THE UNIVERSITY of TENNESSEE KNOXVILLE

Gaussian Processes and Bayesian Optimization

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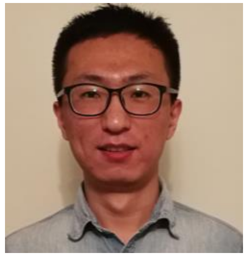

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The dance of policies and rewards

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?

Bayesian paradigm in science

- Bayes' theorem can be usefully re-written for science as:

Posterior: probability of the model given the data

Likelihood: probability of the data given the model

Prior: probability of the model

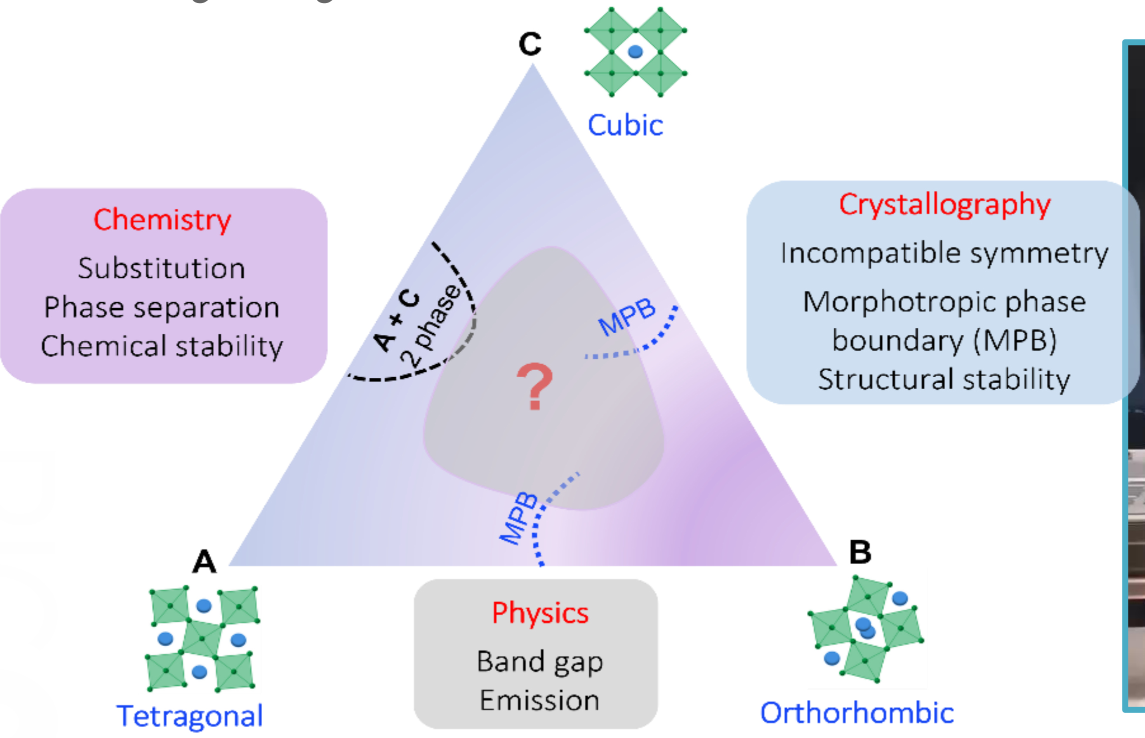
$$P(\text{model}|\text{data}) = \frac{P(\text{data}|\text{model})\, P(\text{model})}{P(\text{data})}$$

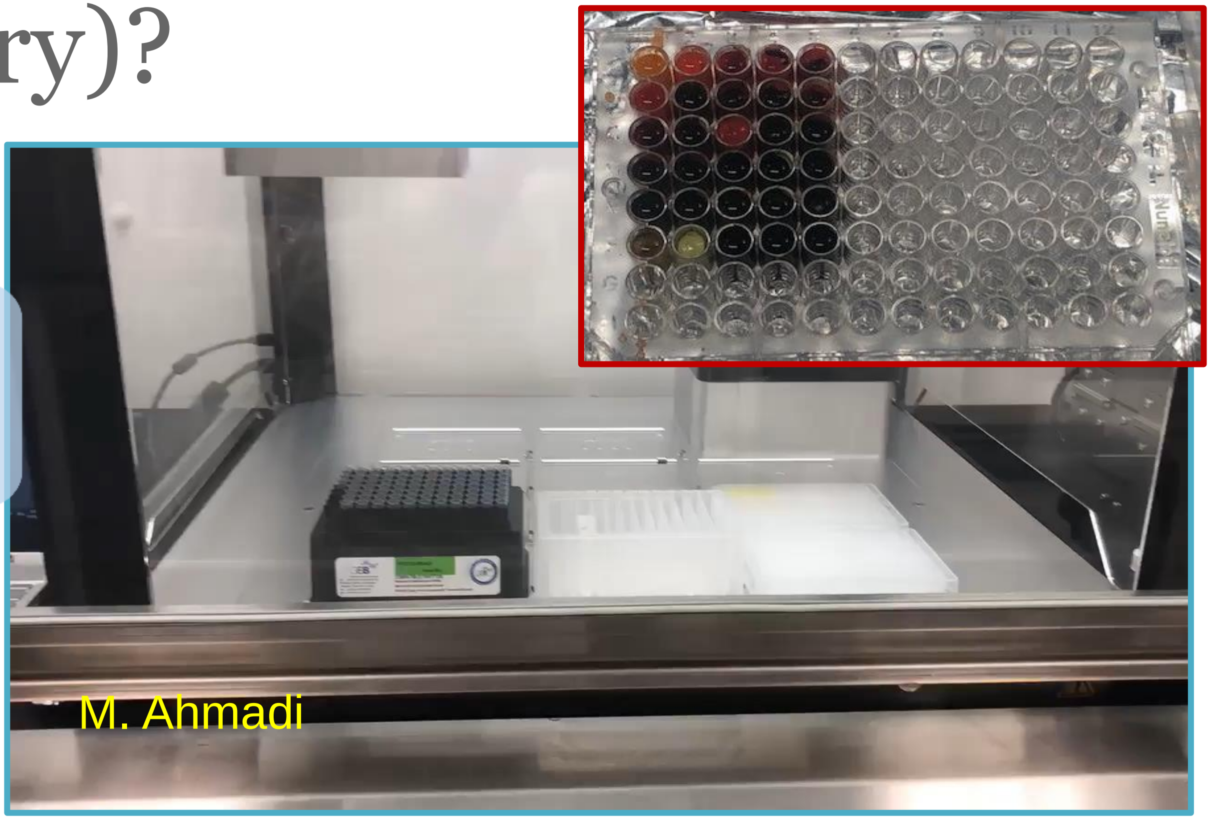
Evidence [can typically be absorbed into the normalization of the posterior]

- Prior
- Posterior
- Belief

Gaussian Processes

Why synthesis (or theory)?

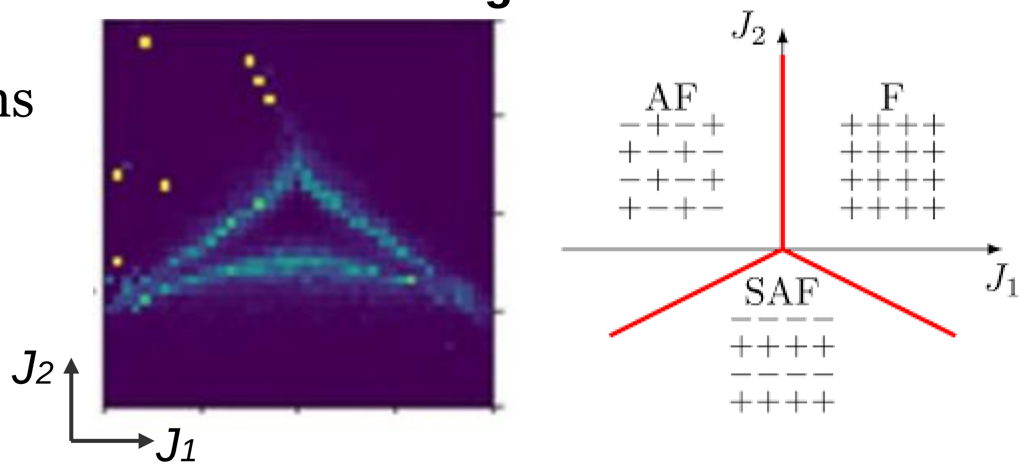




- Automated synthesis in its simplest form requires some way to navigate phase diagrams
- In more complex form, processing space.
- Ideally, incorporate physical knowledge

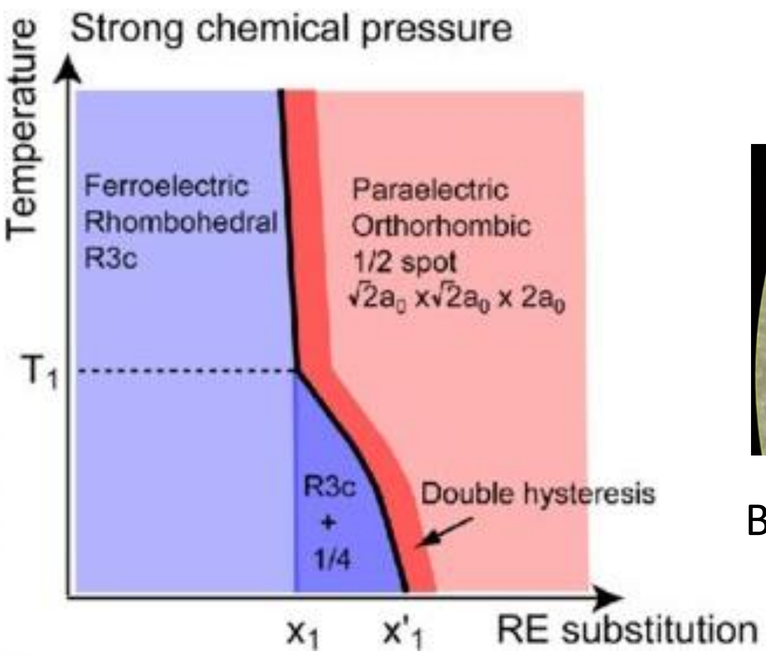
- Similar problem - theory

Ising model

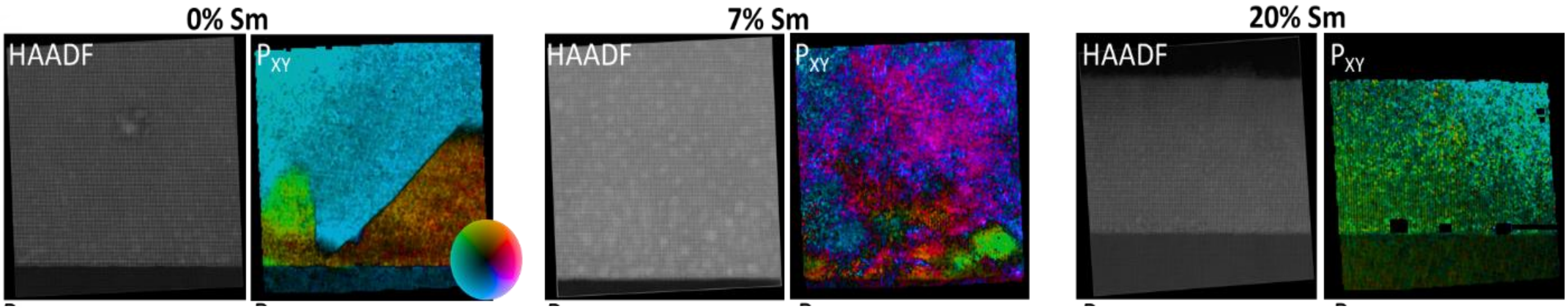


Combinatorial library

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

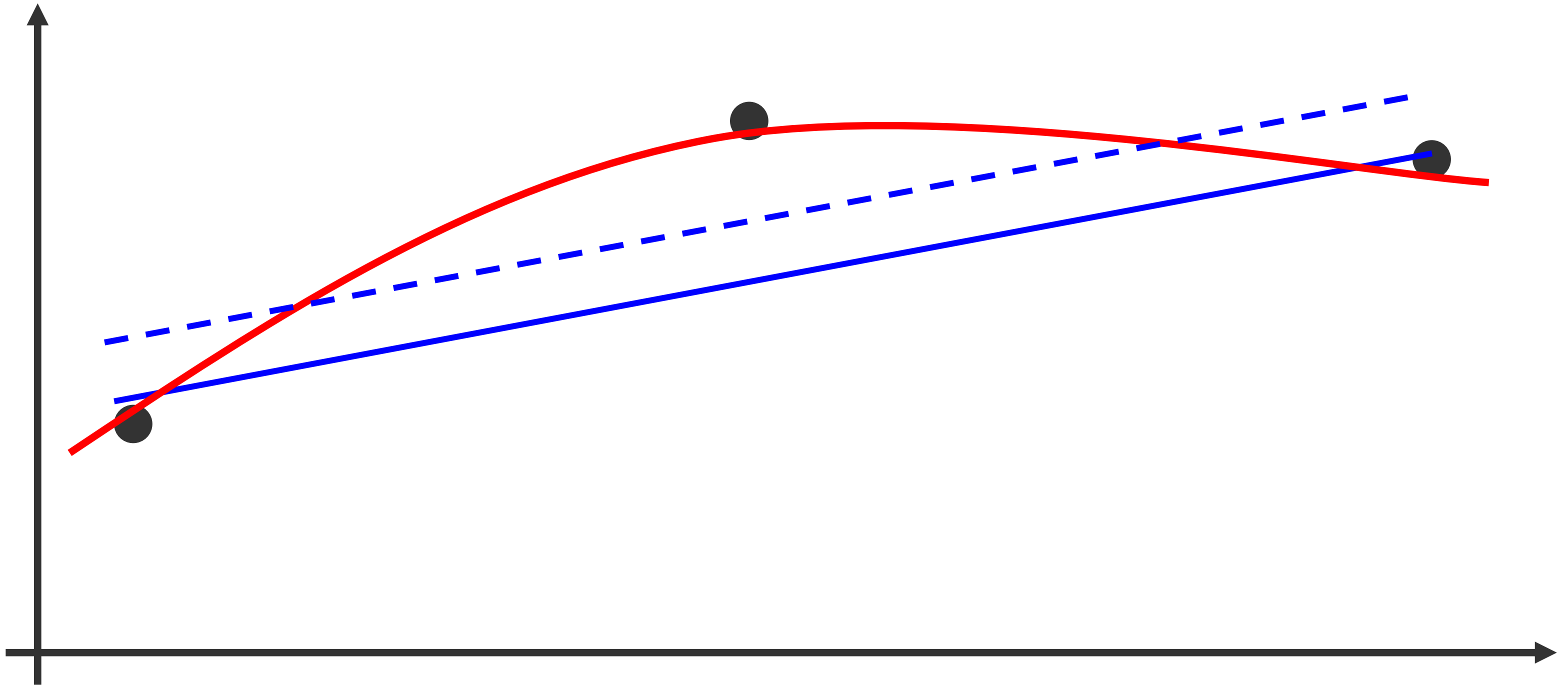


$BiFeO_3$ Linear est. 7%Sm $BiFeO_3$ 20%Sm $BiFeO_3$



arXiv:2004.11817

What do we know if we do not know anything?



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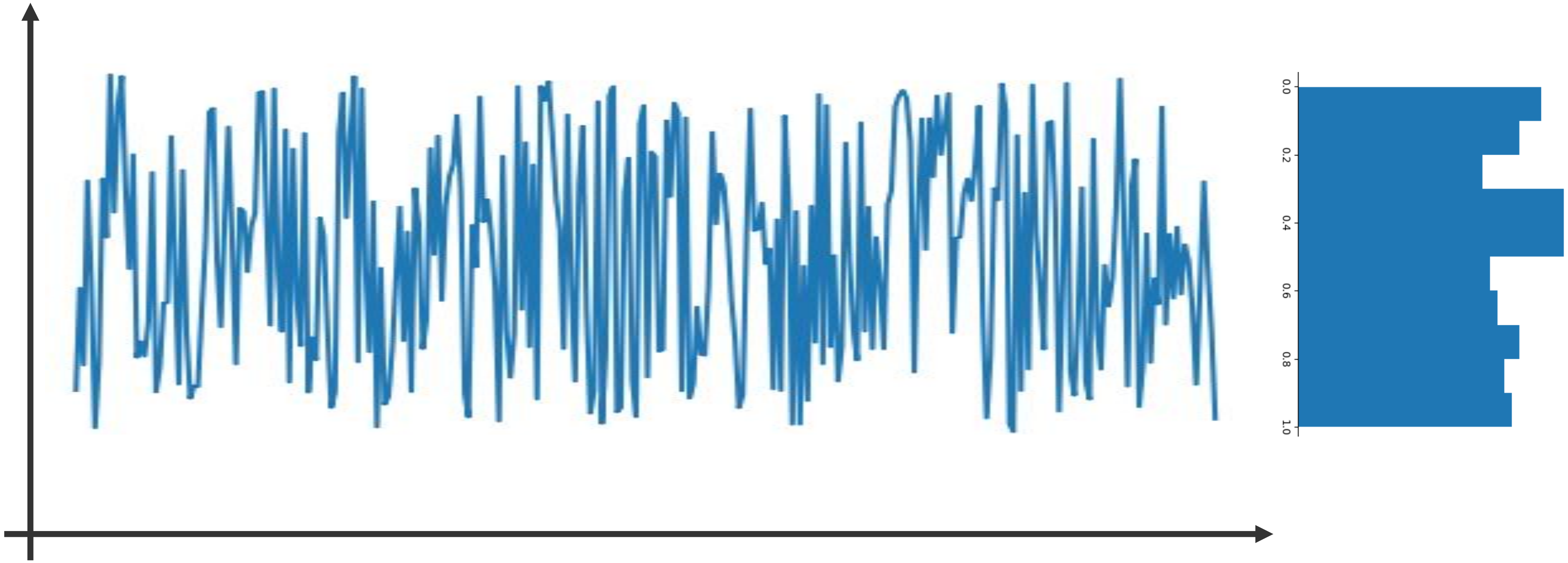
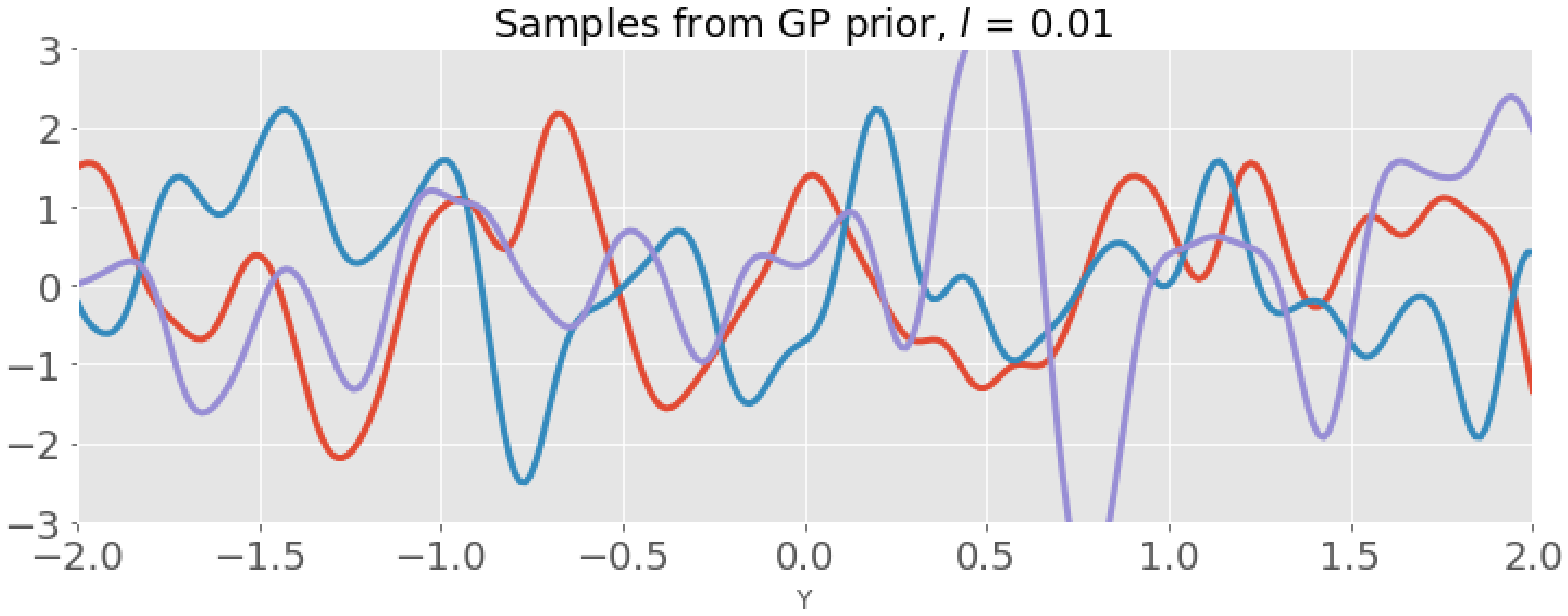


Gaussian Process Regression

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

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



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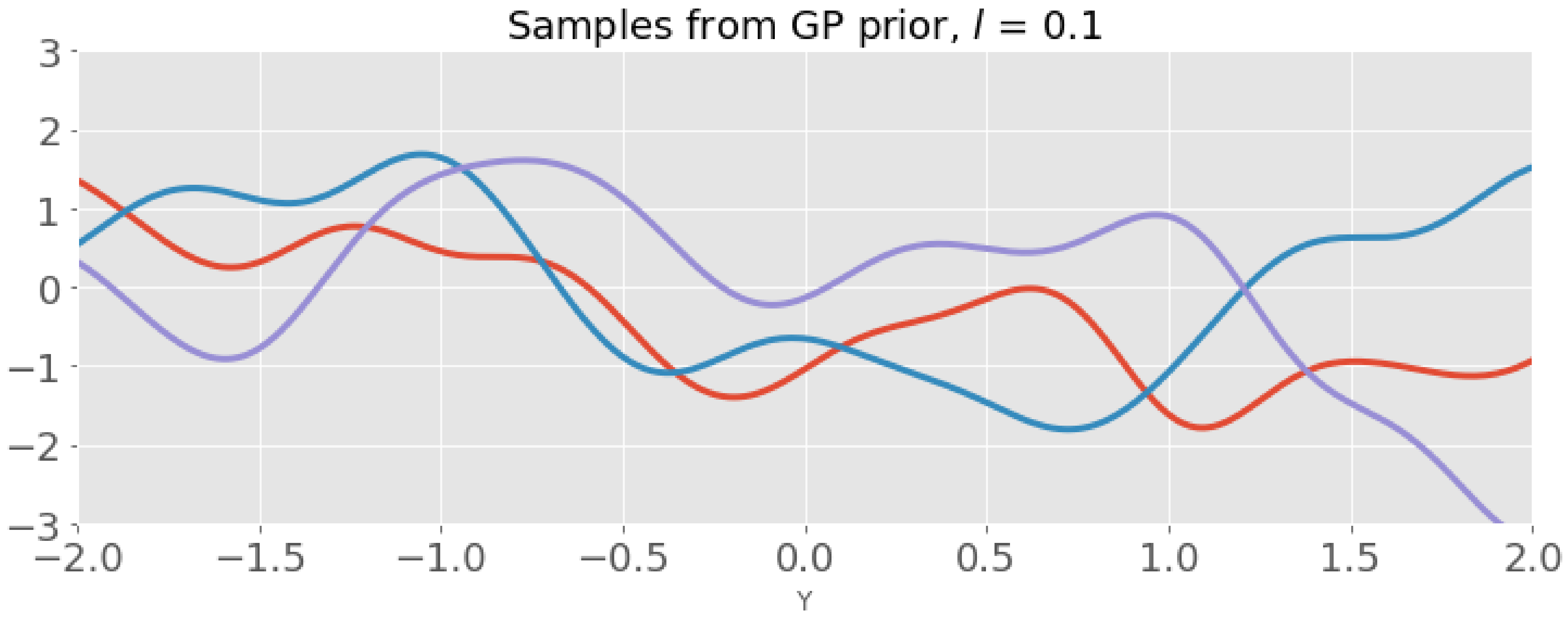
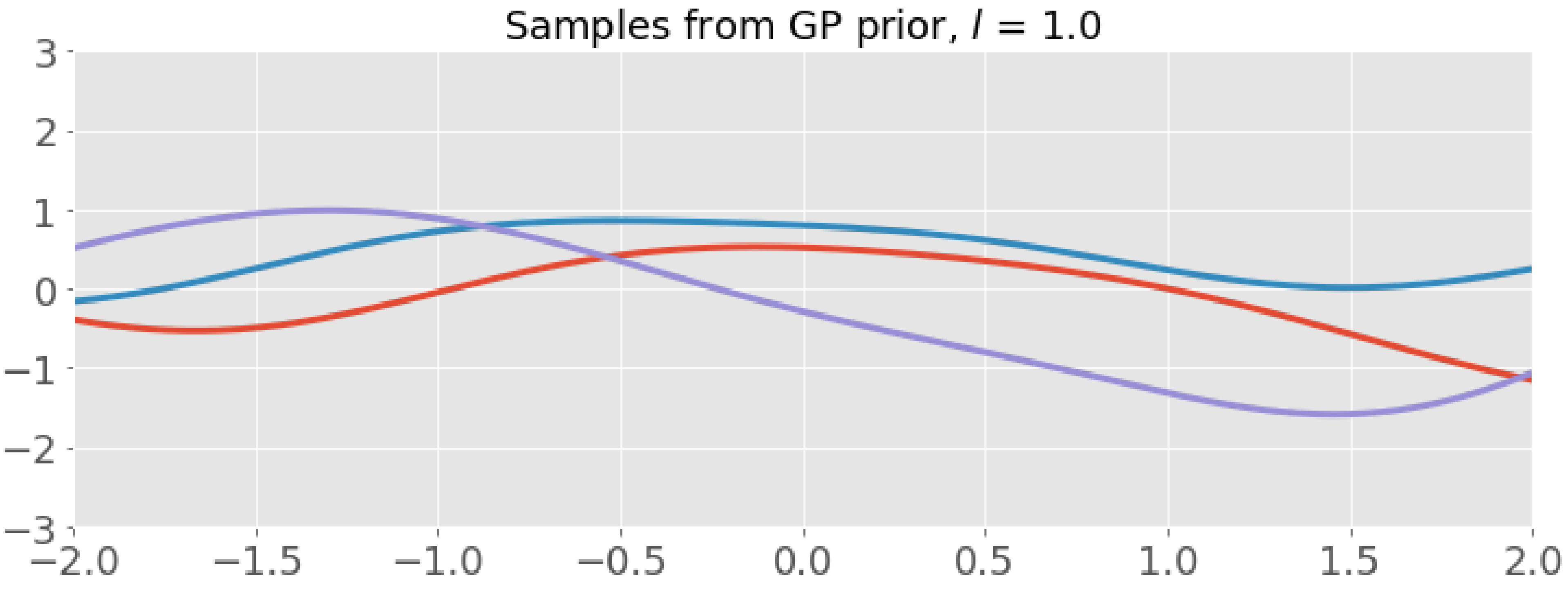


Gaussian Process Regression

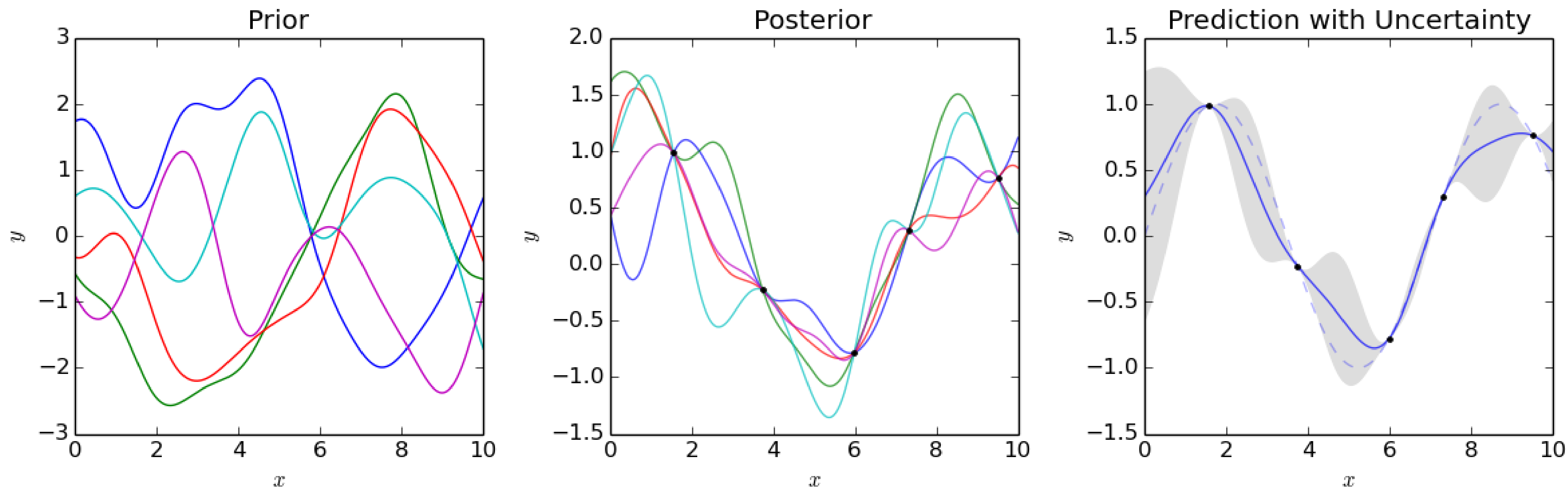
- Covariance matrix (kernel) determines what type of functions we will allow.

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



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

Gaussian Process Regression

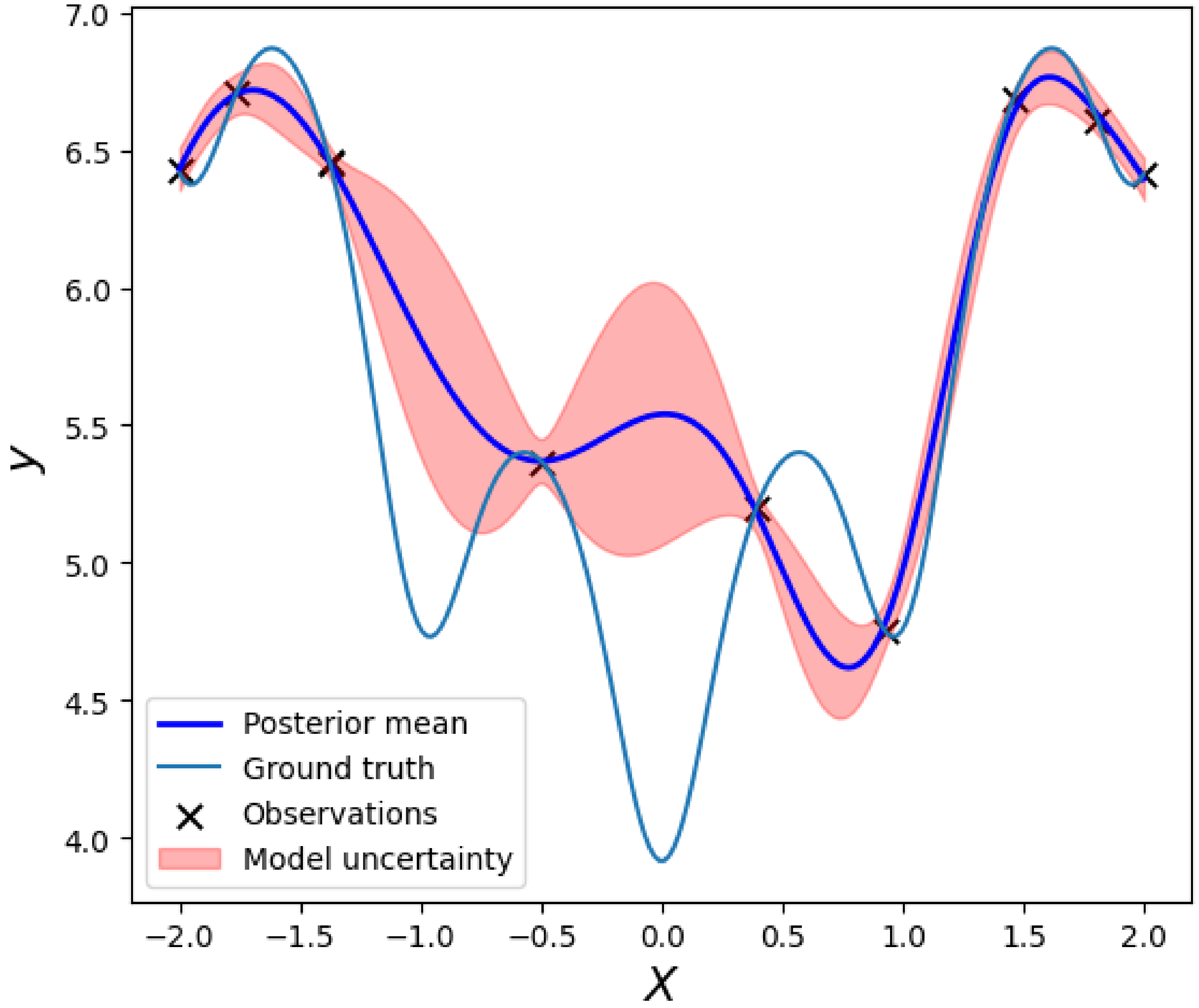


Prior:	What can the function be before the measurement
Data:	Measurement
Posterior:	What can the function be after measurement
Policy:	How do we balance exploration and exploitation (acquisition function)

GP Vocabulary

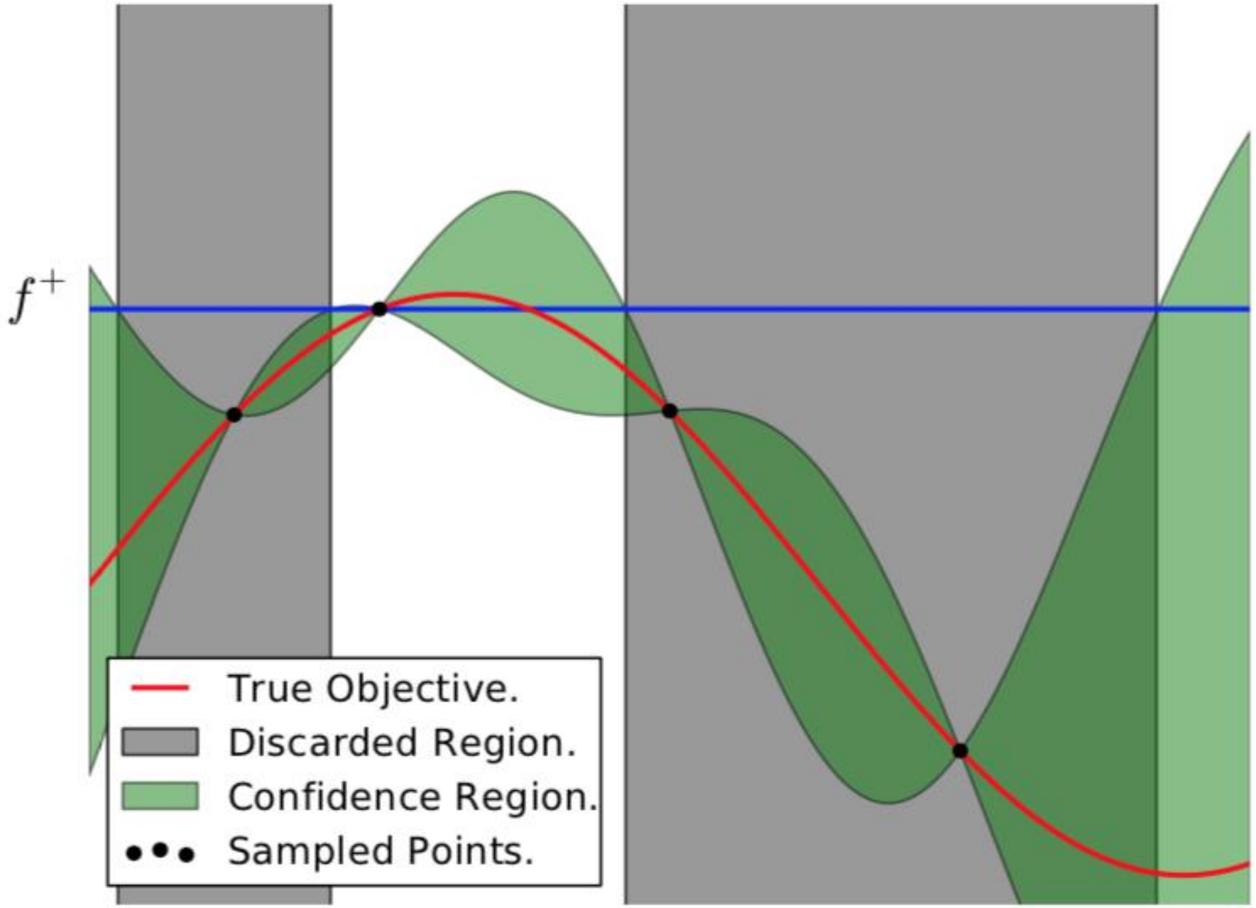
- Gaussian Process
- Kernel and kernel parameters
- Kernel Priors
- Noise Priors
- Posteriors

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

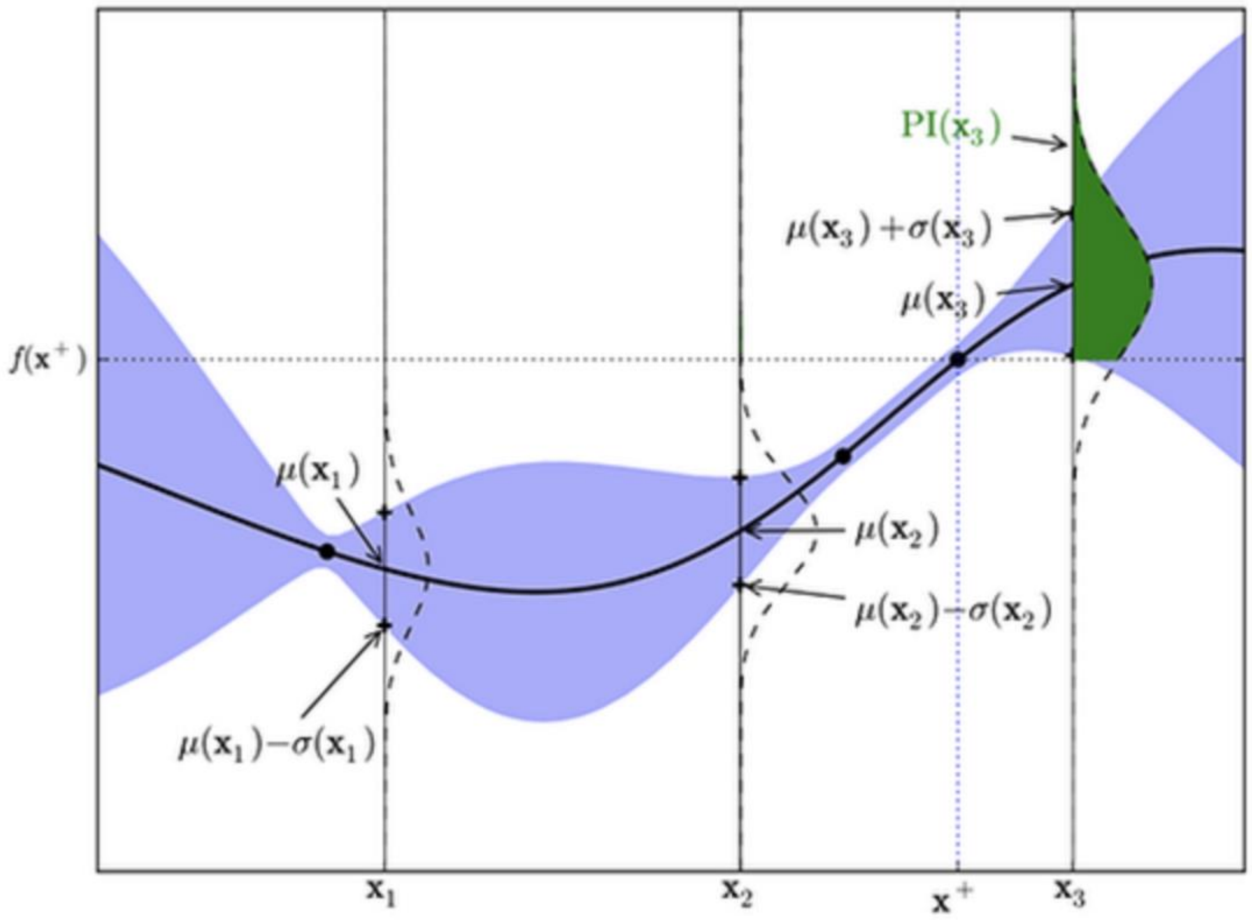


- We have some measurements in space X, and we want to maximize some property f(X).
- How can we decide what point to measure next to best maximize f?
- We need to balance the exploration of the space with exploitation of regions near we have already know

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



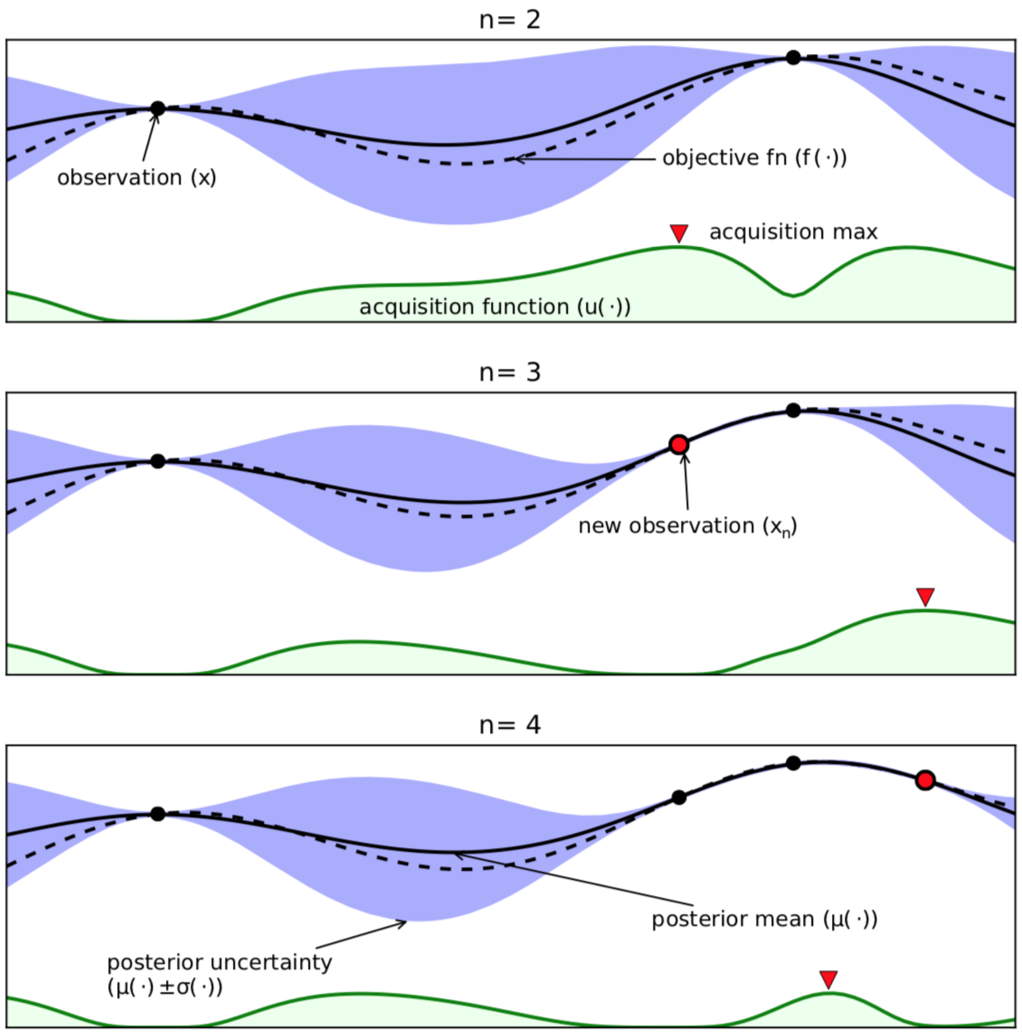
1. **Upper confidence bound:** simplest possible - just take the upper confidence bound from the prediction

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

Obtain/update training data *D*

↓

Train surrogate model $X \rightarrow y$

↓

Make prediction $X_* \rightarrow \bar{f}_*, \mathbb{V}[f_*]$

↓

Acquisition function $acq(\bar{f}_*, \mathbb{V}[f_*])$

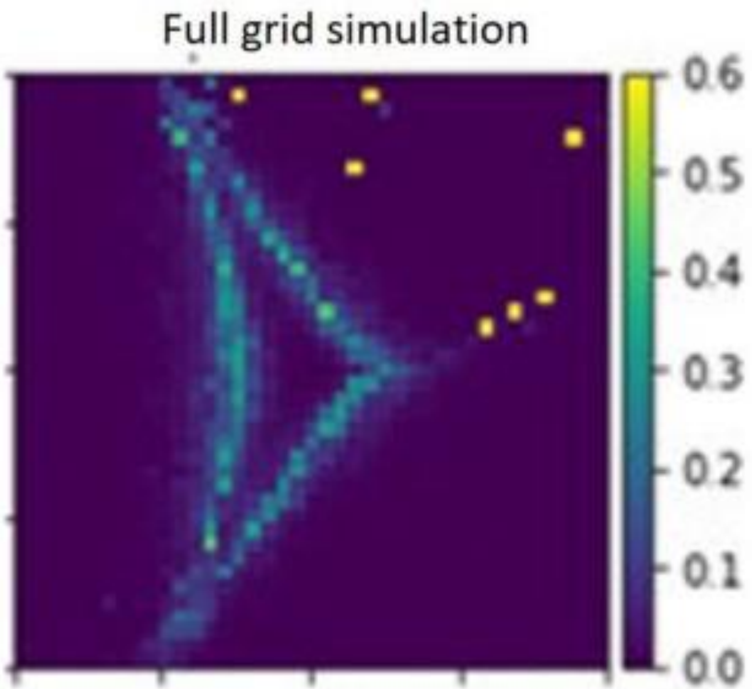
Evaluate new point

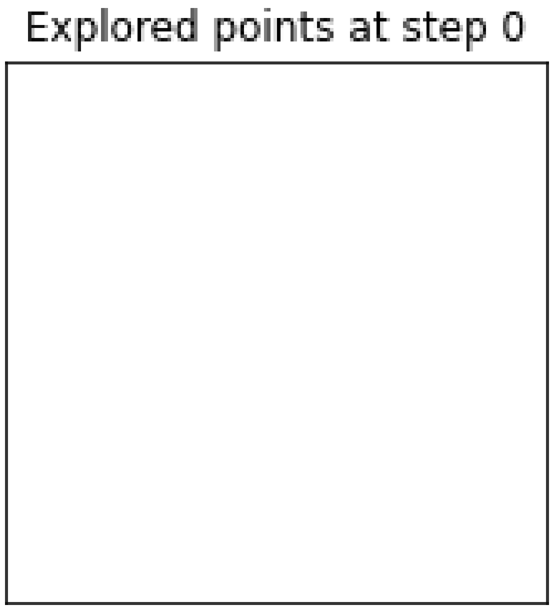
$X_*^i = \mathrm{argmax}(acq)$

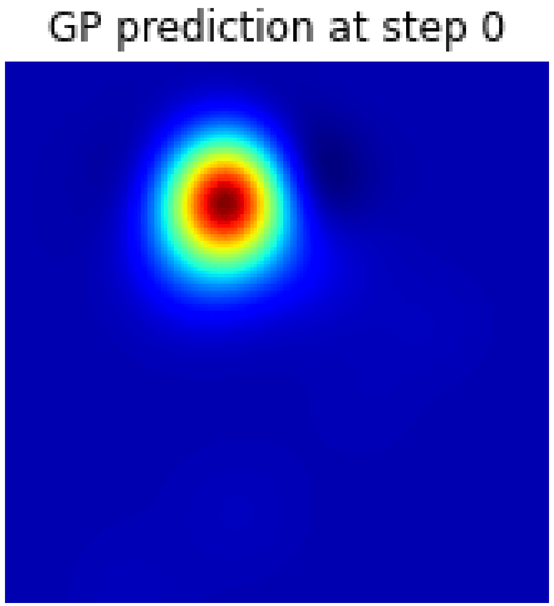
N. de Freitas et al., Taking the Human Out of the Loop: A Review of Bayesian Optimization , *Proceedings of the IEEE* **104**, 148 (2015)

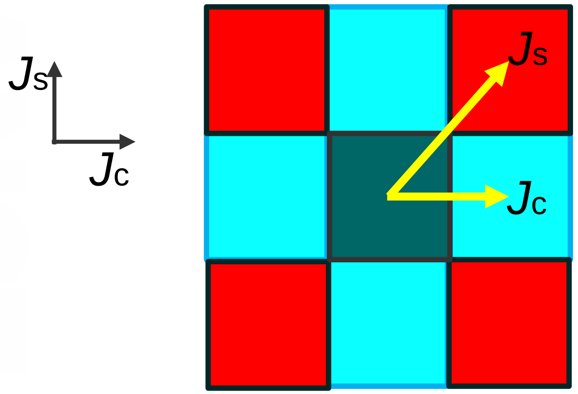
Bayesian Optimization for physical discovery

Discovering regions where heat capacity is maximized in NNN Ising model

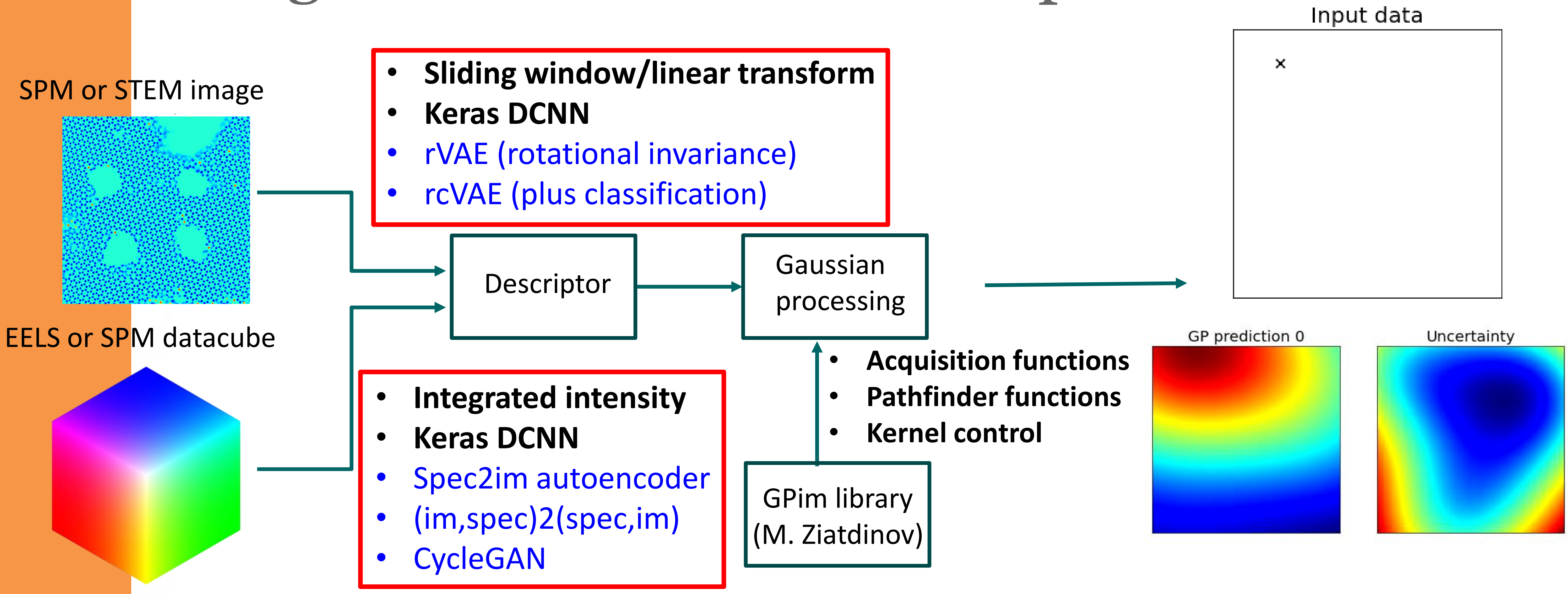






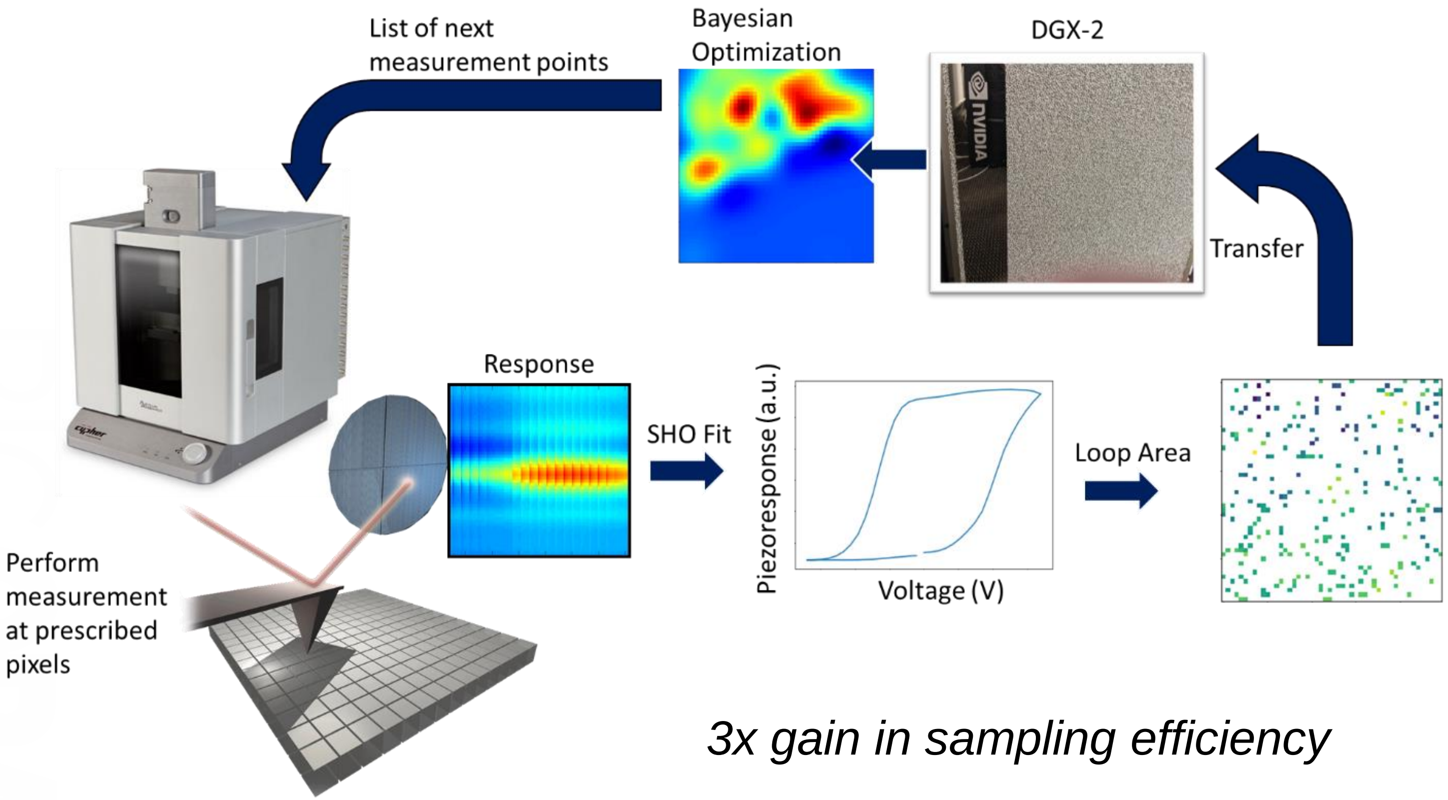


Going real time: automated experiment



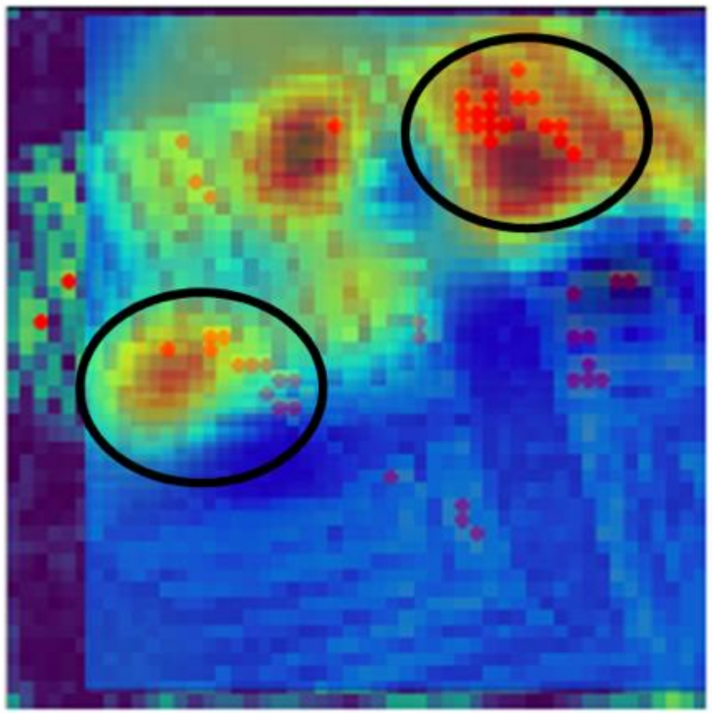
- AE based on structural analysis for STEM data
- AE based on spectral data in PFM
- AE based on DL for EELS data
- Feature of interest finding for mesoscopic images

BO for Self-Driving Microscopy



3x gain in sampling efficiency

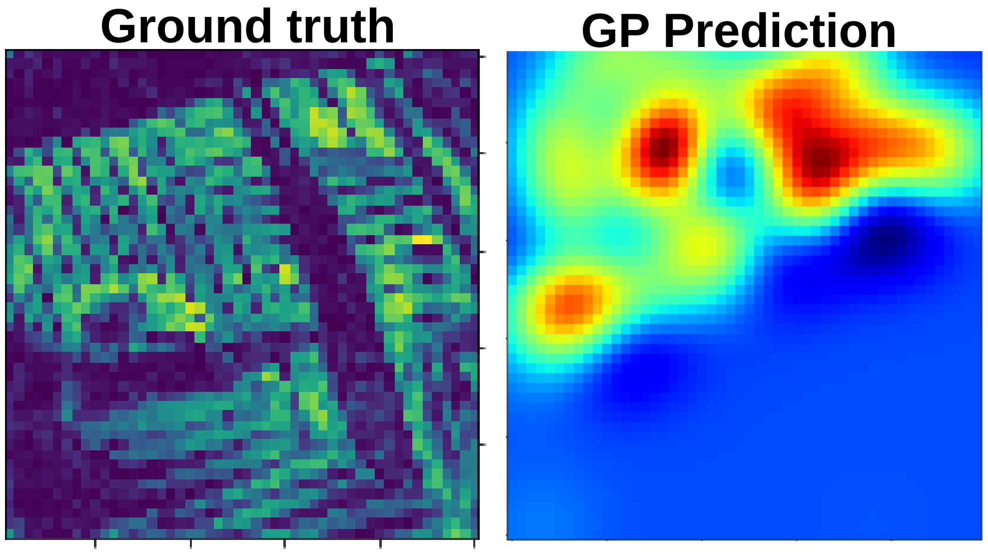
Comparison with "ground truth"



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)