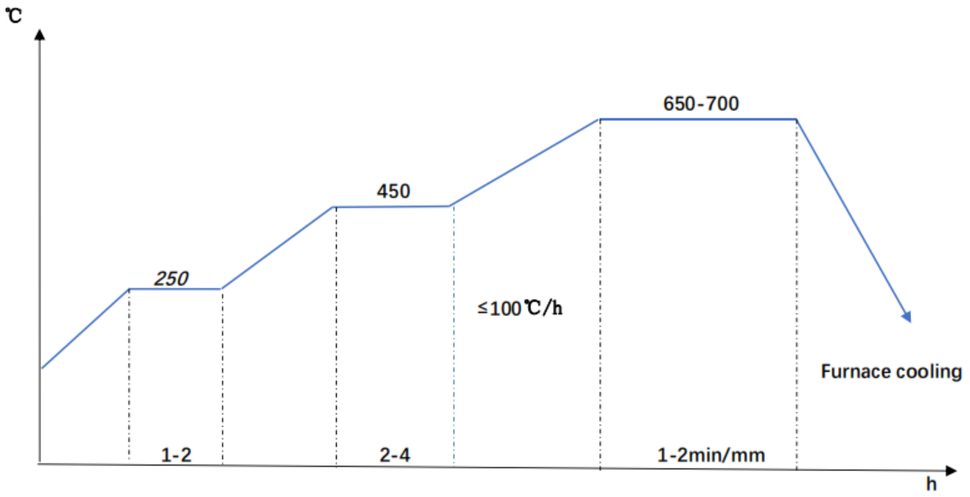
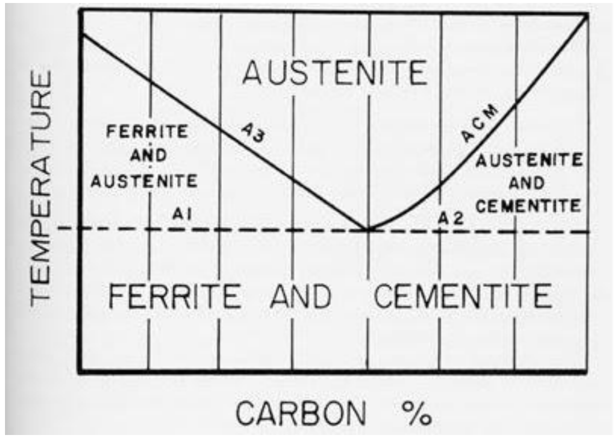
THE UNIVERSITY of TENNESSEE KNOXVILLE

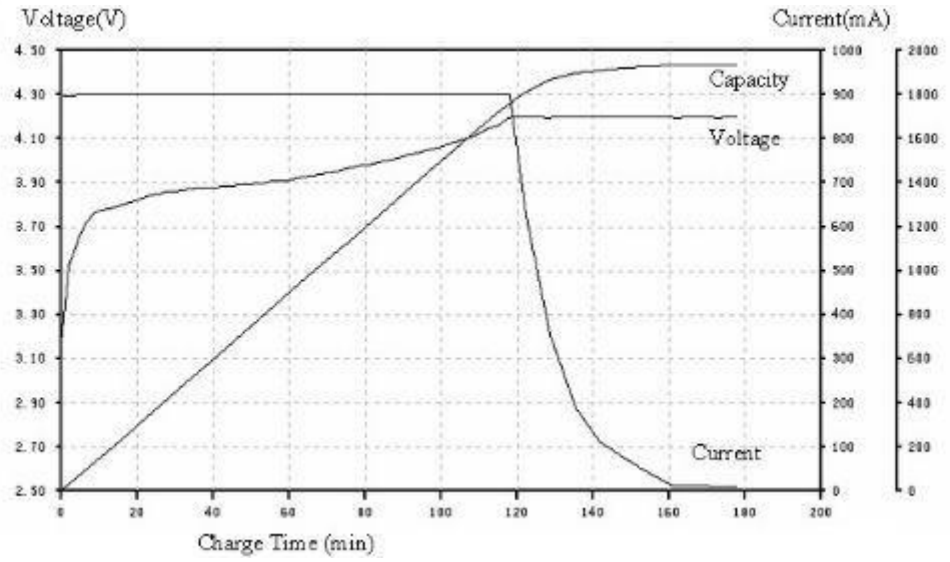
Lecture 25: Deep Kernel Learning for Automated Experiment in Imaging

Instructor: Sergei V. Kalinin

Making materials: process trajectories



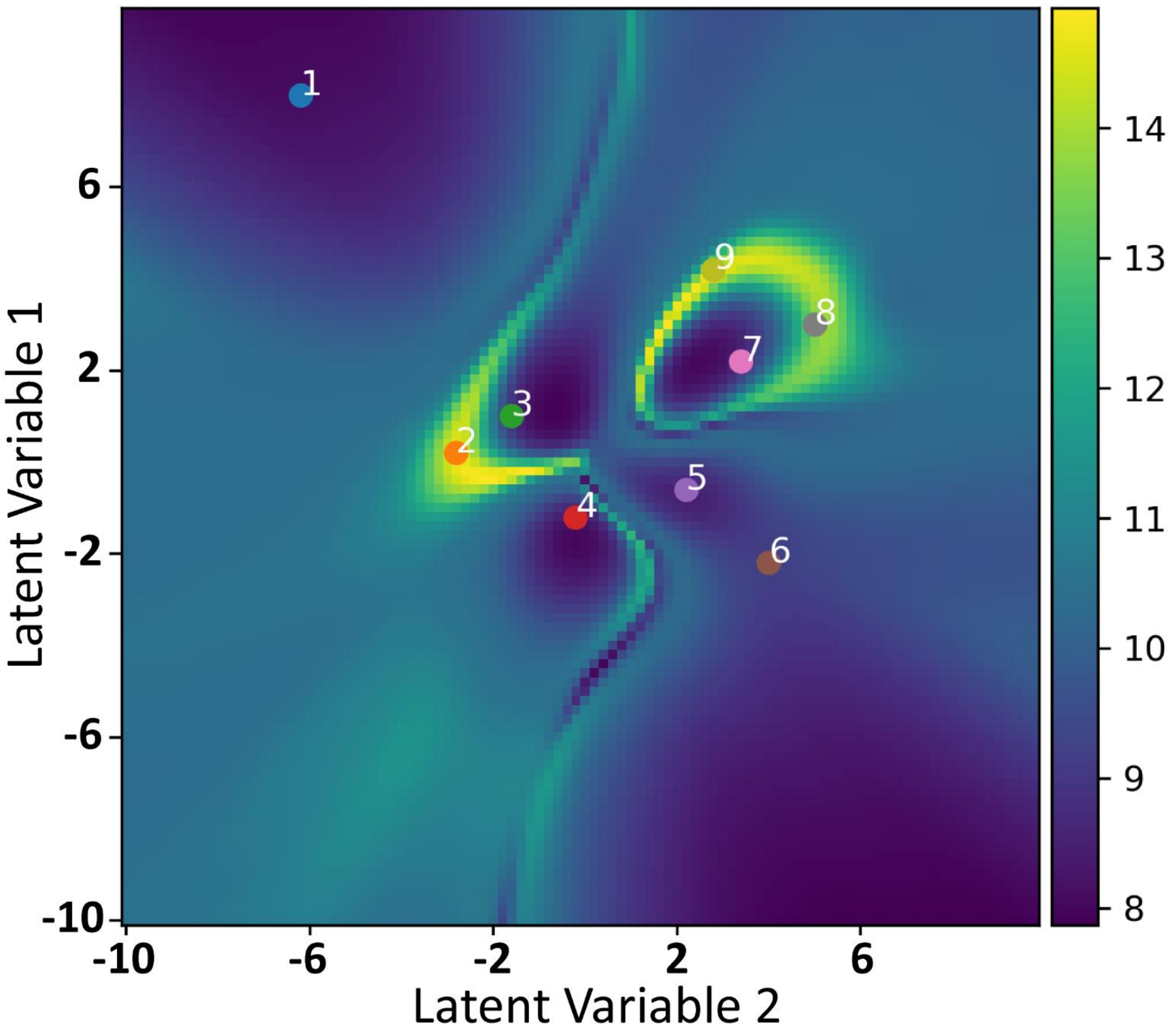




- Making steel: complicated and took a lot of time optimize
- Charging battery: obvious economic impact
- Manufacturing: Annealing hybrid perovskite thin films
- Poling ferroelectric

How do we optimize trajectories if we have (a) only limited or no mechanistic information, (b) our experimental budgets are limited, but (c) we have some access to domain expertise?

What determines success?

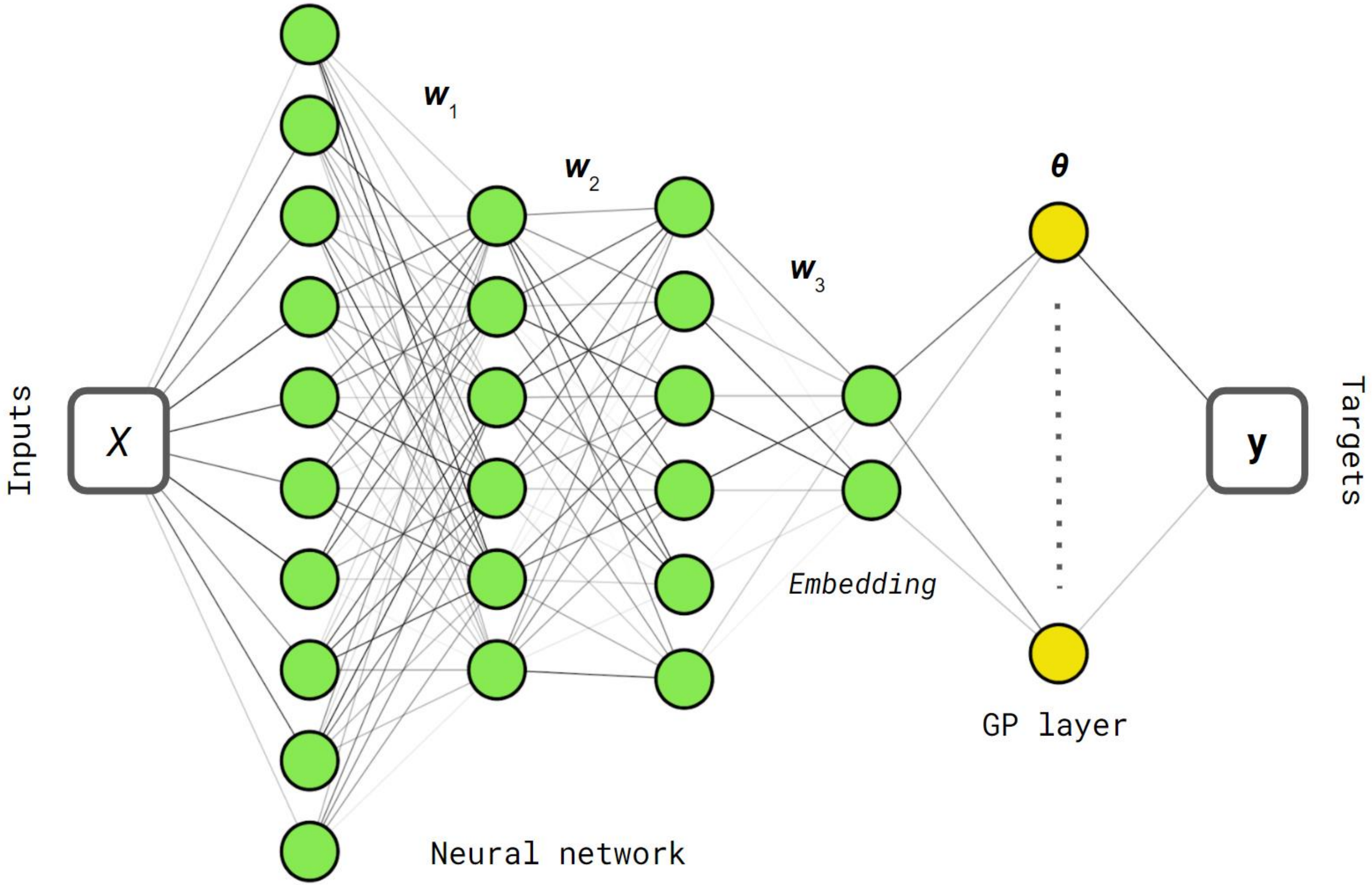


The success of the BO in the latent space clearly depends on the shape on the manifold that points of interest form.

For VAE, the shape of the manifold is determined by the properties of the data only, including

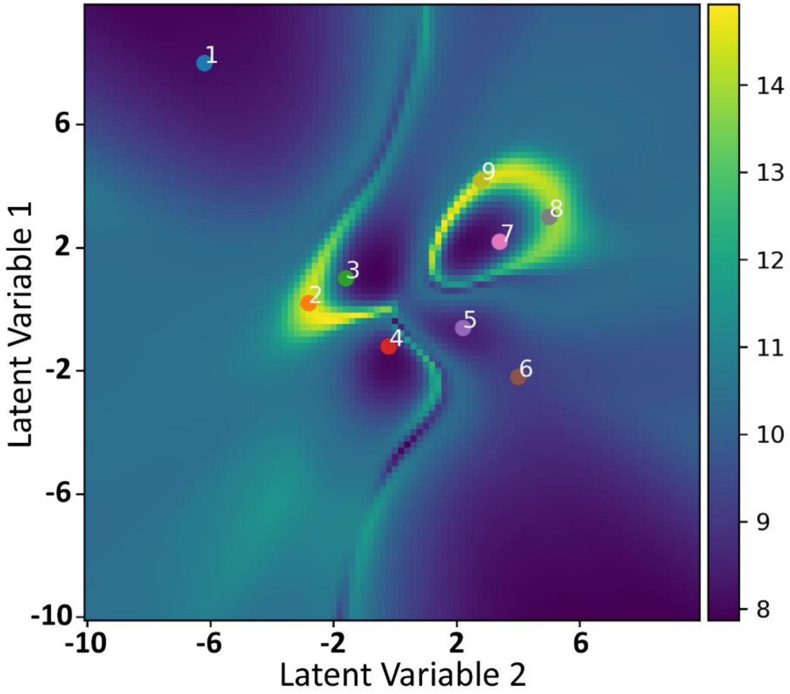
(a) how strong correlations in data reflect in correlation in properties and
(b) weight of the “good” trajectories

Deep Kernel Learning

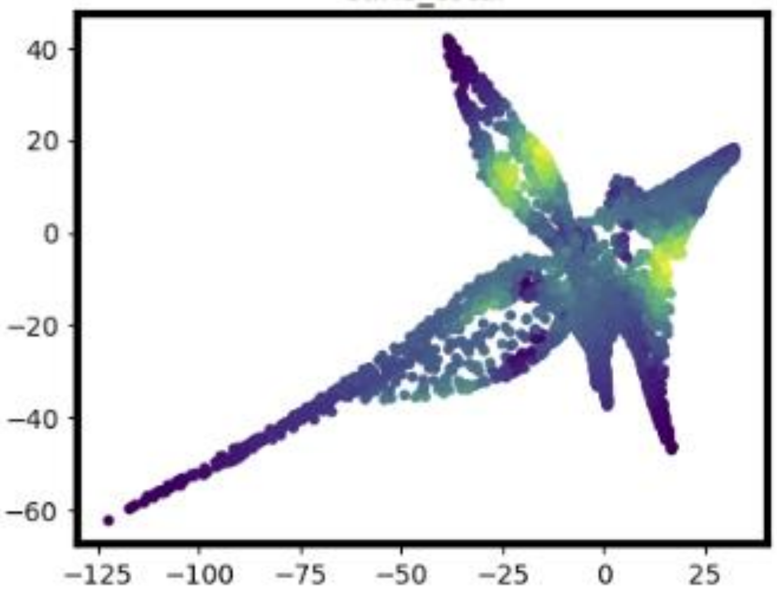


Comparing VAE BO, Static DKL, and Active DKL

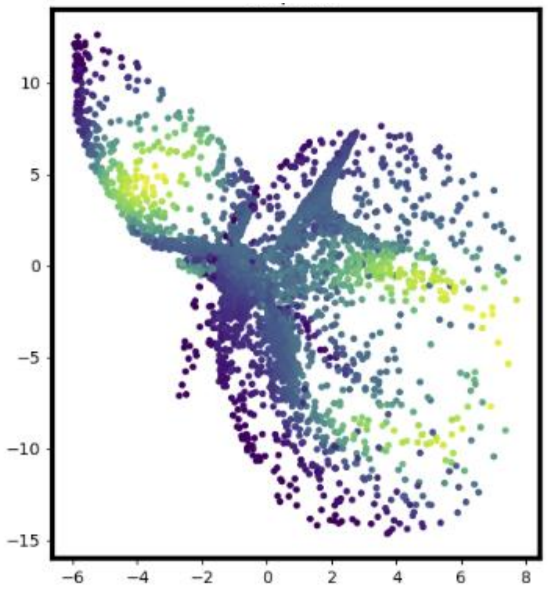
VAE - BO



Static DKL



Active DKL



Summary:

- Manifold structure determines how fast can the unsupervised or active learning work
- For VAEs, the latent structure is determined by the data only. Sometimes property are forming convenient manifolds, most of the time not.
- Static DKL forms much better organized manifolds
- … Active learning produces best manifolds!

BO in Imaging

ML for Automated Microscopy?

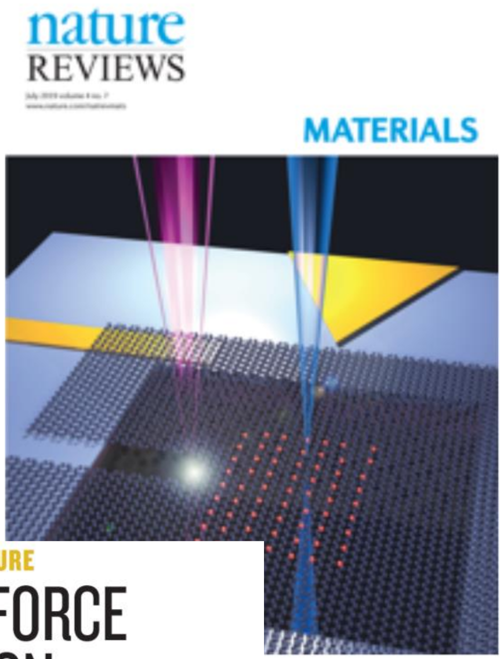
Microscopy today:

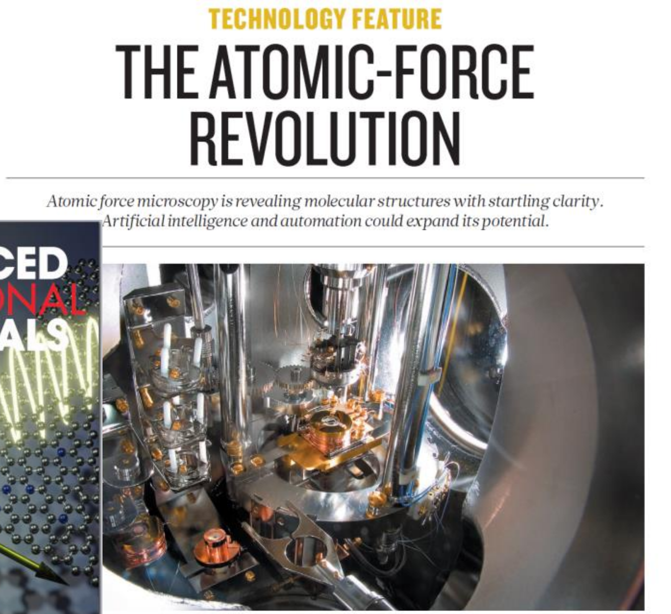
- Primary component of research in materials, physics and biology
- 1000s of high-end (S)TEM platforms, ~10,000 overall
- 1000s of high-end UHV SPMs, >50,000 ambient
- Chemical and mass-spectrometric imaging

What do microscopists do?

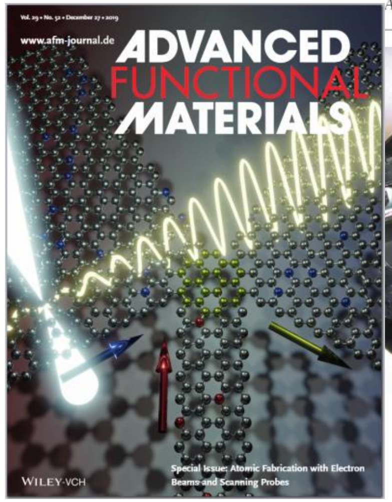
- Most of the time - sit alone in the dark room and turn knobs 😌
- Limited amount of collected data
- Case for automation: CryoEM

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





July 2019



Reward functions in imaging

Imaging Optimization

Physical laws discovery

Image-based reward functions

- Human selected objects (DCNNs)
- Equal sampling of feature space
- Equal sampling of parameter space (combi library)

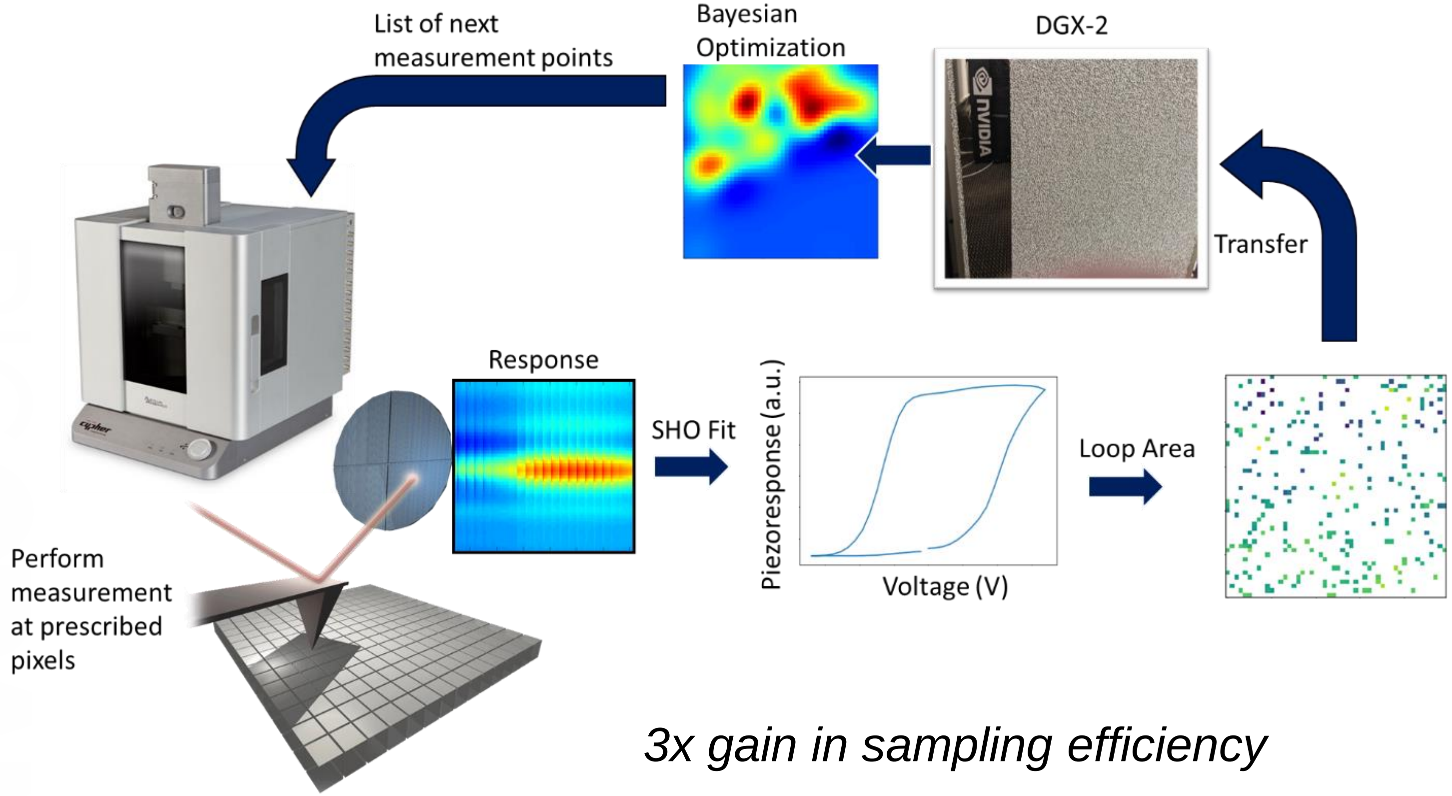
Structure property relationship discovery

- Reward definition (with cost)
- Tuning curiosity
- Human in the loop DKL

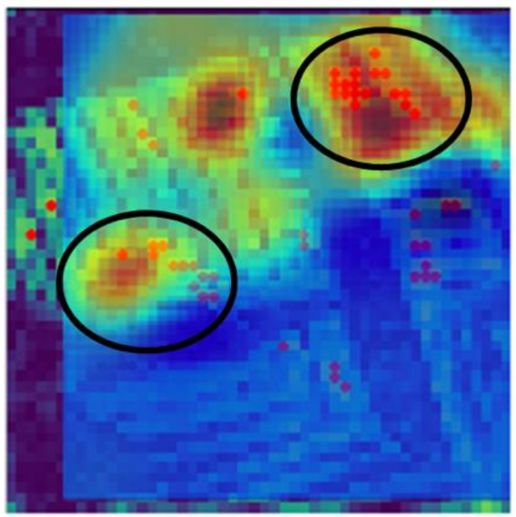
Co-orchestration multiple tools
Co-navigation between theory and experiment

BO for Self-Driving Microscope

First implementation of self-driving microscope: 2020

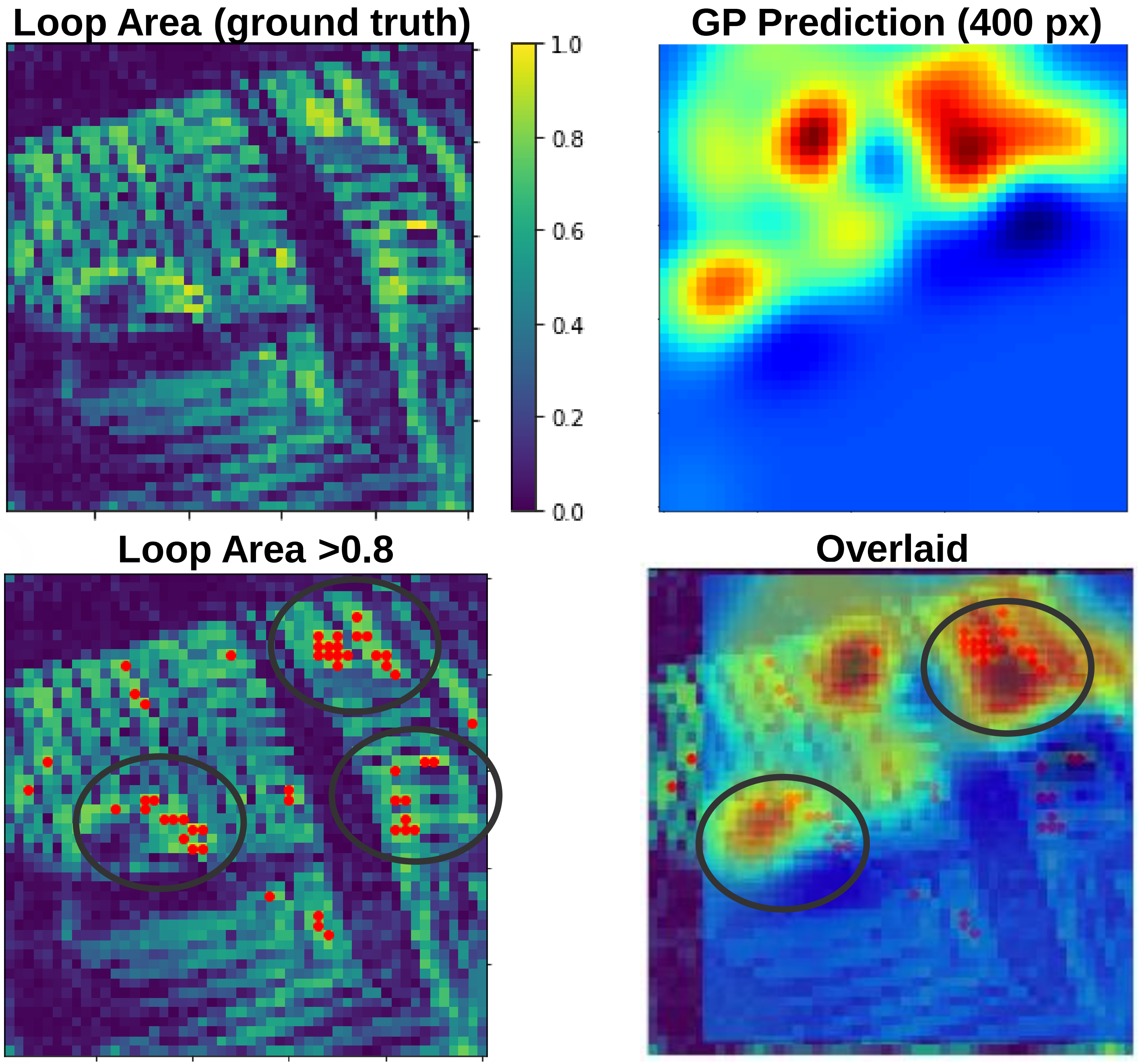


Comparison with "ground truth"



3x gain in sampling efficiency

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



[arXiv:2103.12165](https://arxiv.org/abs/2103.12165)

[arXiv:2011.13050](https://arxiv.org/abs/2011.13050)

The application of simple data-driven GP for real world scenarios did not work particularly well.

We were solving wrong problem!

Two modes of operations

Structural imaging (Cheap)

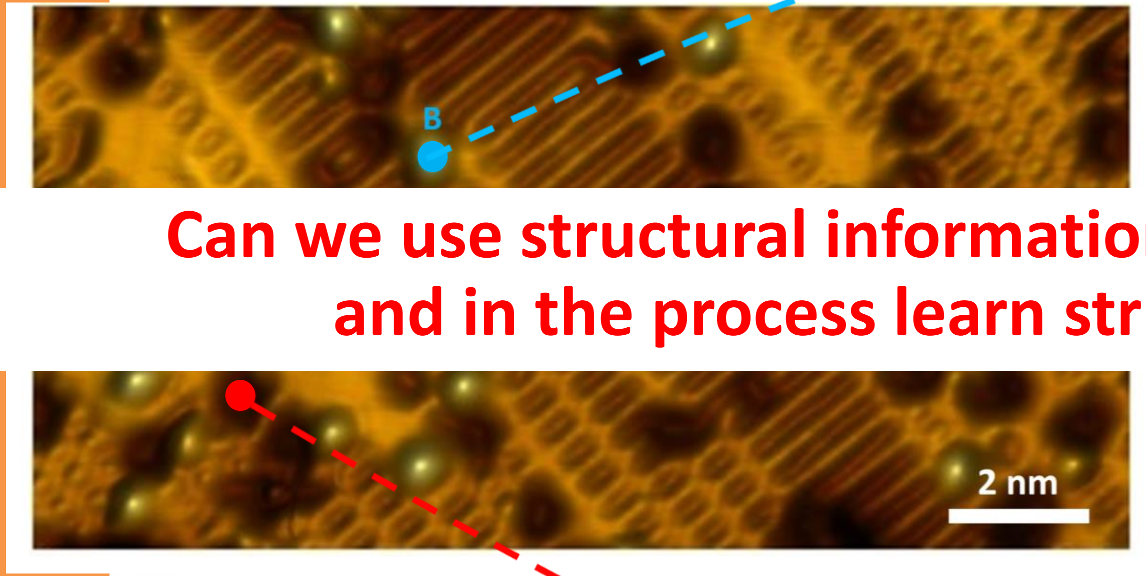
Topography in STM, amplitude/phase in SPM, (HA)ADF-image in STEM, etc.

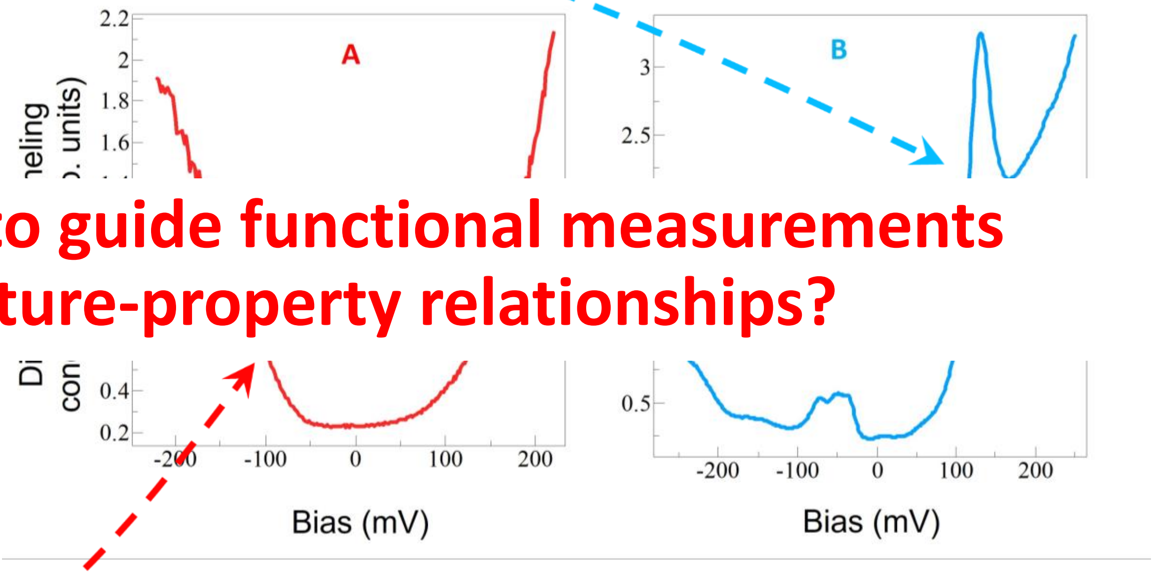
These are FAST measurements (from seconds to minutes)

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)

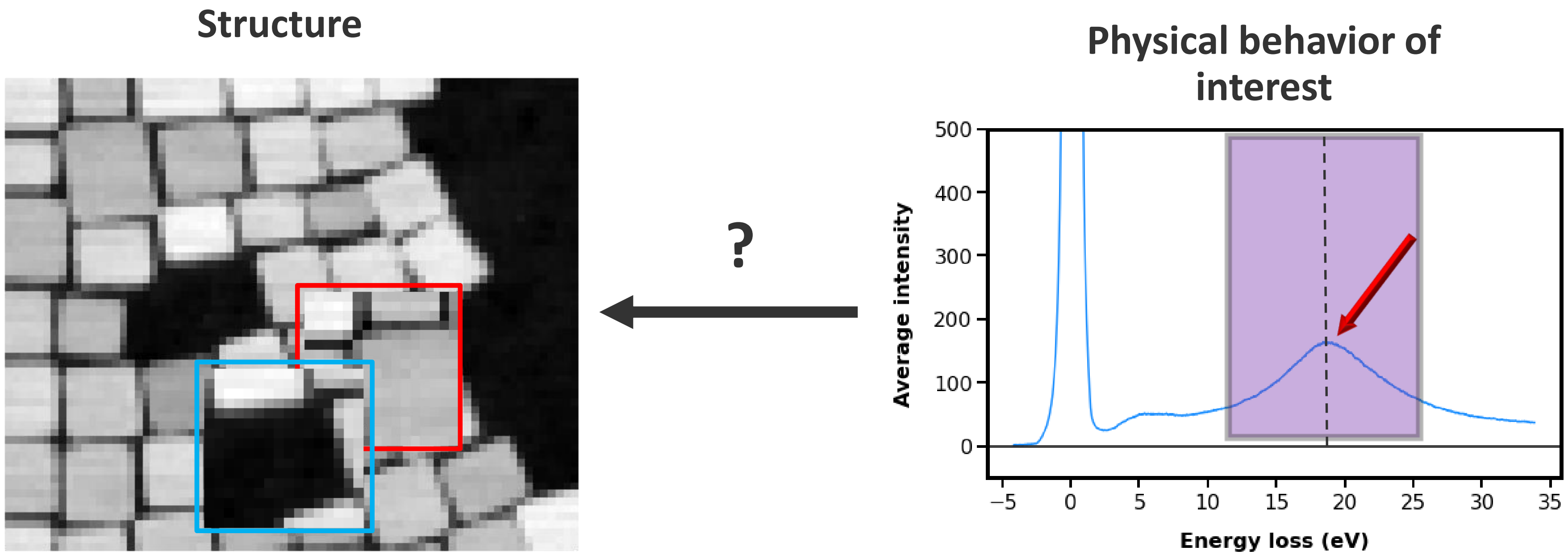




Can we use structural information to guide functional measurements and in the process learn structure-property relationships?

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**)

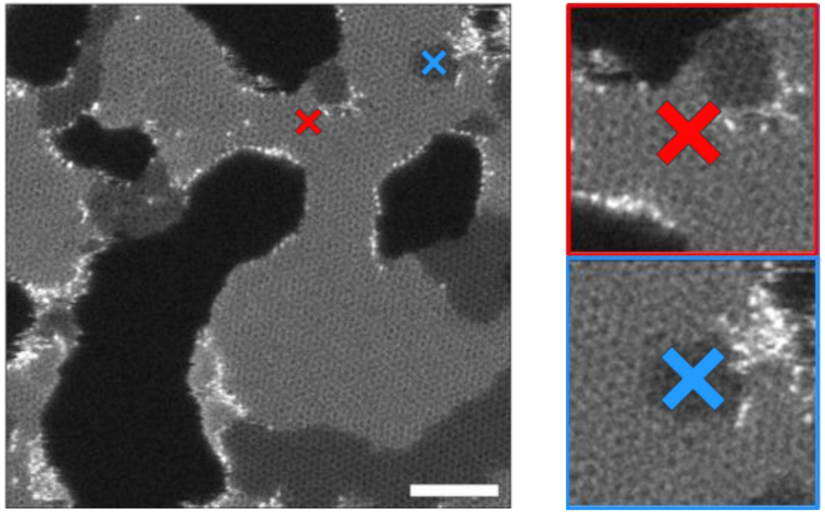


Deep Kernel Learning: Step 1

A1

Full image

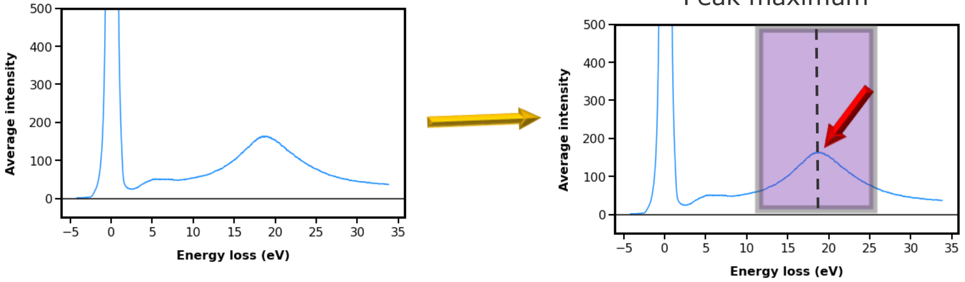
Image patches



A2

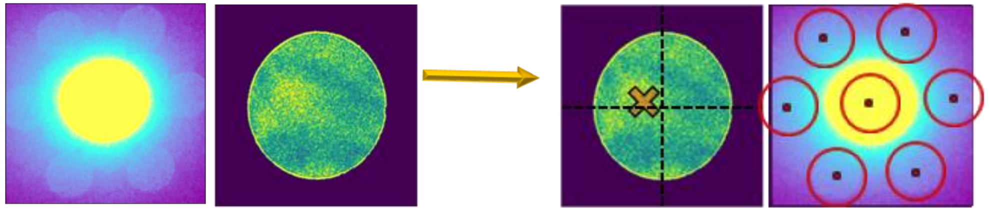
Measurement (1D or 2D)

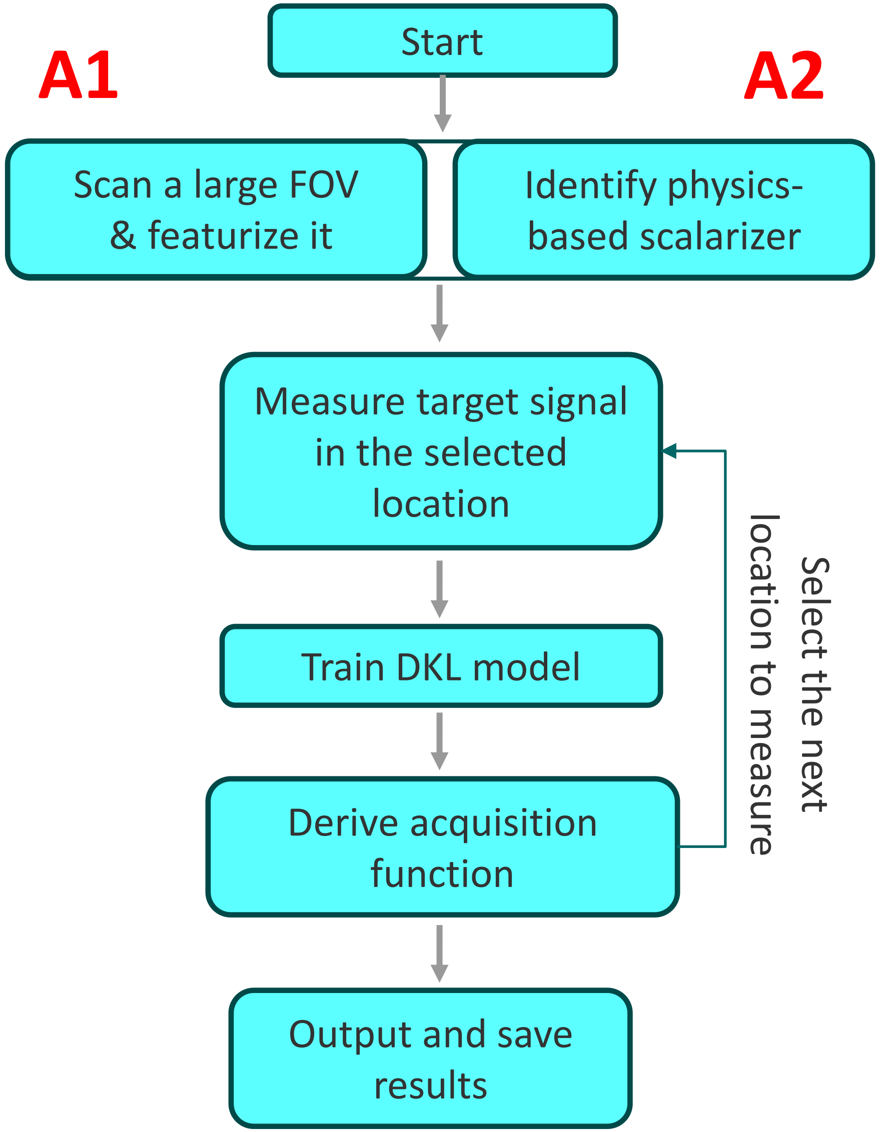
Scalar target



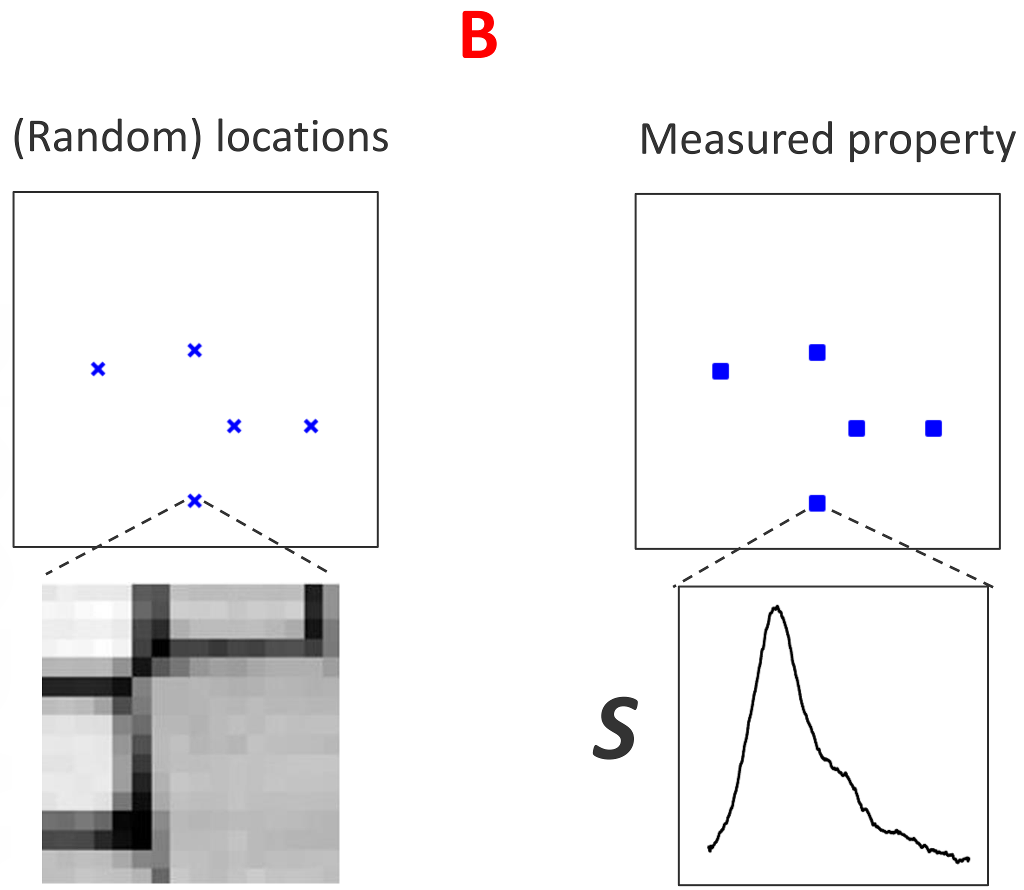
Center of Mass

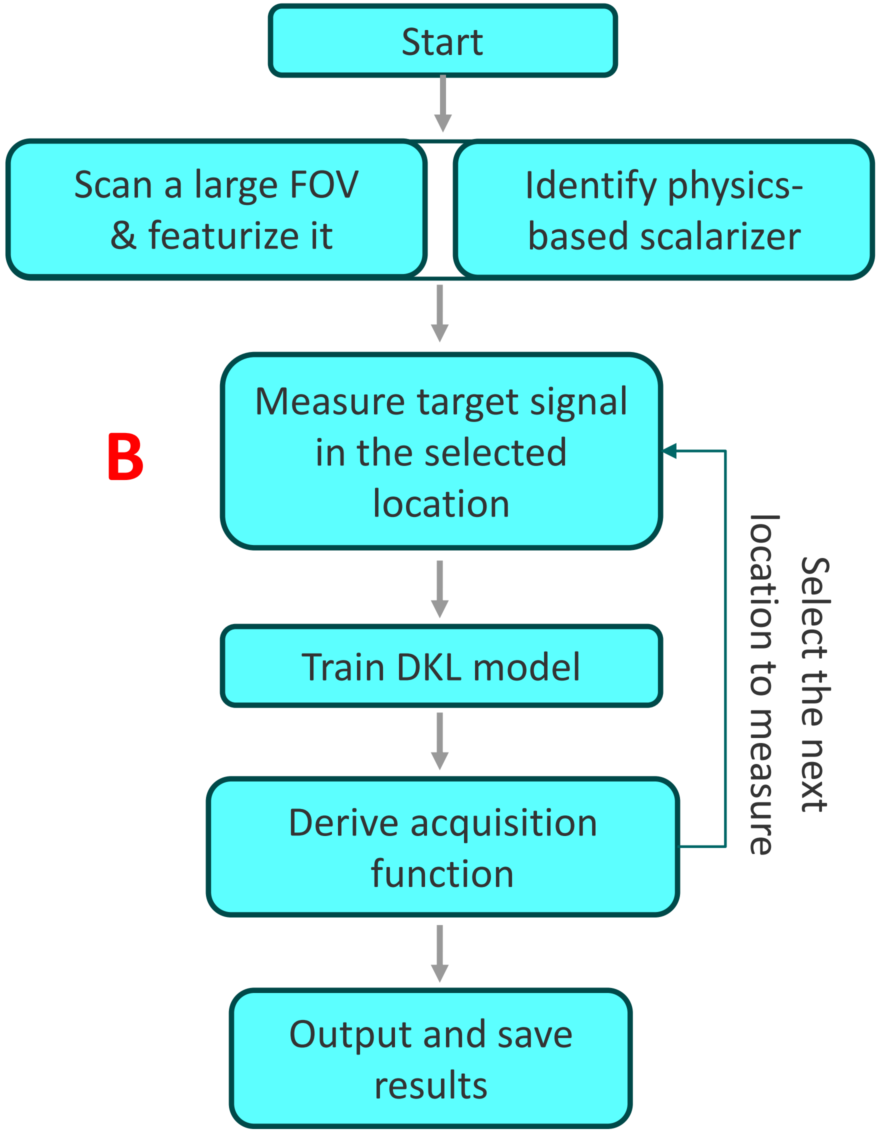
Disc fitting



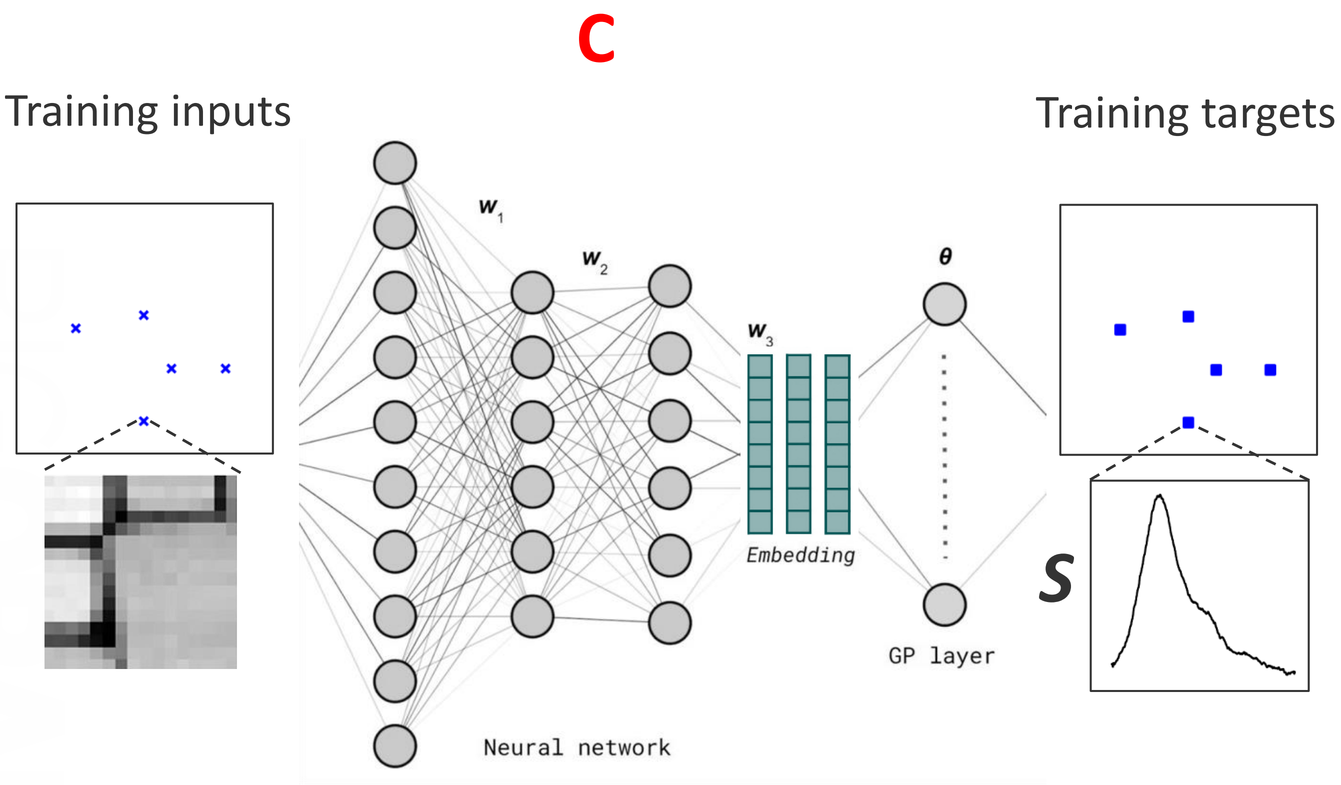


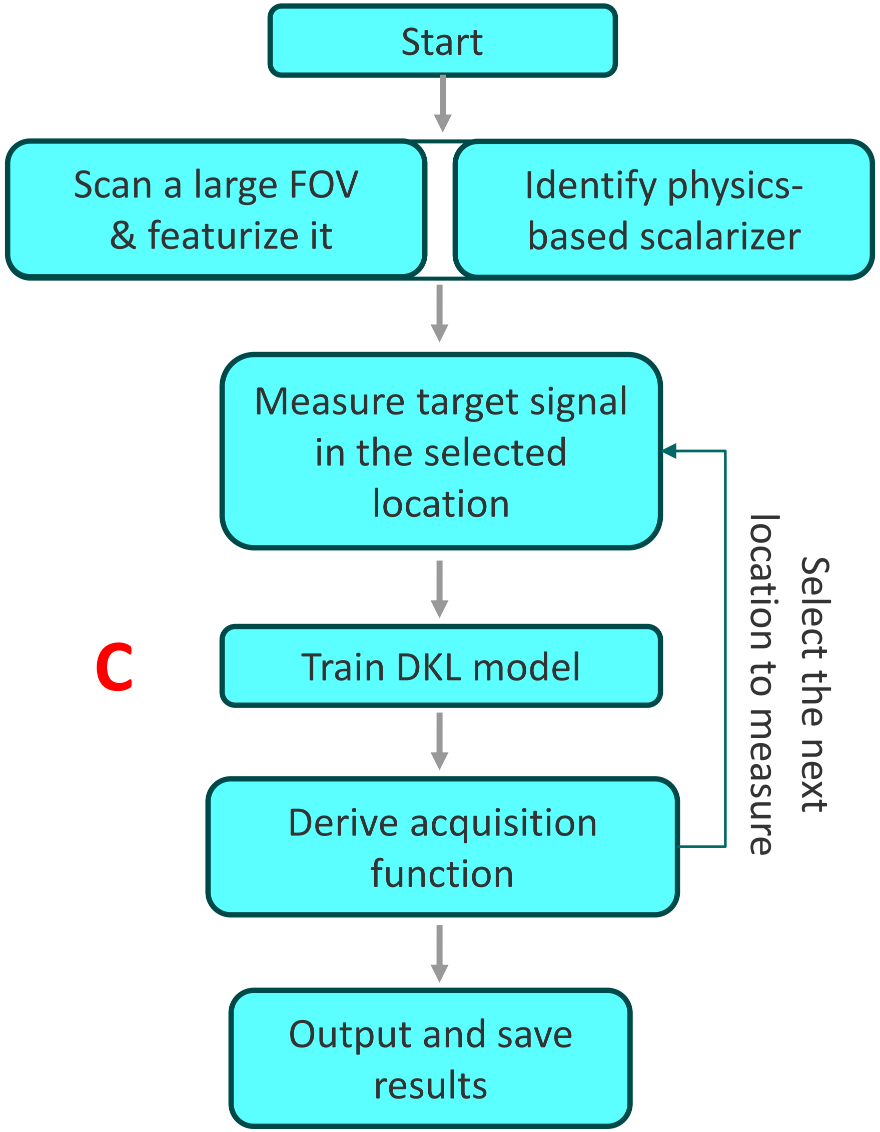
Deep Kernel Learning: Step 2





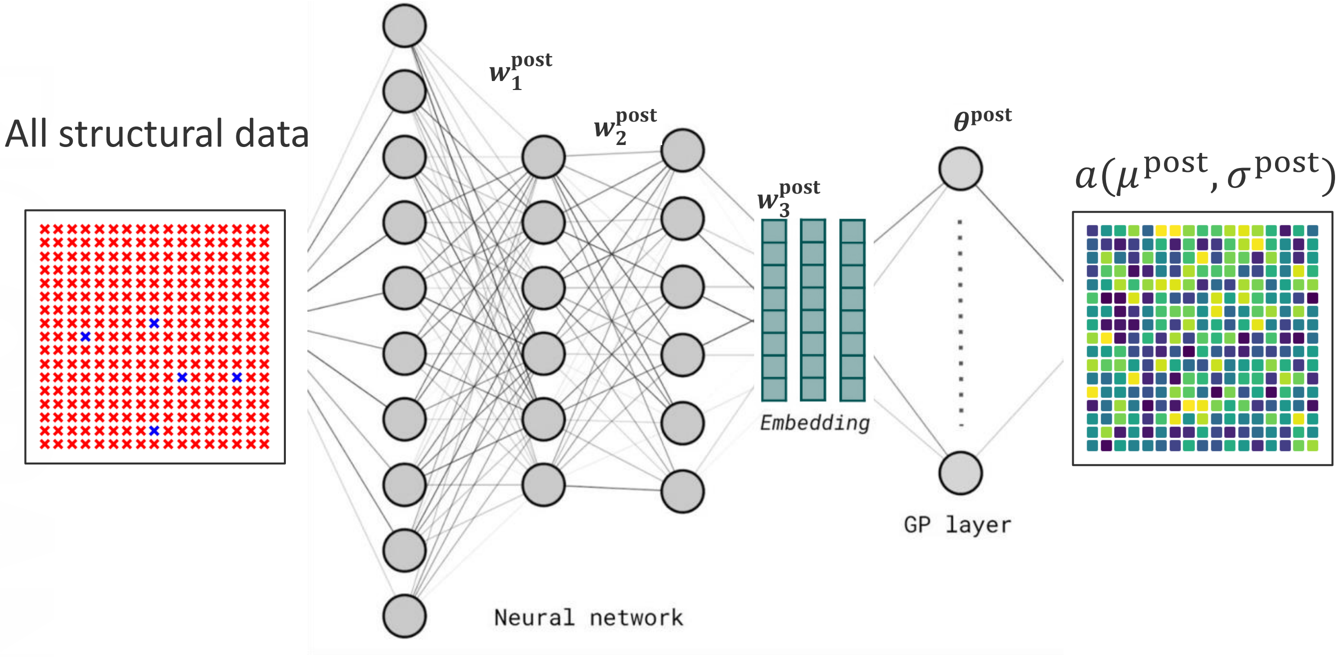
Deep Kernel Learning: Step 3





Deep Kernel Learning: Going Active

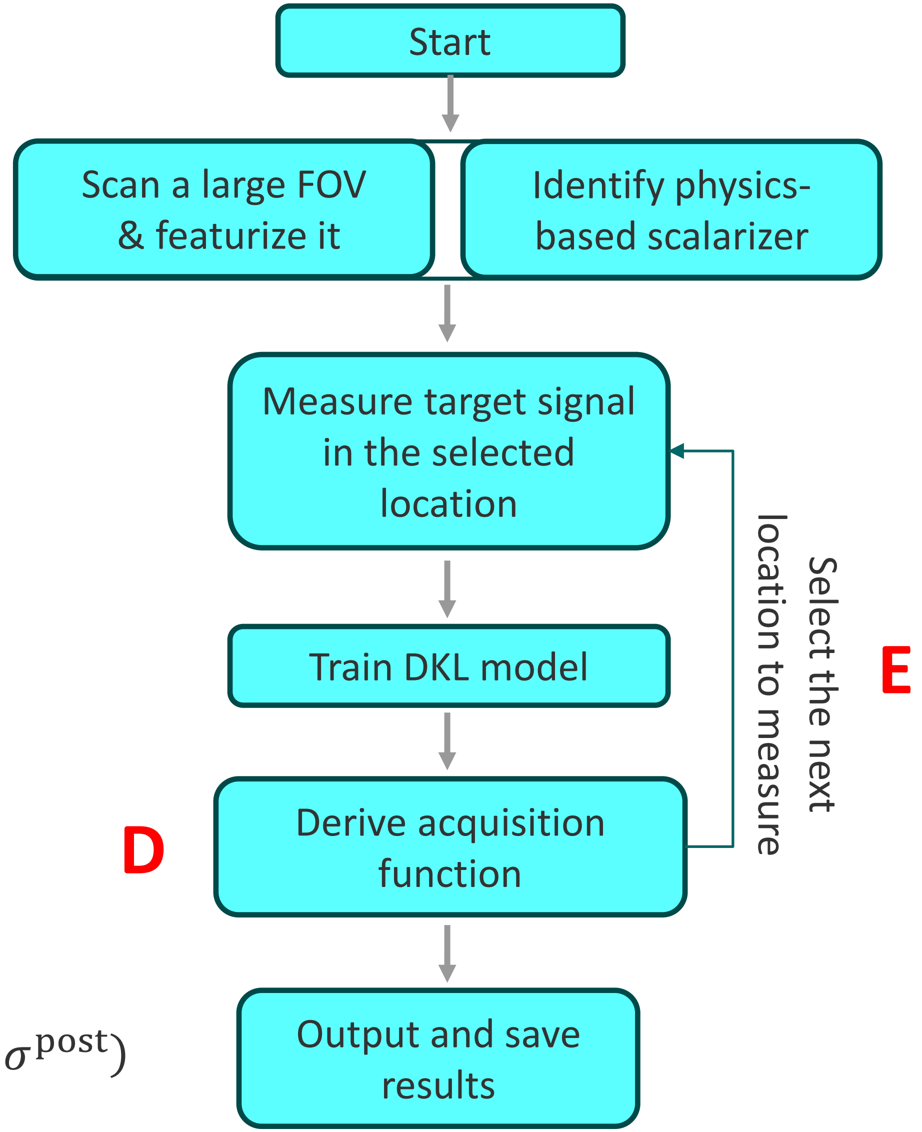
D



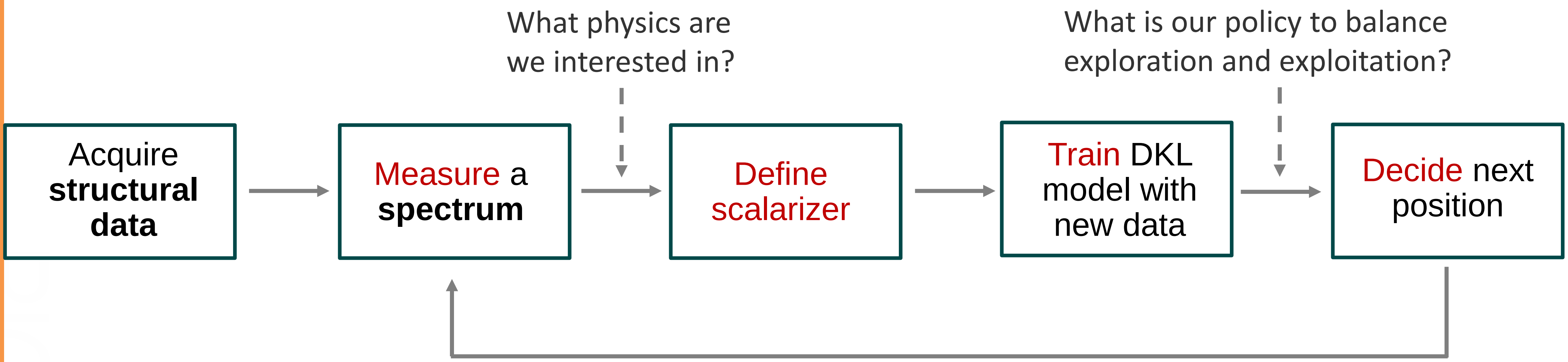
E

Next point to measure:

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



Deep Kernel Learning based BO

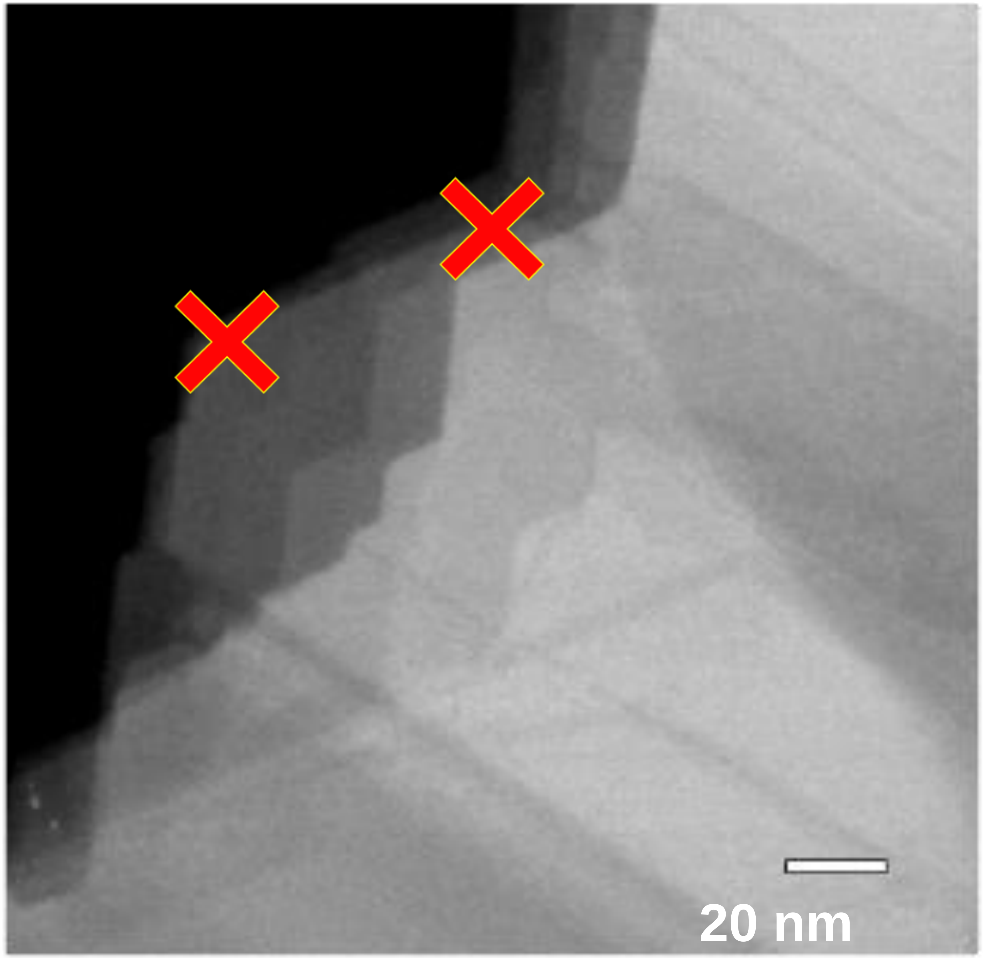


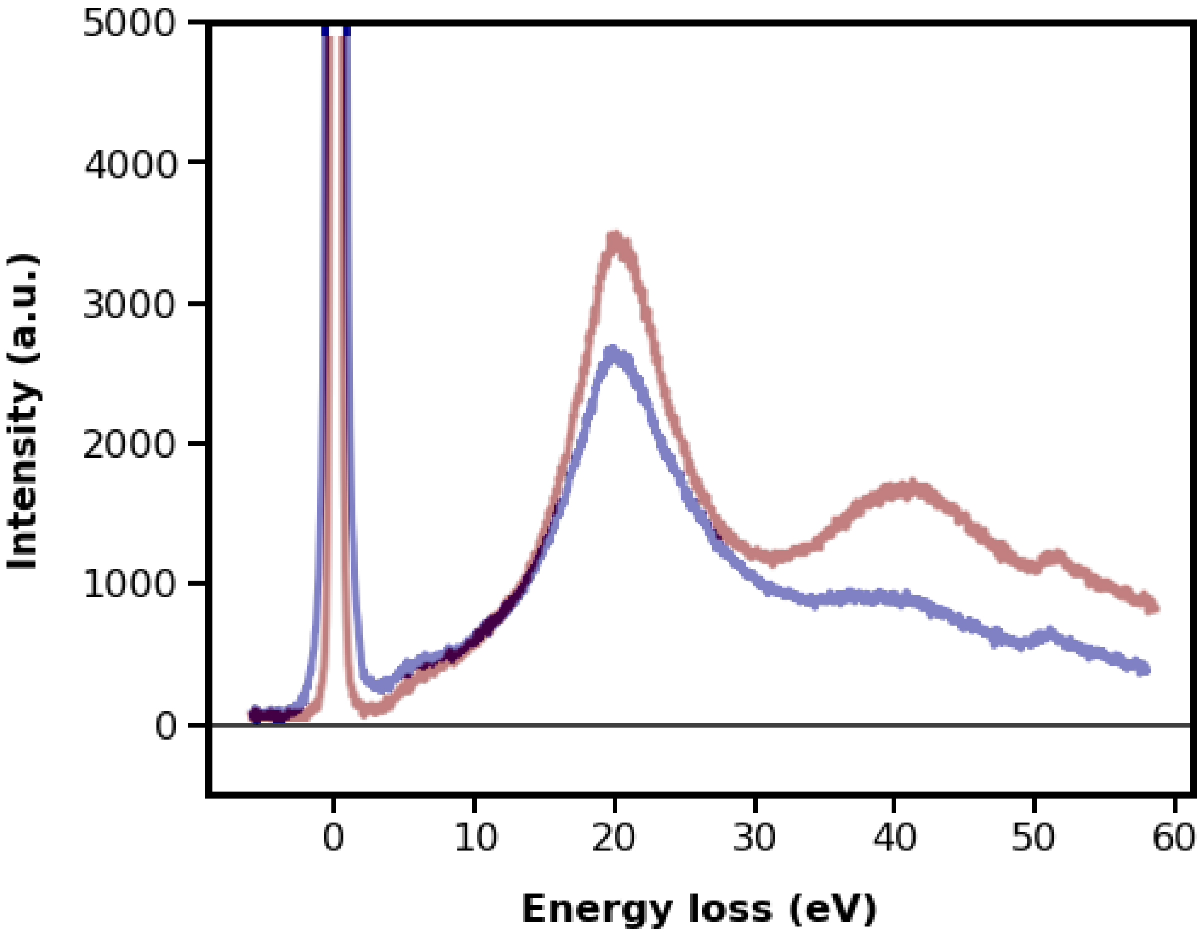
Key concepts:

- **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



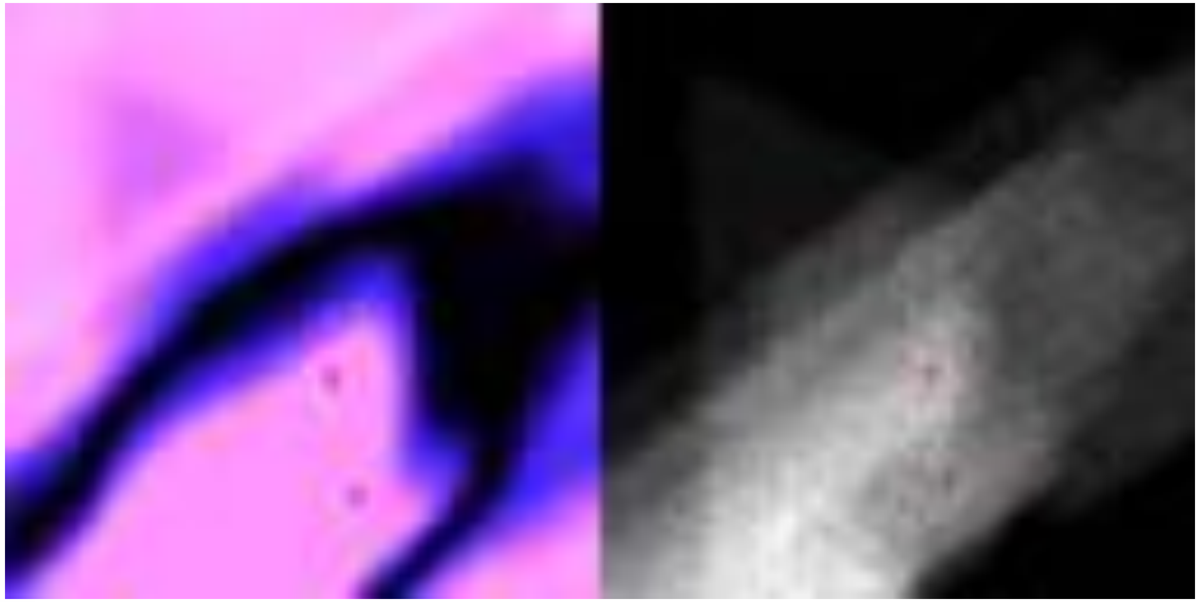


1. What if we have full access to structural information
2. And want to choose locations for (EELS, 4D STEM, CL, EDX) measurements
3. So as to **learn** relationship between structure and spectrum fastest
4. Or **discover** which microstructural elements give rise to specific **desired** spectral features?

Discovering Regions with Interesting Physics

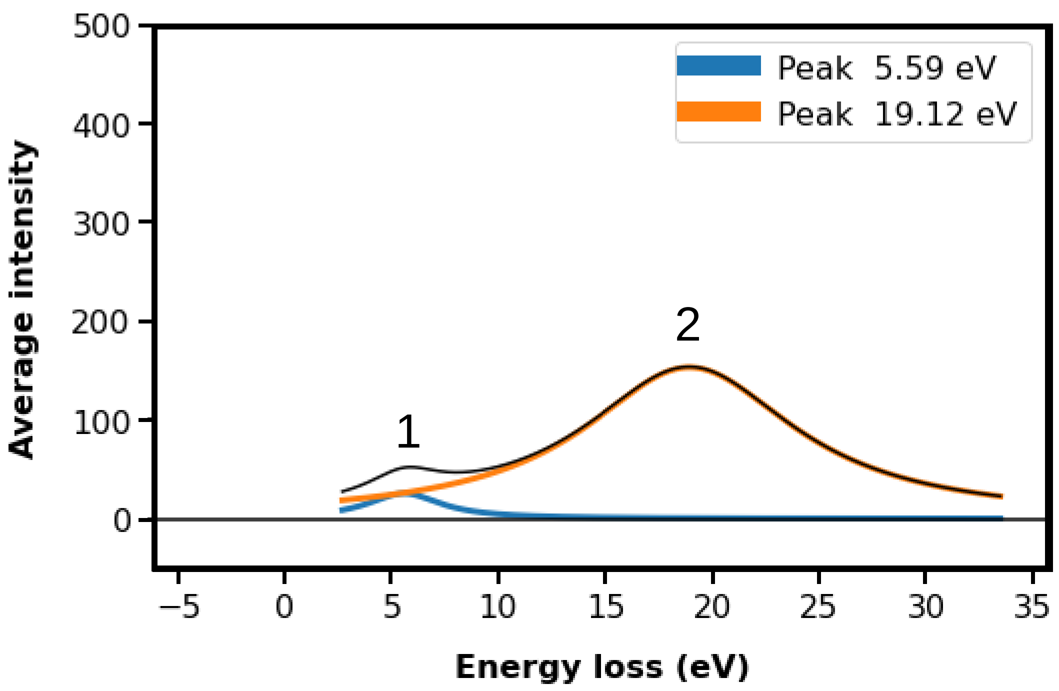
- **Discovering physics in a "new" material $MnPS_3$**
- **Curve fitting** to help enforce physical processes

Acquisition function **HAADF-STEM**

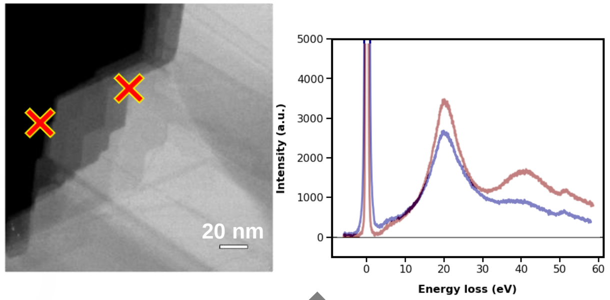


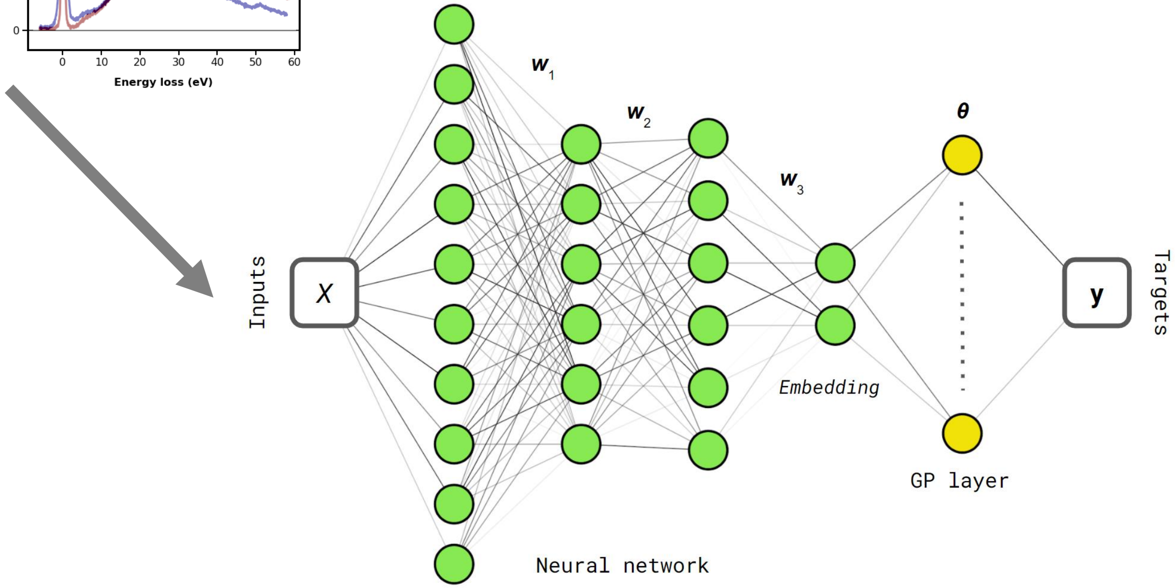
Physics search criteria:

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



Deep Kernel Learning





Specify physics criteria

Acquire
structural data

↓

Measure a
spectrum

↓

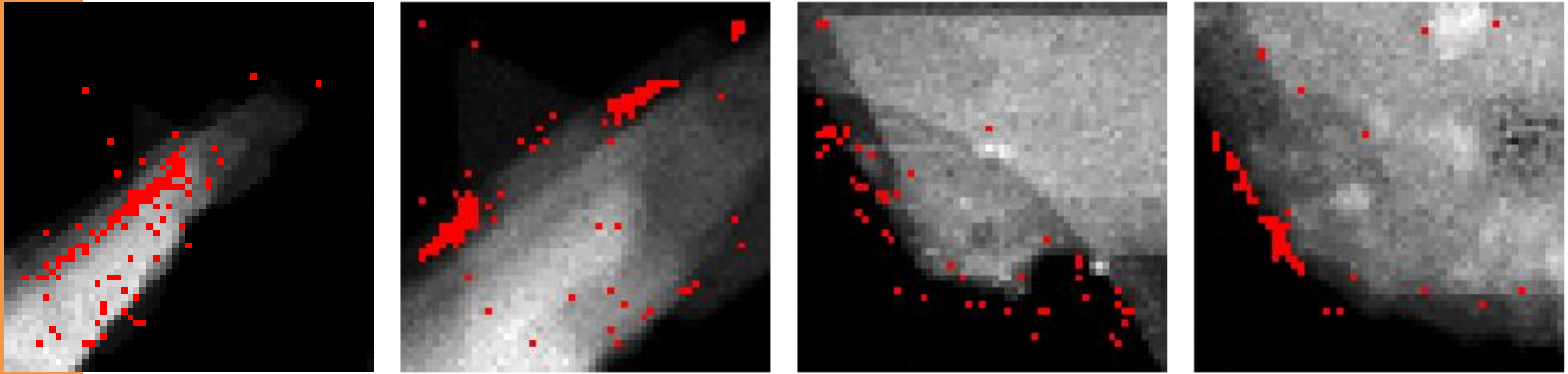
Train DKL
model with new
data

↓

Decide next
position (optimize
physics criteria)

Allows navigation of the system to search for physics

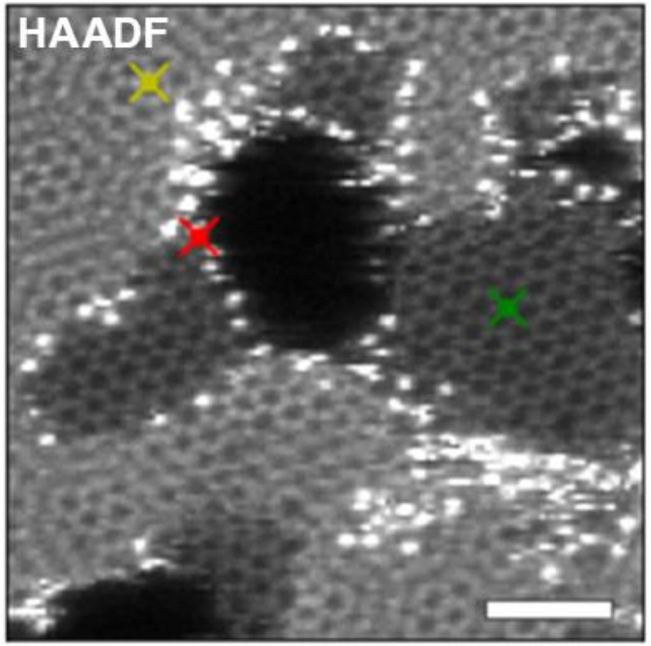
More Examples of Physics Discovery

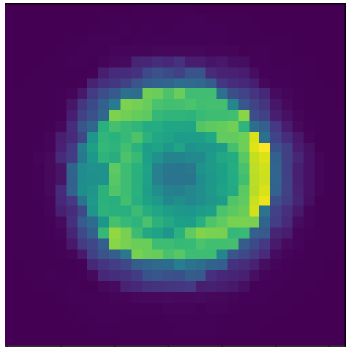


Discovery pathway depends on the reward structure (scalarizer that defines signature of physics we want to discover)!

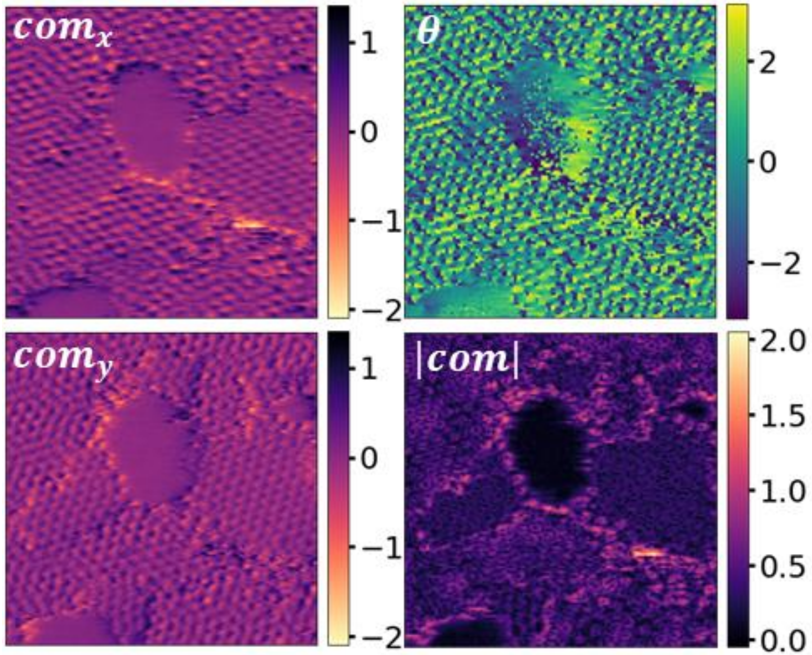
- Currently, we run 4D STEM measurements on a grid.
- What if we want to explore smarter workflows – where microscope chooses where to take 4D STEM measurements?
- **Direct:** We can do it for a priori known objects of interest
- **Inverse:** Or we can aim to discover objects which have predefined signatures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

4D STEM: Grid, Direct, and Inverse





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

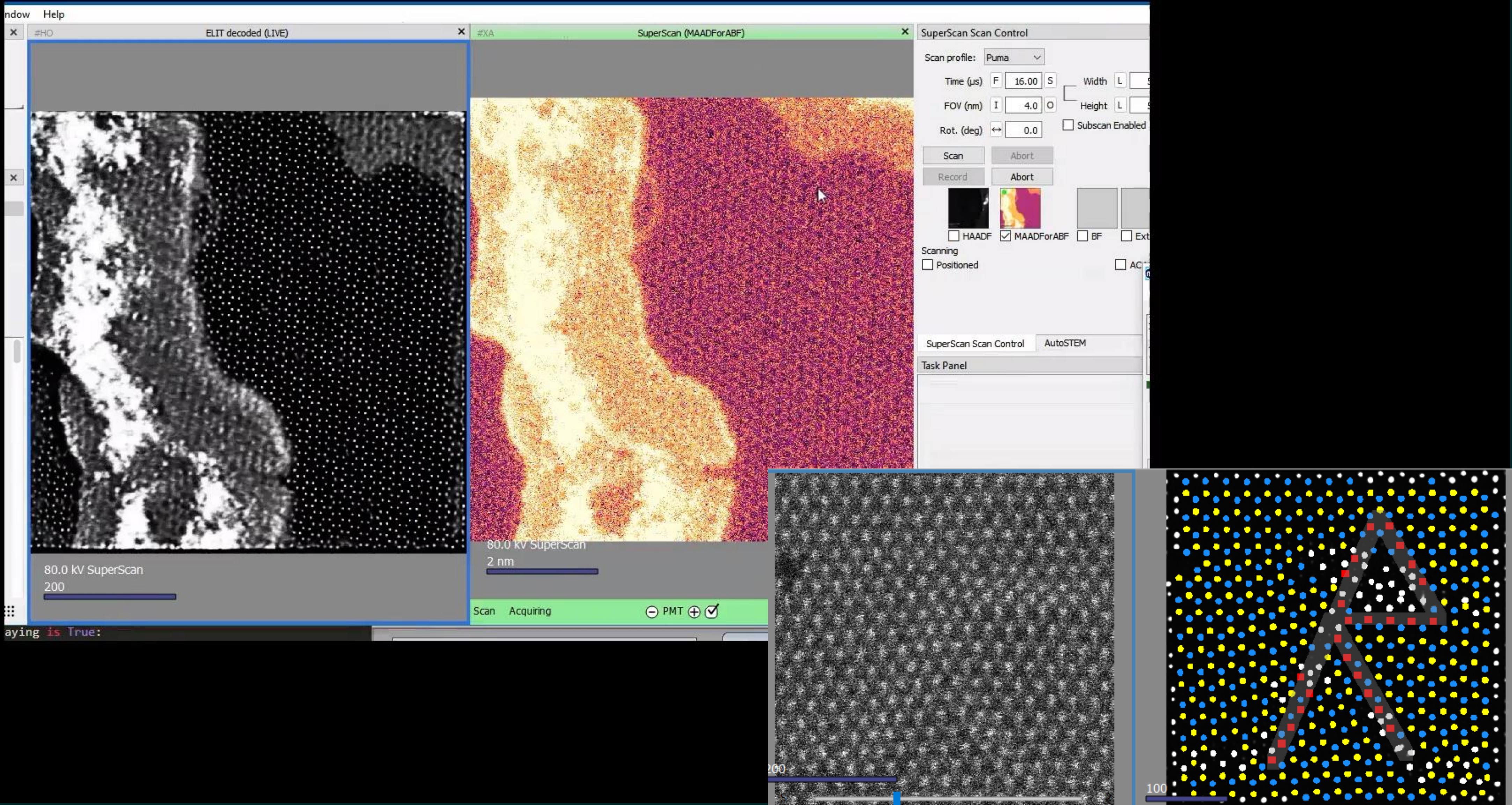


- What can we say about structure?
- Interesting functionalities are expected at the certain structural elements
- We can guess some; we have to discover others
- Multiple goals while running experiment

- **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

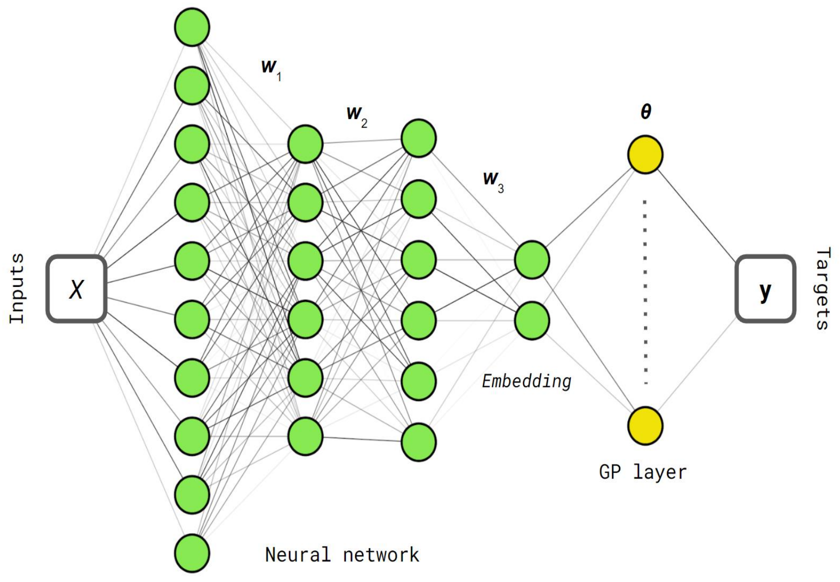
What physics are we interested in?

What is our policy to balance exploration and exploitation?

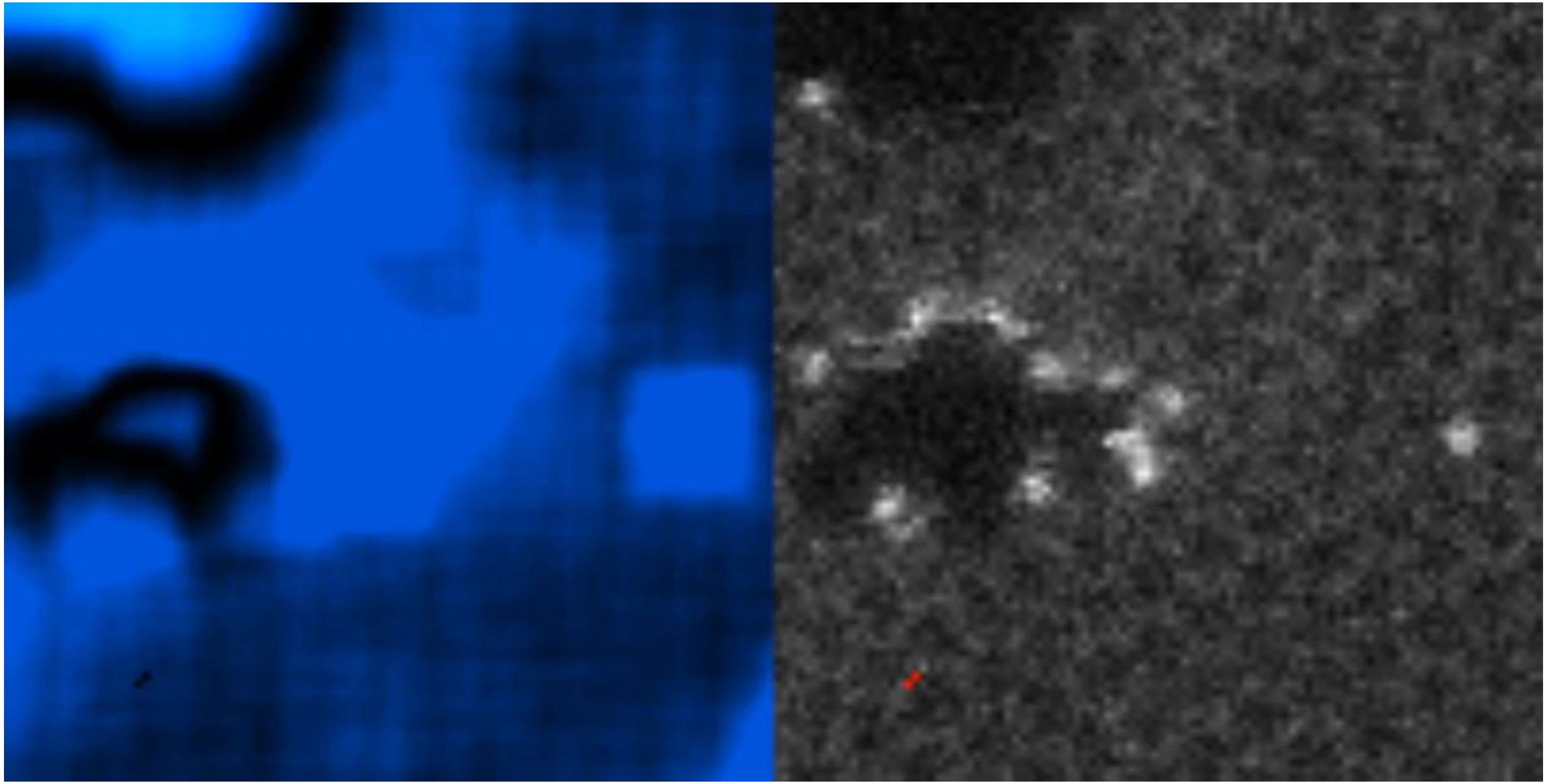
Acquire **structural data** → Measure a **ronchigram** → Define scalarizer → Train DKL model with new data → Decide next position → (back to Measure a **ronchigram**)

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 Active Microscope

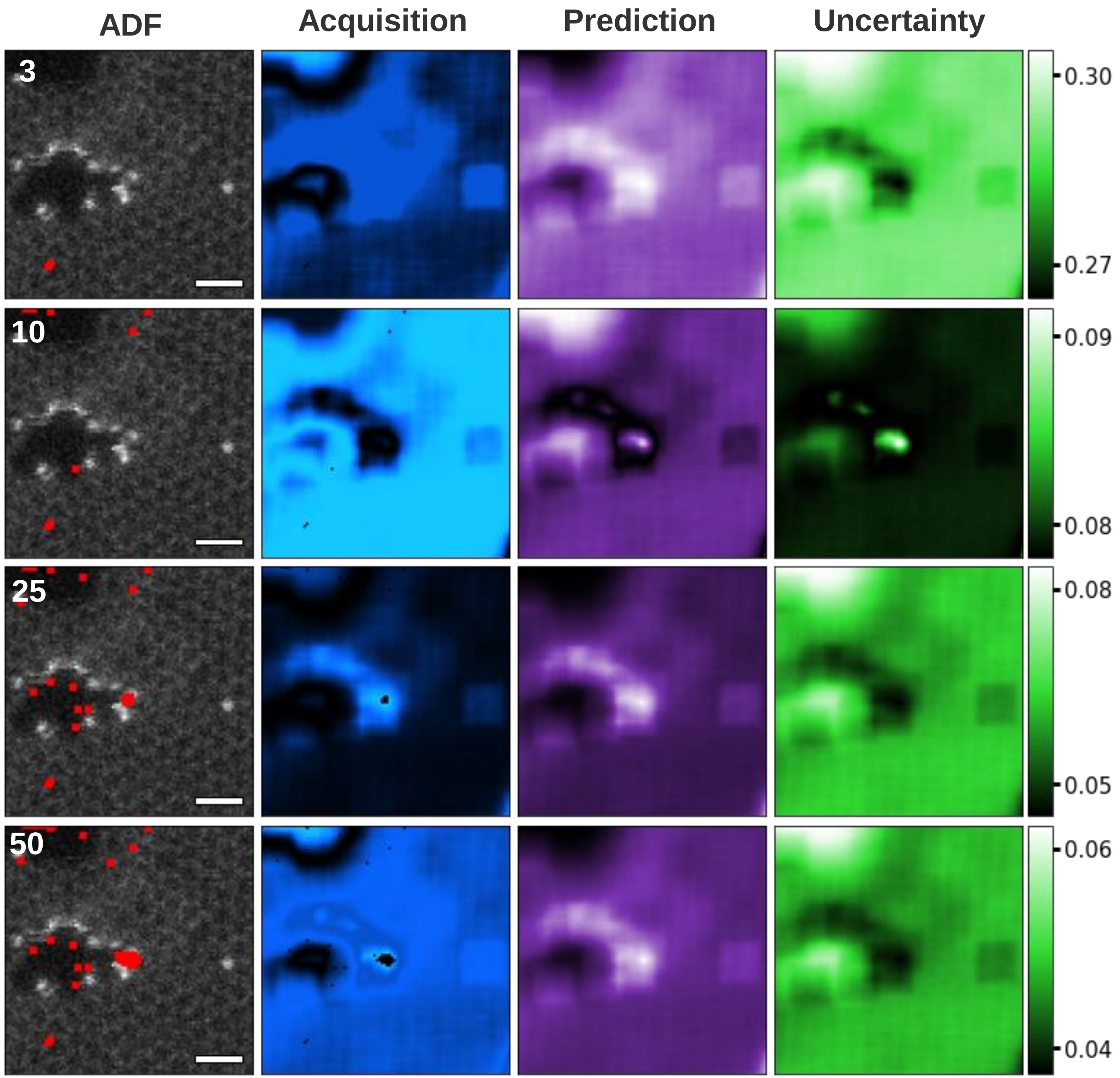


- Different **acquisition functions** can be used:
  - Expected Improvement (**EI**) (usually what was used)
  - Upper Confidence Bound (**UCB**), etc
- Usually based on some combination of **prediction** and **uncertainty**.

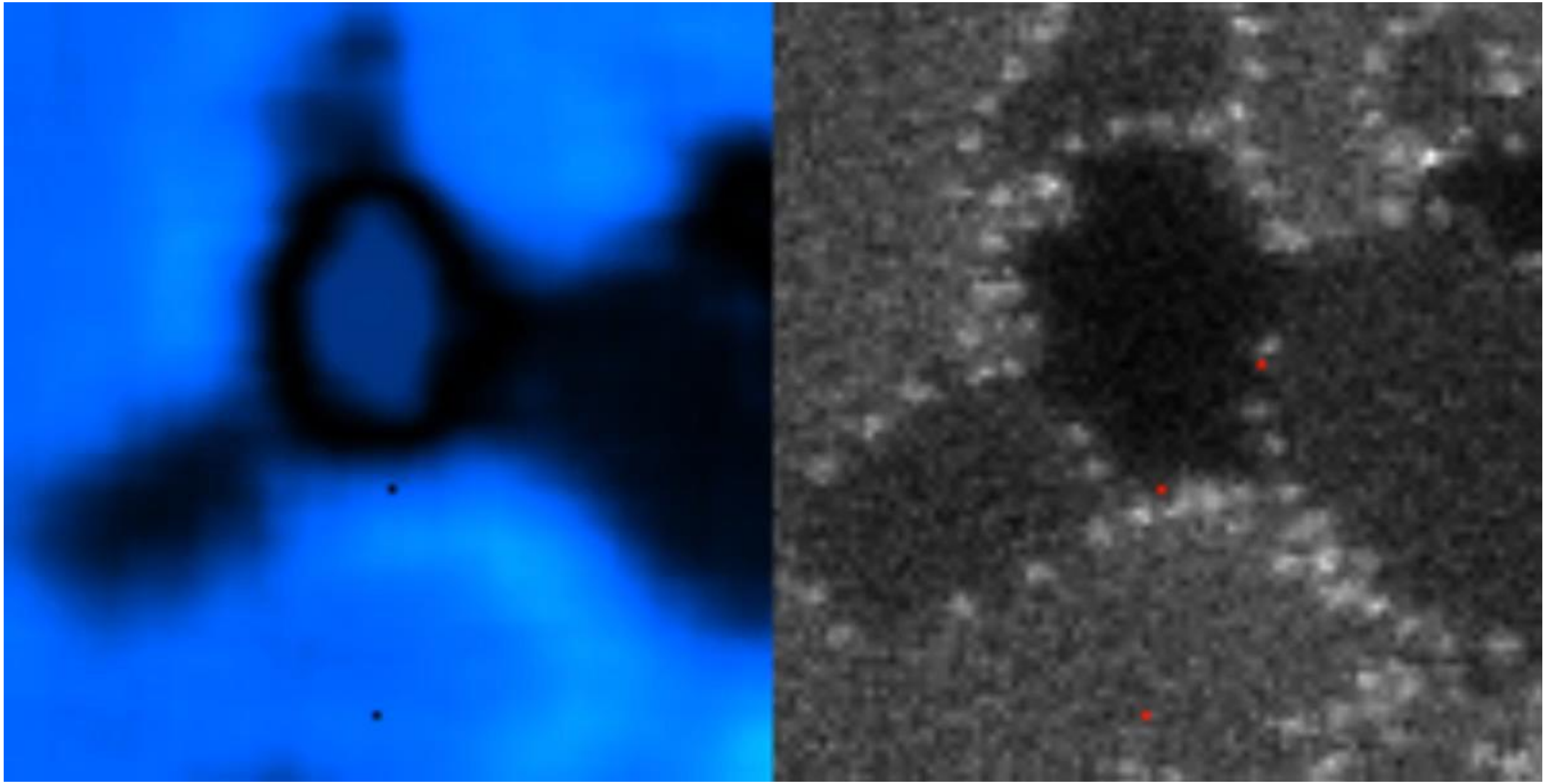
A closer look

Scalarizer: *CoM* magnitude

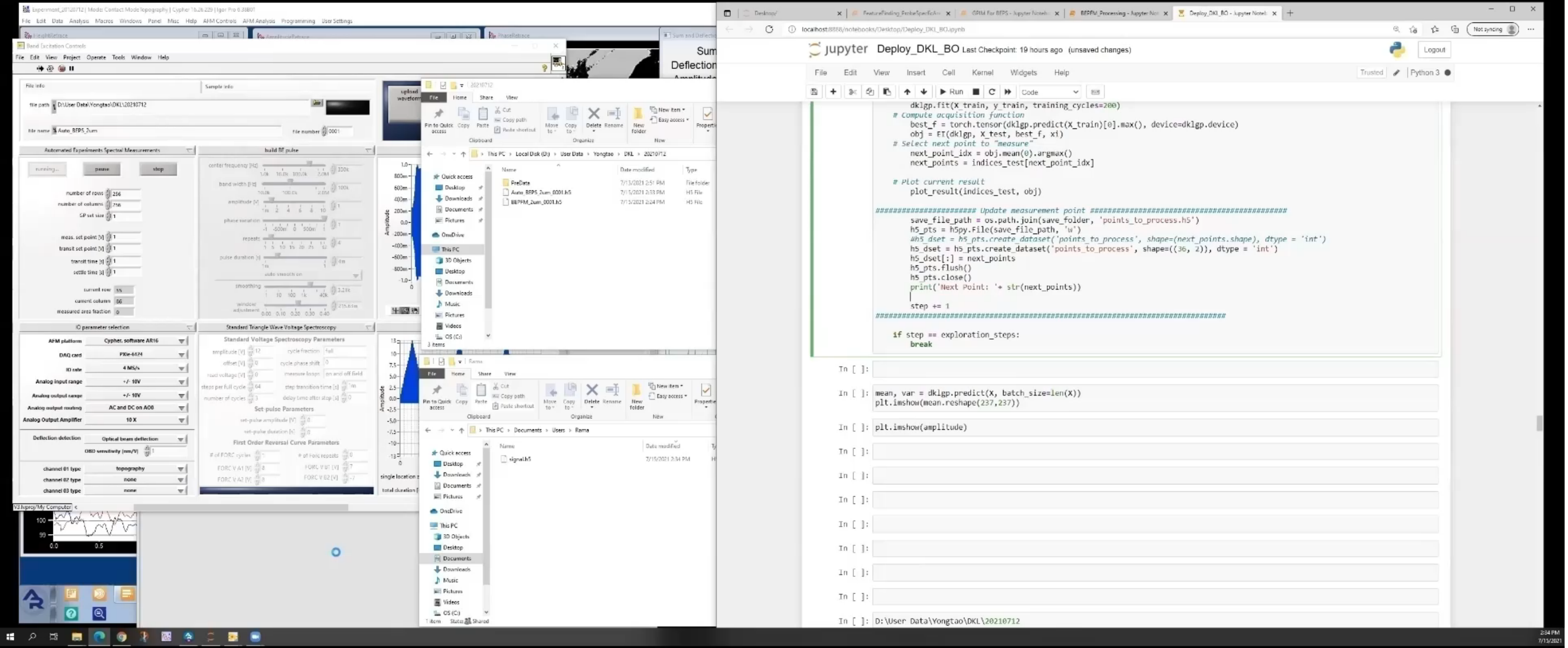
- High uncertainty @ start, but fairly quickly reduces
- Prediction actually doesn't drastically change throughout experiment
    - Structure-property relationship here is fairly rapidly learned
- Note the training can be halted after some criterion is met, making remainder of experiment go much quicker



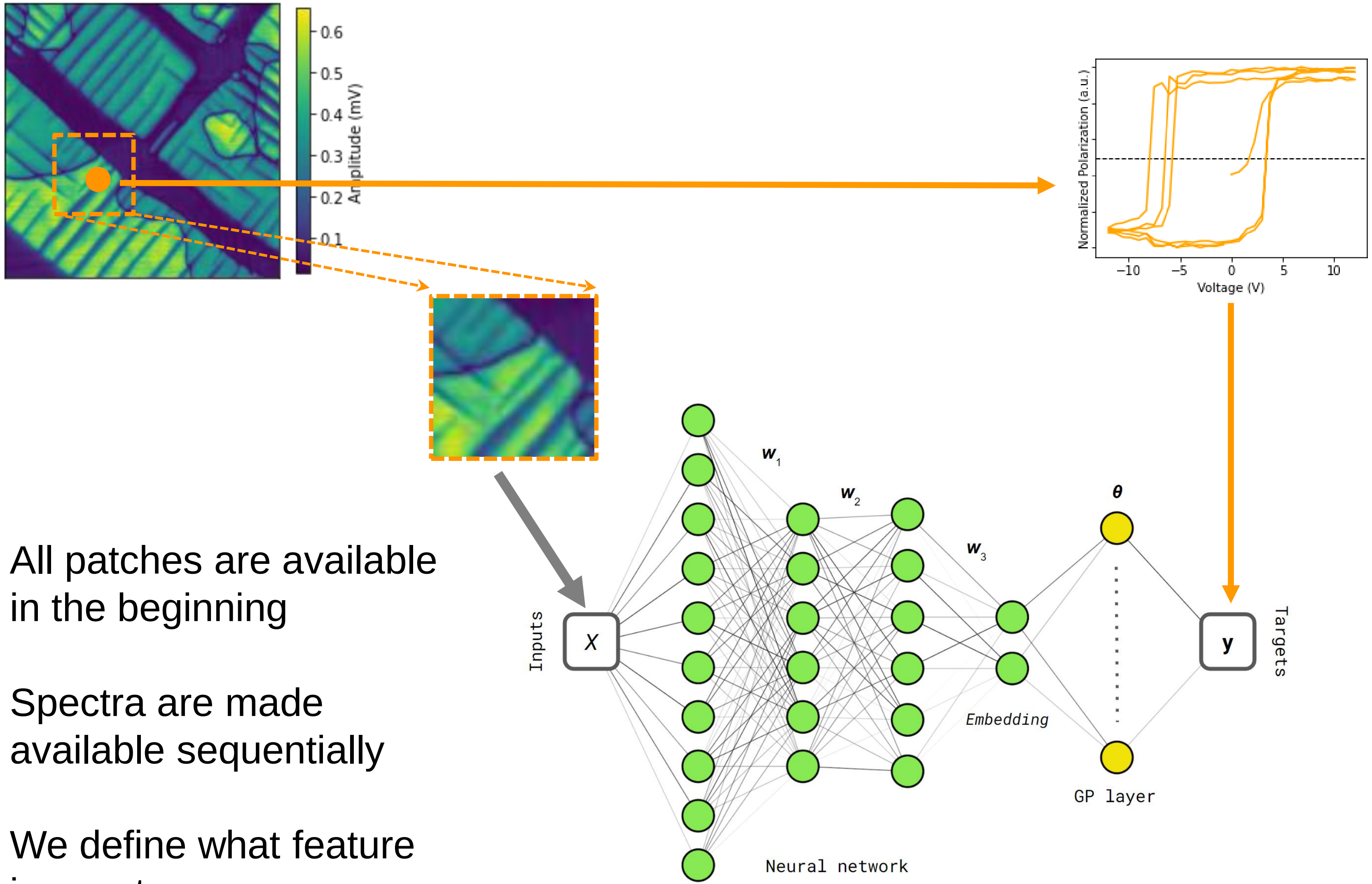
Does it always work?



Deep Kernel Learning AE



Deep Kernel Learning



<u>Specify physics criteria</u>

Acquire **structural data**

Measure a **spectrum**

Train DKL model with new data

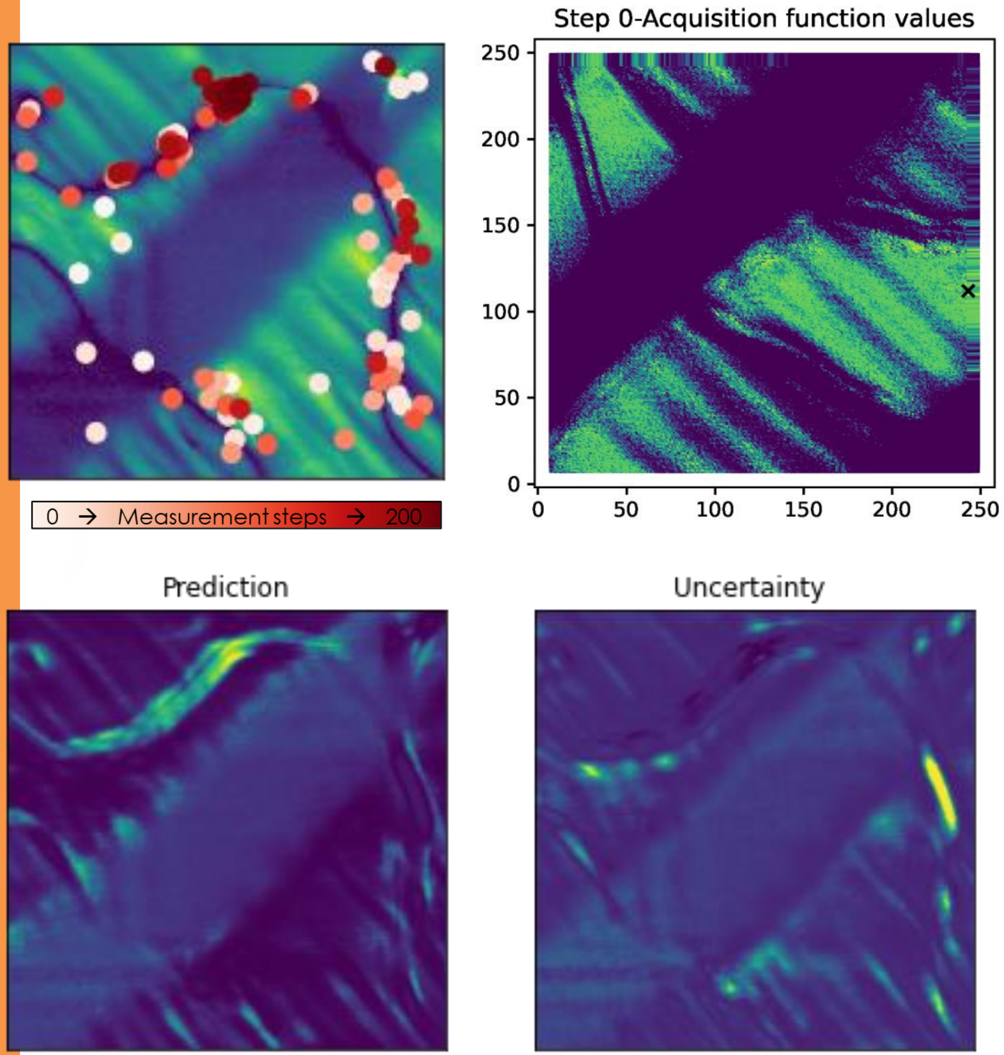
Decide next position (optimize physics criteria)

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

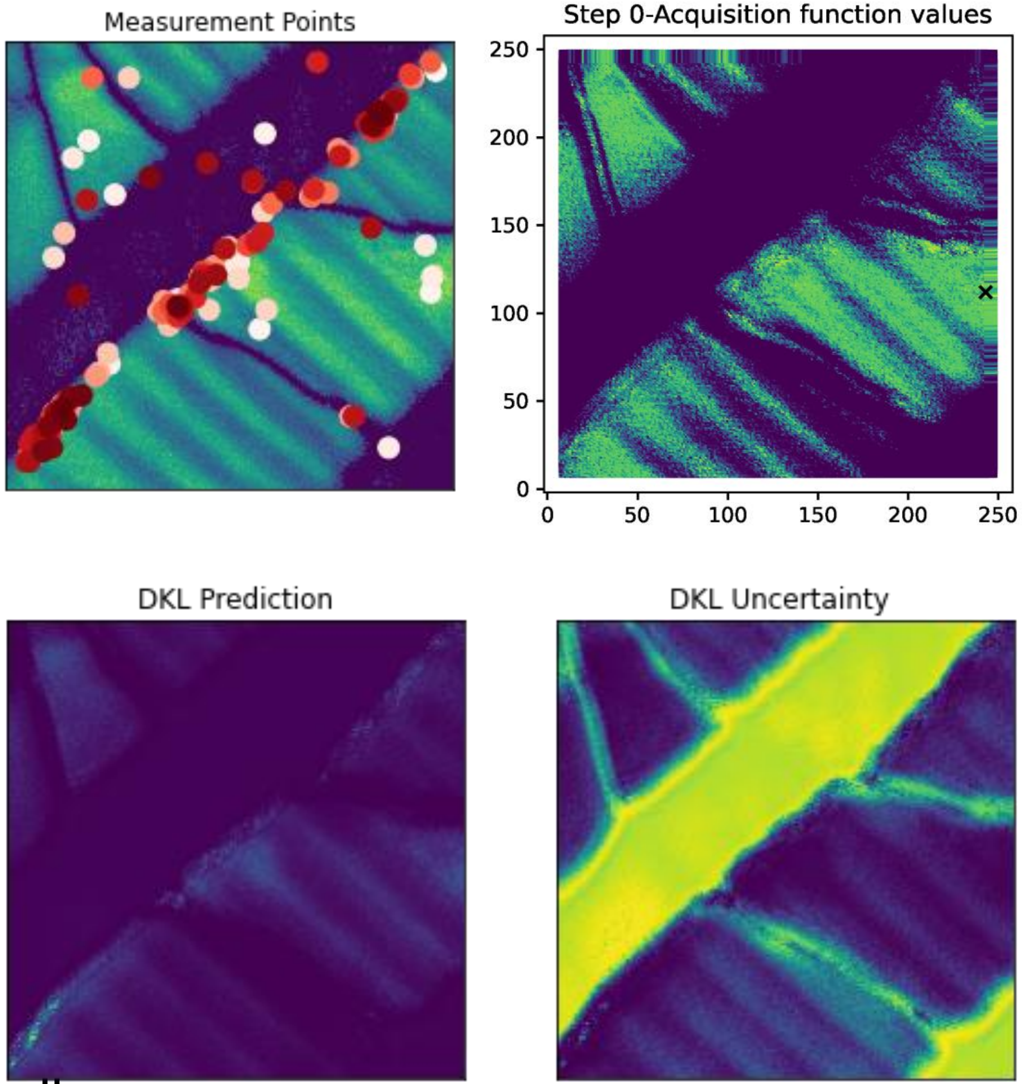
Allows navigation of the system to search for physics

Deep Kernel Learning SPM

Guided by: On field loop area



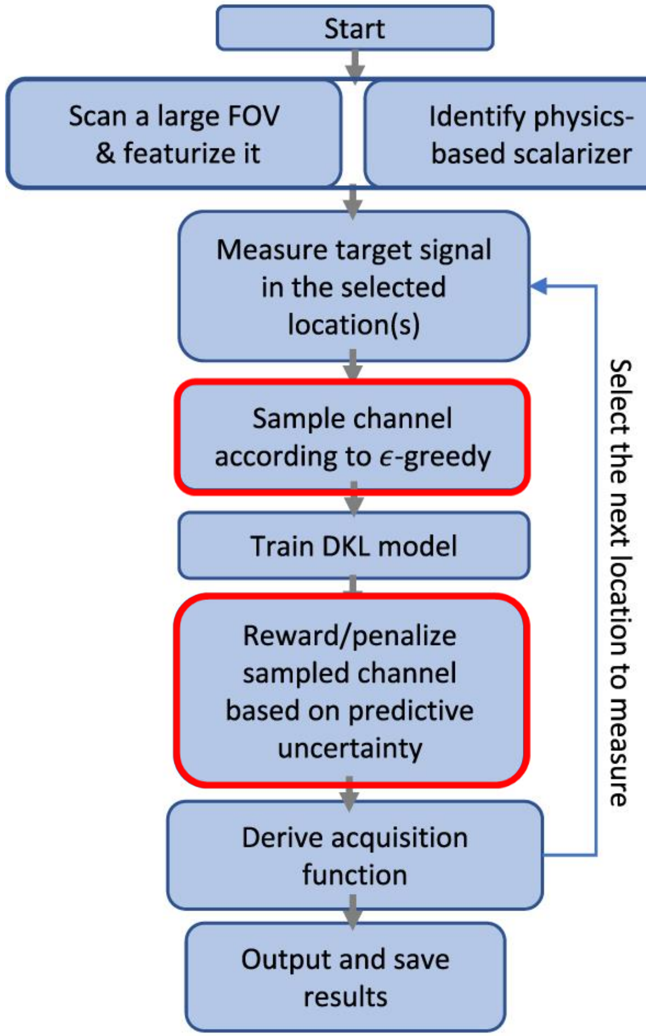
Guided by: Off field loop area

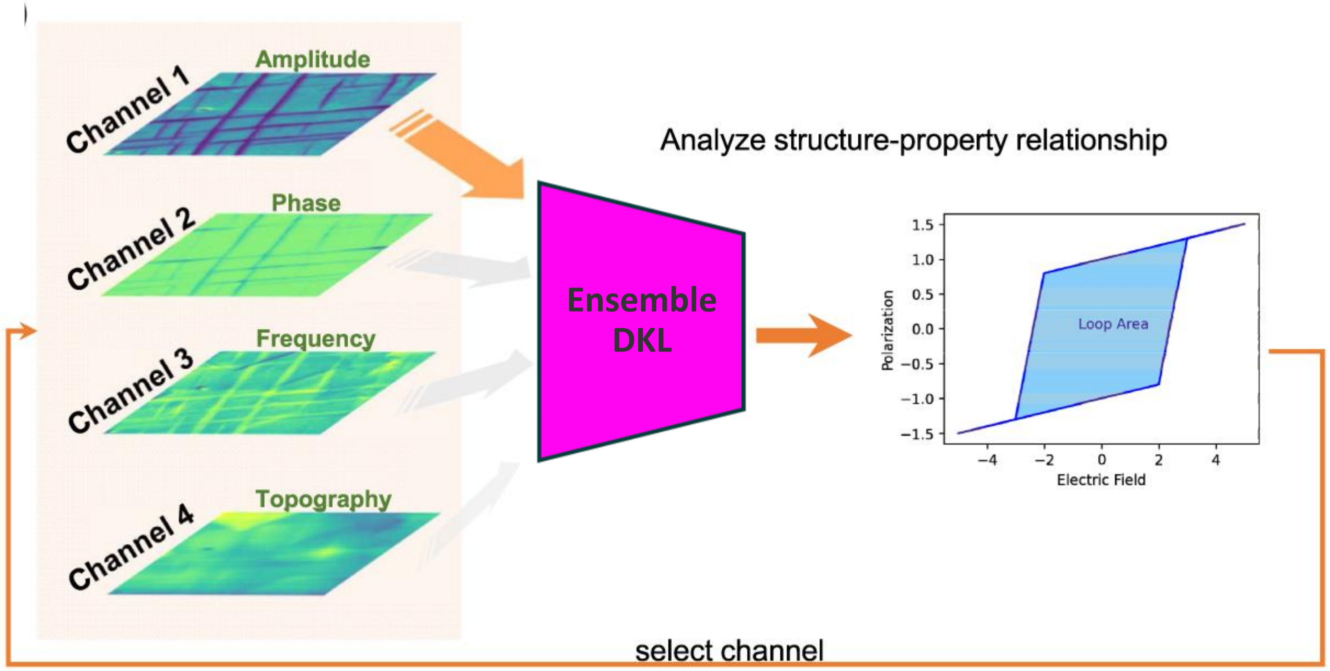


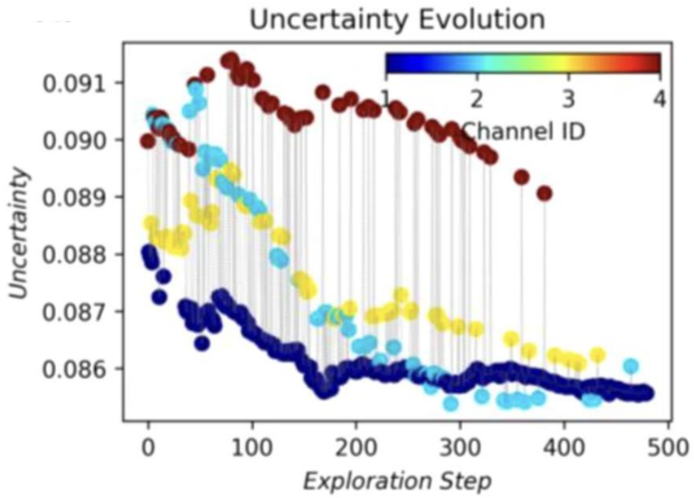
- Large loop opening corresponding 180º domain walls
- This behavior can be attributed to the large polarization mobility of 180º walls

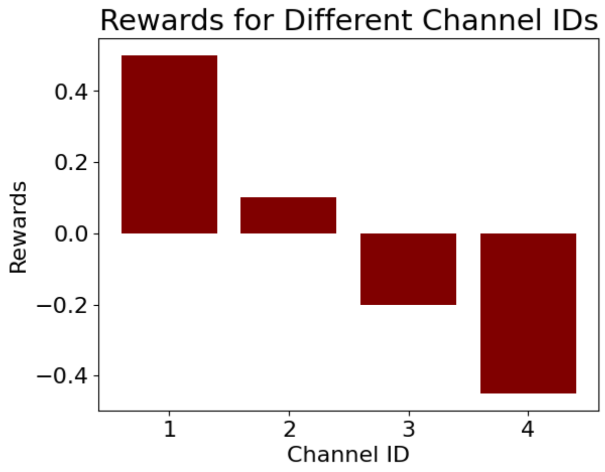
Liu, Yongtao, et al, Nature Machine Intelligence 4, 4 (2022): 341-350.

DKL: Learning the best channel



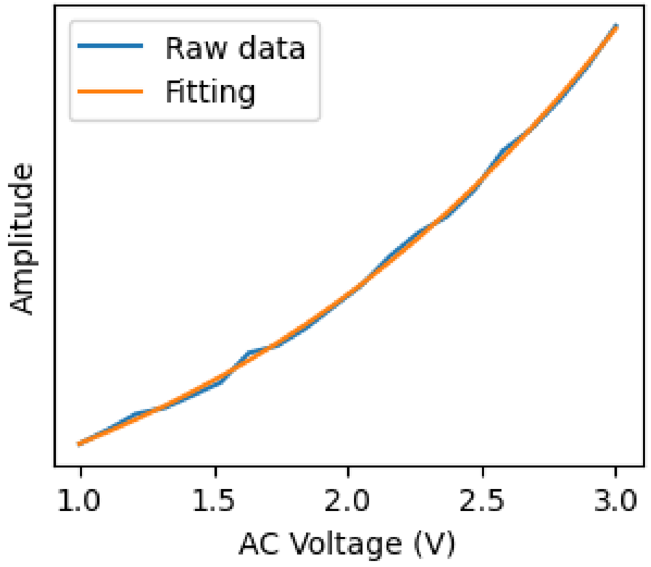


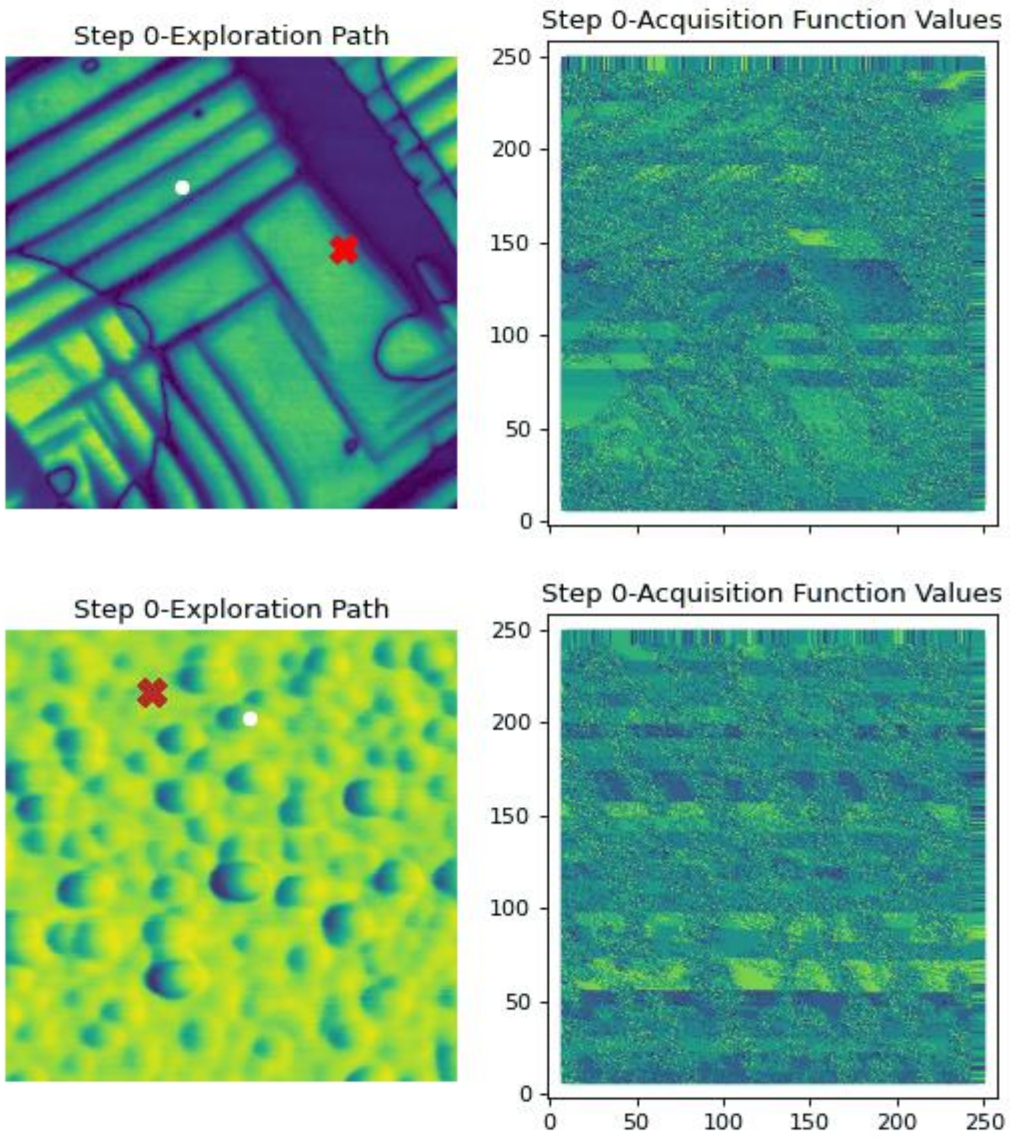




Y. Liu et al. npj Comput Mater **9**, 34 (2023).

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 ?

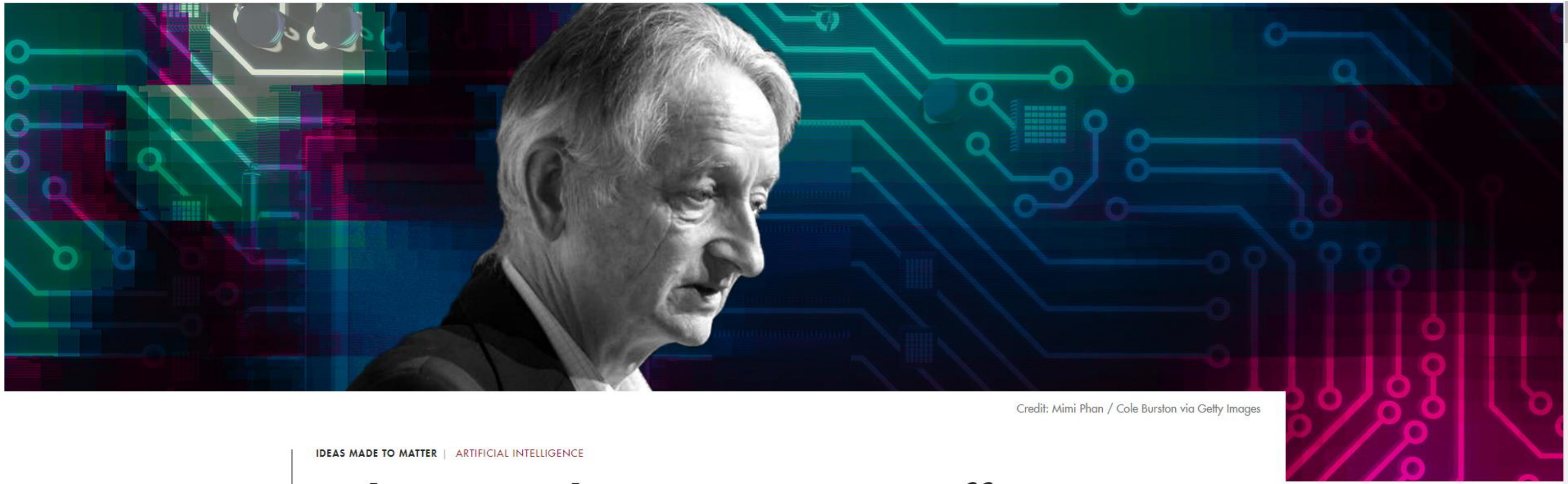
Taking the Human Out of the Loop: A Review of Bayesian Optimization

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

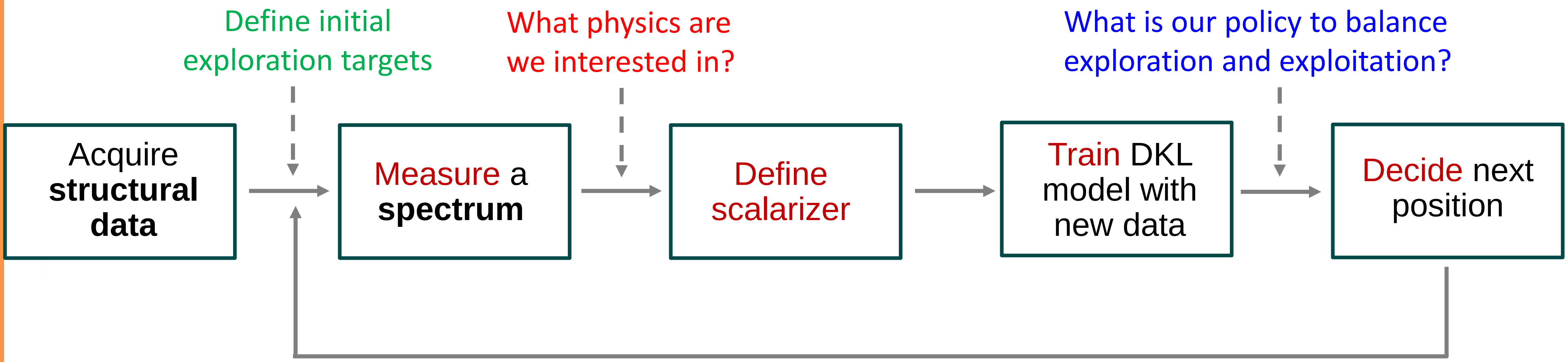


Credit: Mimi Phan / Cole Burston via Getty Images

IDEAS MADE TO MATTER | ARTIFICIAL INTELLIGENCE

Why neural net pioneer Geoffrey Hinton is sounding the alarm on AI

Bringing Human into the Loop



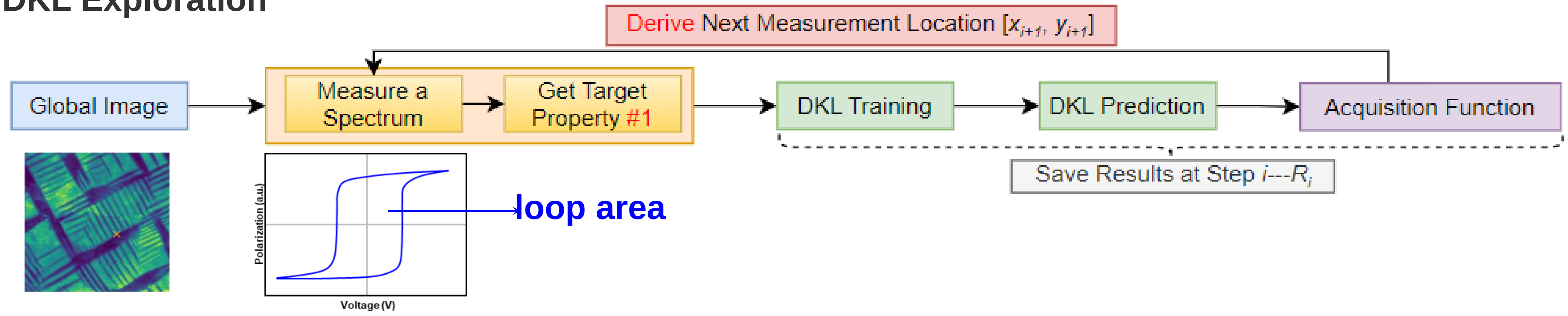
Key concepts:

- **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

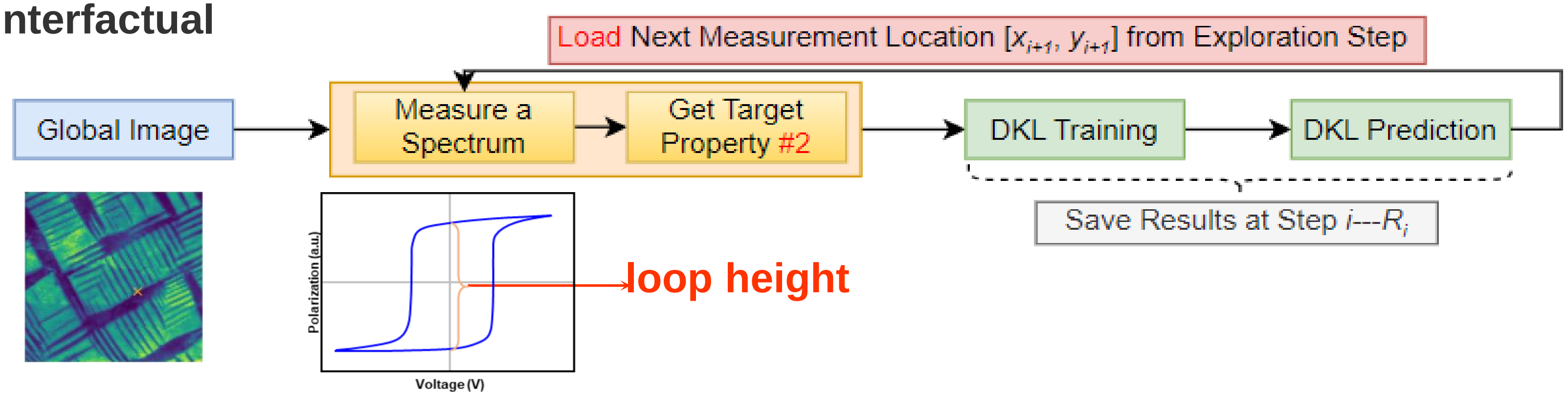
- **Experimental trace:** collection of image patches and associated spectra acquired during experiment. Note that we collect spectra, not only scalarizers

Counterfactual scalarizers

DKL Exploration



DKL Counterfactual



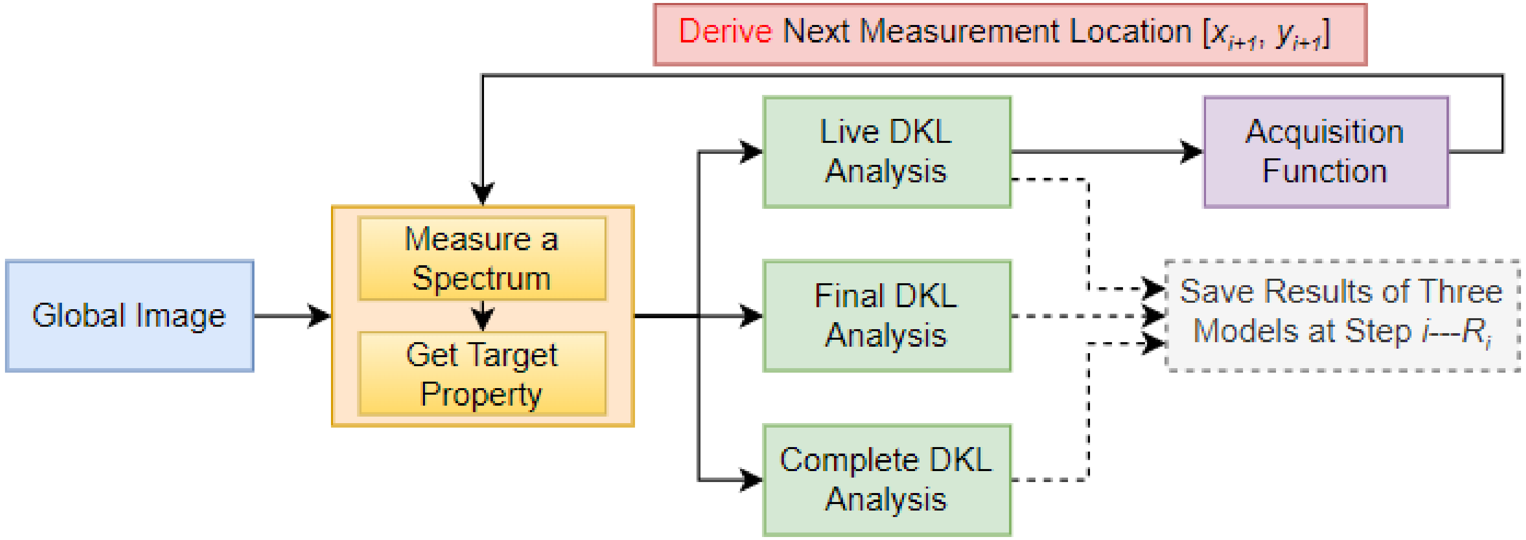
Target properties:

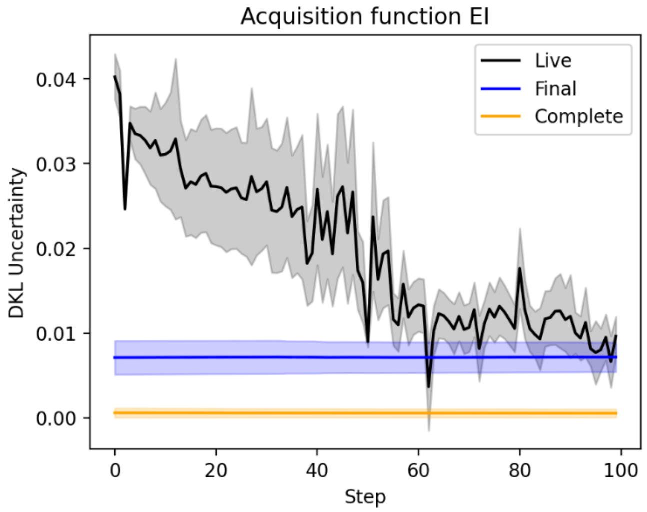
1. Loop Area
2. Loop Height
3. Coercive Field
4. …

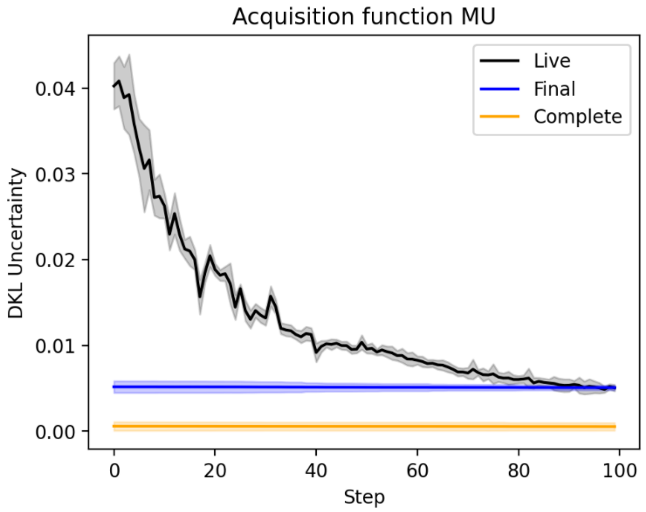
- We save the full experimental trace
- What if we follow the actual experimental path – but calculate alternative (counterfactual) scalarizers?

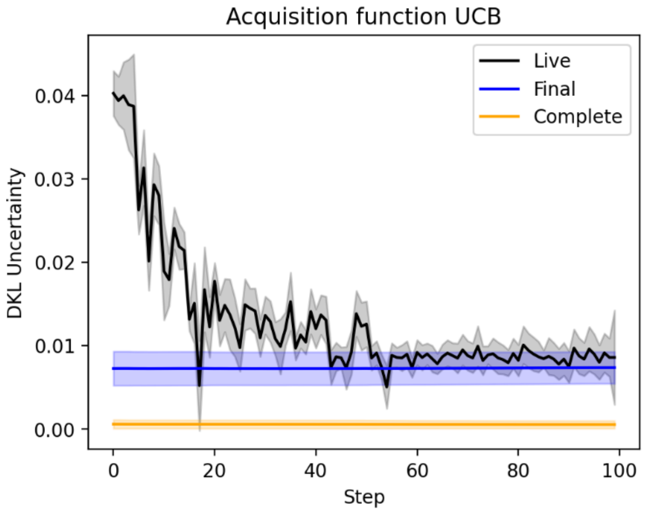
Explainable AE

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

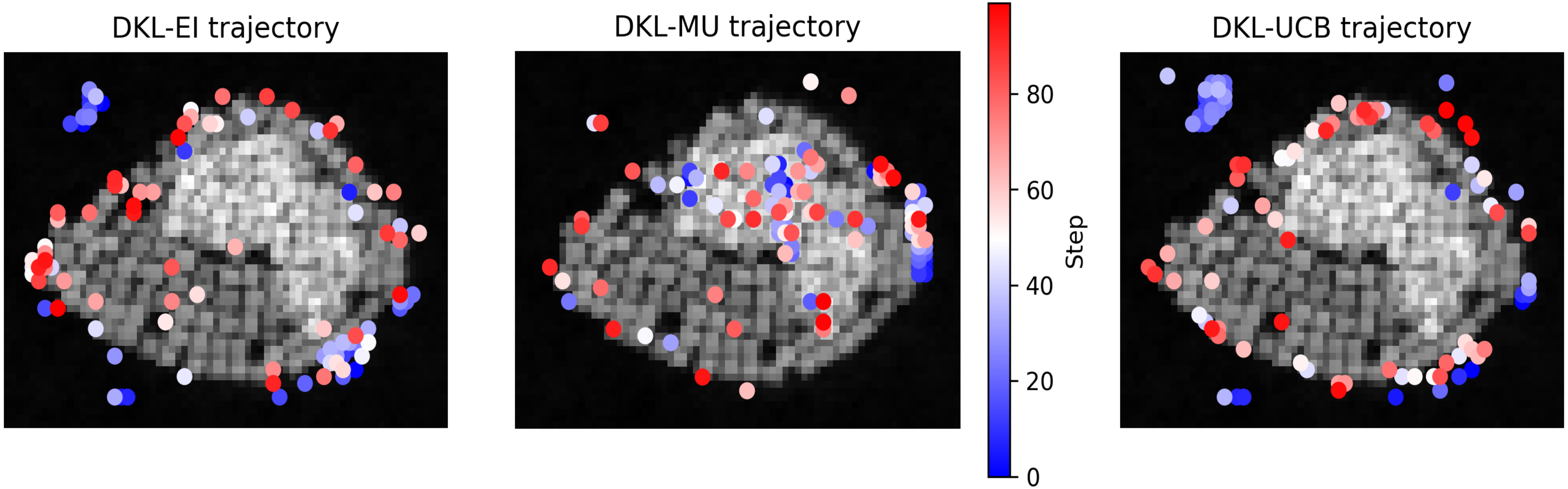






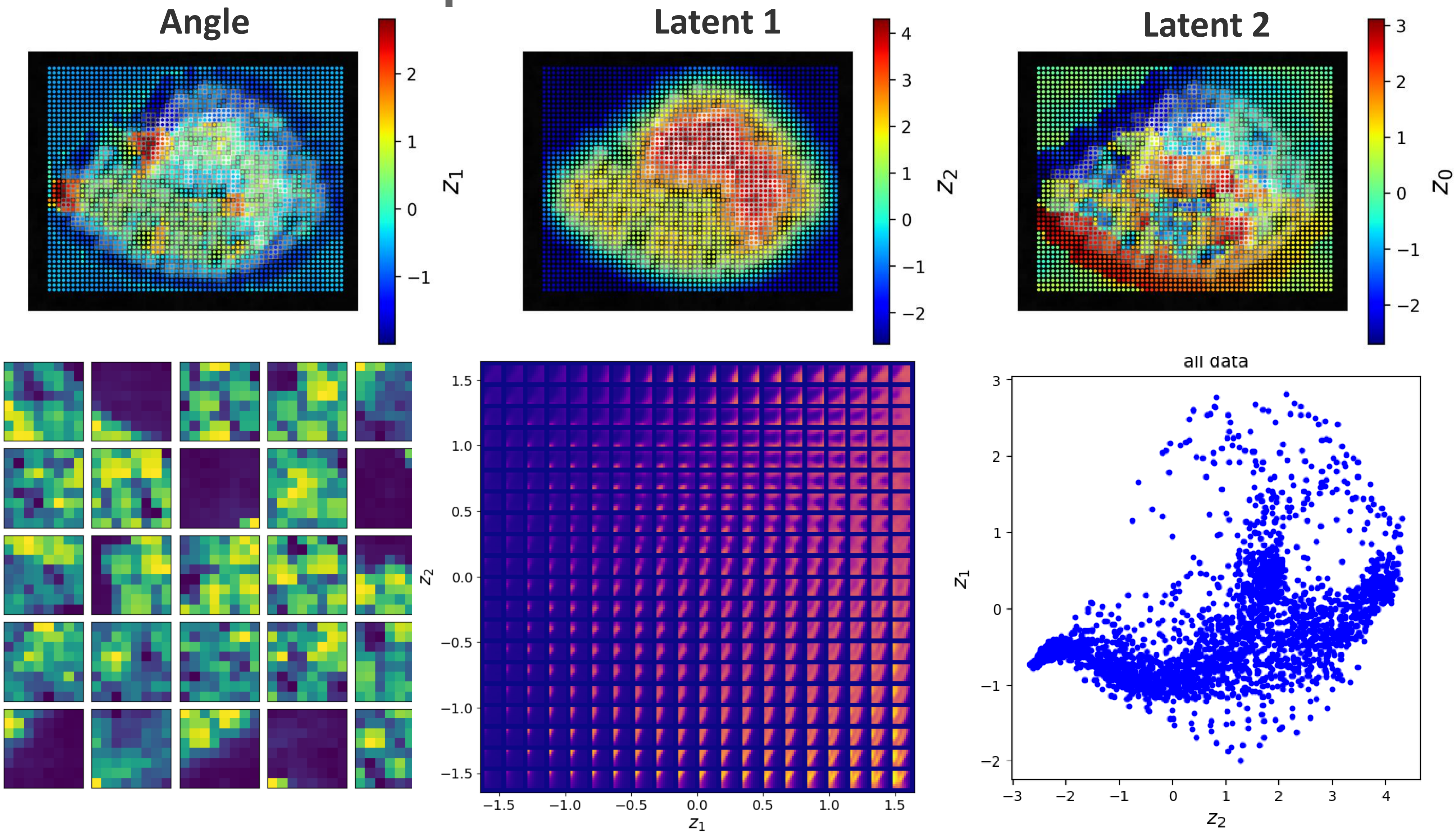


Monitoring the AE



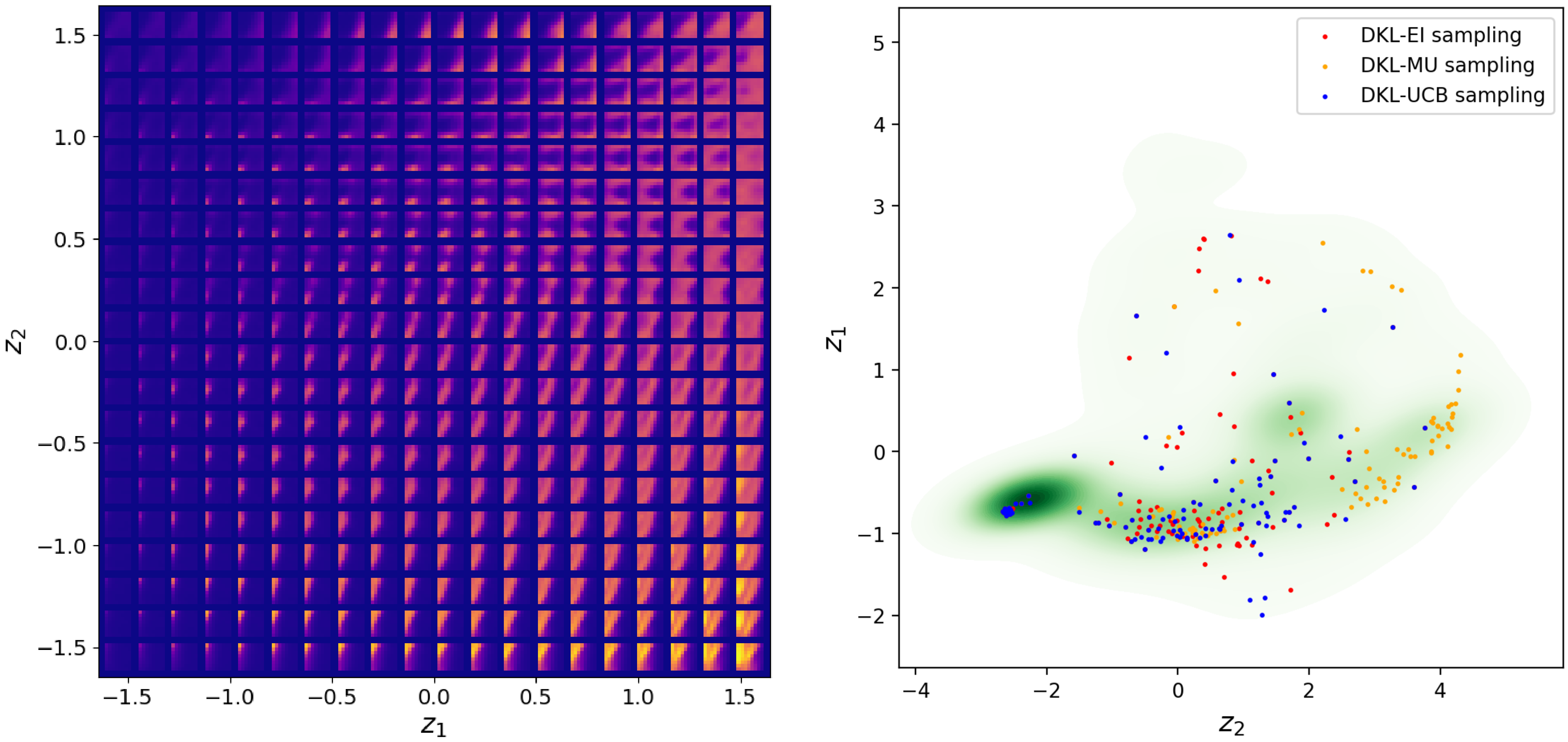
- Different acquisition functions (policies) give different experimental paths for AE
- Can we analyze what is special about points visited?

Global Feature Space

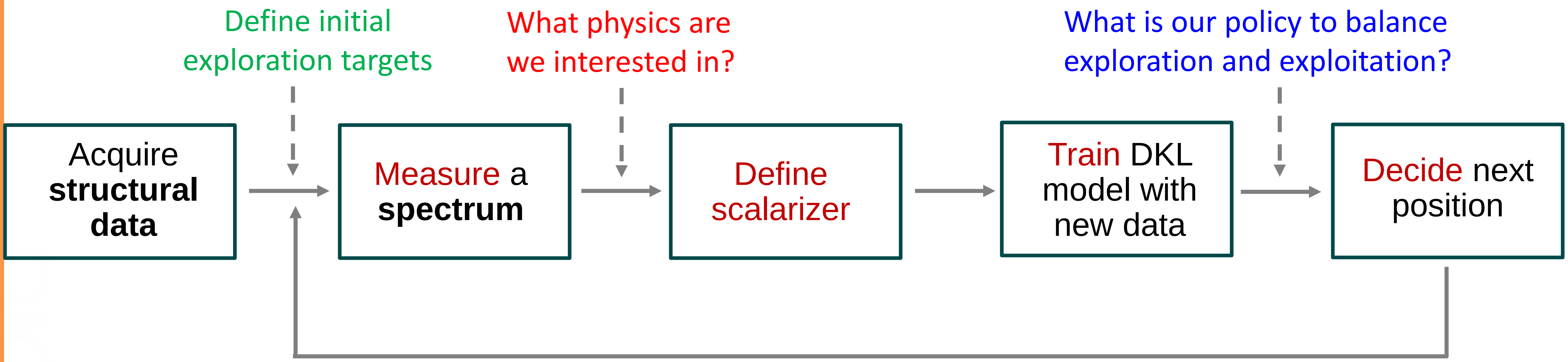


- Global feature space is available from the beginning of the experiment and is stationary
- Experimental trajectories can be visualized in the global feature space

VAE approach: full feature space



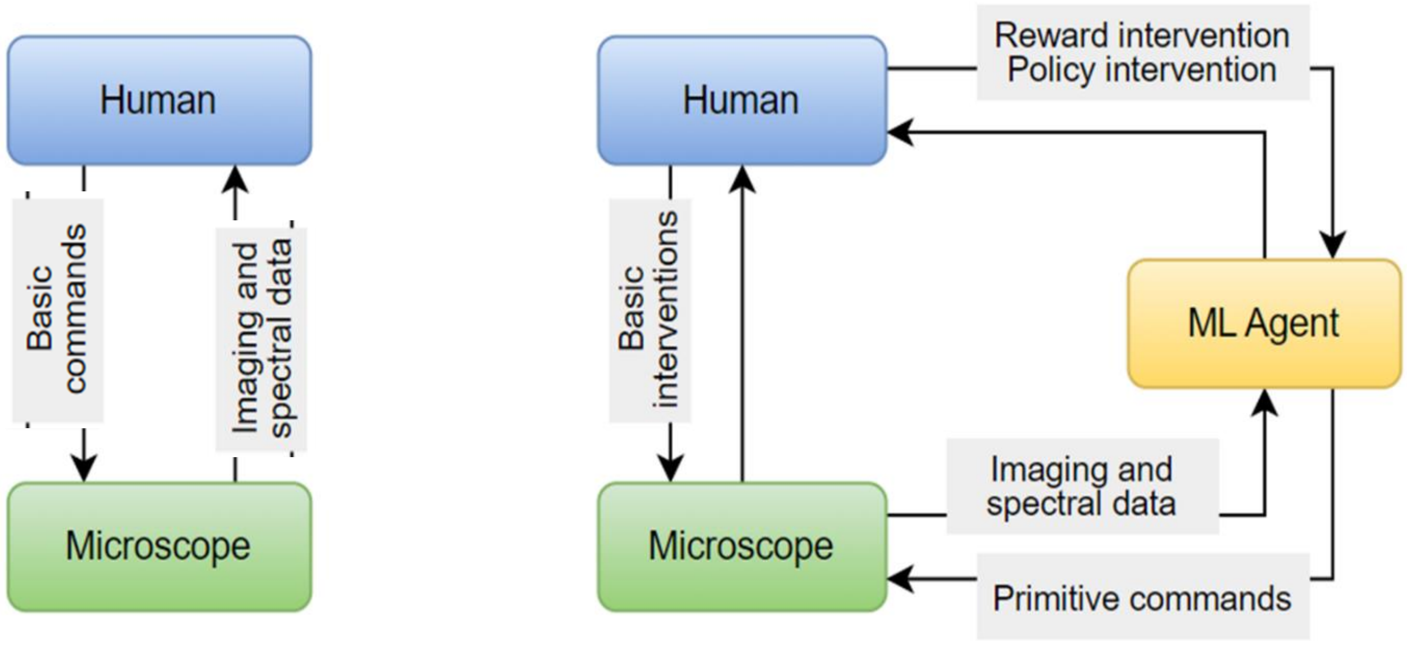
Bringing Human into the Loop

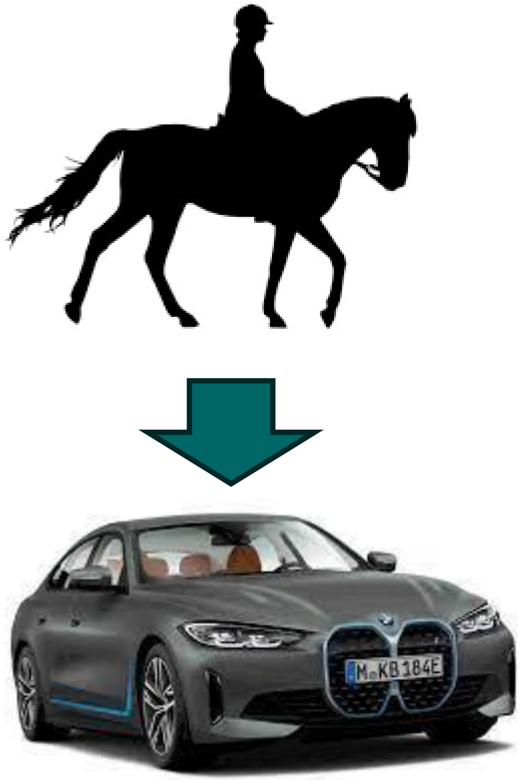


We can intervene on:

- **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

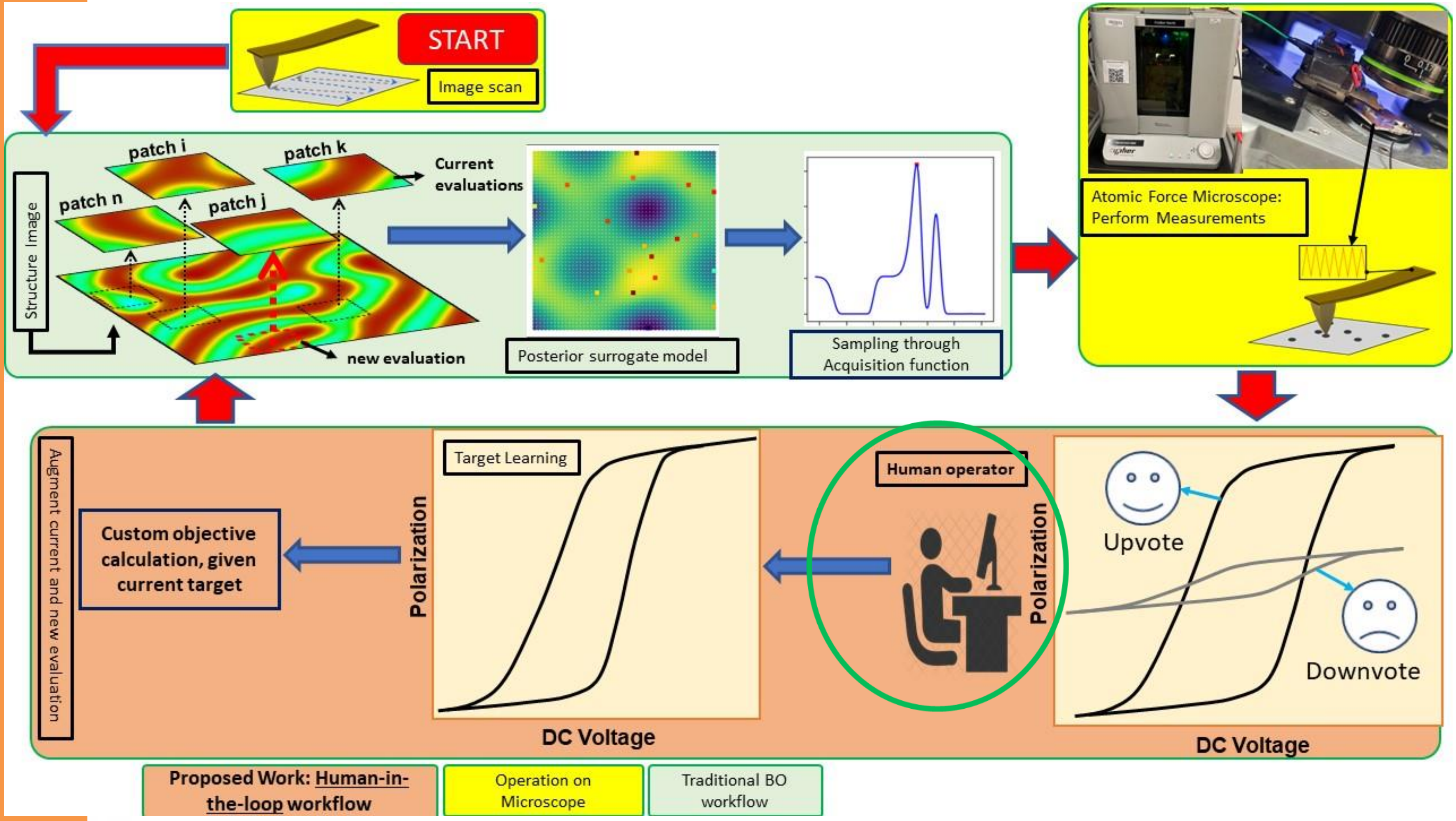




We can intervene on:

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

BOARS: Human (partially) in the loop



Arpan Biswas

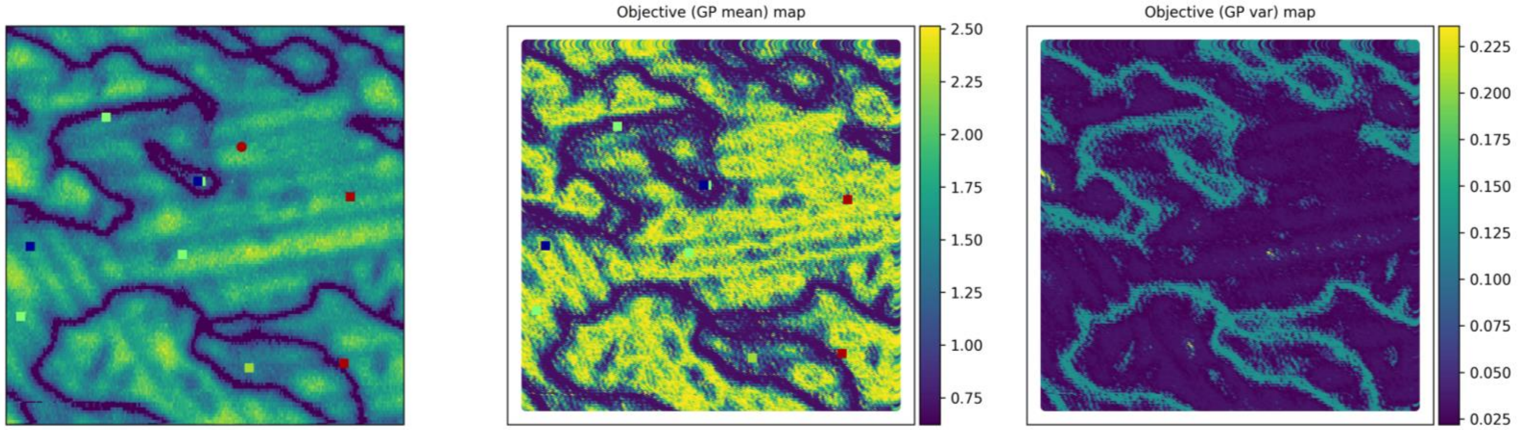
Bayesian optimized Active Recommender System (BOARS)

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)
```

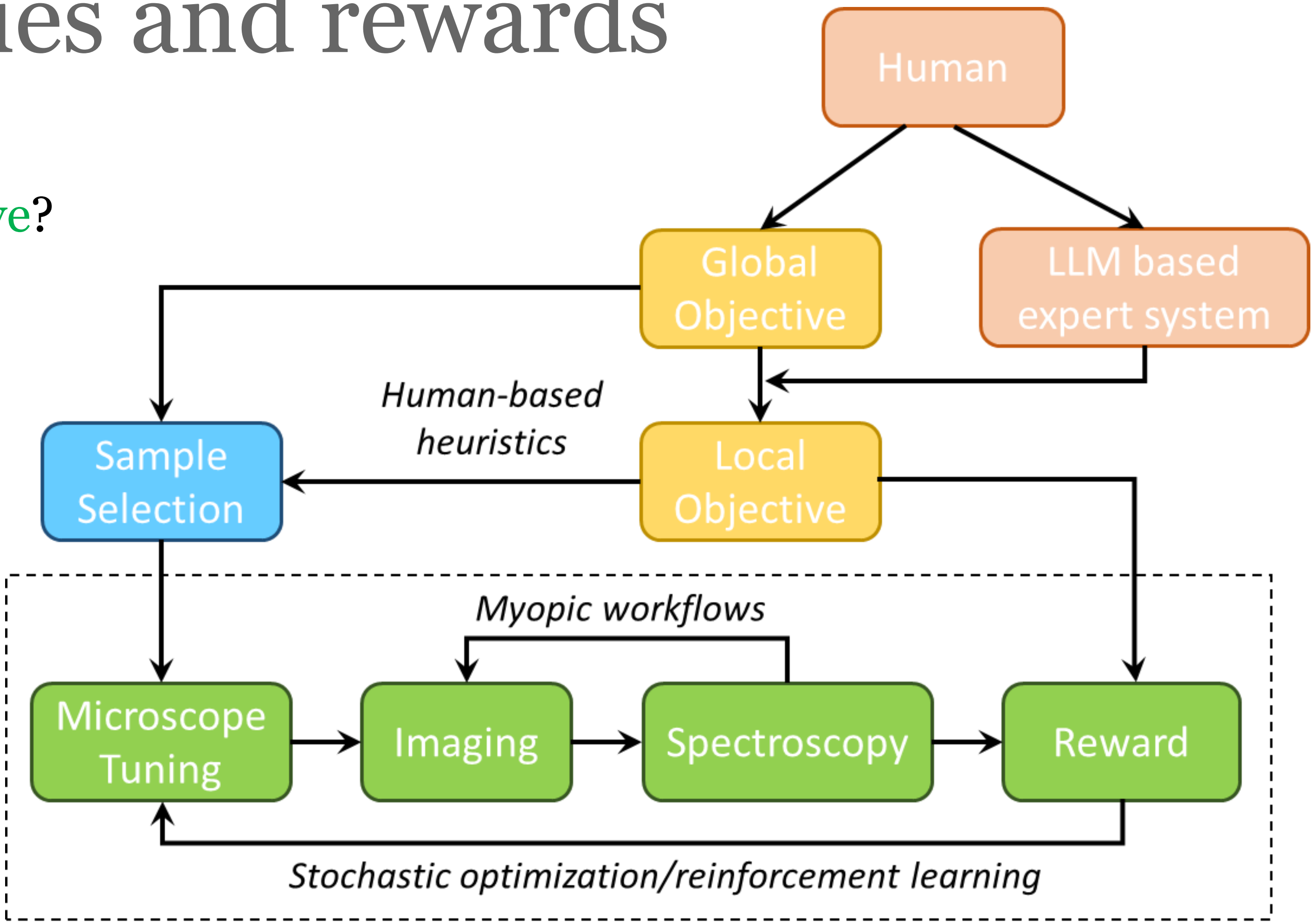
An implementation to SPM



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?