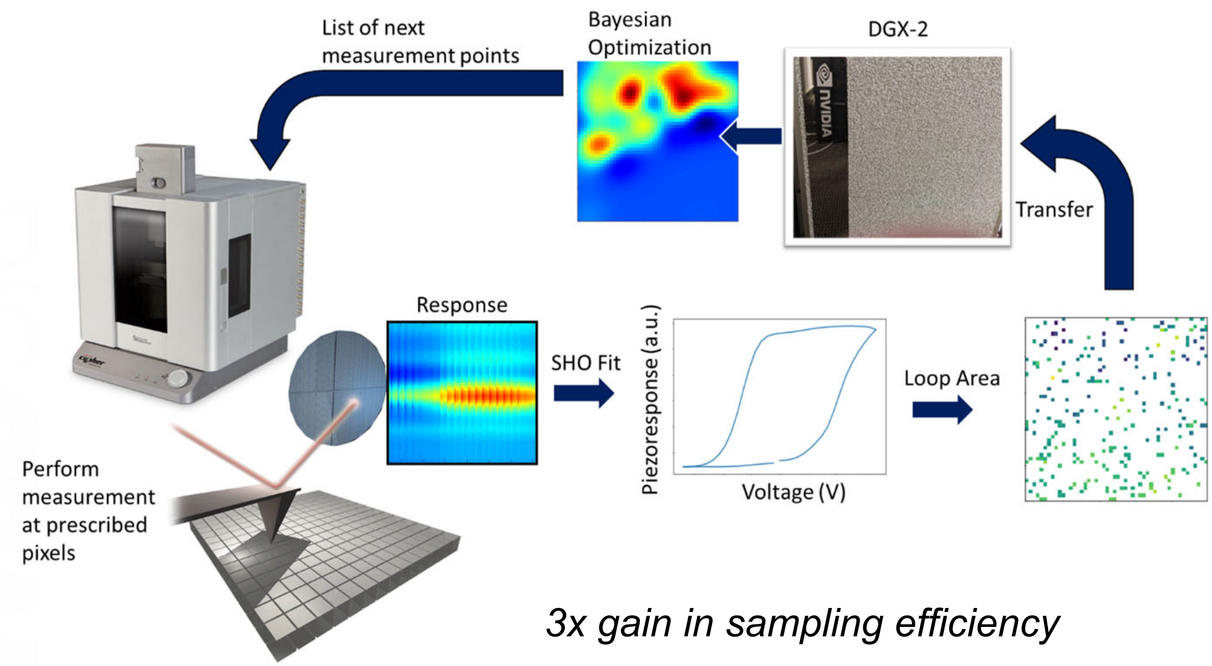
THE UNIVERSITY of TENNESSEE KNOXVILLE

Day 5: Deep Kernel Learning and Human in the Loop Automated Experiment

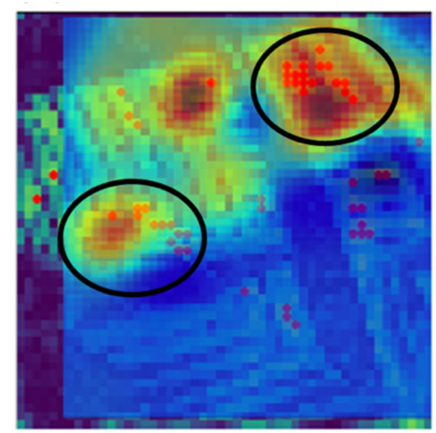
Instructor: Sergei V. Kalinin

BO for Self-Driving Microscope

First implementation of self-driving microscope: 2020

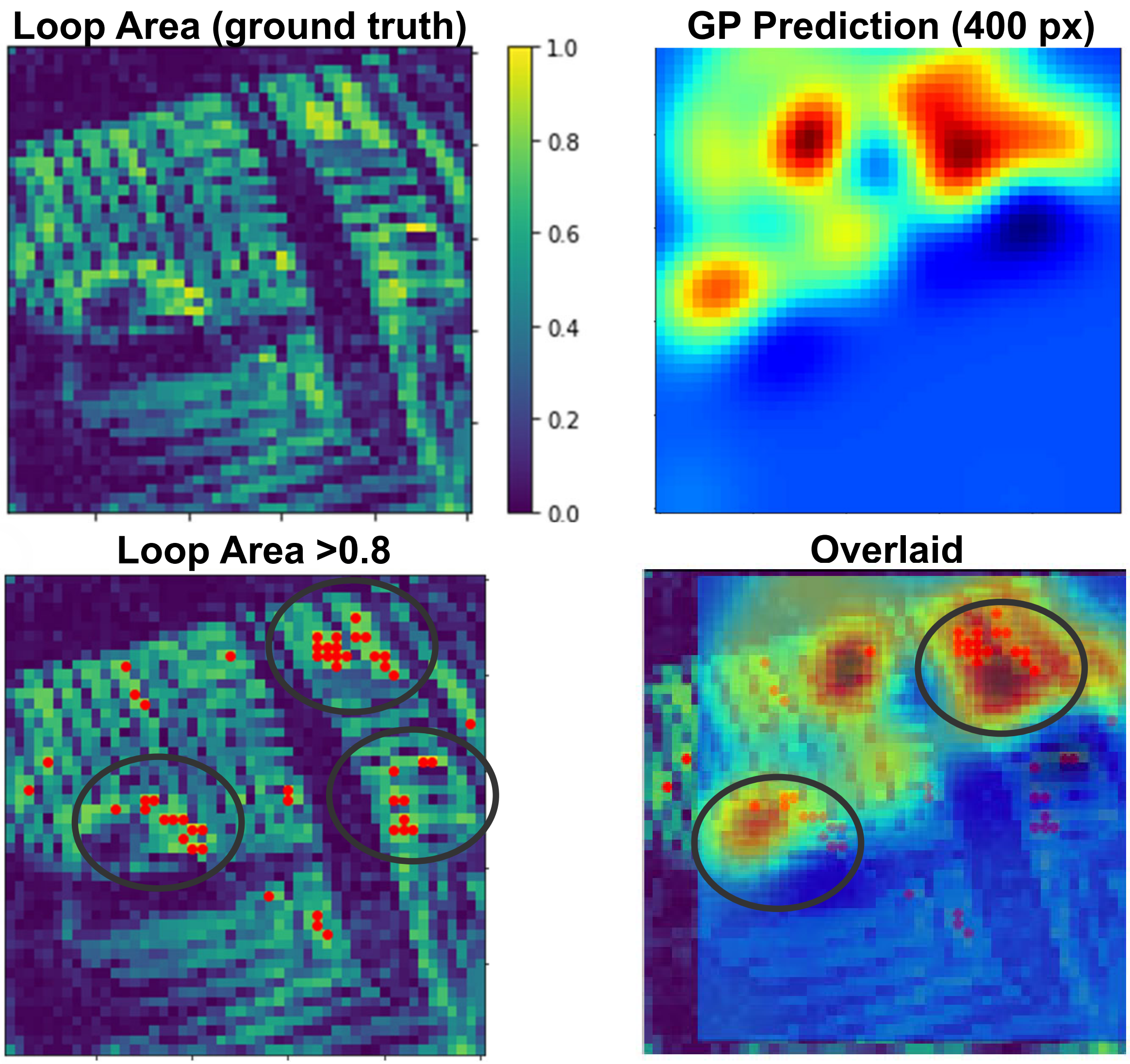


Comparison with "ground truth"



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*



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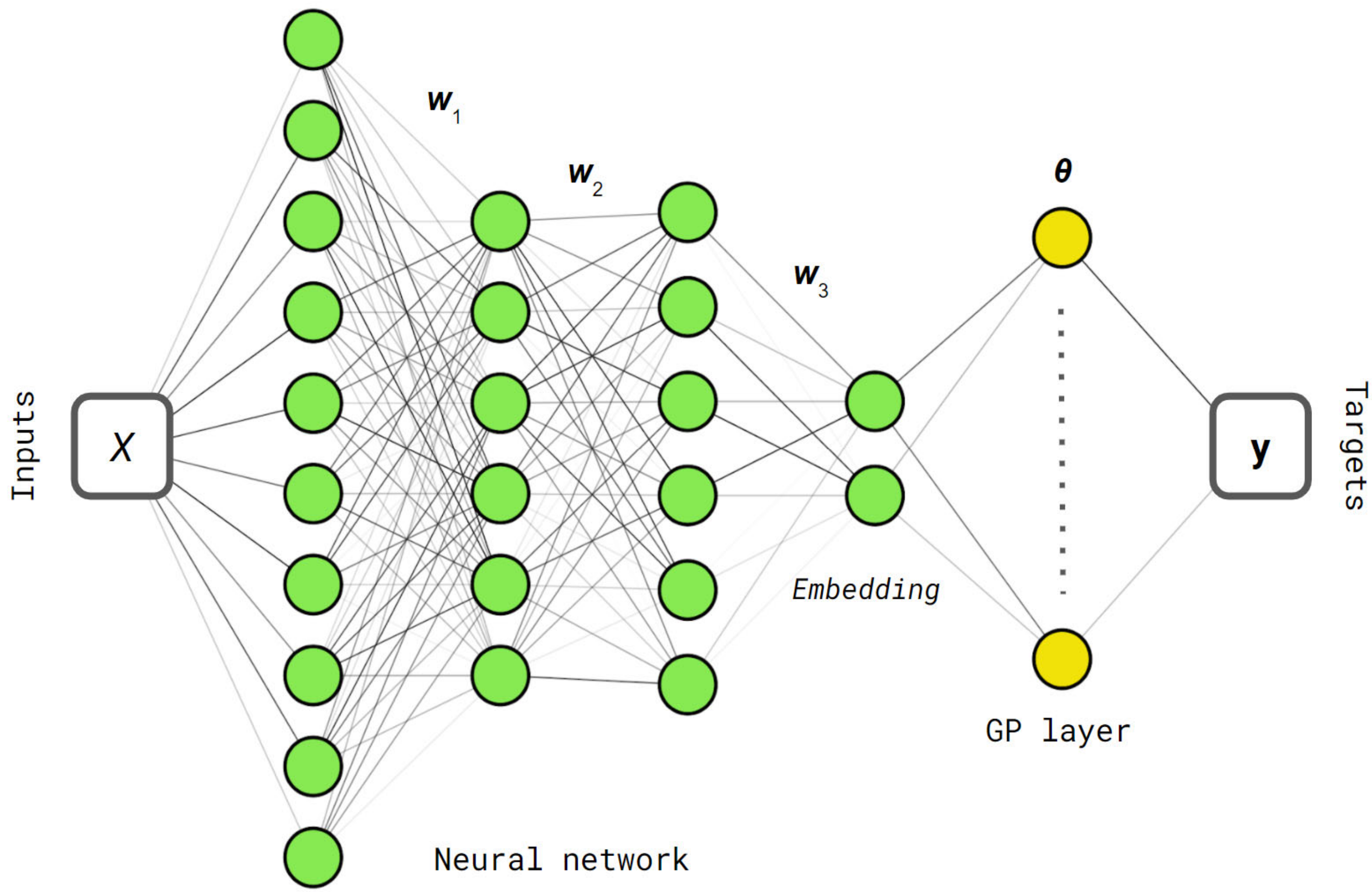
The application of simple data-driven GP for real world scenarios did not work particularly well.

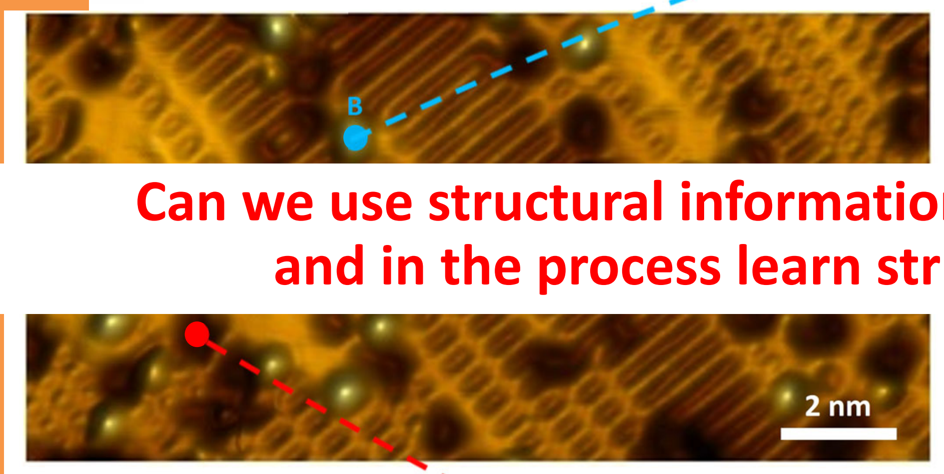
We were solving wrong problem!

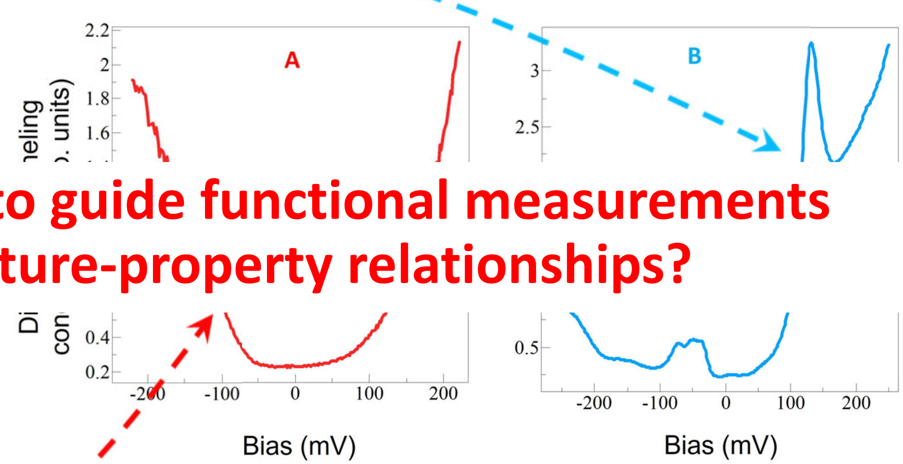
Deep Kernel Learning



Two modes of operations

Structural imaging (Cheap)	**Functional imaging (Costly)**
Topography in STM, amplitude/phase in SPM, (HA)ADF-image in STEM, etc.	Scanning tunneling spectroscopy (STS), polarization loops in SPM, EELS in STEM, etc.
These are FAST measurements (from seconds to minutes)	These are SLOW and/or DESTRUCTIVE measurements (from minutes to days)

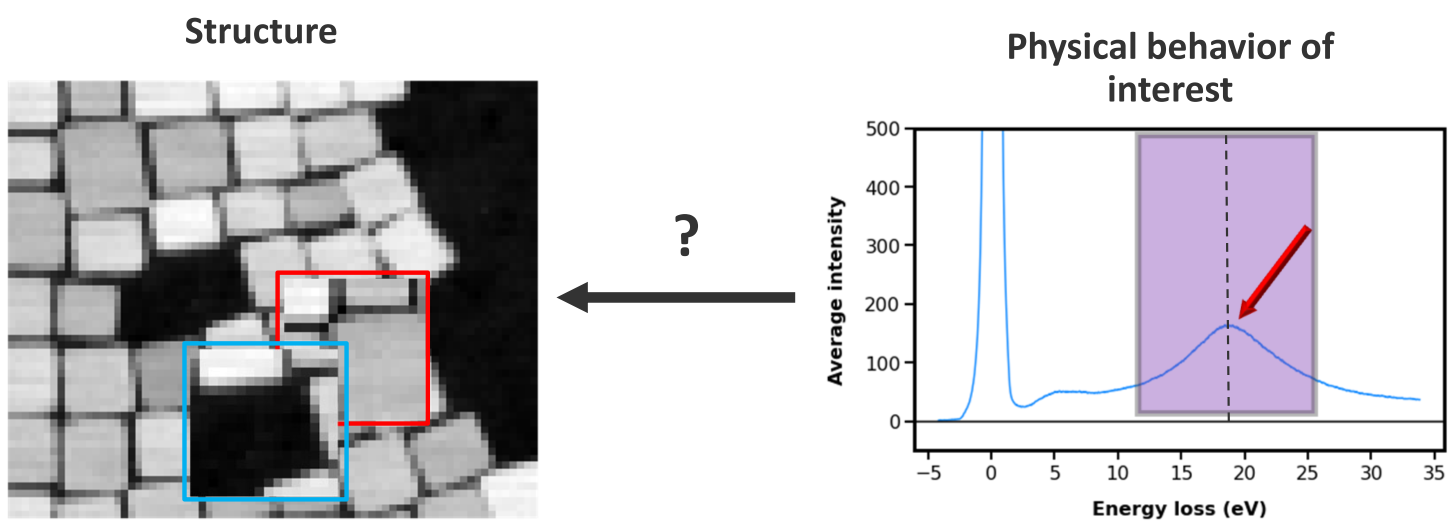




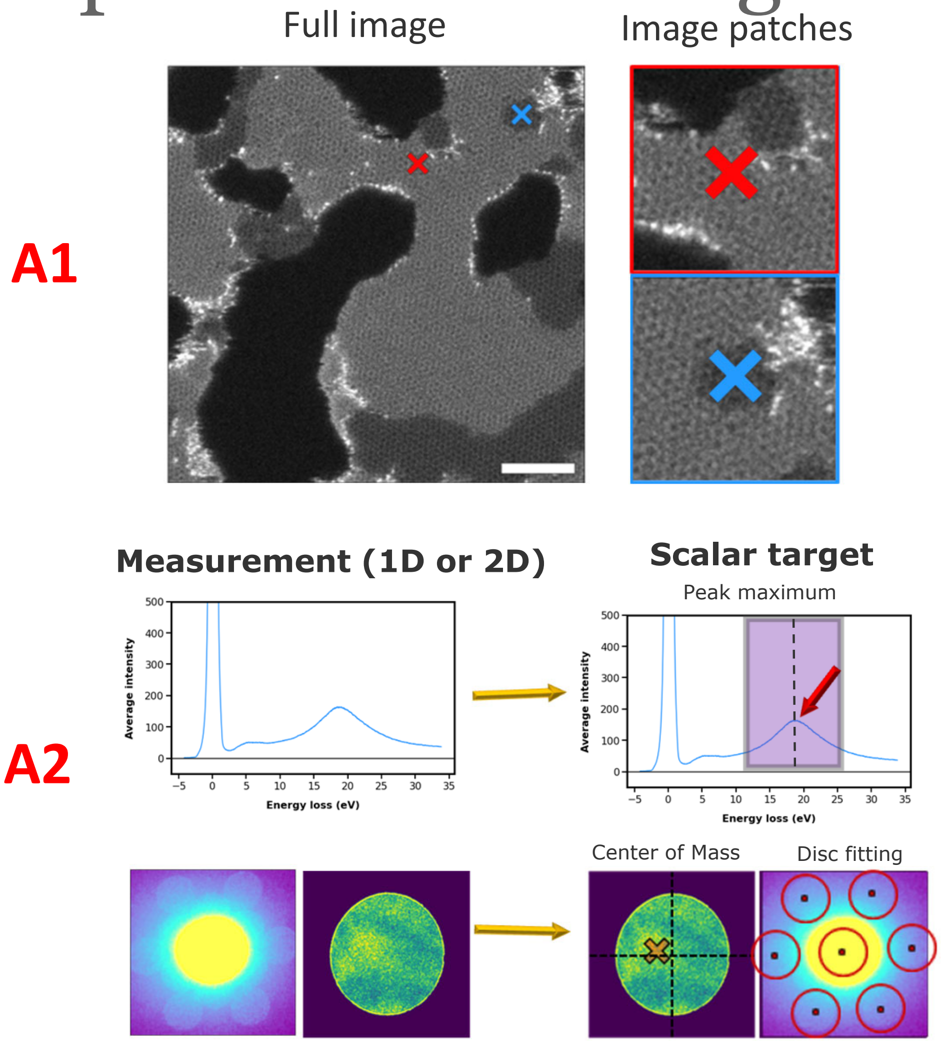
Can we use structural information to guide functional measurements and in the process learn structure-property relationships?

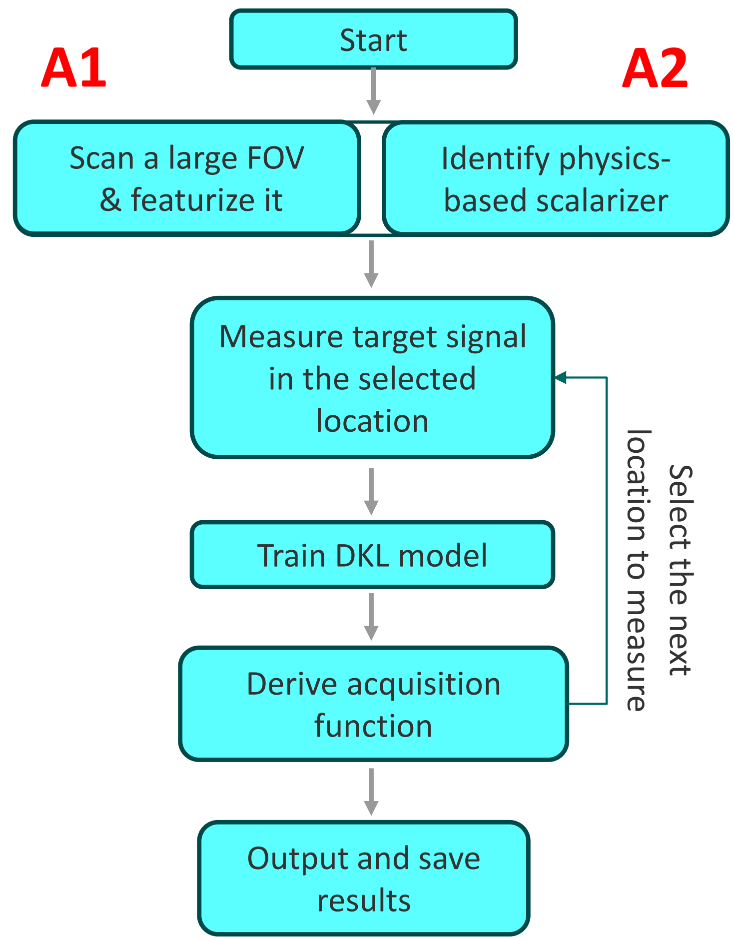
Physics discovery in active experiments

- 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**)

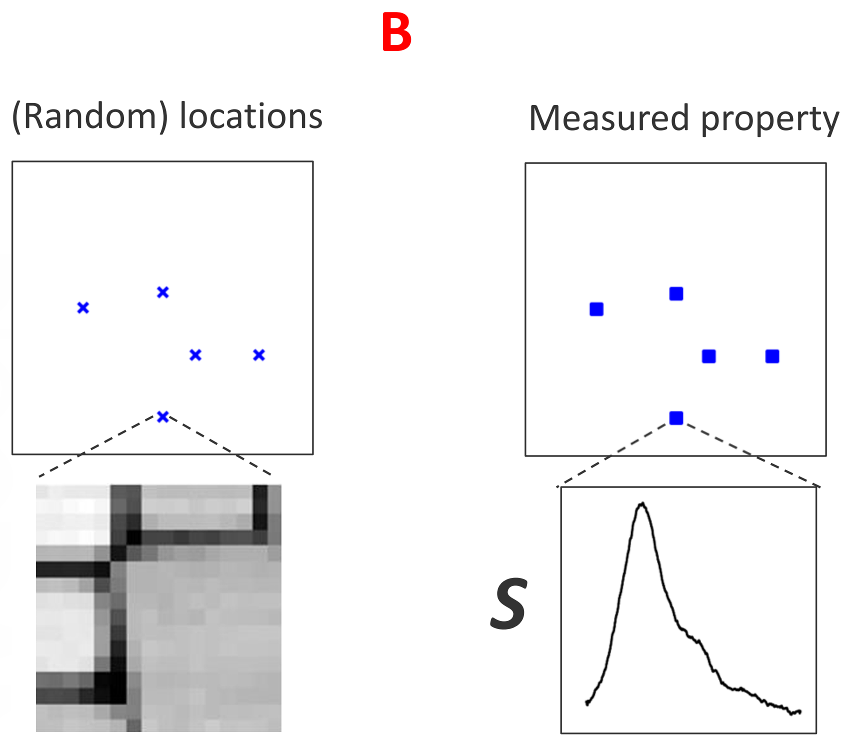


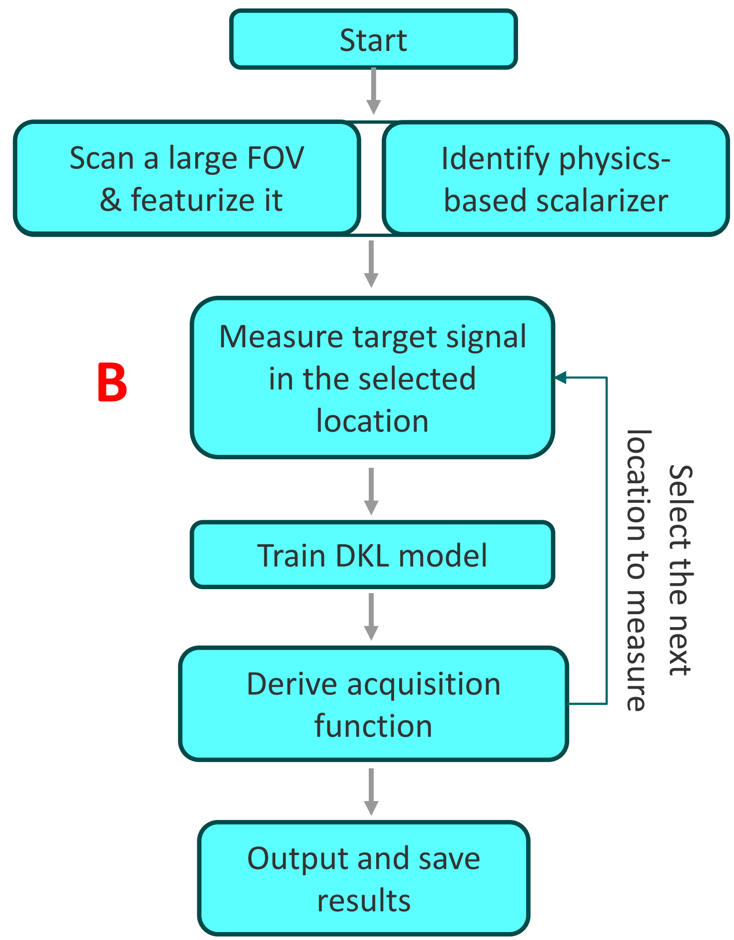
Deep Kernel Learning: Step 1



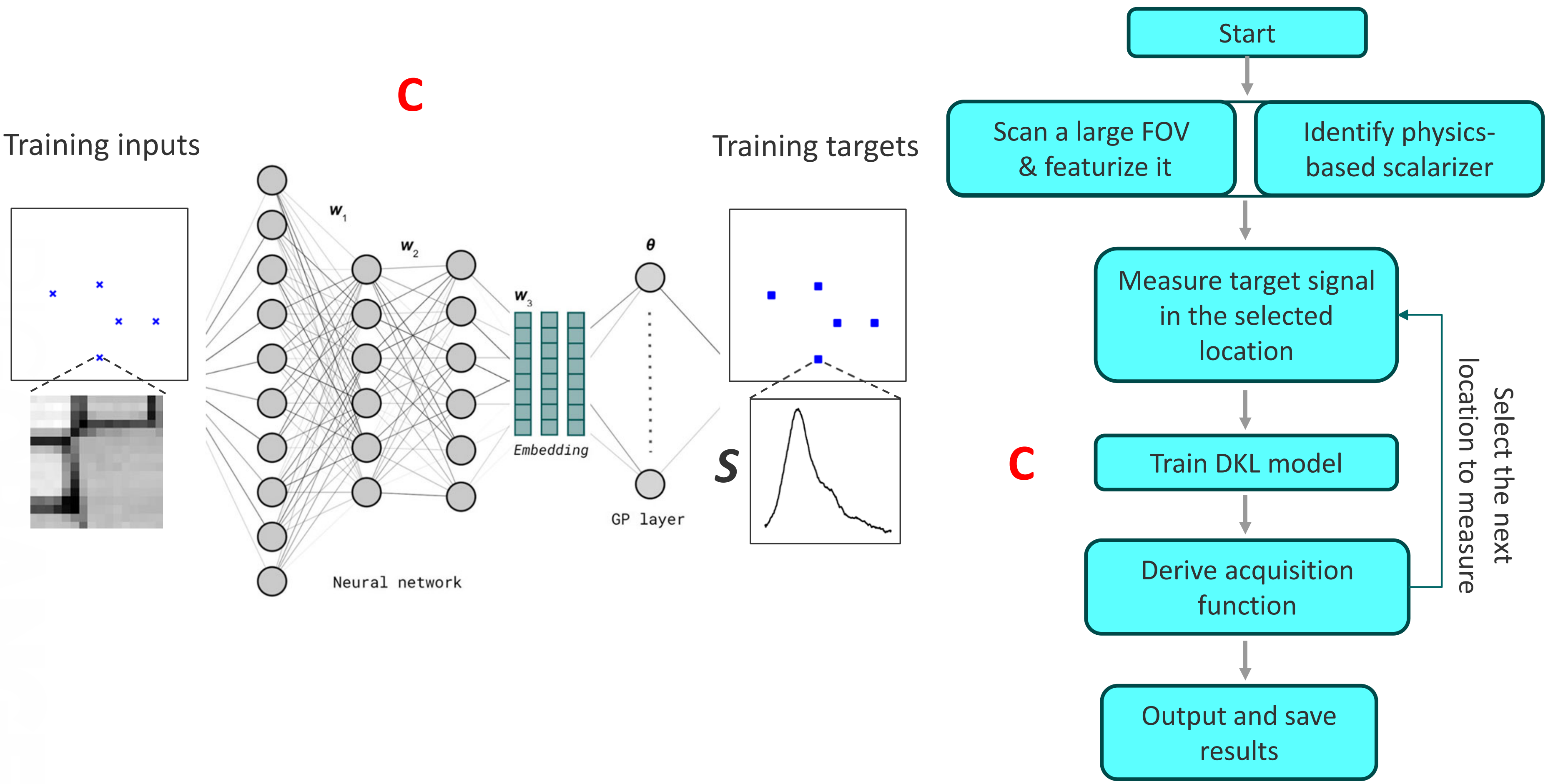


Deep Kernel Learning: Step 2





Deep Kernel Learning: Step 3



Deep Kernel Learning: Going Active

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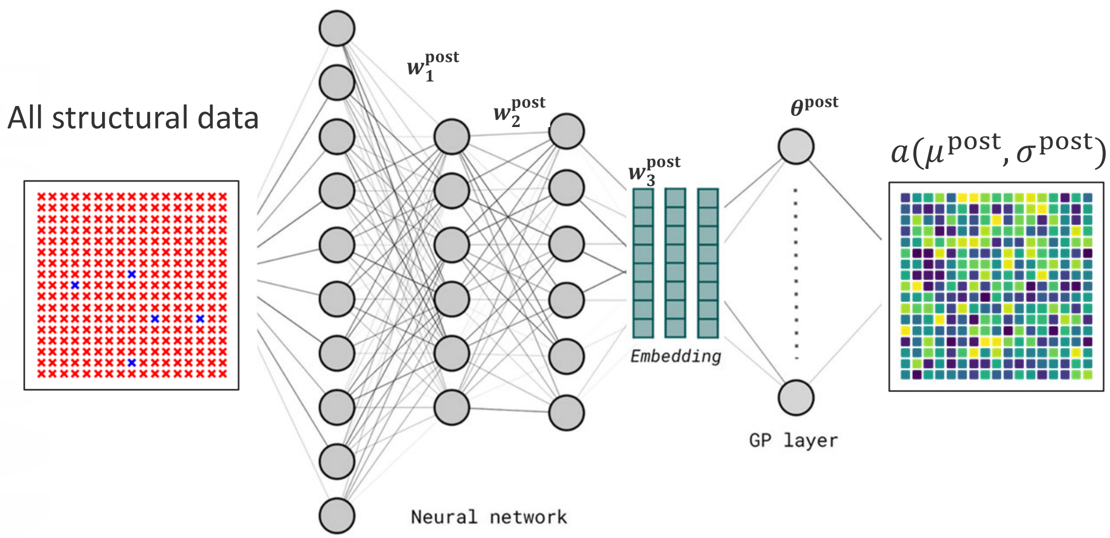

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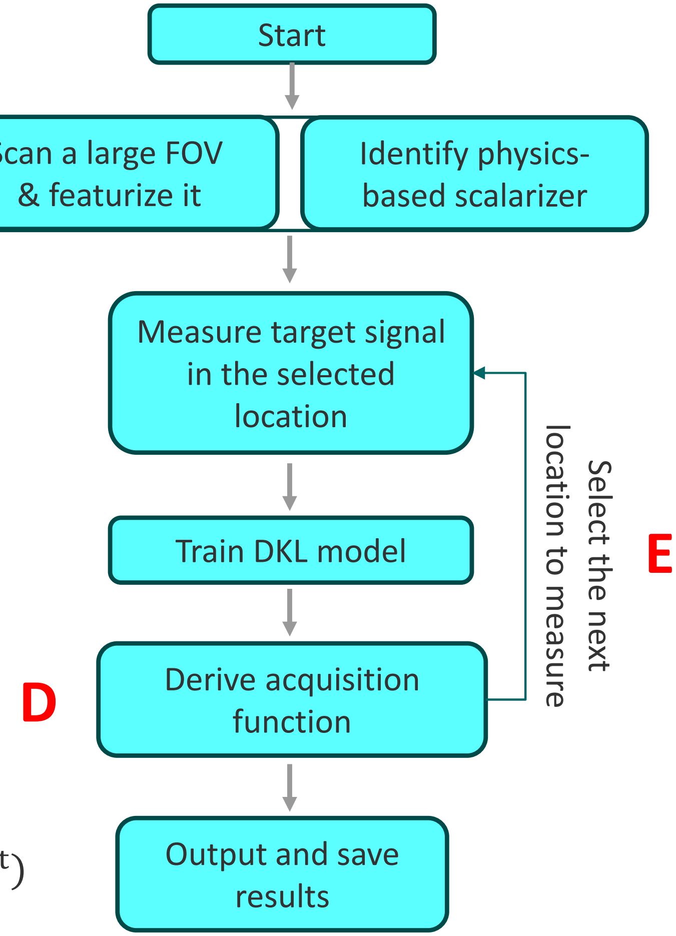
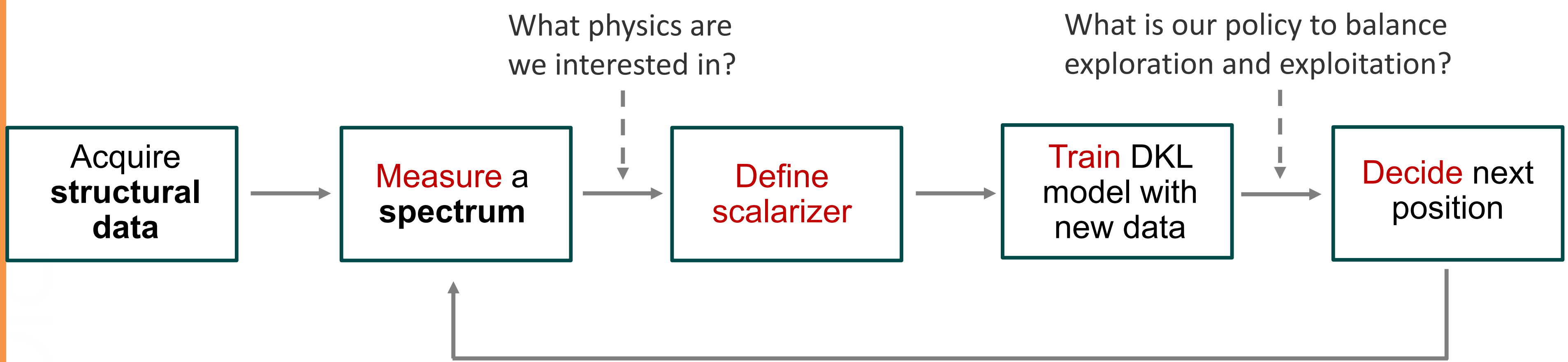
Next point to measure:

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



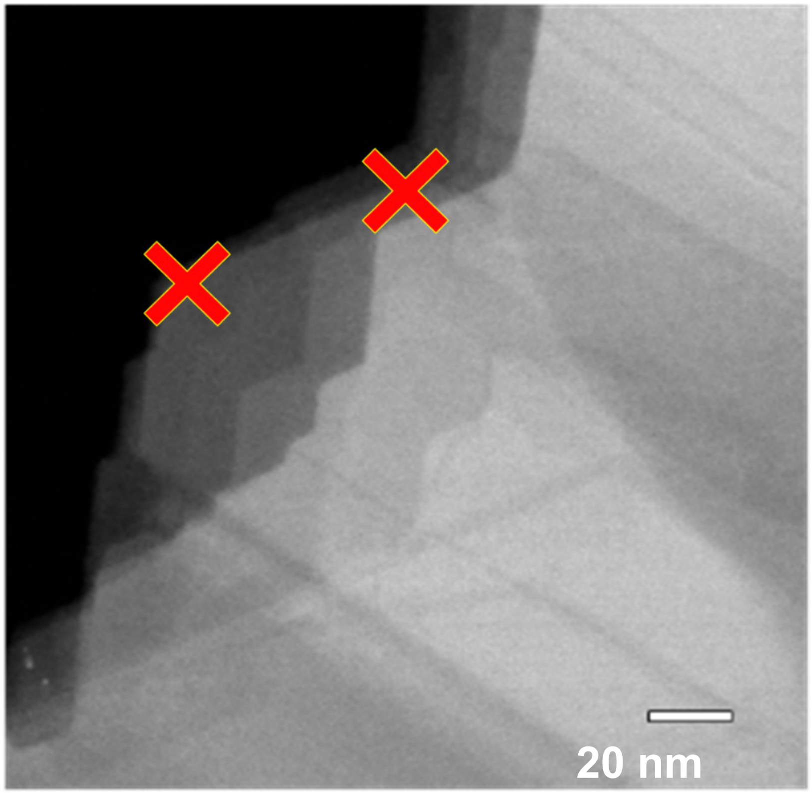
Deep Kernel Learning based BO

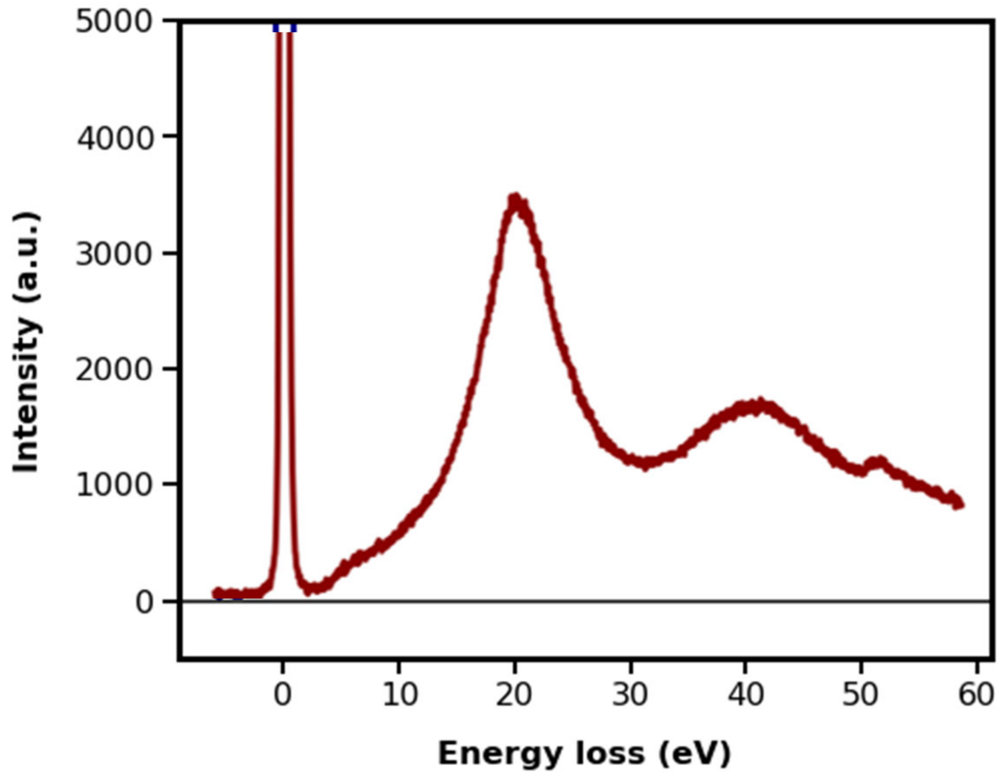


Key concepts:

- **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





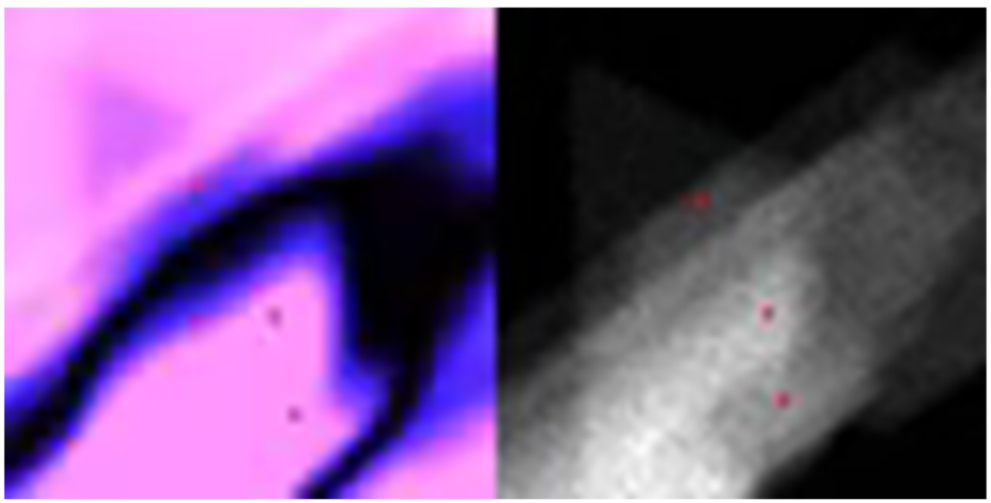
1. What if we have full access to structural information
2. And want to choose locations for (EELS, 4D STEM, CL, EDX) measurements
3. So as to **learn** relationship between structure and spectrum fastest
4. Or **discover** which microstructural elements give rise to specific **desired** spectral features?

Discovering Regions with Interesting Physics

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

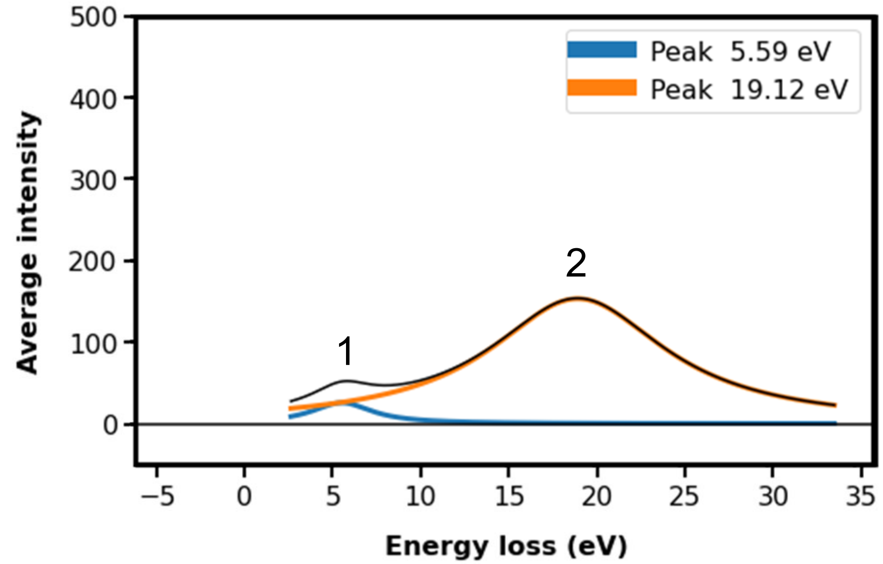
Acquisition function

HAADF-STEM

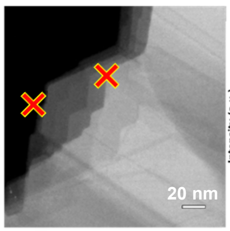


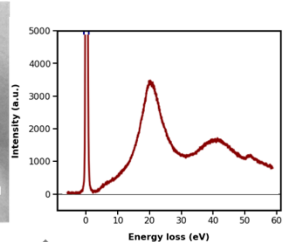
Physics search criteria:

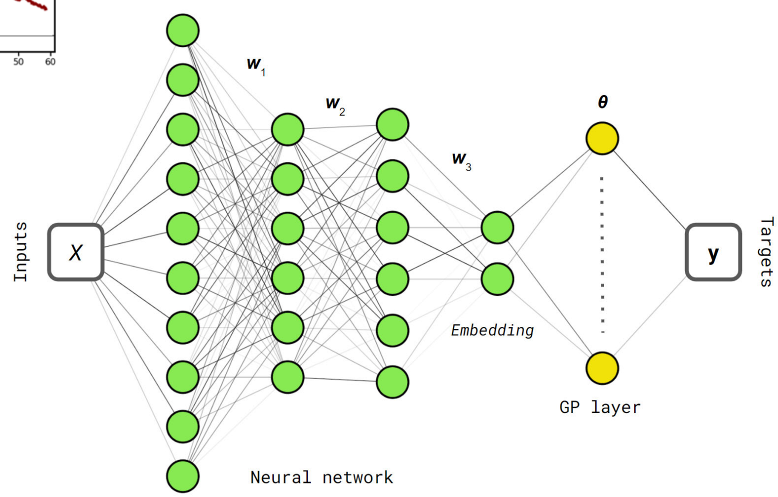
$$\boldsymbol{Ratio = Peak\,1\,/\,peak\,2}$$



Deep Kernel Learning







<u>Specify physics criteria</u>

Acquire
structural data

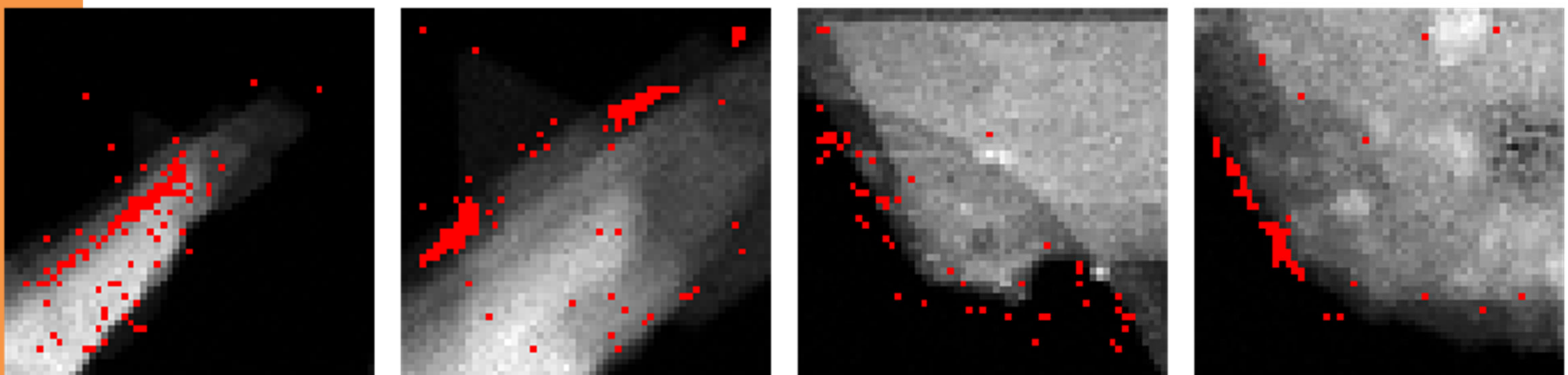
Measure a
spectrum

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)!

Changing the scalarizer

- (Same region) **Simple physics search:** peak max in selected region

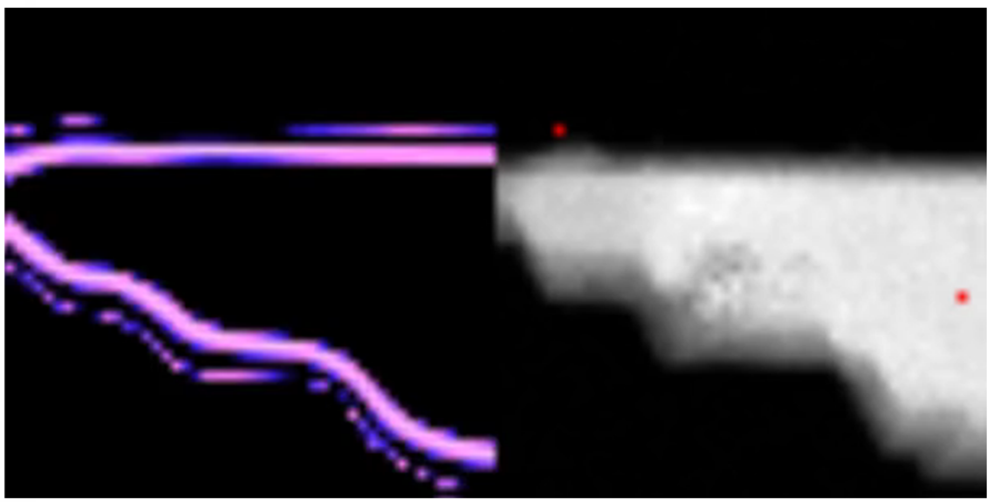
Physics search criteria:

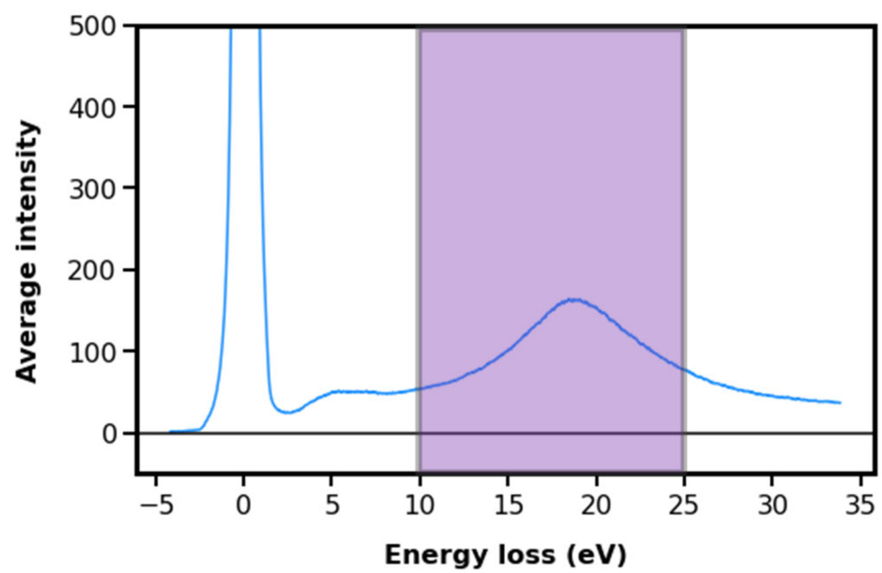
$$\boldsymbol{Maximize(f)}$$

(Specific peak intensity)

Acquisition function

HAADF-STEM + points visited





What were we doing?

- **Objective(s):**
  - Understanding the emergence of the nanoplasmonic behaviors
  - For understanding physics, optical interfaces to quantum devices, etc.
  - … and so on. For fundamental research, vey often impact is clear later!

- **Reward:**
  - Minimizing uncertainty in structure-property relationships (can we predict expensive EELS from cheap structure)
  - Discovering structures that maximize certain aspect of nanophotonic behavior (have maximal intensity of certain peak, peak area ratio, etc.)

- **Value:** expected reward. Here – predicted scalarizer

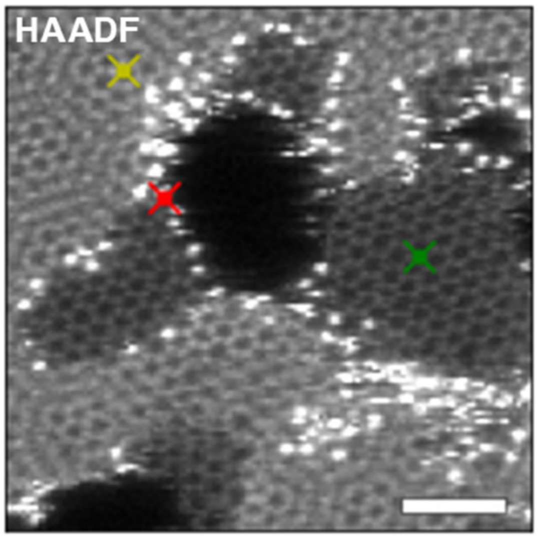
- **Action:** position STEM probe at a given location, take EELS spectrum

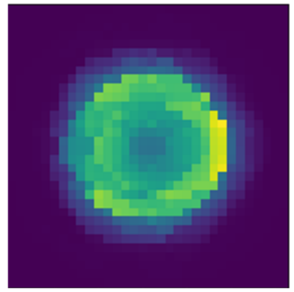
- **State:** image patch

- **Policy:** myopic optimization (actually, upper confidence bound) with defined exploration-exploitation balance

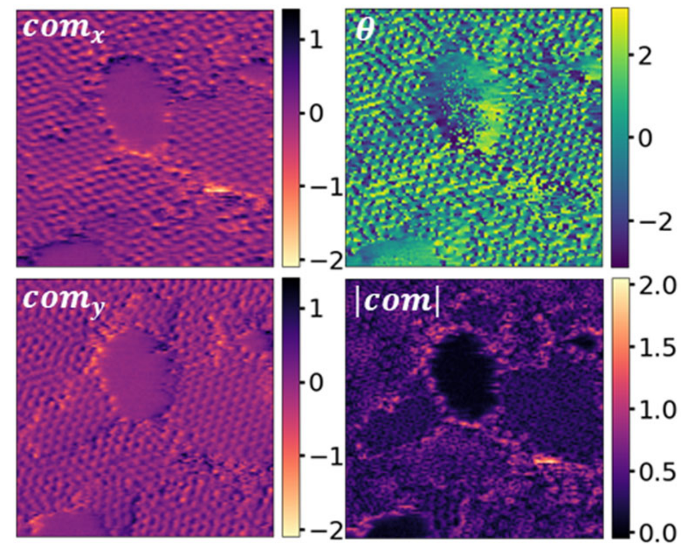
- Currently, we run 4D STEM measurements on a grid.
- What if we want to explore smarter workflows – where microscope chooses where to take 4D STEM measurements?
- **Direct:** We can do it for a priori known objects of interest
- **Inverse:** Or we can aim to discover objects which have predefined signatures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

4D STEM: Grid, Direct, and Inverse





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

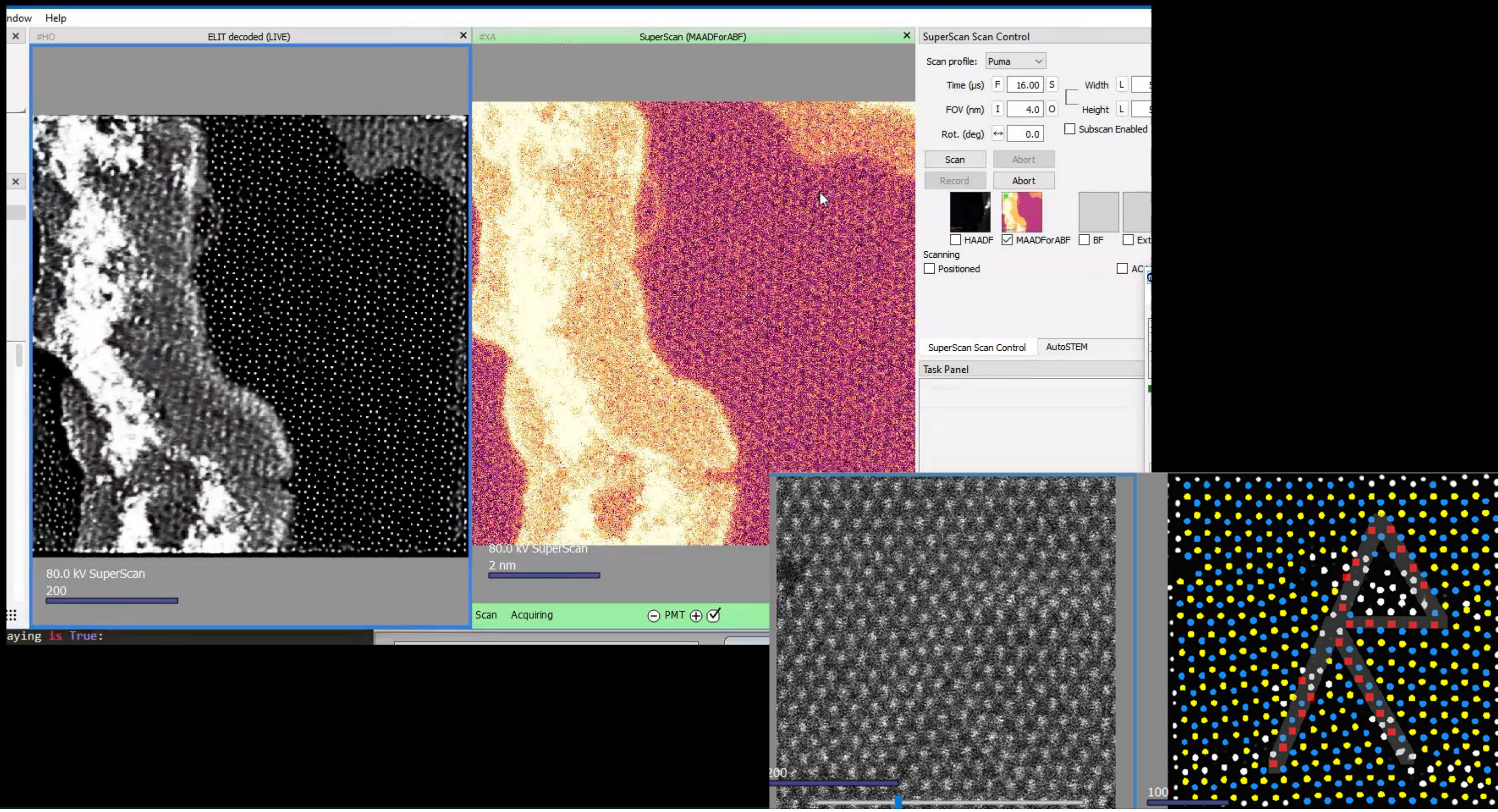


- What can we say about structure?
- Interesting functionalities are expected at the certain structural elements
- We can guess some; we have to discover others
- Multiple goals while running experiment

- **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?

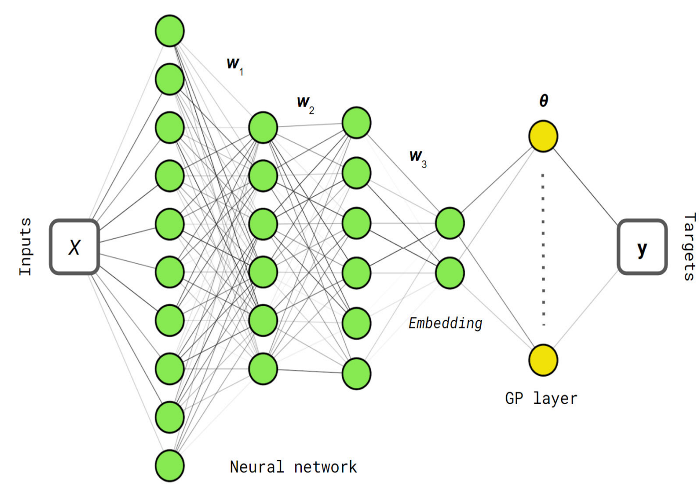
What is our policy to balance exploration and exploitation?

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

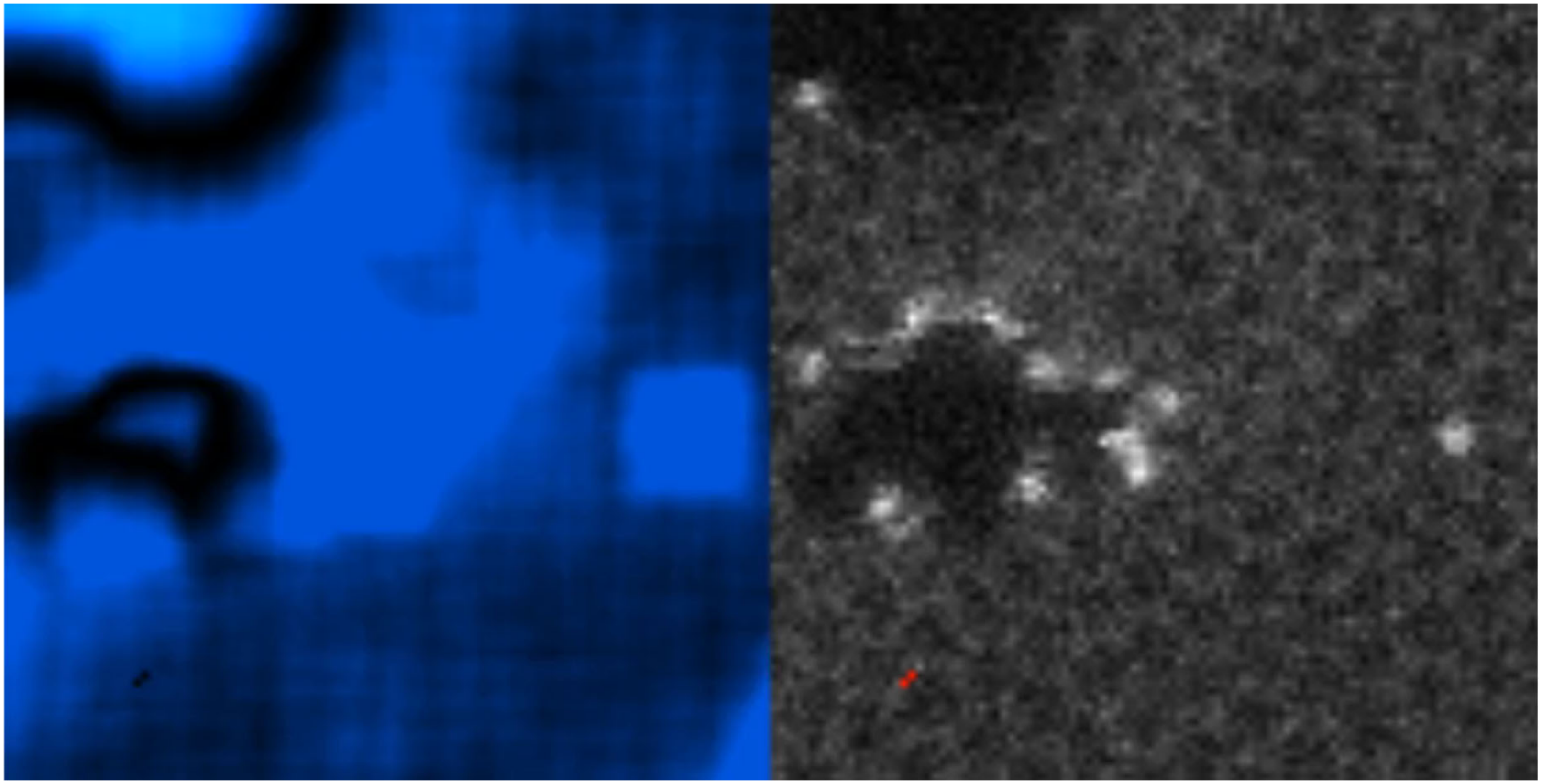
Key concepts:

- **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

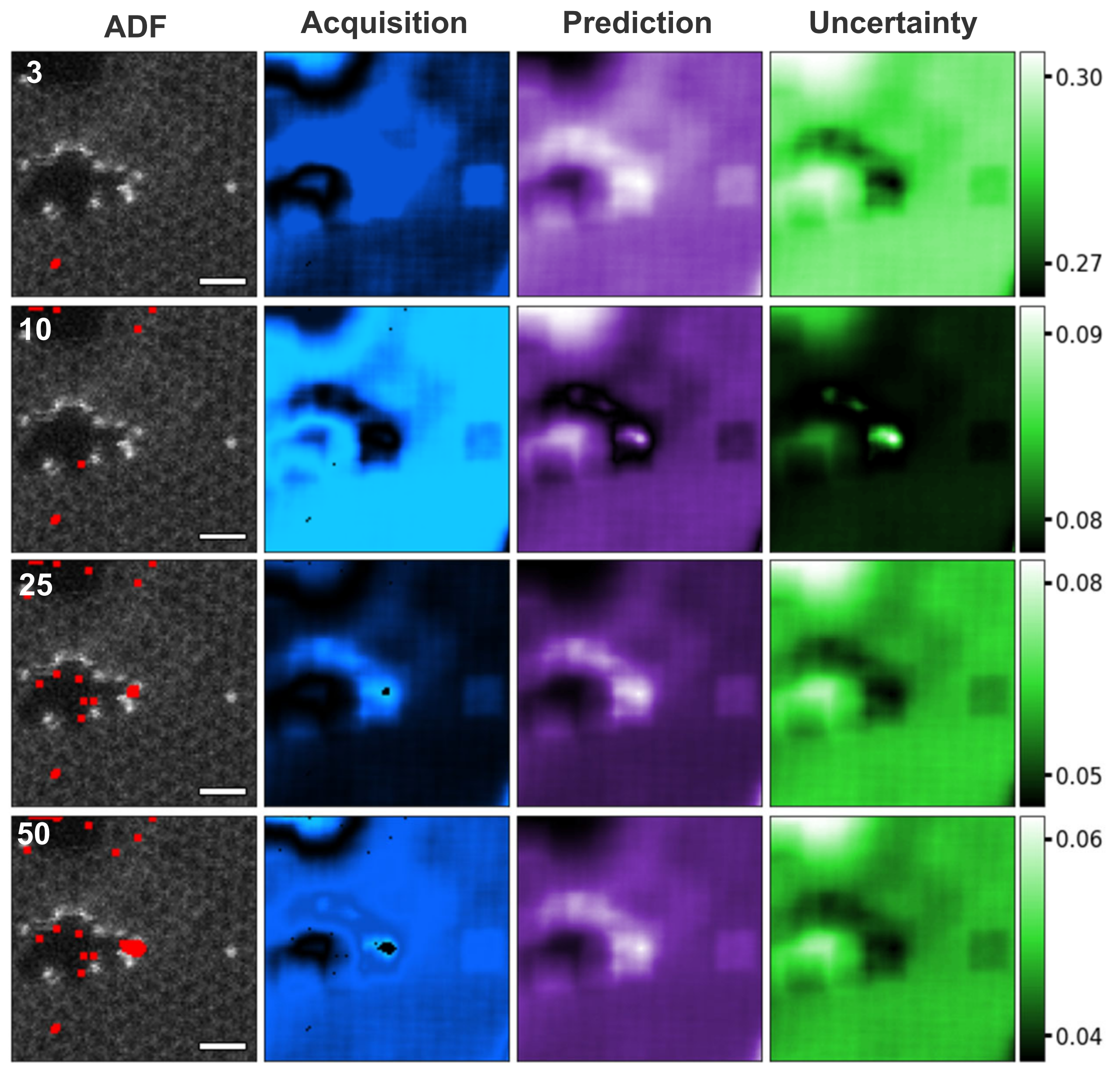


- Different **acquisition functions** can be used:
  - Expected Improvement (**EI**) (usually what was used)
  - Upper Confidence Bound (**UCB**), etc
- Usually based on some combination of **prediction** and **uncertainty**.

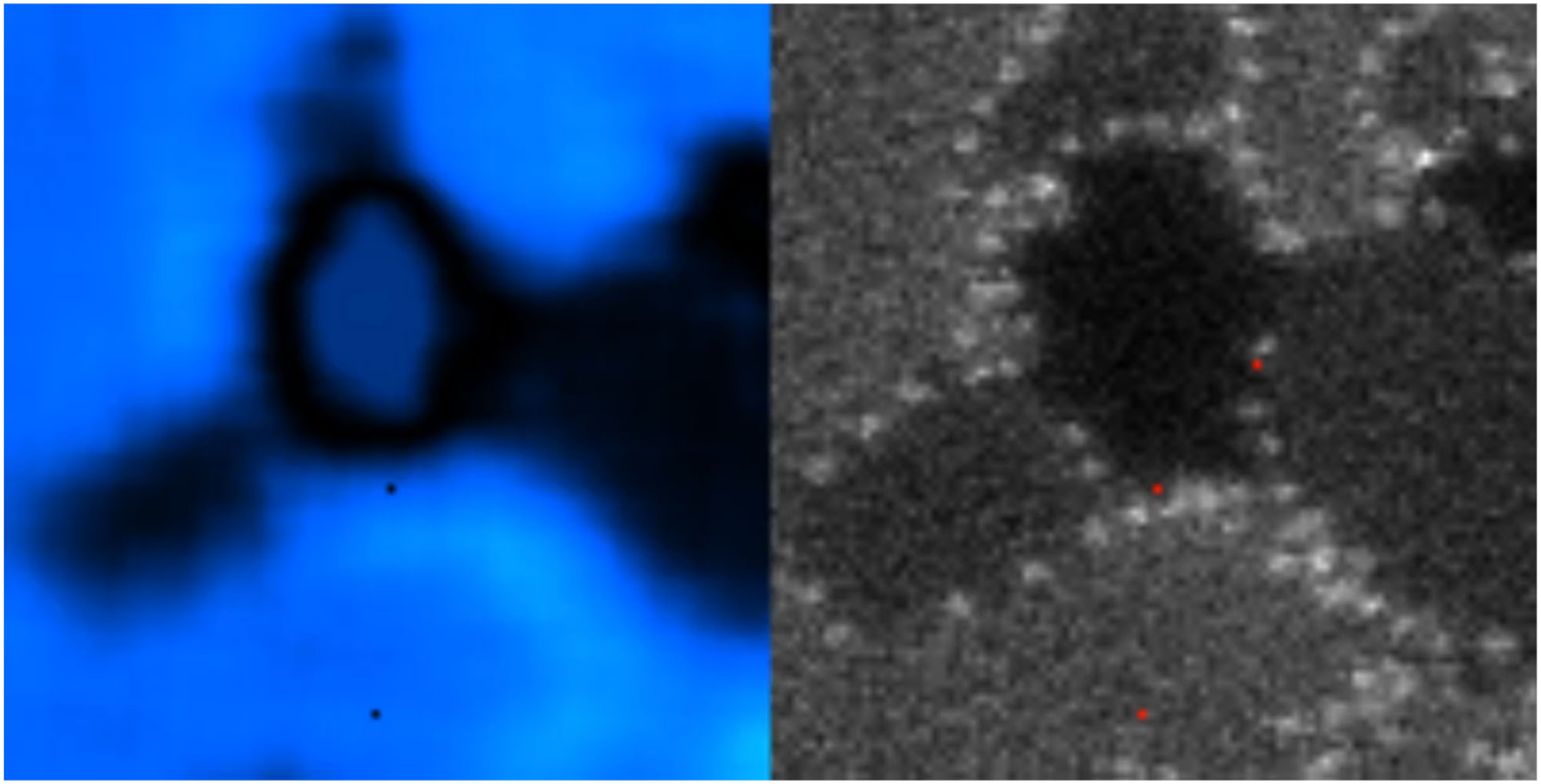
A closer look

Scalarizer: CoM magnitude

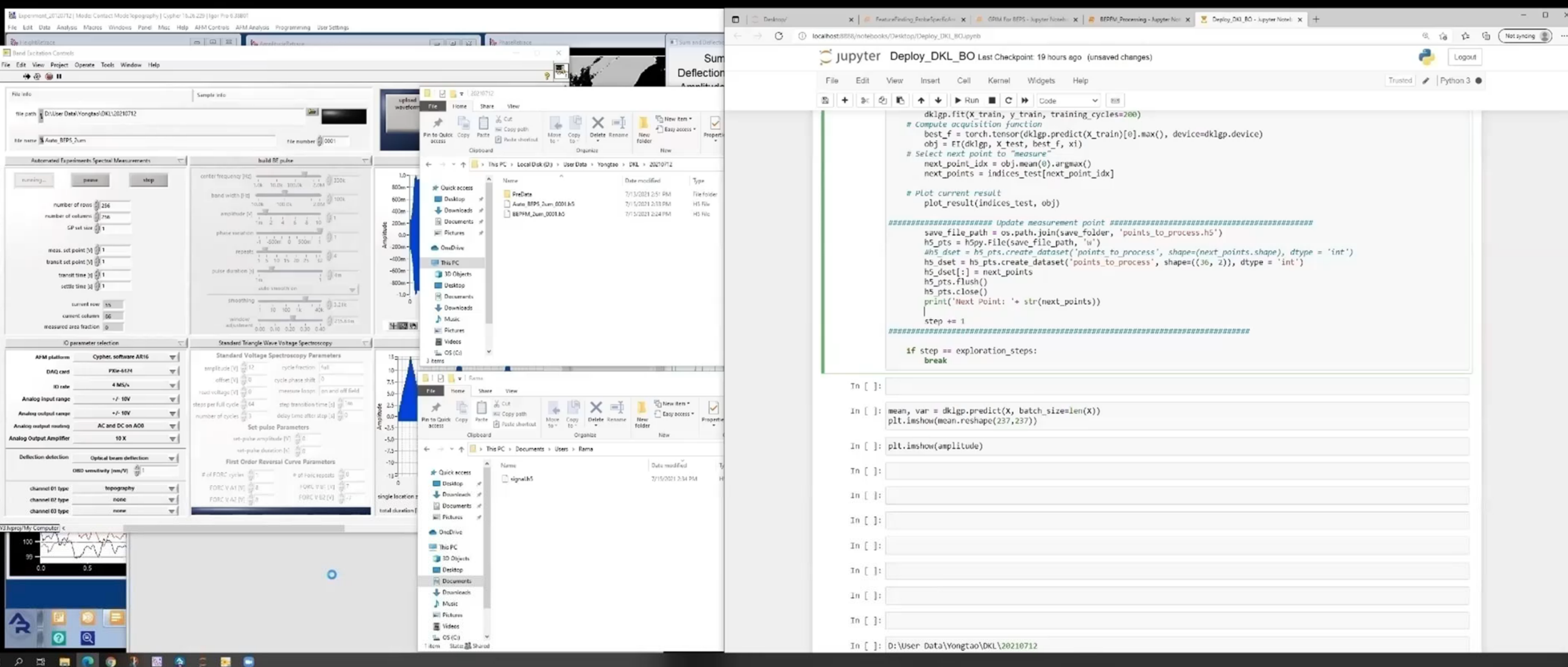
- High uncertainty @ start, but fairly quickly reduces
- Prediction actually doesn't drastically change throughout experiment
    - Structure-property relationship here is fairly rapidly learned
- Note the training can be halted after some criterion is met, making remainder of experiment go much quicker



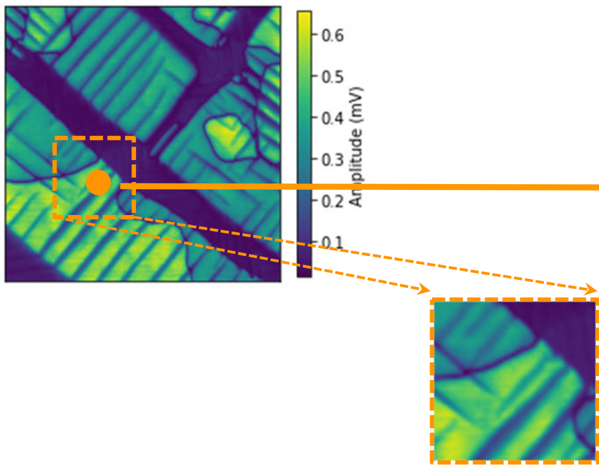
Does it always work?

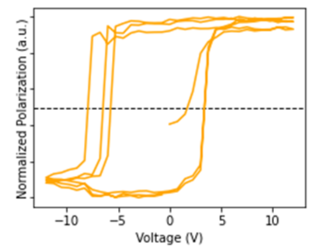


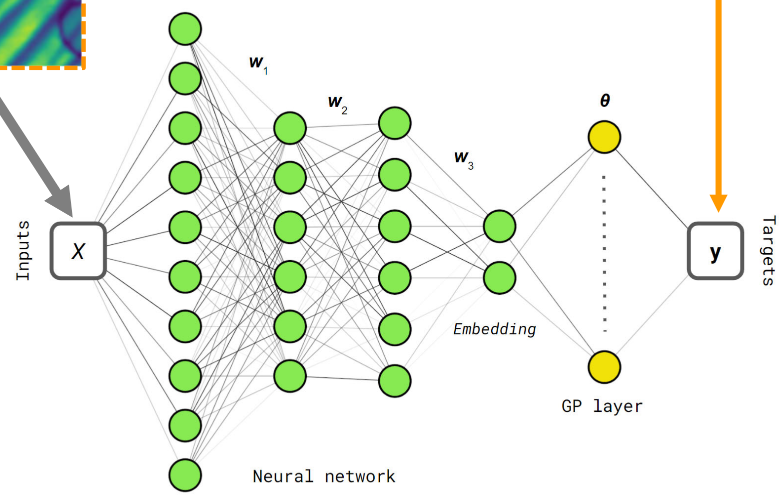
Deep Kernel Learning AE



Deep Kernel Learning







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

<u>Specify physics criteria</u>

Acquire **structural data**

Measure a **spectrum**

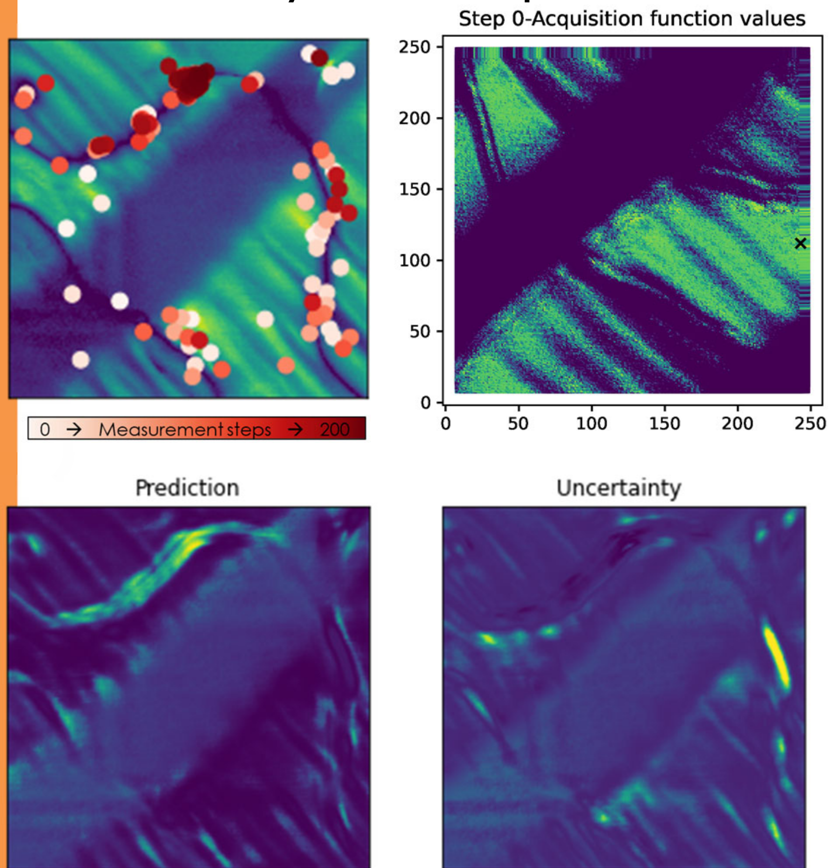
Train DKL model with new data

Decide next position (optimize physics criteria)

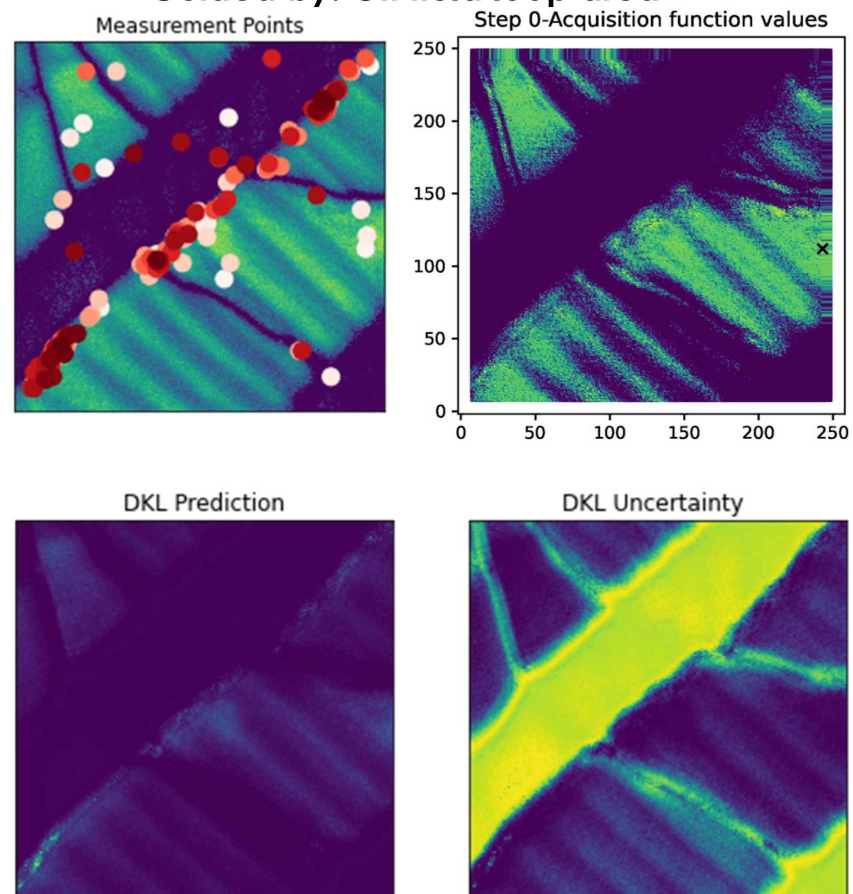
Allows navigation of the system to search for physics

Deep Kernel Learning SPM

Guided by: On field loop area



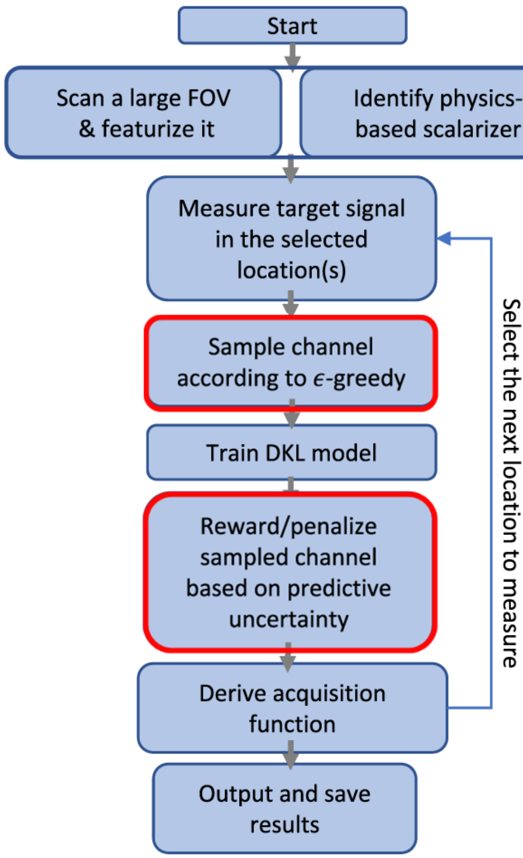
Guided by: Off field loop area

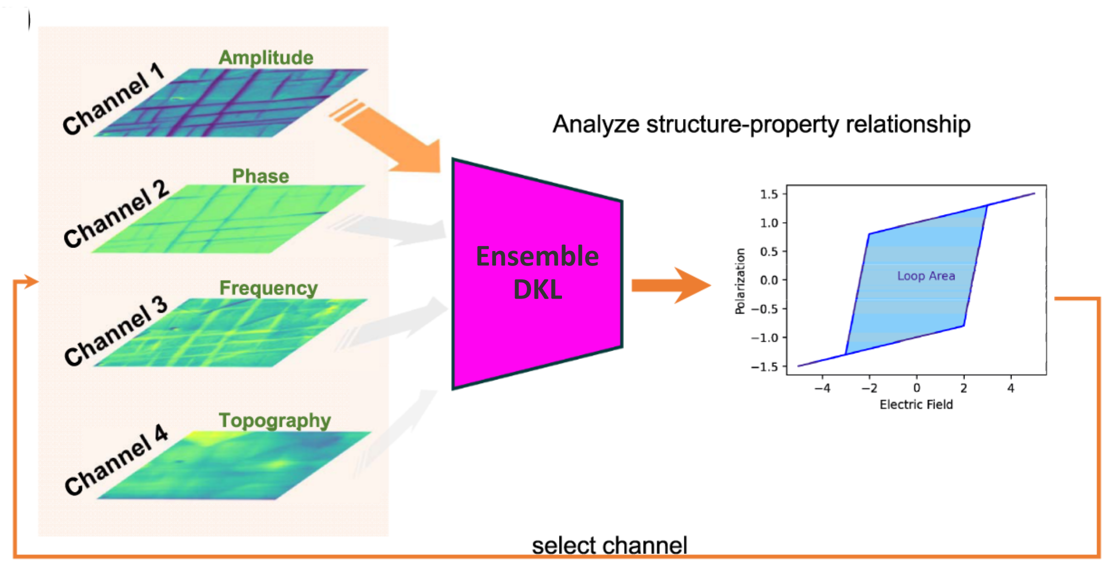


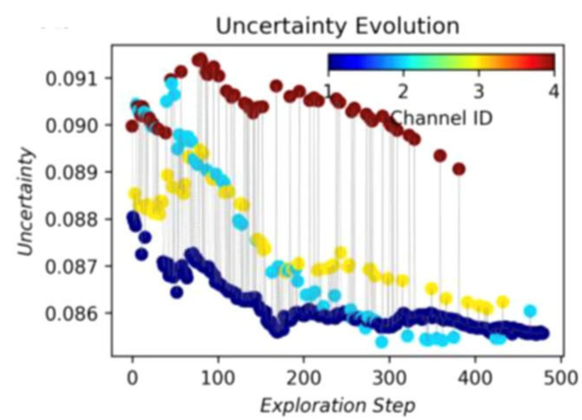
- ➢ Large loop opening corresponding 180° domain walls
- ➢ This behavior can be attributed to the large polarization mobility of 180° walls

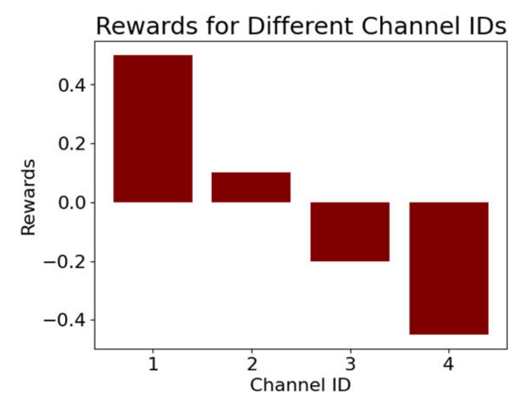
Liu, Yongtao, et al, Nature Machine Intelligence 4, 4 (2022): 341-350.

DKL: Learning the best channel



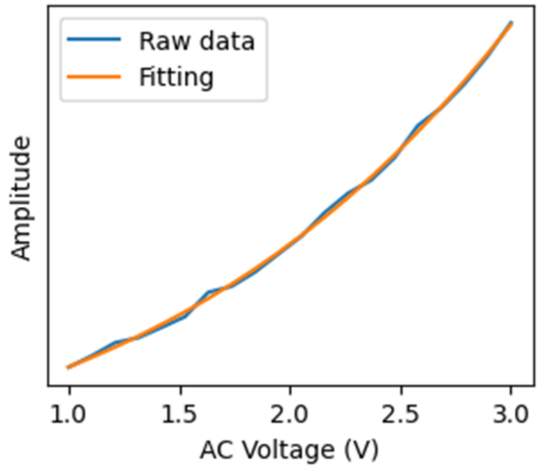


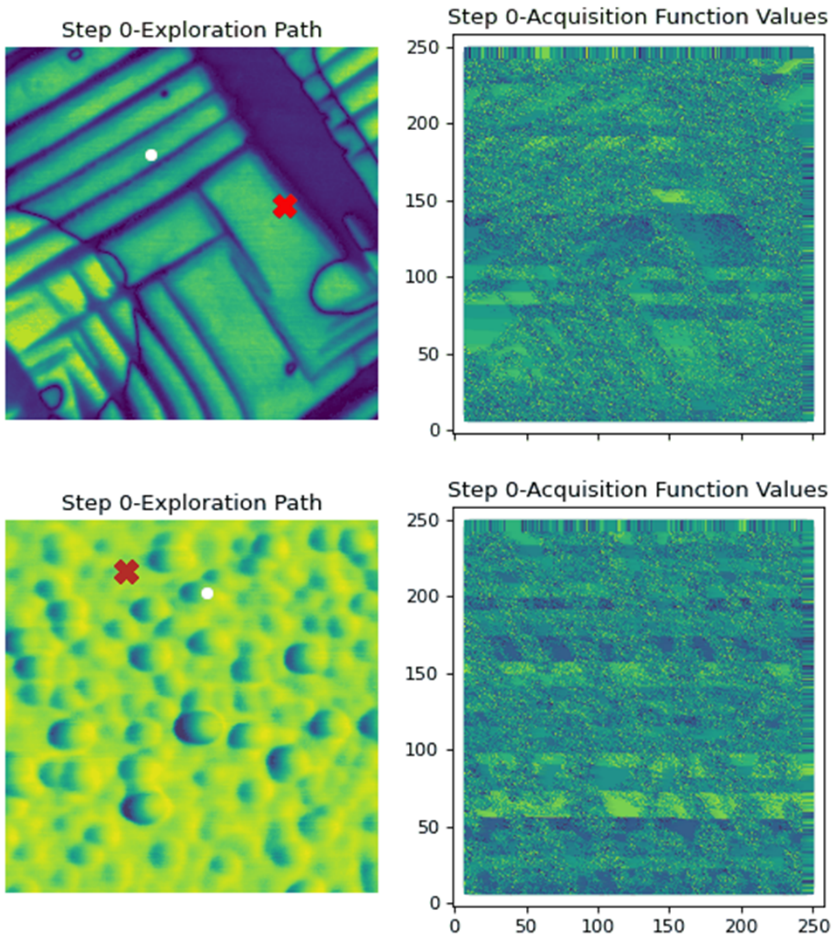




Y. Liu et al. npj Comput Mater **9**, 34 (2023).

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

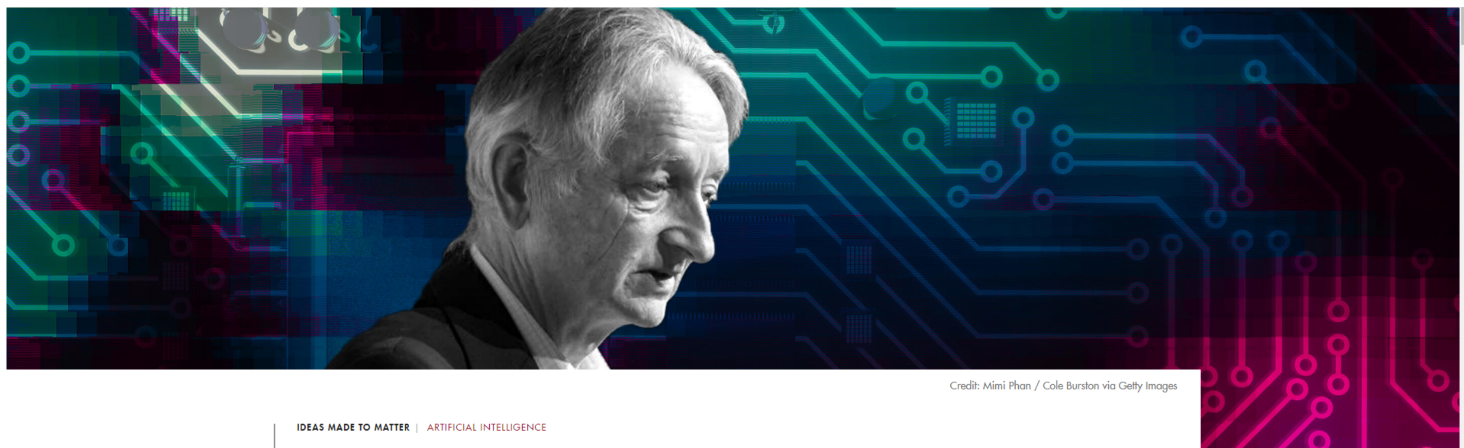
Taking the Human Out of the Loop: A Review of Bayesian Optimization

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

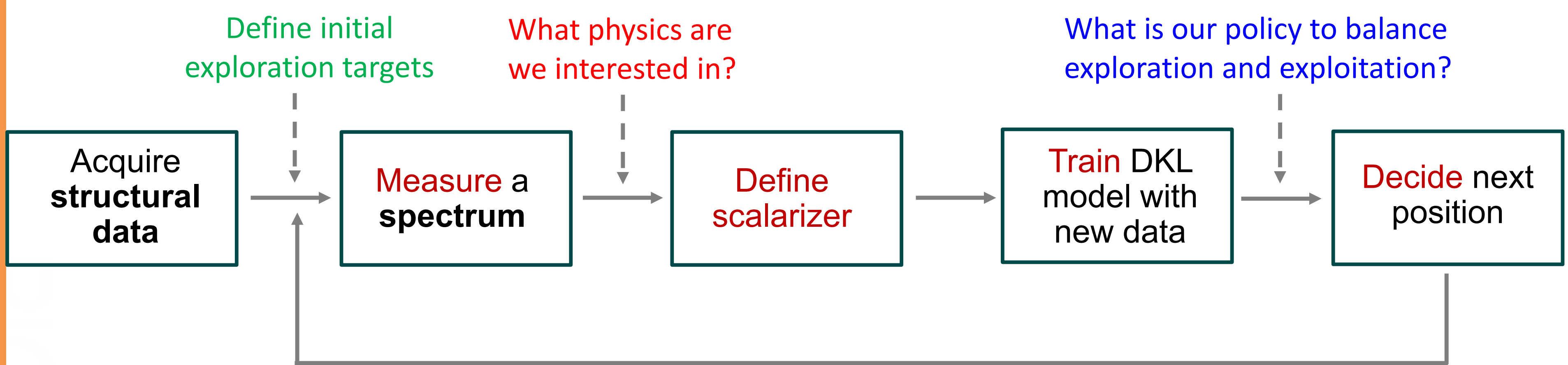


Credit: Mimi Phan / Cole Burston via Getty Images

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:

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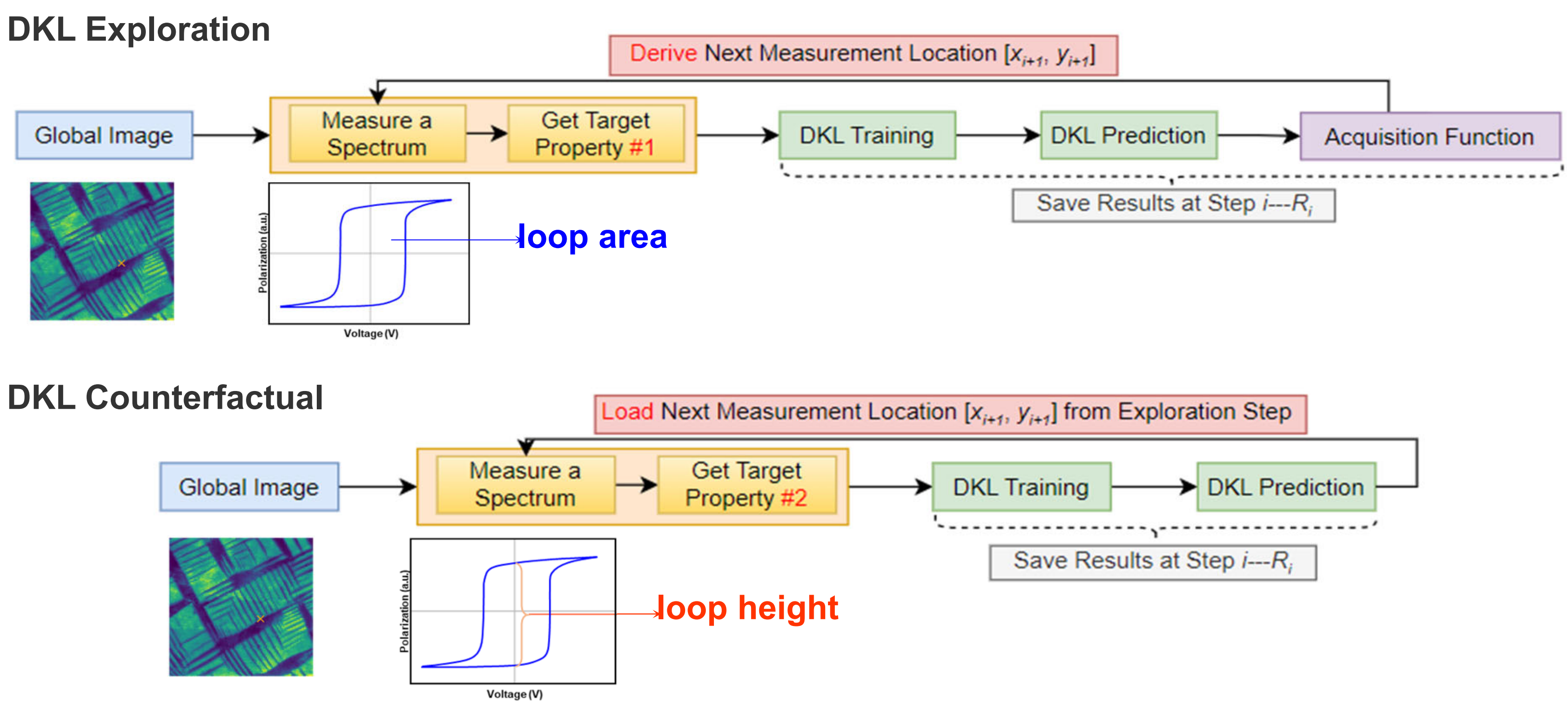
Counterfactual scalarizers



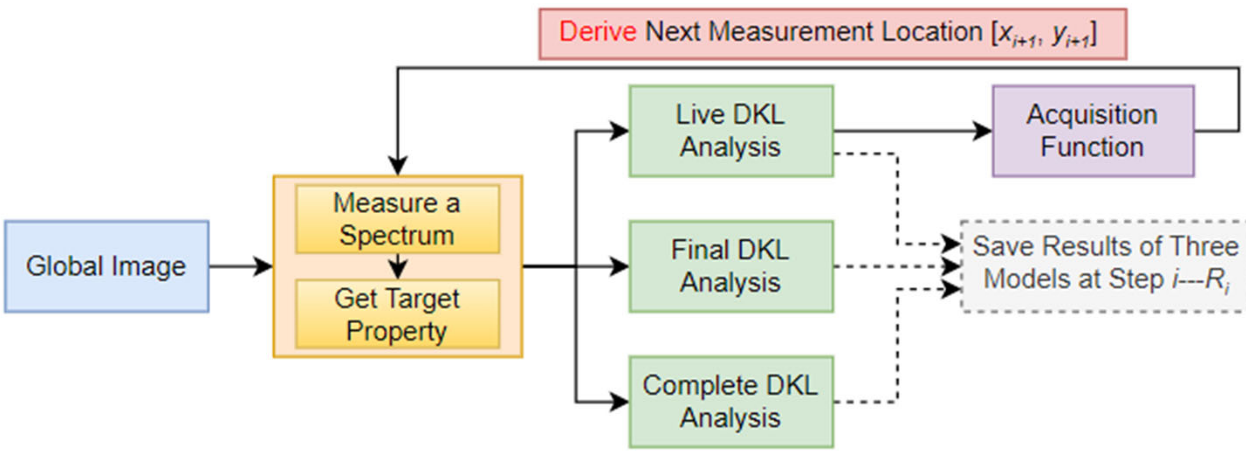
Target properties:

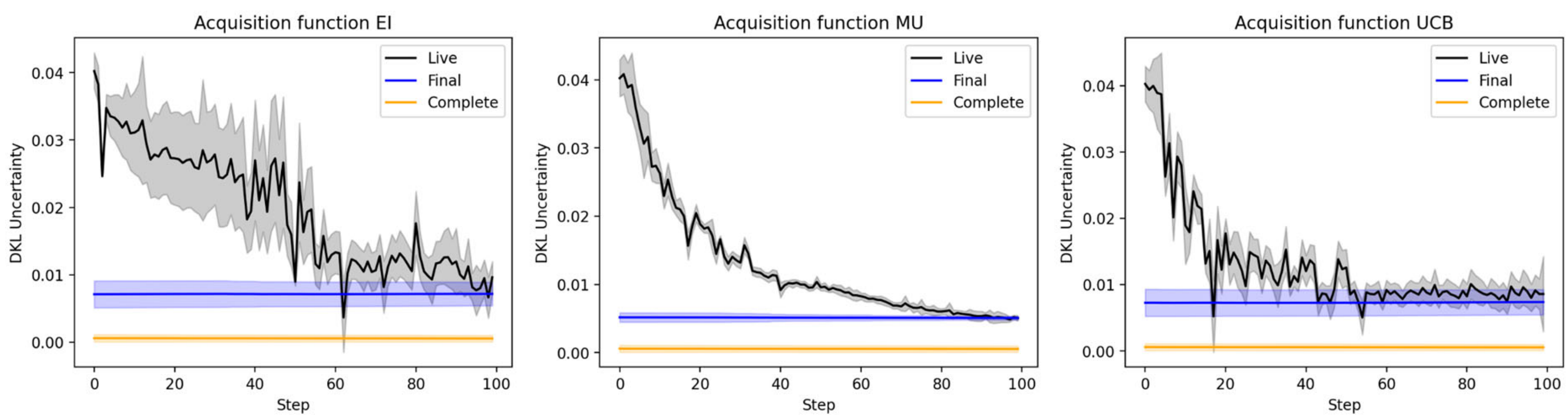
1. Loop Area
2. Loop Height
3. Coercive Field
4. …

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

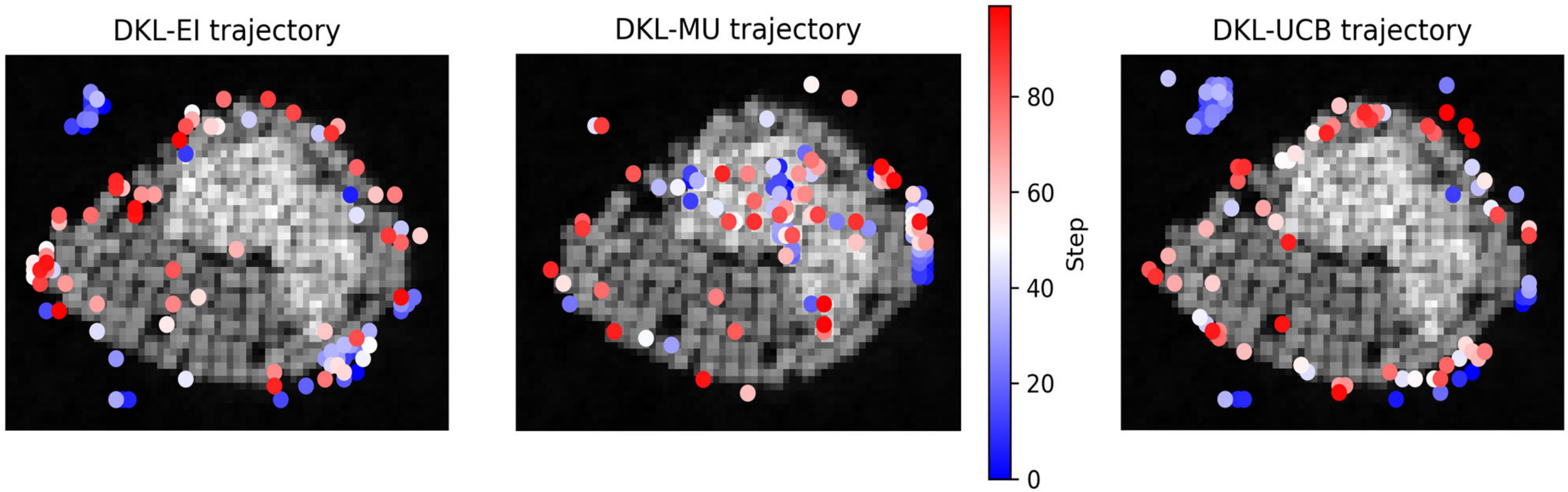
Explainable AE

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



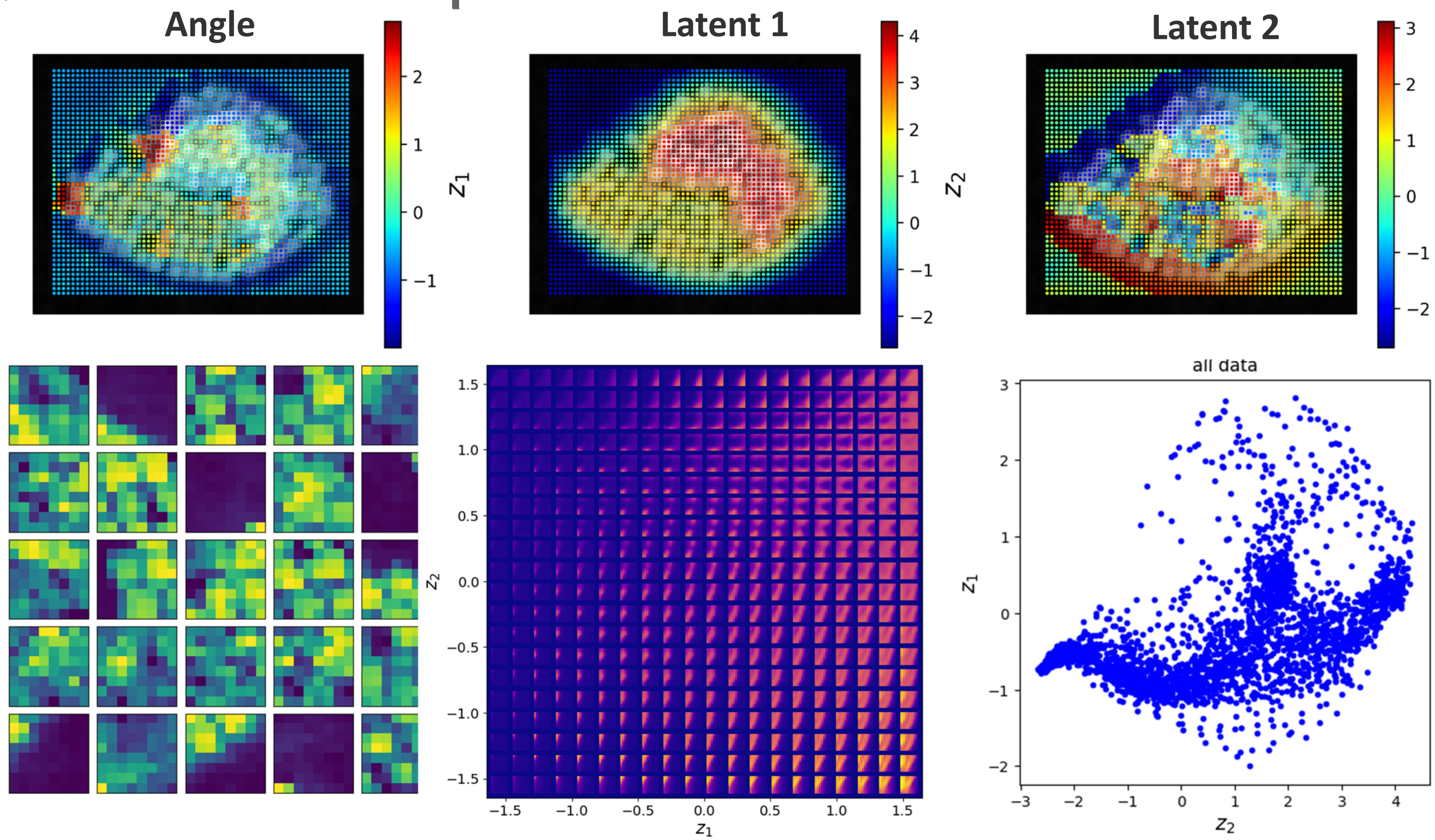


Monitoring the AE



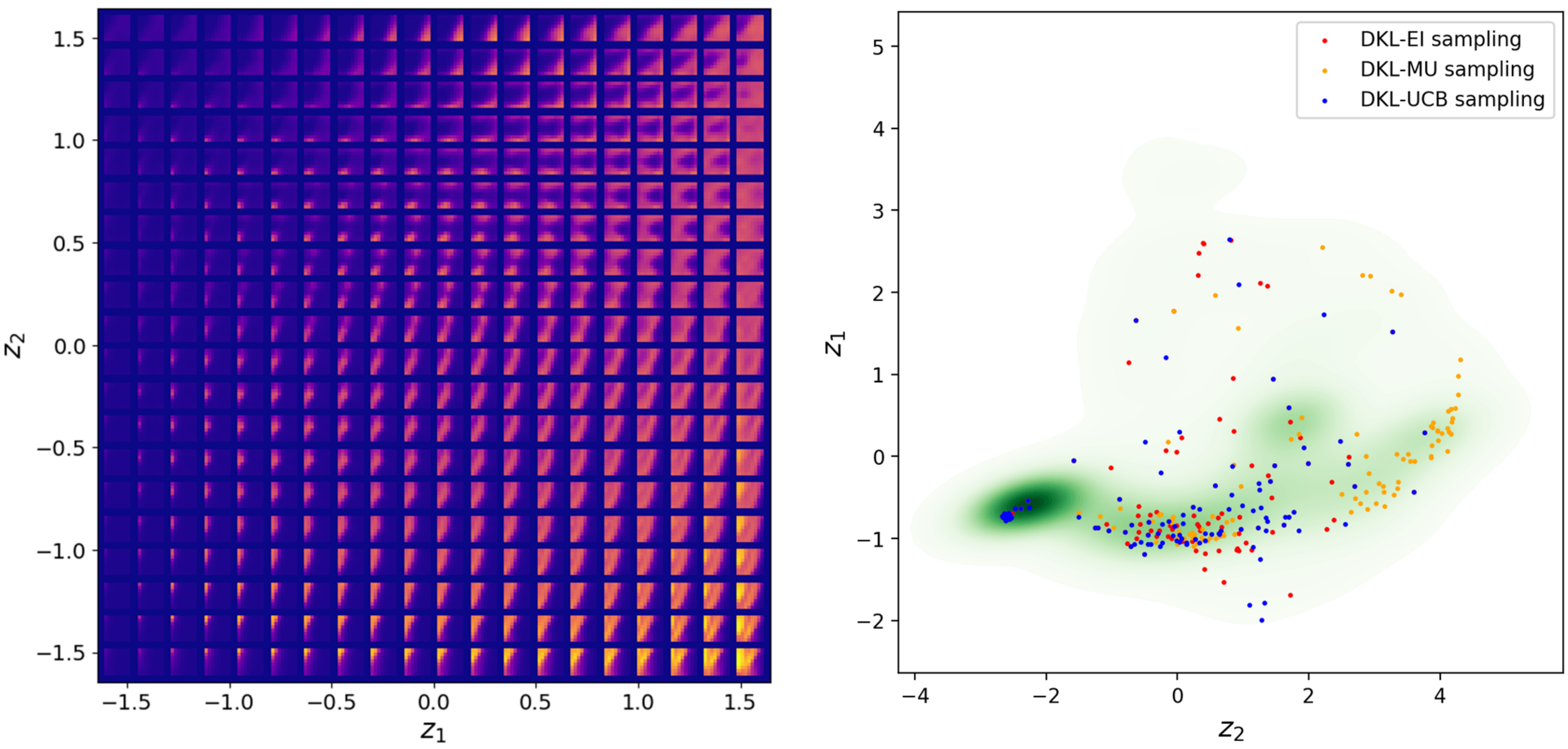
- Different acquisition functions (policies) give different experimental paths for AE
- Can we analyze what is special about points visited?

Global Feature Space

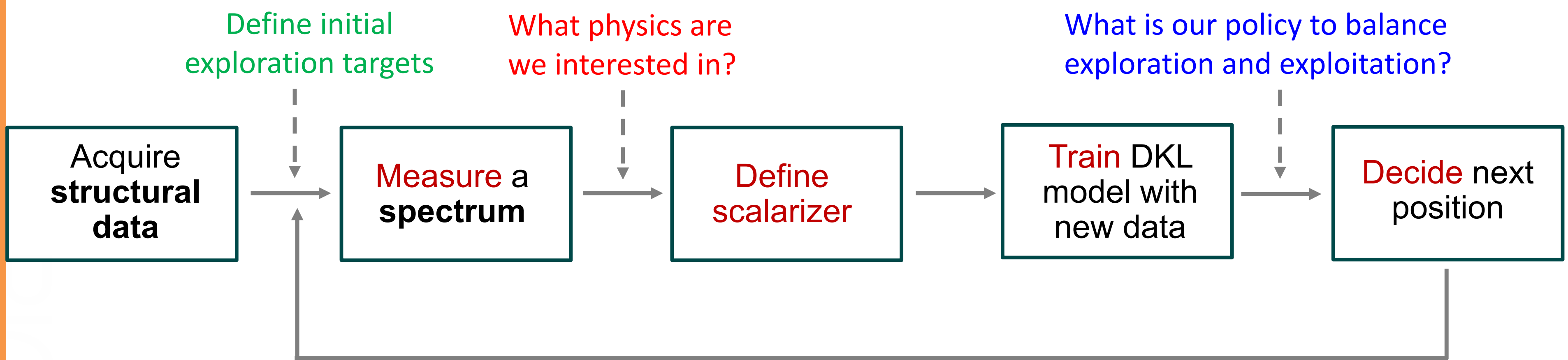


- Global feature space is available from the beginning of the experiment and is stationary
- Experimental trajectories can be visualized in the global feature space

VAE approach: full feature space



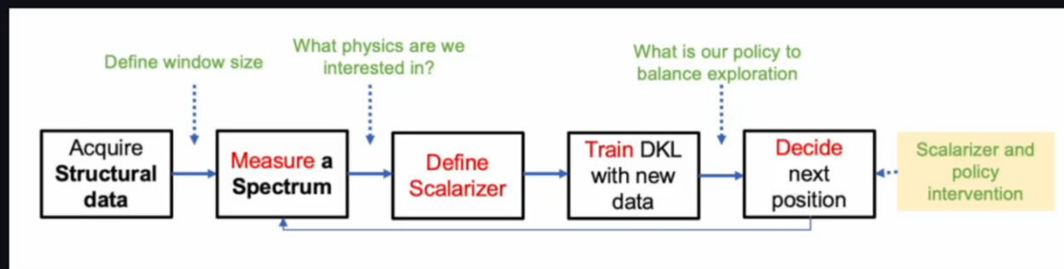
Bringing Human into the Loop



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

h(human)AE interface



Configure the parameters for the experiment

Save Directory

out_data_sl/

Window Size

16 − +

Data Path

in_data_sl/Plasmonic_EELS_FITO0_edgehole_01

Budget

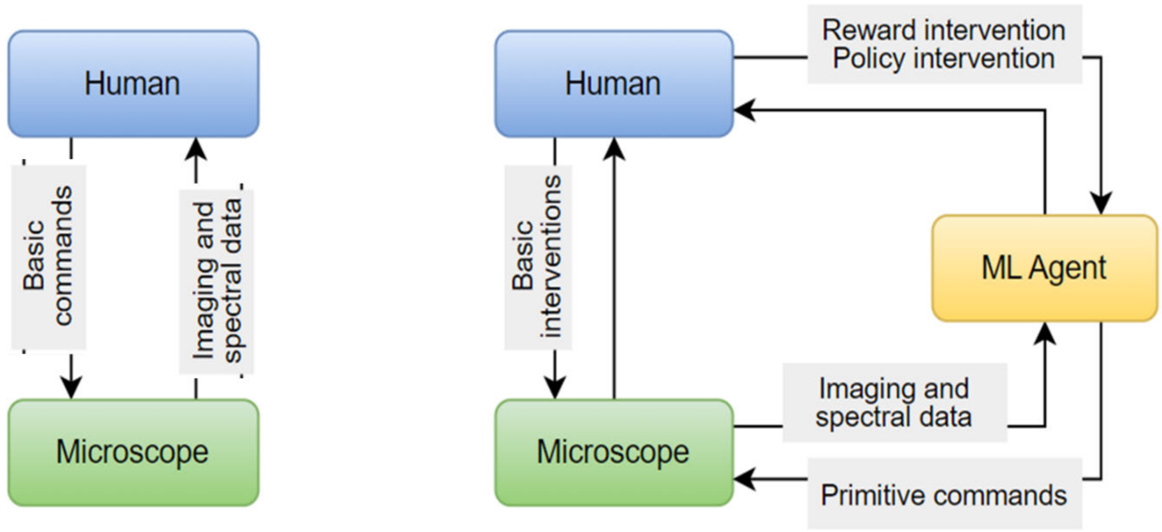
20 − +

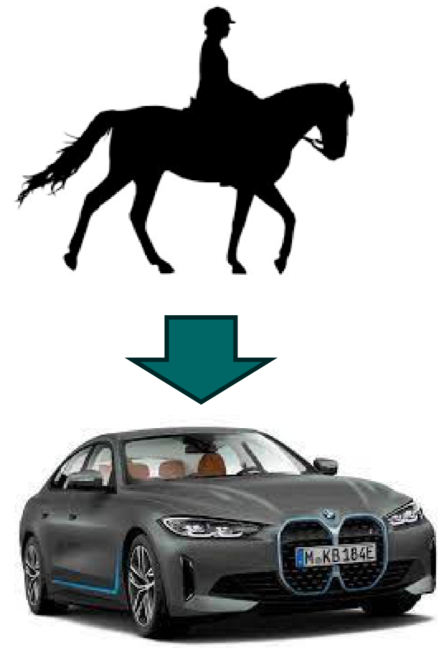
HAADF Exposure

0.00 − +

HAADF Resolution

Human in the loop AE



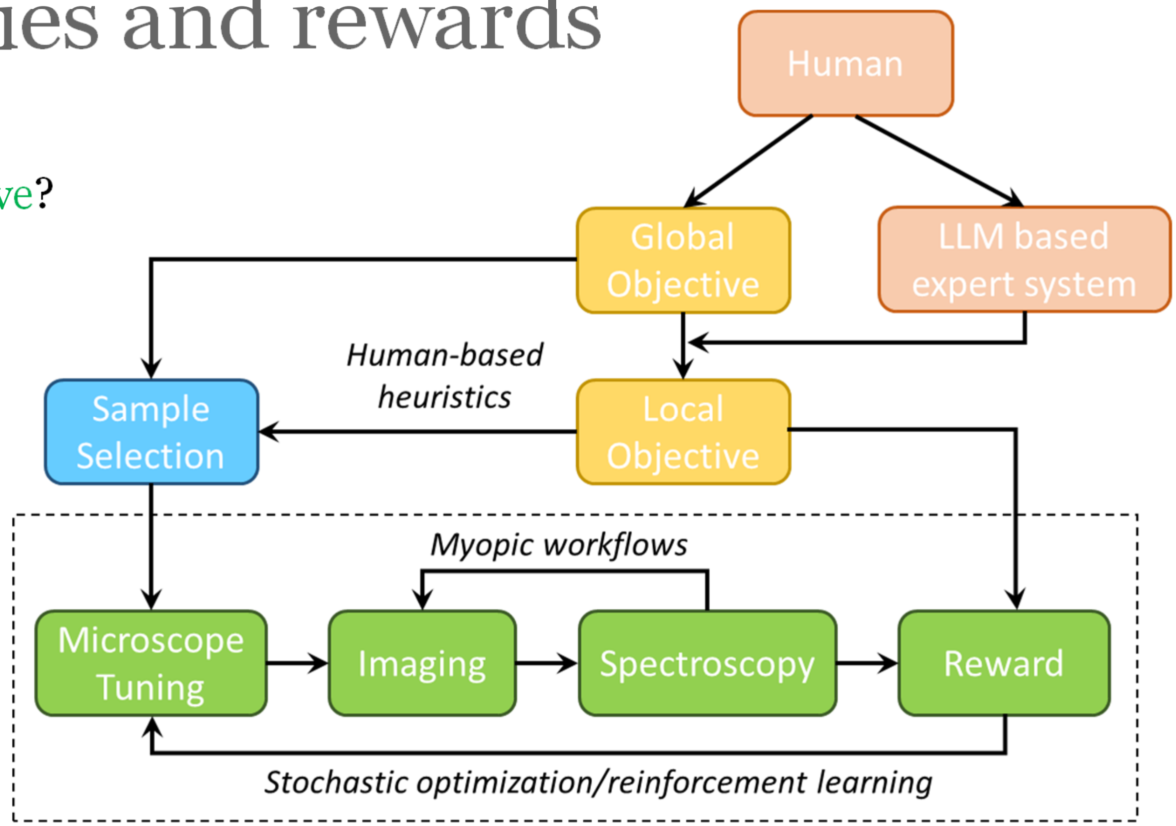


We can intervene on:

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

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?