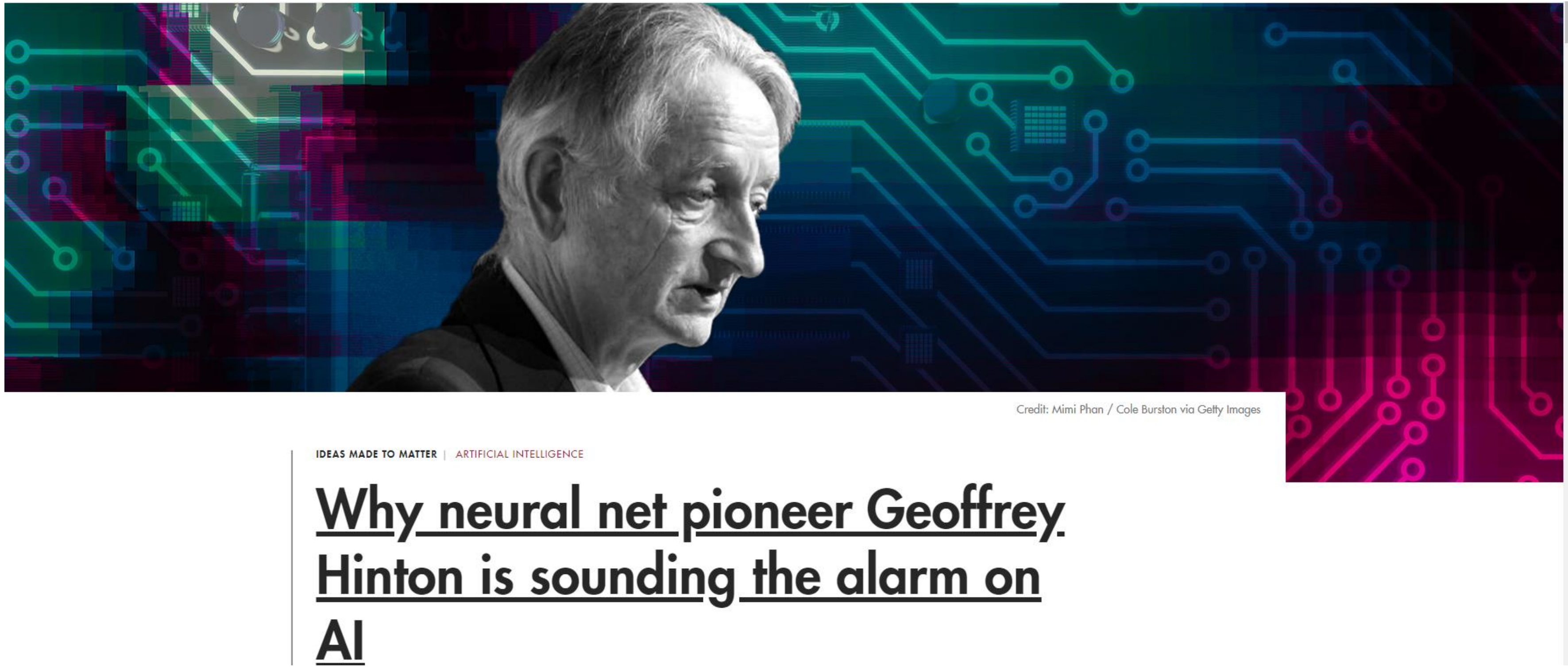
Taking the Human Out of the Loop: A Review of Bayesian Optimization

Citation

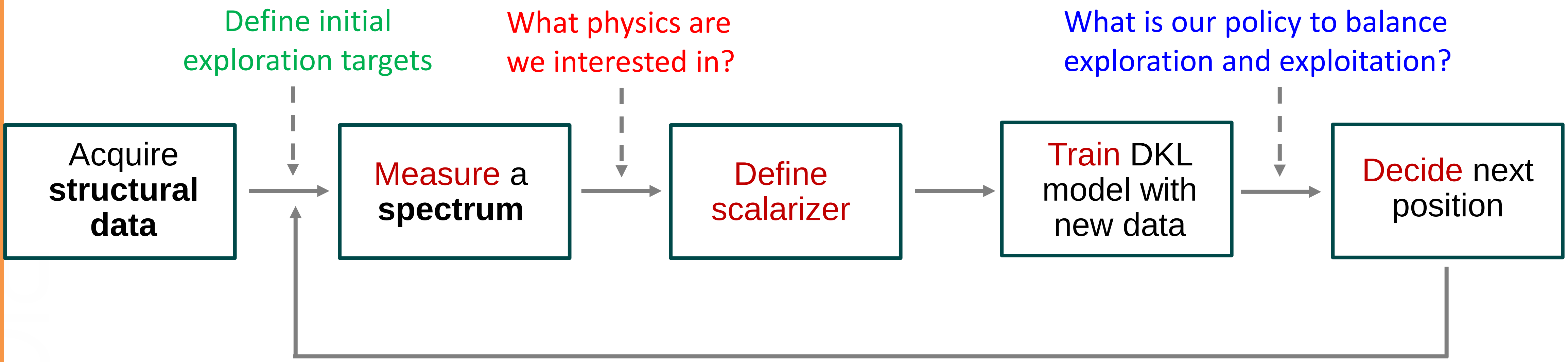
Shahriari, Bobak, Kevin Swersky, Ziyu Wang, Ryan P. Adams, and Nando de Freitas. 2016. "Taking the Human Out of the Loop: A Review of Bayesian Optimization." Proc. IEEE 104 (1) (January): 148–175. doi:10.1109/jproc.2015.2494218.

Published Version

doi:10.1109/JPROC.2015.2494218



Bringing Human into the Loop



Key concepts:

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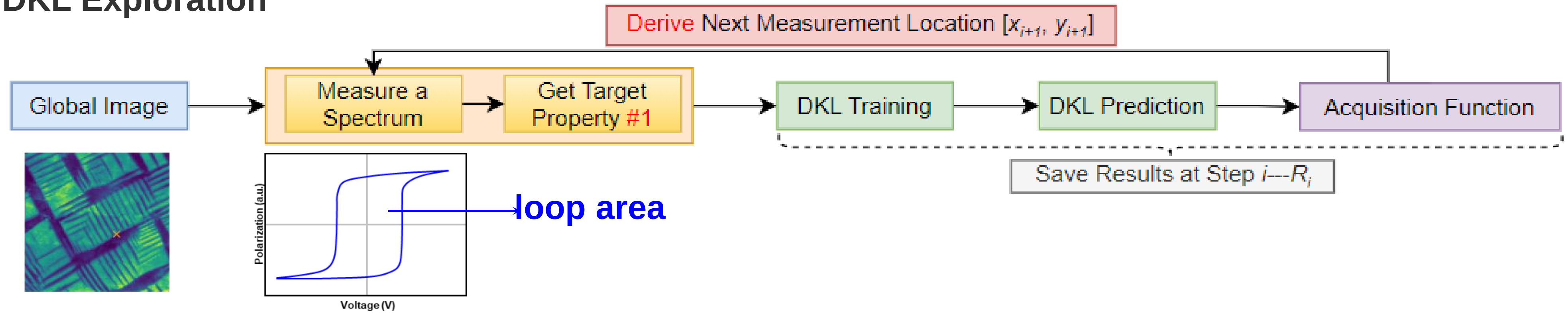
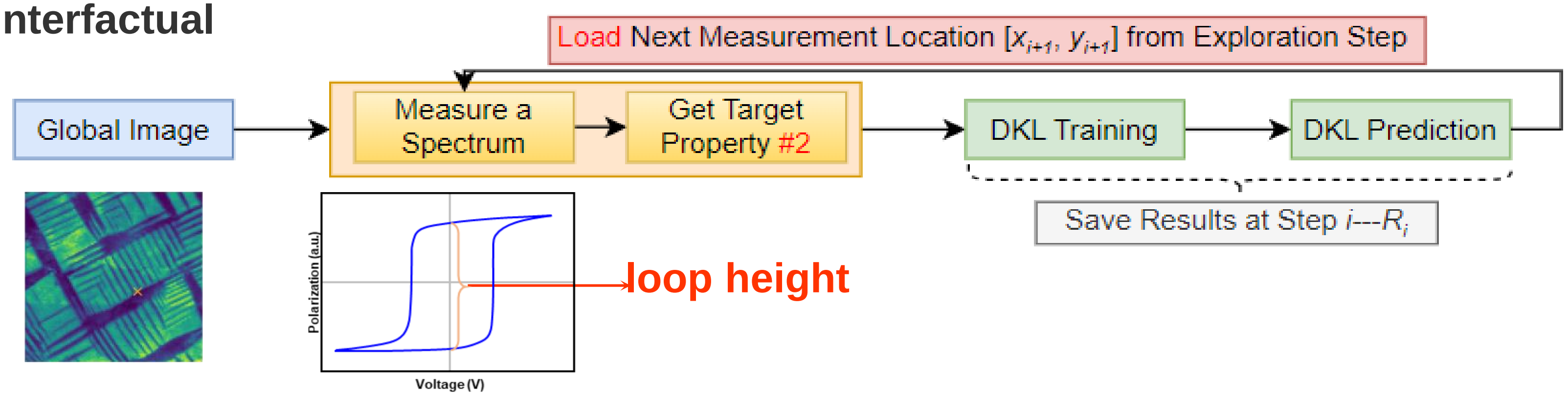
Counterfactual scalarizers

DKL Exploration



DKL Counterfactual



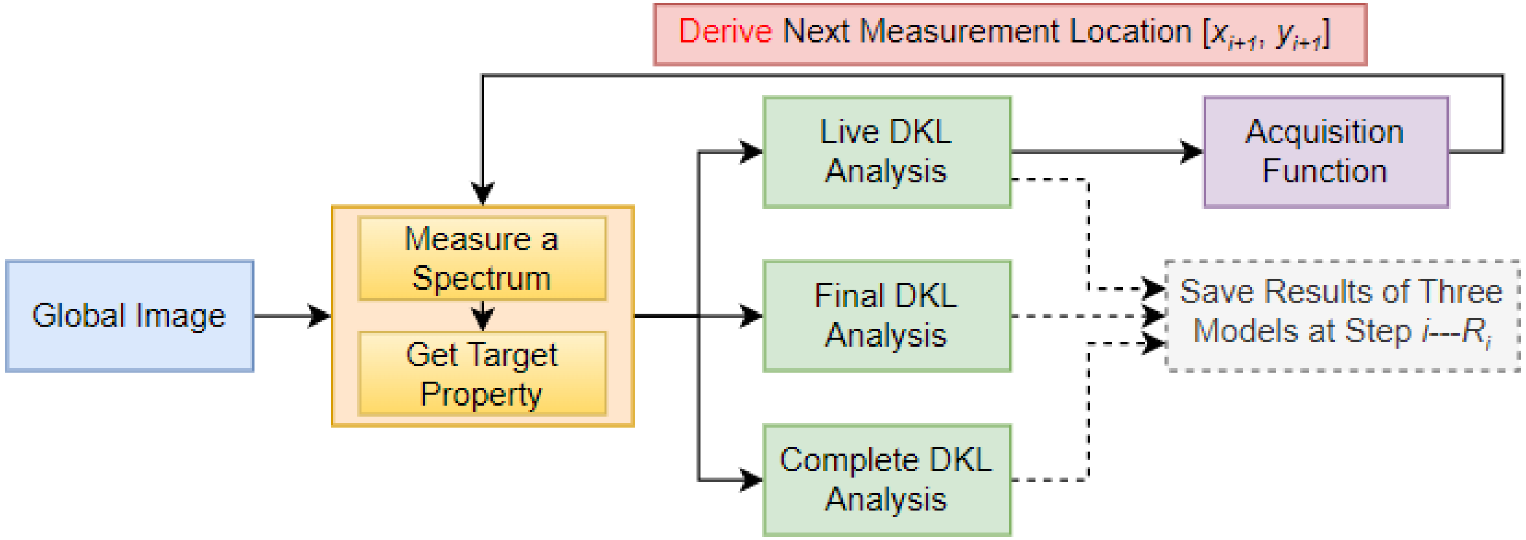
Target properties:

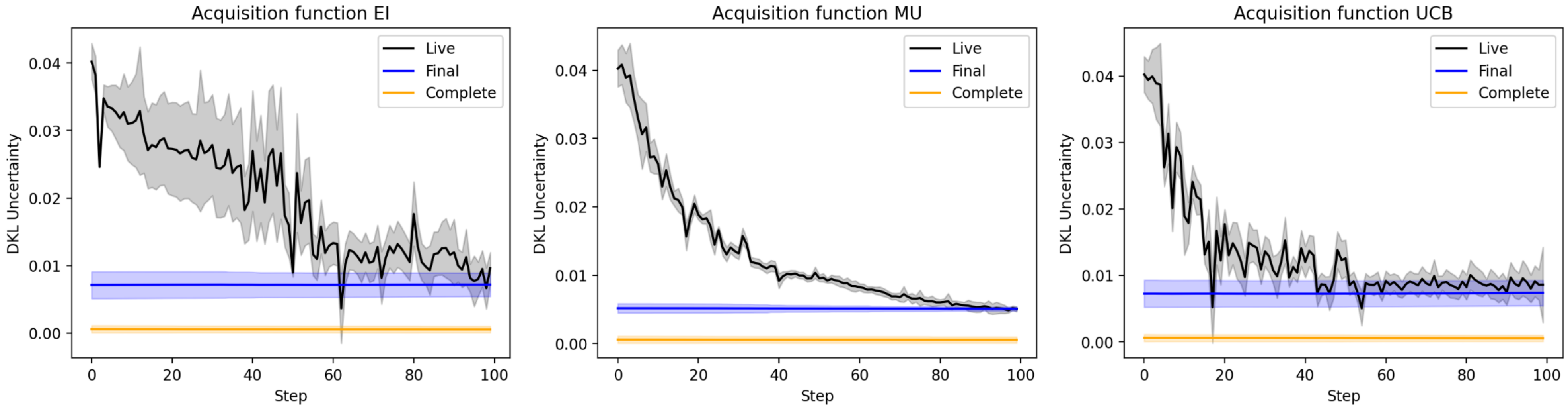
1. Loop Area
2. Loop Height
3. Coercive Field
4. …

- We save the full experimental trace
- What if we follow the actual experimental path – but calculate alternative (counterfactual) scalarizers?

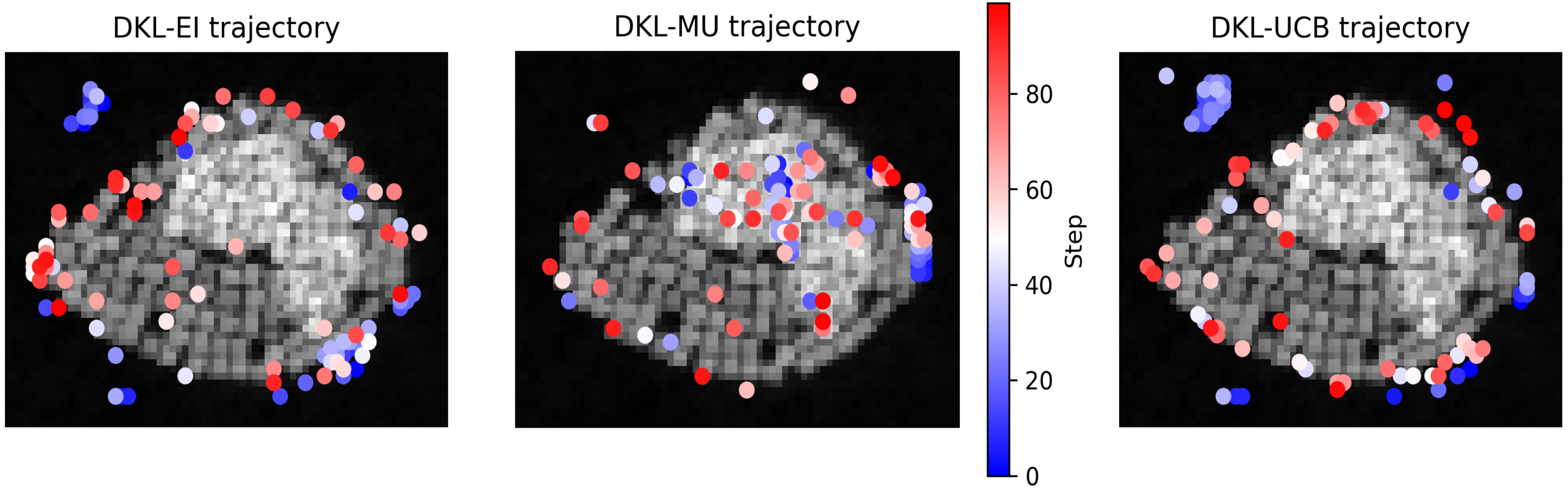
Explainable AE

- During the AE, model learns structure-property relationships.
- What if we retrace the experimental steps – using the fully trained model?



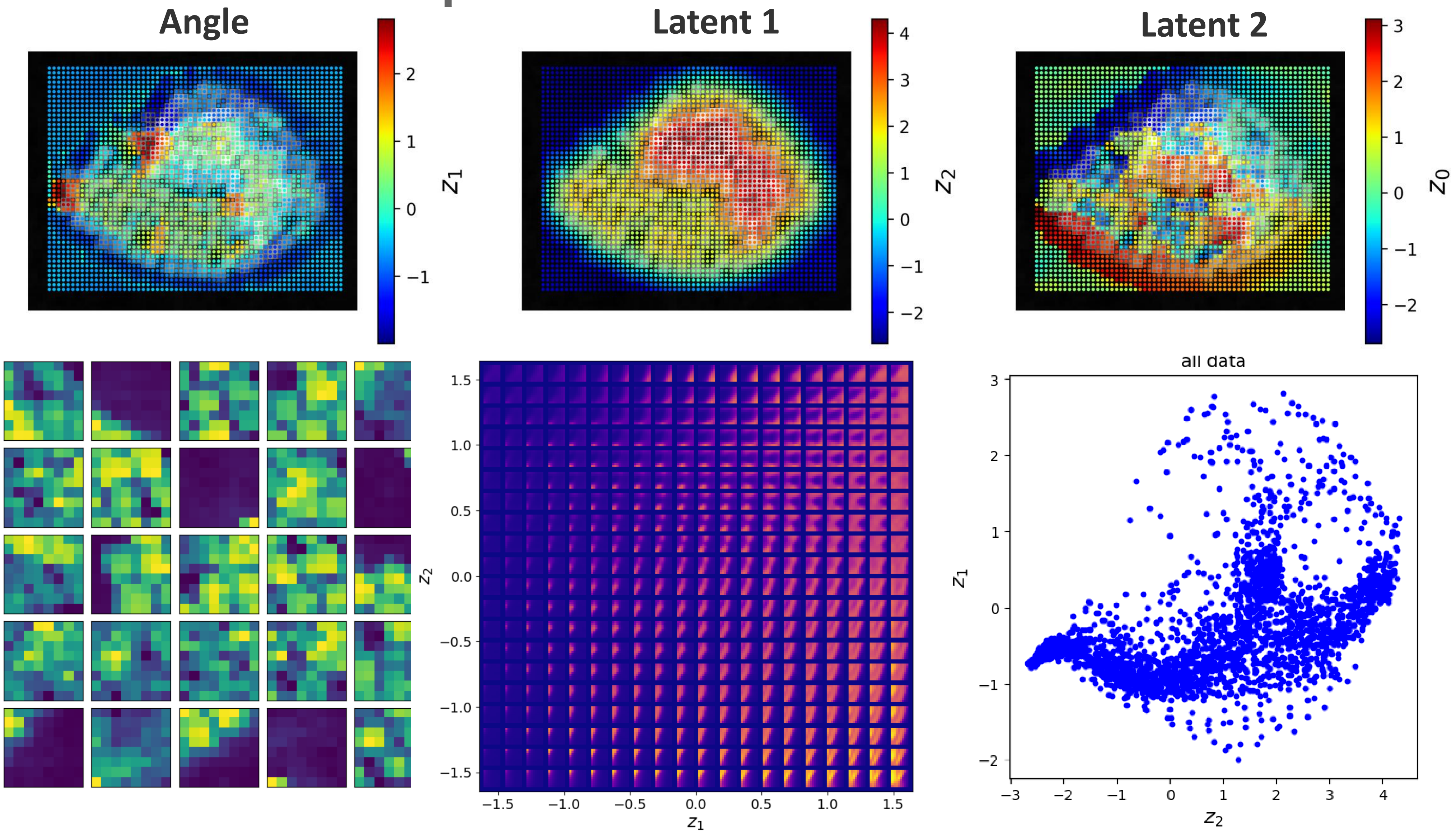


Monitoring the AE



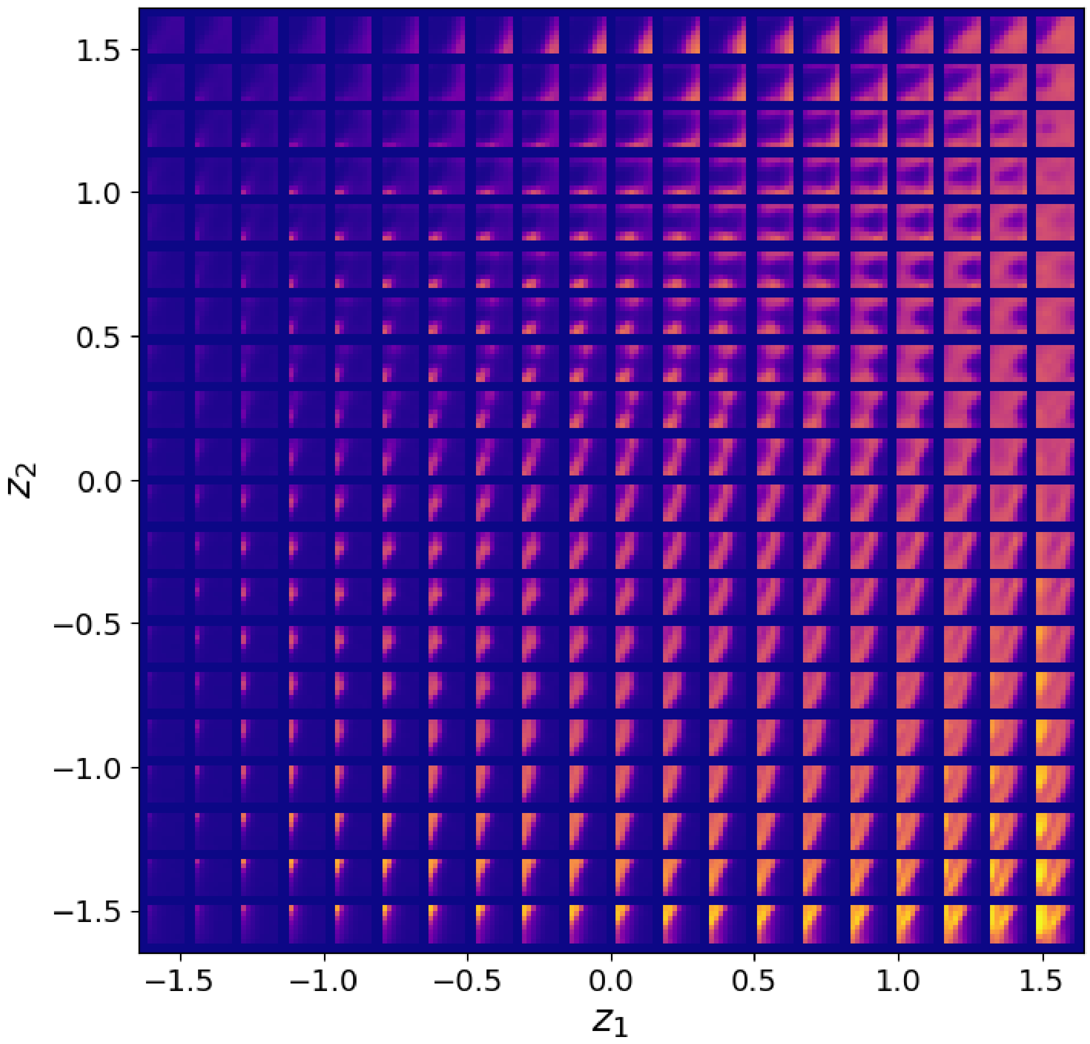
- Different acquisition functions (policies) give different experimental paths for AE
- Can we analyze what is special about points visited?

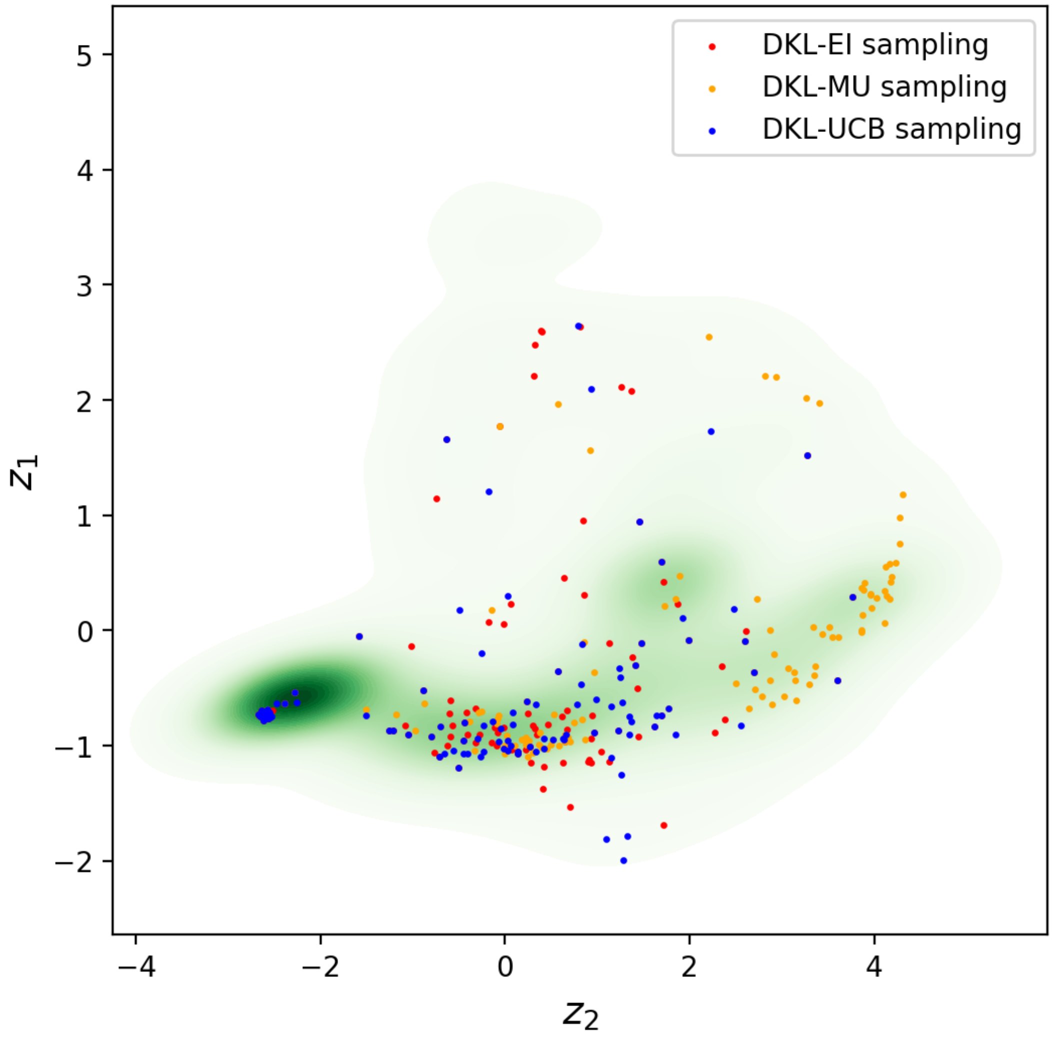
Global Feature Space



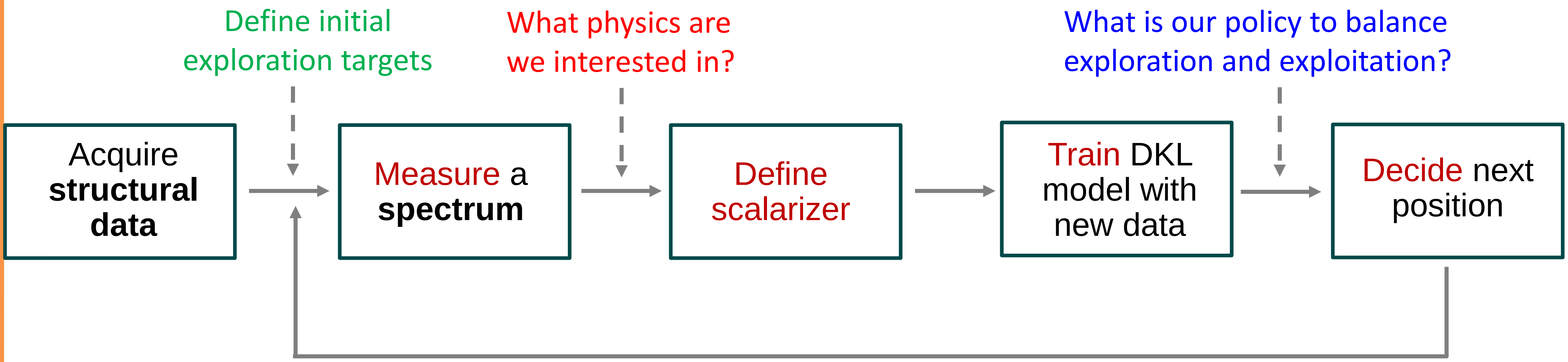
- Global feature space is available from the beginning of the experiment and is stationary
- Experimental trajectories can be visualized in the global feature space

VAE approach: full feature space





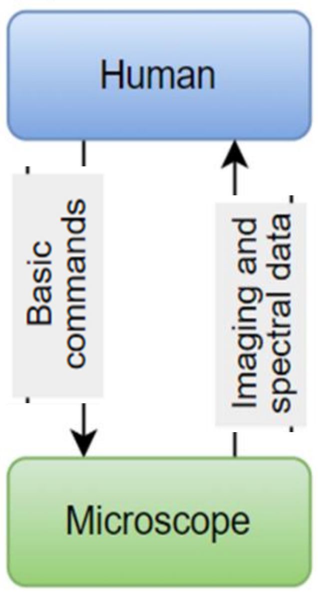
Bringing Human into the Loop

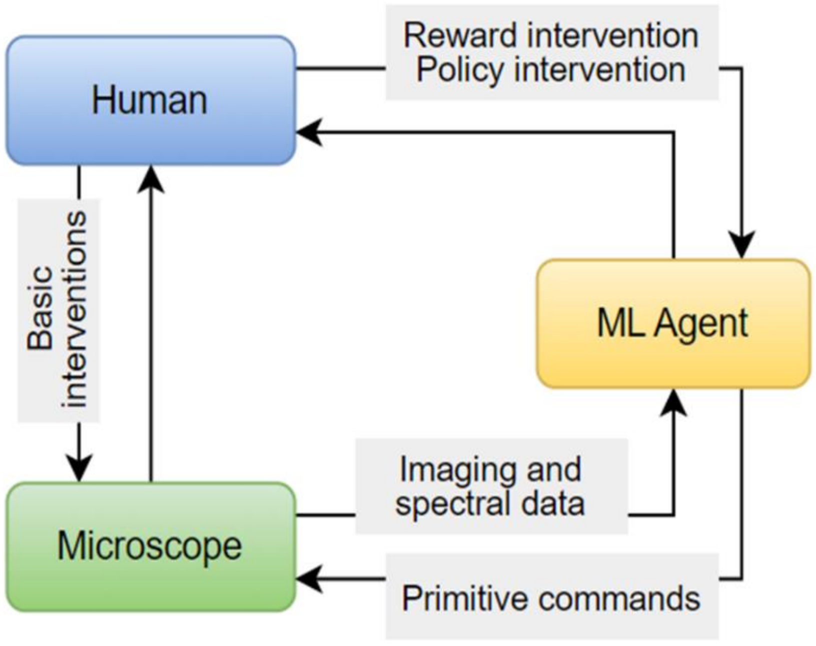


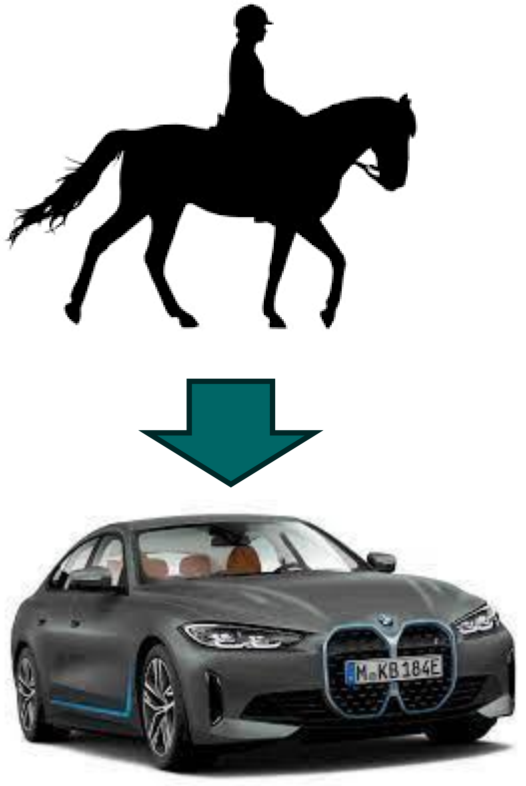
We can intervene on:

- **Policies** (acquisition functions): type and parameters
- **Scalarizers:** what physics are we interested in - type and parameters
- **Knowledge injection:** what microstructures are we interested in?
- **Cost and latencies:** trivial via acquisition functions

Human in the loop AE



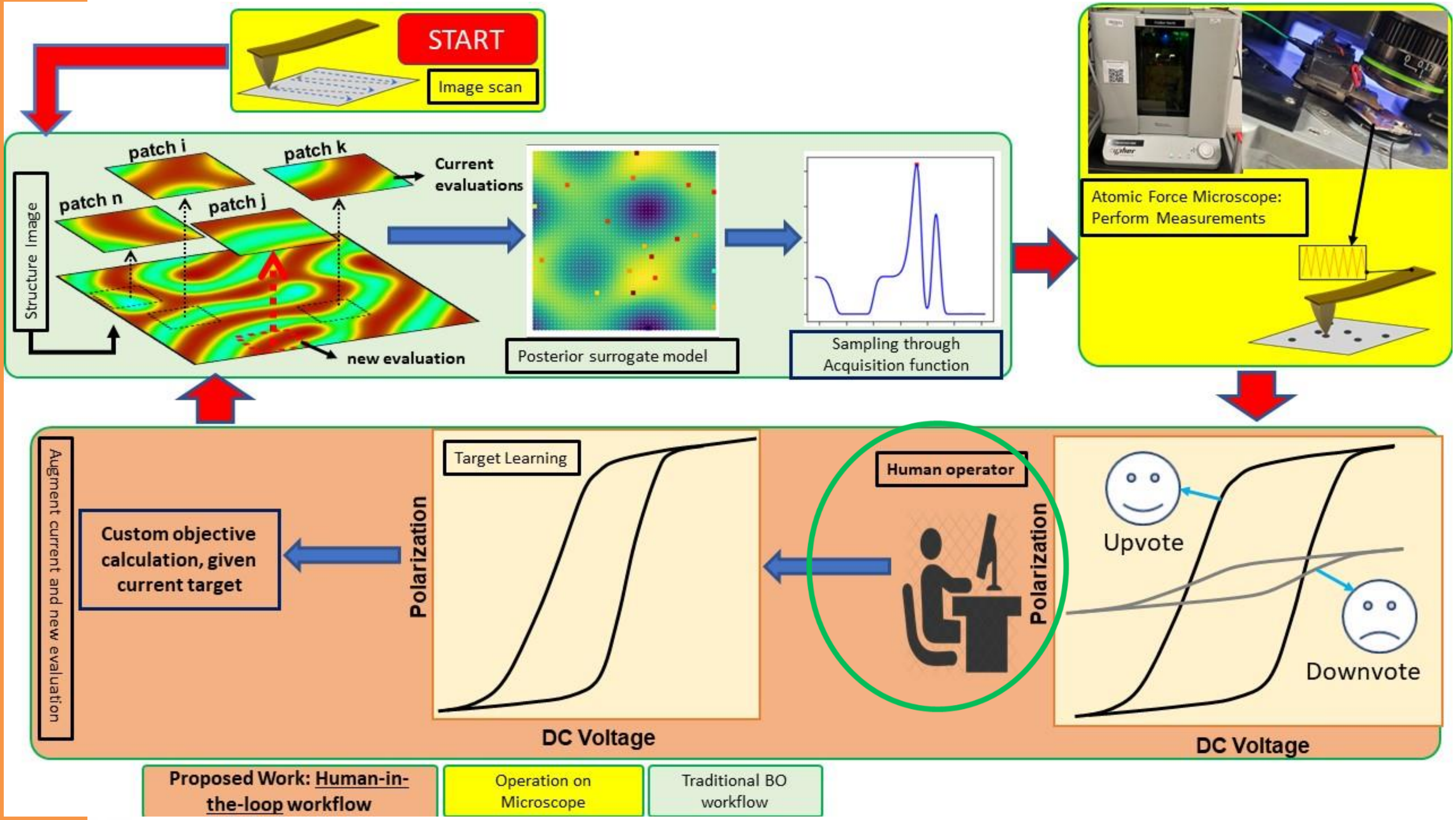




We can intervene on:

- Policies (acquisition functions): type and parameters
- Scalarizers (physics descriptors): type and parameters
- Knowledge injection
- Direct operation

BOARS: Human (partially) in the loop



Bayesian optimized Active Recommender System (BOARS)

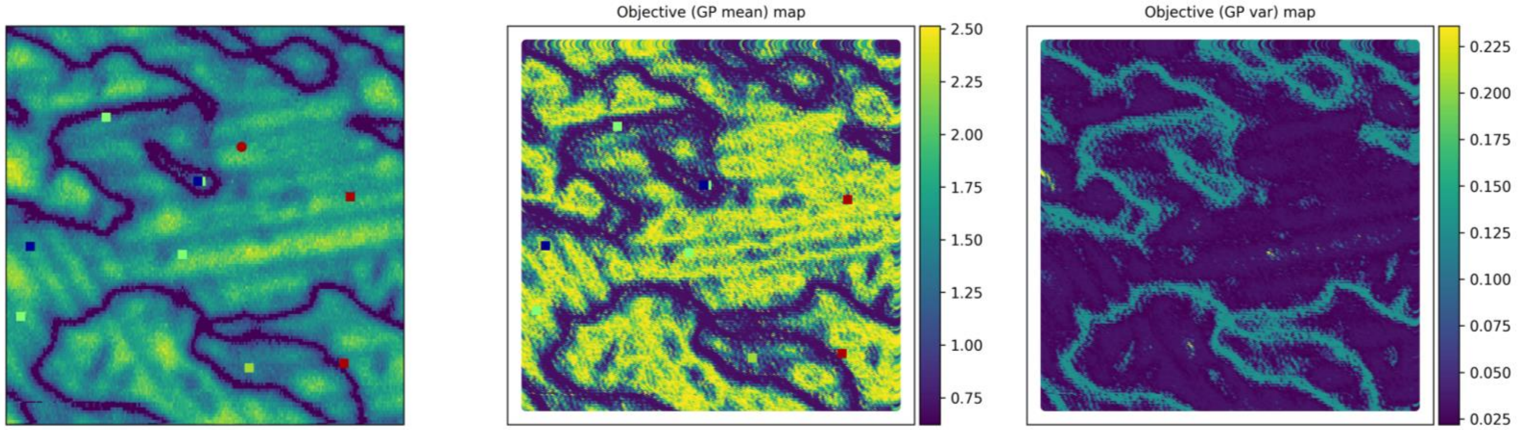
Arpan Biswas

BOARS: Human partially in the loop

Human Assessment within loop

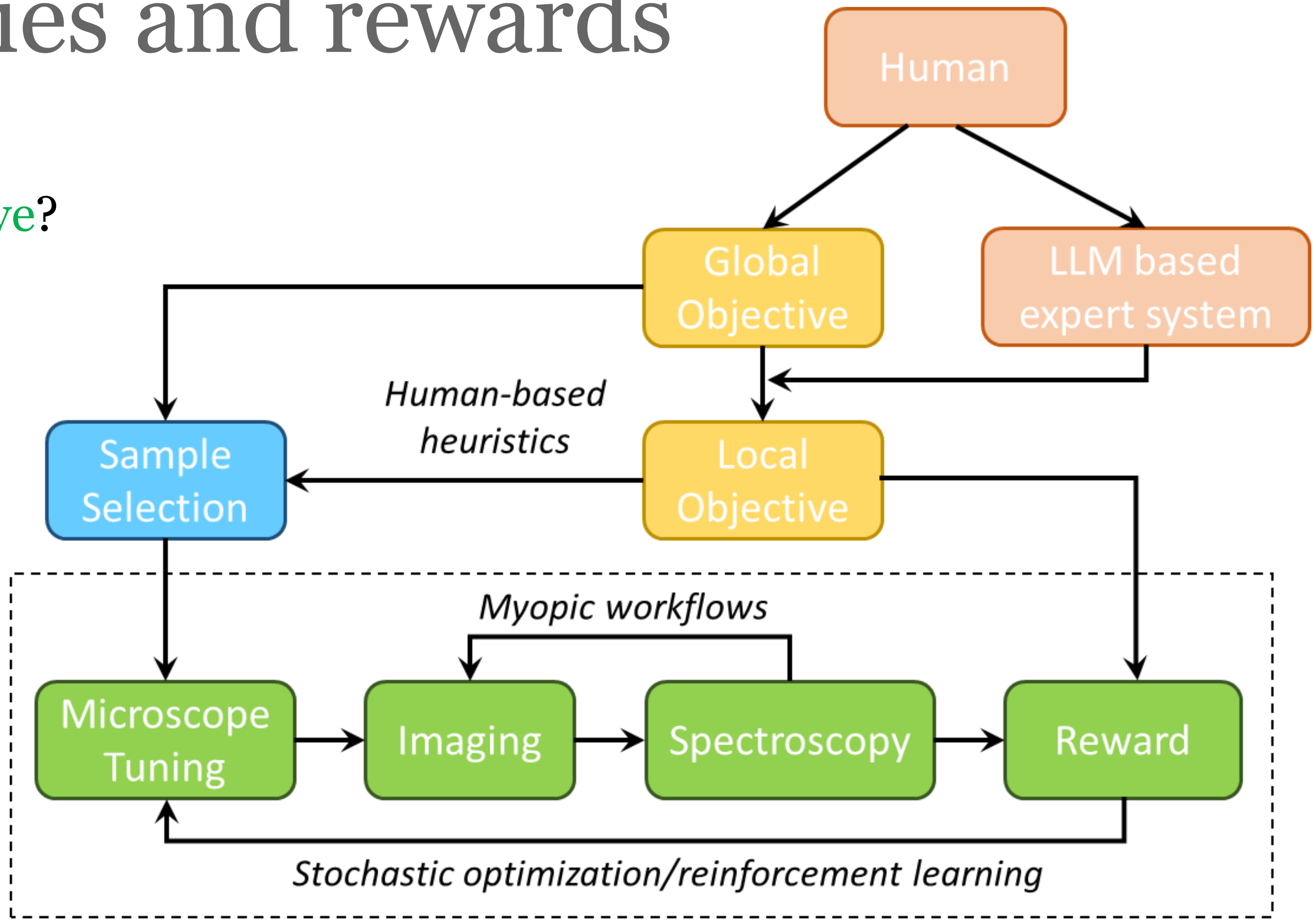
```
    return ampdat_masked, points_measured

# set parameters
IV = np.copy(amp_masked)
points_measured = np.array(idx)
last_points_measured = np.array(points_measured)
vdc = vdc_vec

train_Y = torch.empty((num_start, 1))
pref = torch.empty((num_start, 1))
init_spec = torch.empty((num_start, spec_length))
# Define a sparse grid to store evaluated spectral locations
eval_spec_y = torch.zeros(img.shape[0],img.shape[0],spec_length)
#Evaluate initial training data
x = torch.empty((1,2))

# First generate target loop, based on initial training data
wcount_good= 0
target_func = torch.zeros(spec_length)

mask = np.isin(points_measured, last_points_measured, invert = True)
new_points_measured = points_measured[mask]
last_points_measured = np.append(last_points_measured, new_points_measured)

for i in range(0, num_start):

    ##*****************experiment start*****************##
    time.sleep(0.1)
```

An implementation to SPM



The dance of policies and rewards

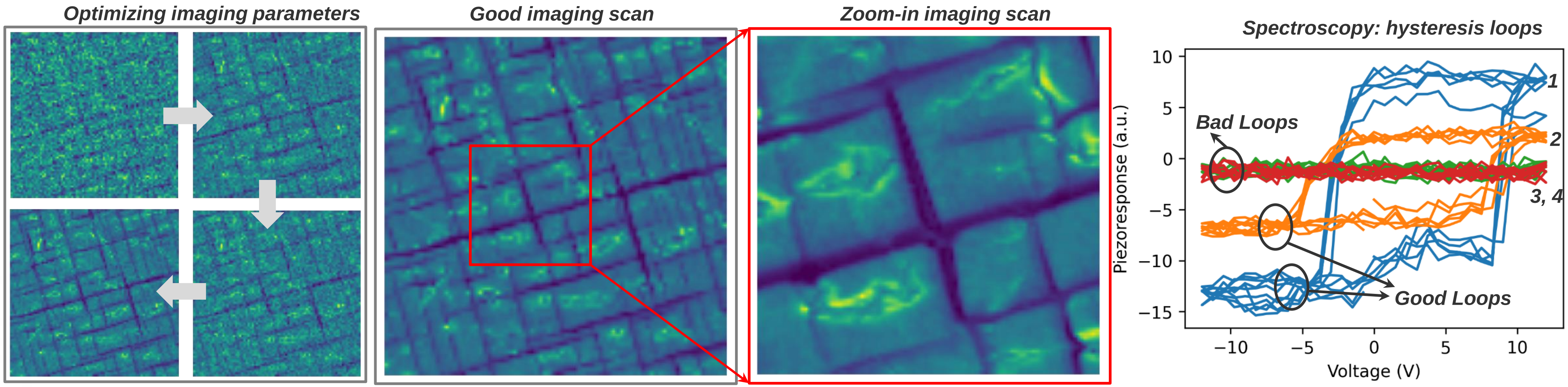
Rewards and objectives:
- What is our (hierarchical) objective?
- Can we define reward(s)?

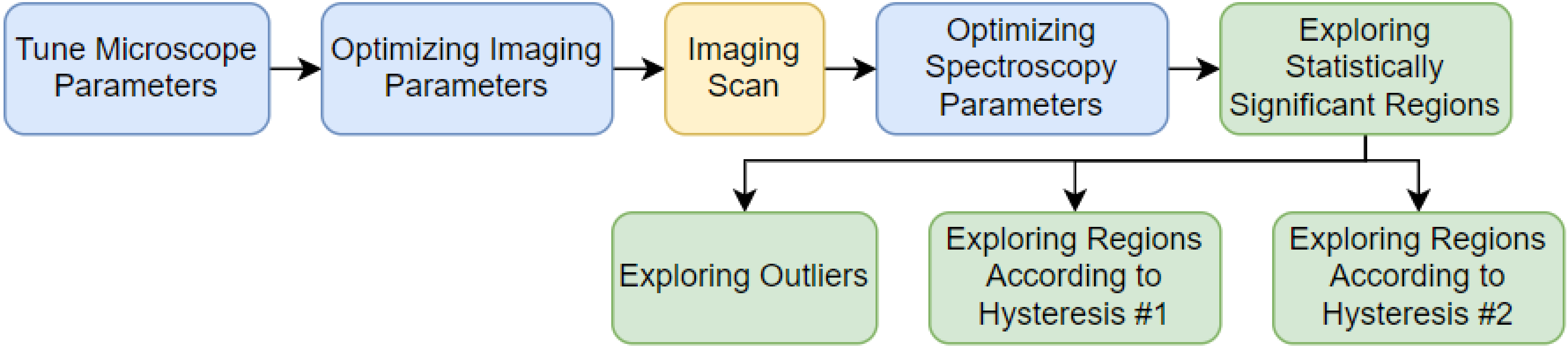


Inferential biases:
- What do we know before the experiment?
- What do we (hope to) learn after the experiment?

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?

Future: full workflow optimization





Characteristic	Definition	Availability
Global image	Initial structural data set available before DKL experiment. Used to create patches for DKL training	Before
DKL latents	The latent variables encoding the structural information in the patches	During**
Scalarizer function	Function defining what characteristic of spectrum guides Bayesian Optimization	Before*
Acquisition function	Function combining DKL prediction and uncertainty of the scalarizer function	Before*
Policy	Principle for selection of next path. Simplest policy is maximization of acquisition function, but can be more complex including epsilon-greedy or switch between multiple scalarizers or acquisition functions. Human in the loop intervention tunes some aspect of the policy	Before*
Experimental trace	Collection of patches (and their coordinates) and spectra derived during experiment. Trace and global image are the results of AE SPM.	During

Characteristic	**Definition**	**Availability**
Live DKL model	DKL model in the state corresponding to the *n*-th experimental step	During
Final DKL model	DKL model in the state corresponding to the end of the experiment	After
Complete DKL model	DKL model trained on the full data set (if available from grid measurements, etc).	
Regret analysis	The difference between predictions of live DKL model and final DKL model after the whole experiment (i.e., after 200 steps in this work)	During** and After
Learning curve	Change of the DKL uncertainty (mean and deviation), indicative of the predictability of the patch-scalarizer relationship	During
Counterfactual scalarizer	The availability of full spectral data as a part of experimental trace allows to estimate what the BO step would be if scalarizer were chosen to be different	During
Trajectory analysis	Real-time trajectory of the probe that can be represented in the global image plane	During
Feature discovery	Analysis of the latent variables and latent representations of image patches and spectra in the trace. Here, we realize only patch analysis but extension to spectra is straightforward.	After
Latent trajectory analysis	Analysis of the experimental trajectory in the latent space of the full collection of the image patches derived from the global image	During**