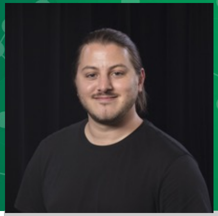
Algorithms and opportunities for self-driving laboratories: model-based control, physics discovery, and co-navigating theory and experiments

Rama Vasudevan, Aditya Vatsavai,
Yongtao Liu, Sumner Harris

Center for Nanophase Materials Sciences,
Oak Ridge National Laboratory

AI in Materials Sciences Workshop, NIST



Sumner
Harris

Yongtao
Liu

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- Mathew Cherukara (ANL)

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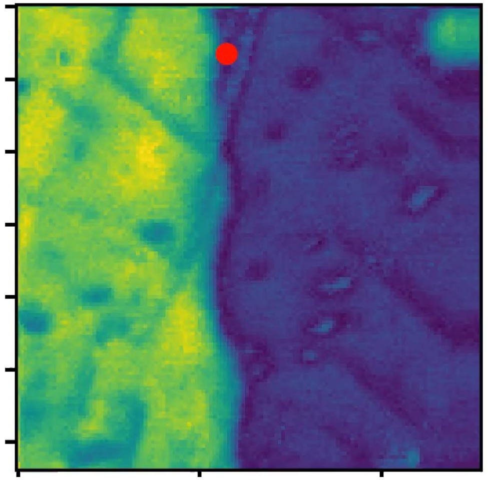
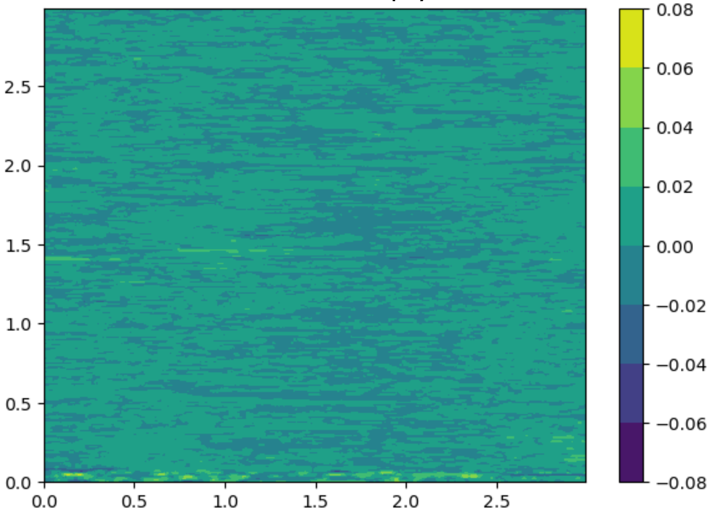
Autonomous Labs: Characterization and Synthesis

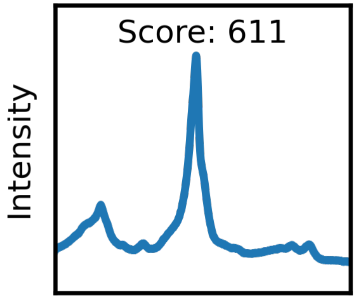
Piezoresponse Force Microscopy

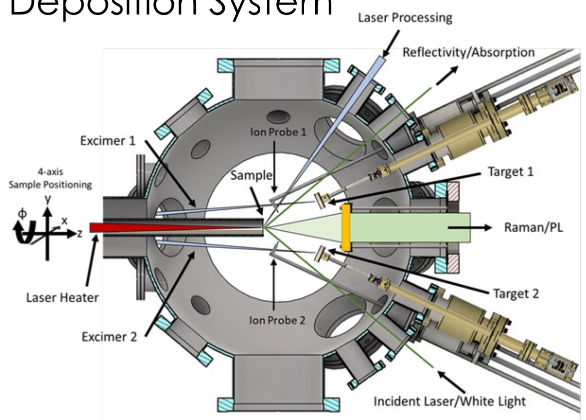


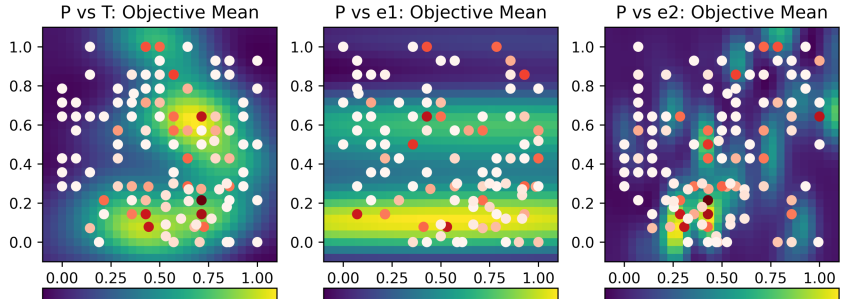
Scanning Tunneling Microscopy



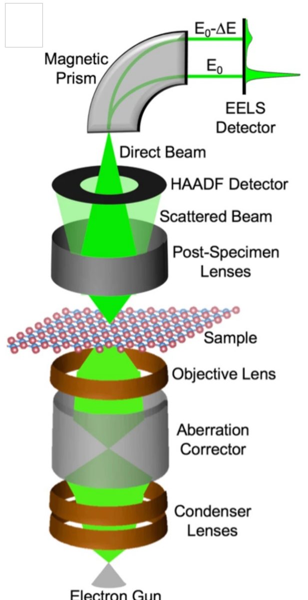
Pulsed Laser Deposition System

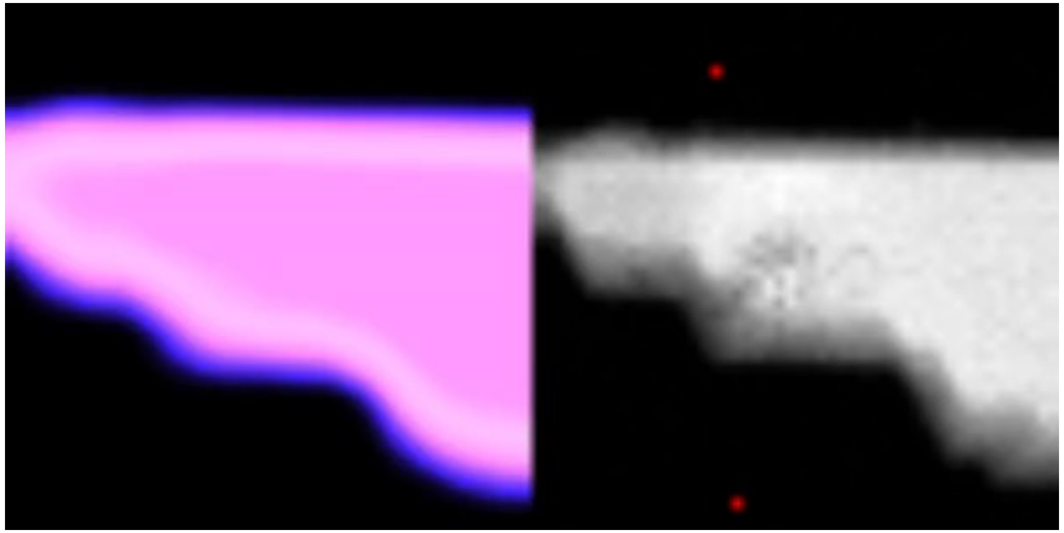




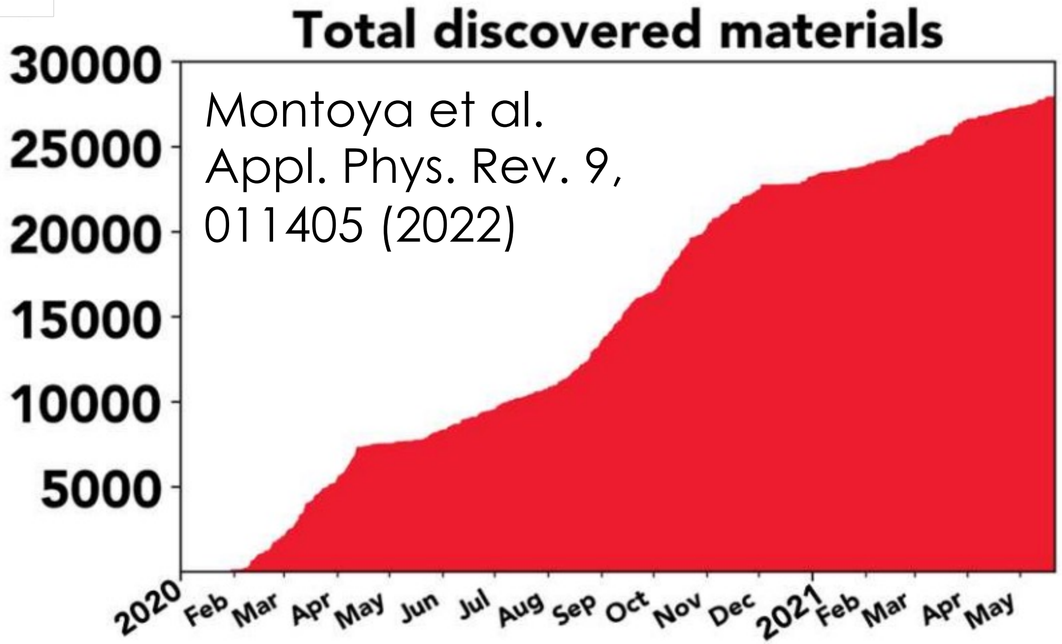


Scanning Transmission Electron Microscopy





Realizing the promise of ML



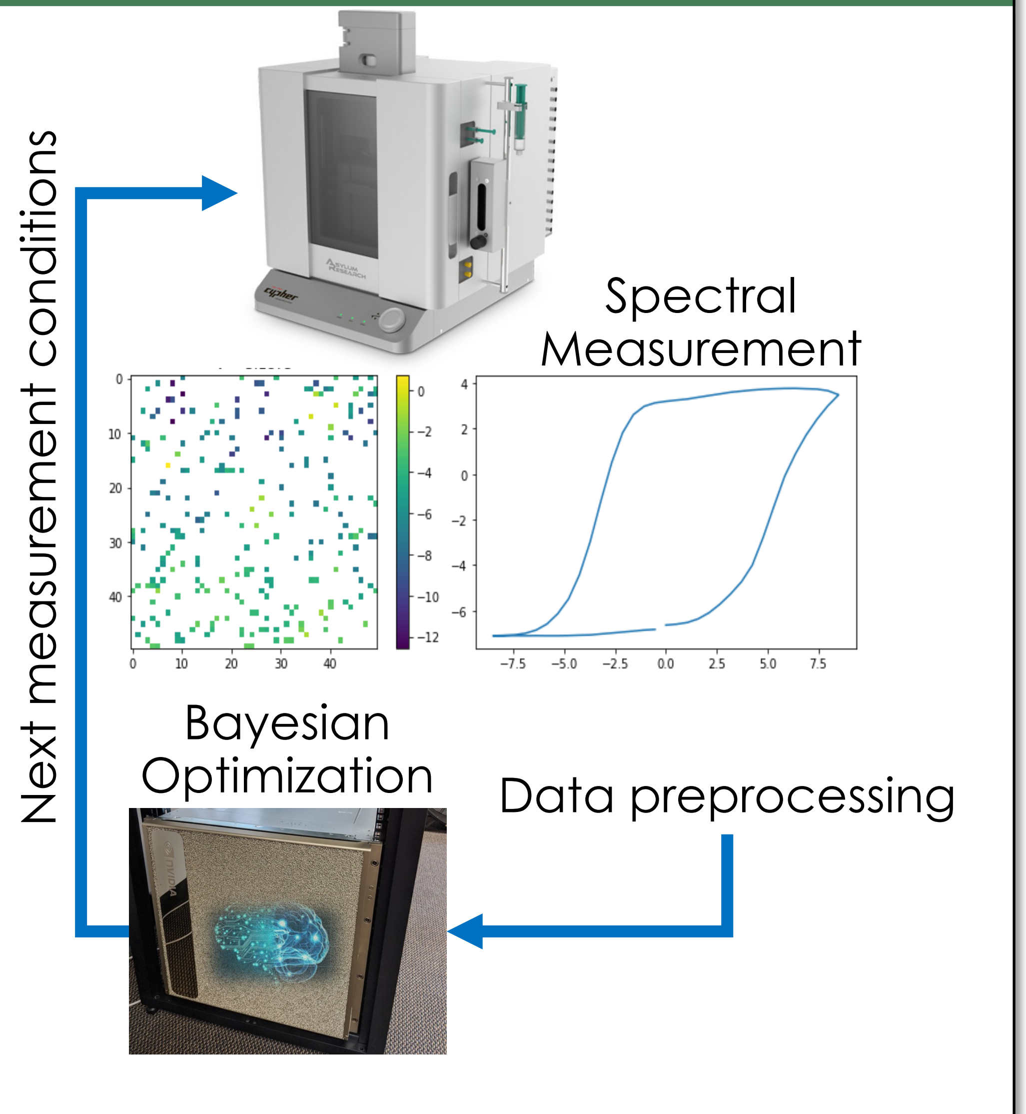
Accumulated new crystal structures in 1374 chemical systems explored by CAMD

- It is possible to predict thousands of structures quite quickly
- We can use data driven methods for property optimization in autonomous workflows
- However:

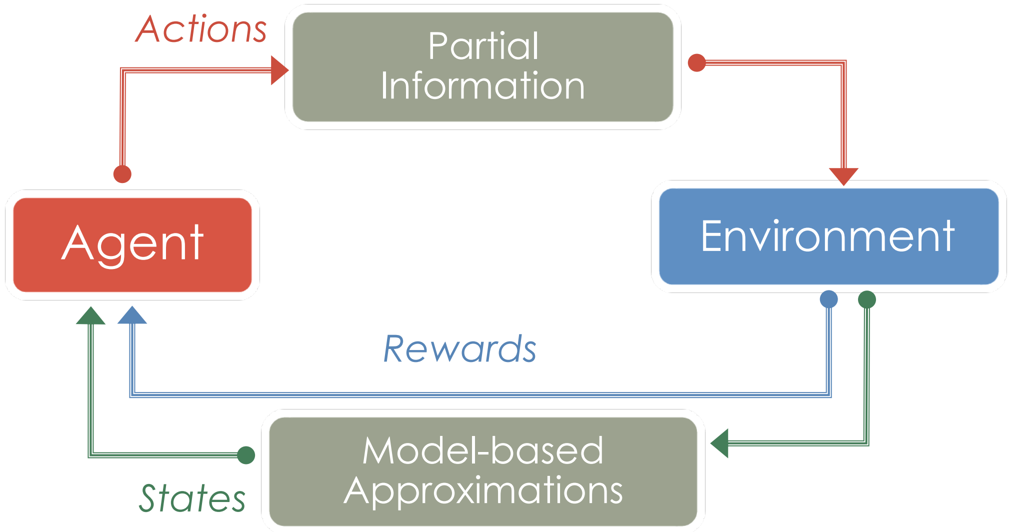
1. Physics Discovery work is less explored
2. Realizing predicted structures in synthesis

Algorithms for autonomous labs focus on properties

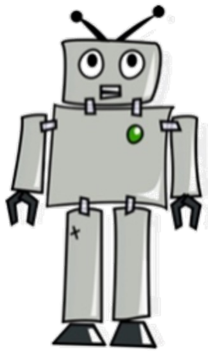
Bayesian Active Learning

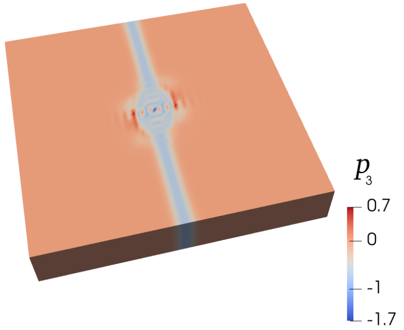


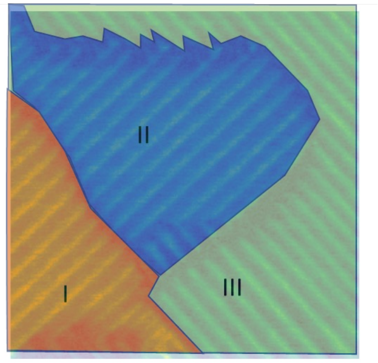
Reinforcement Learning



$$\nabla_\theta J(\theta) = \mathbb{E}_\tau\left[\sum_{t=0}^{T-1} \nabla_\theta \log \pi_\theta(a_t|s_t) G_t\right]$$



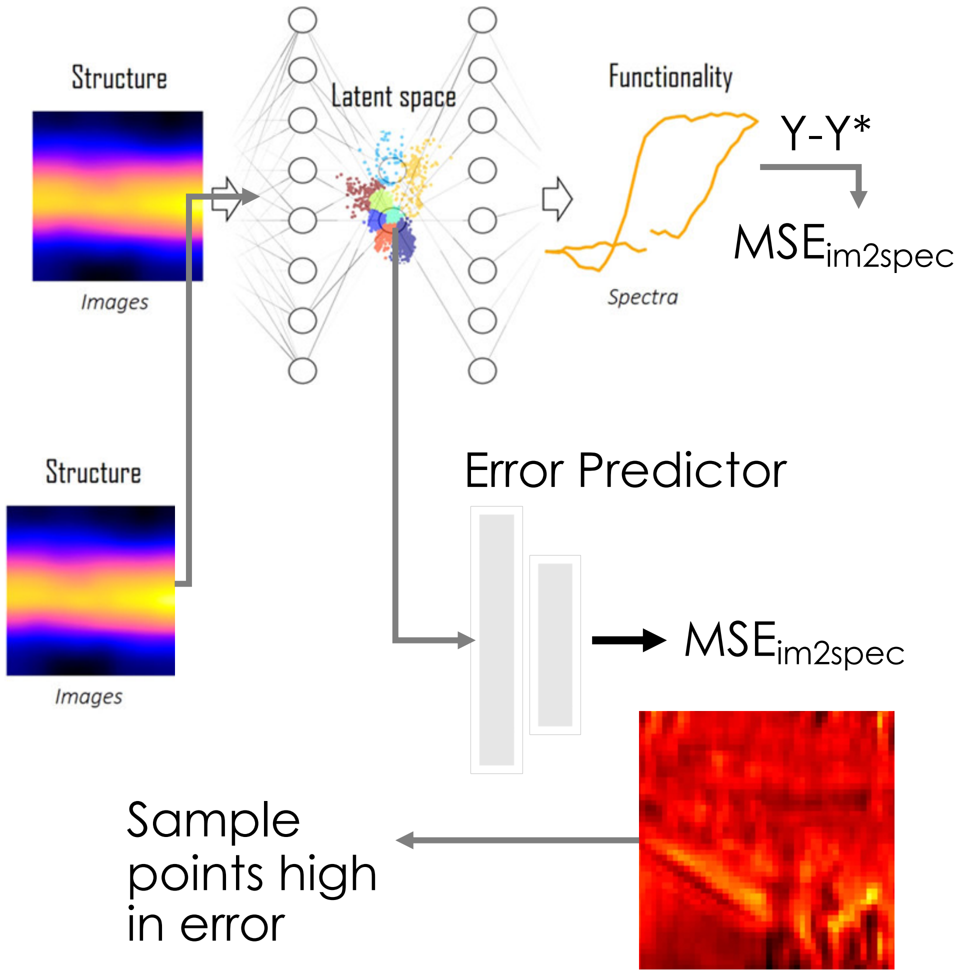




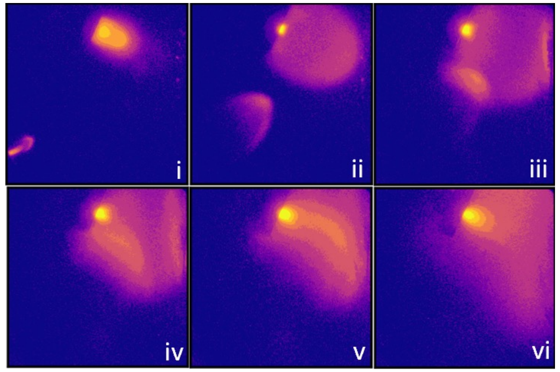
Can we produce alternatives?

"Bayesian optimization-lite" method

ACS Appl. Mater. Inter. 13, 1693 (2021)



Model-based control for synthesis



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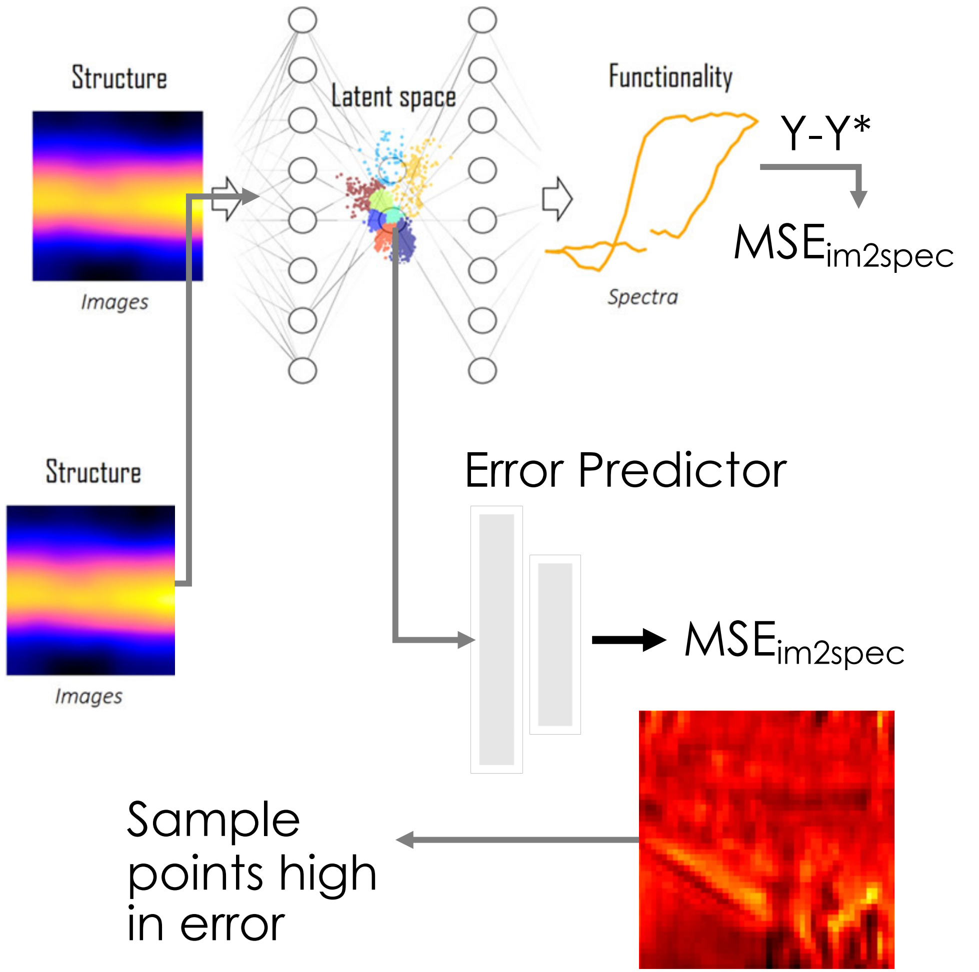
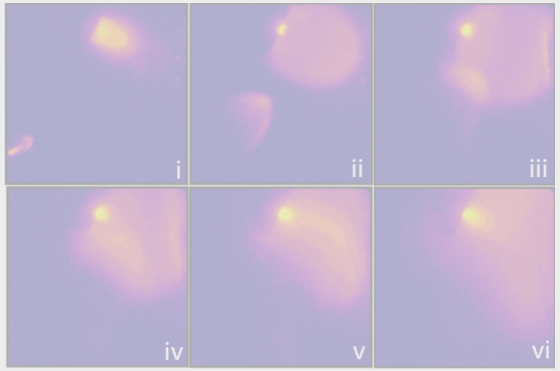
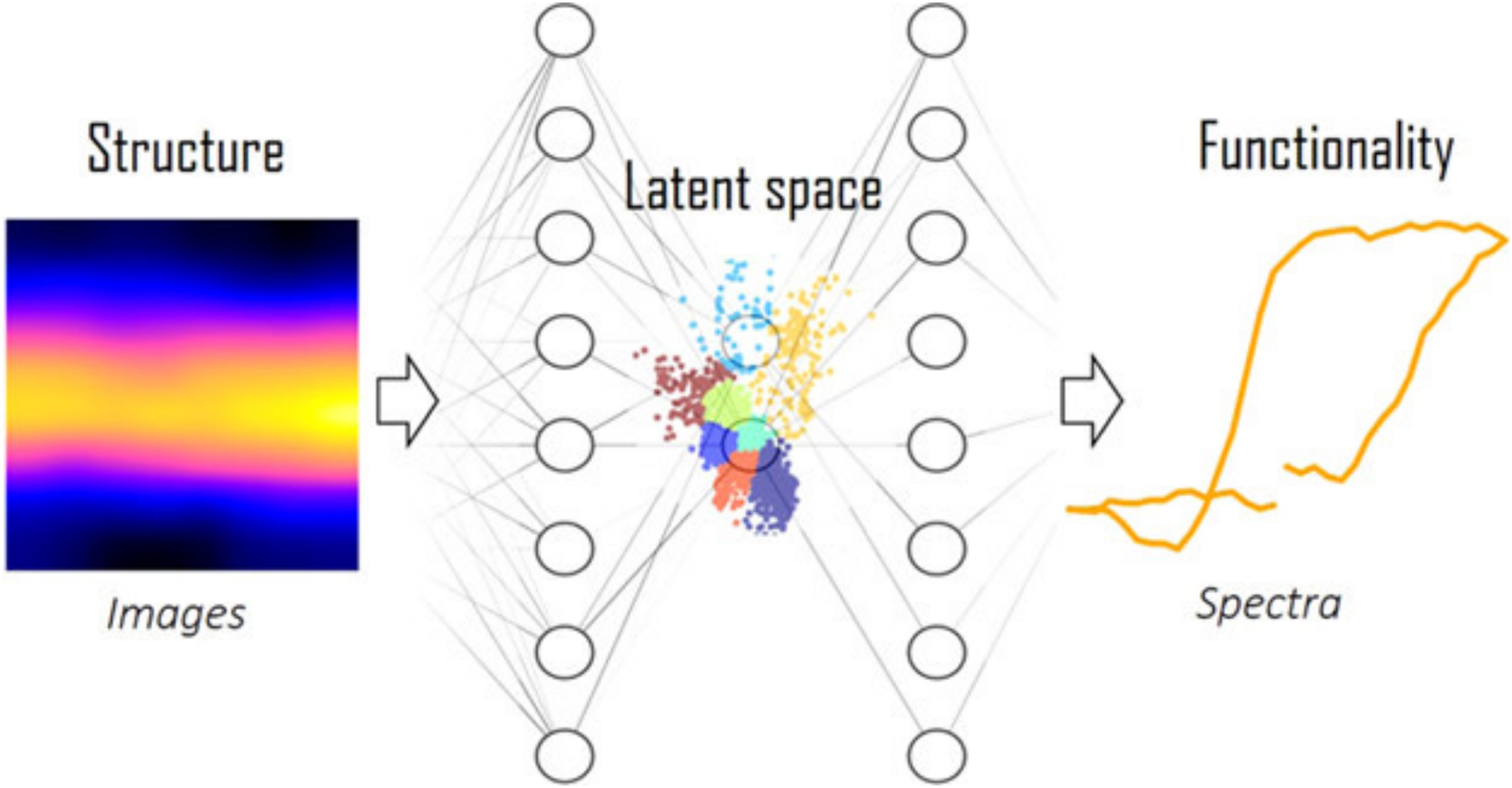


Model-based control for synthesis



Im2Spec: Predicting Spectra from Image Patches

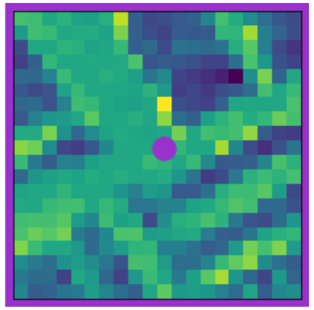
Kalinin et al. ACS Appl. Mater. Inter. 13, 1693 (2021)

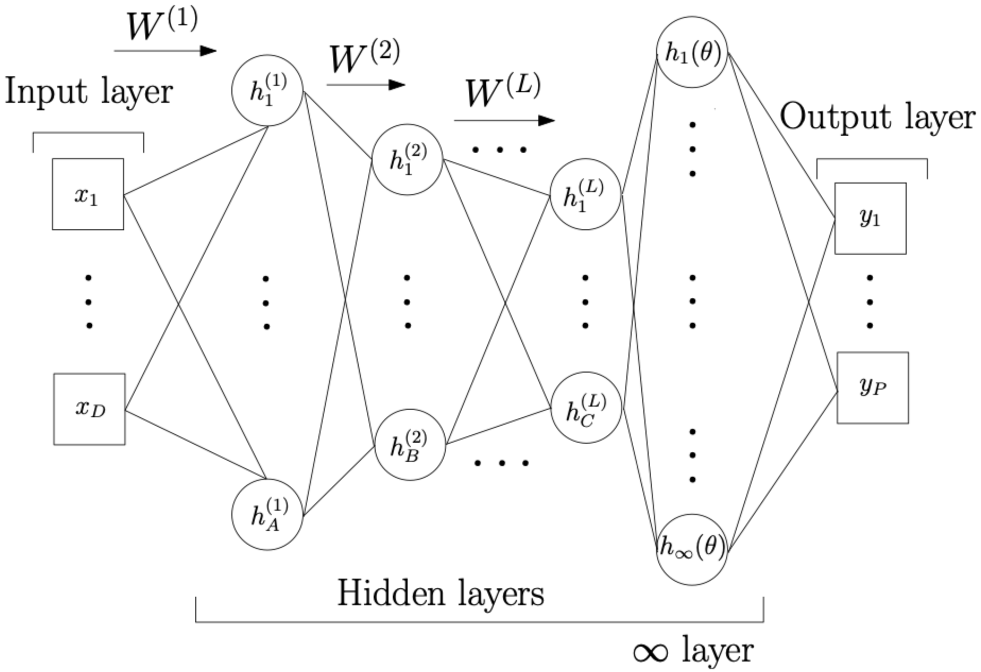


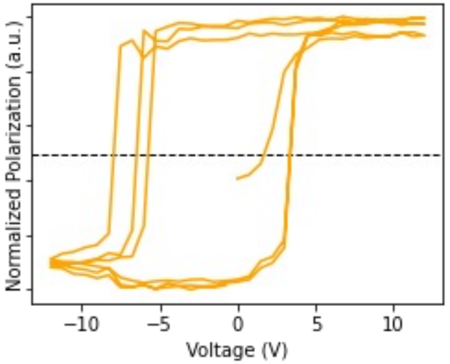
Structure-property correlations in microscopy for unearthing physics

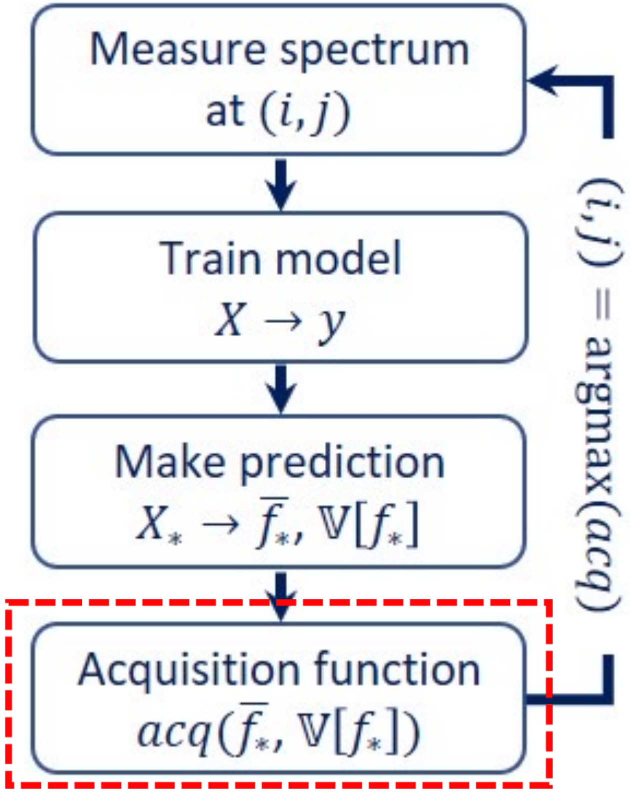
Deep Kernel Learning: More images, better kernels

Deep Kernel Learning
(A.G. Wilson, 2015)









Combine a neural network with GP, learn parameters jointly

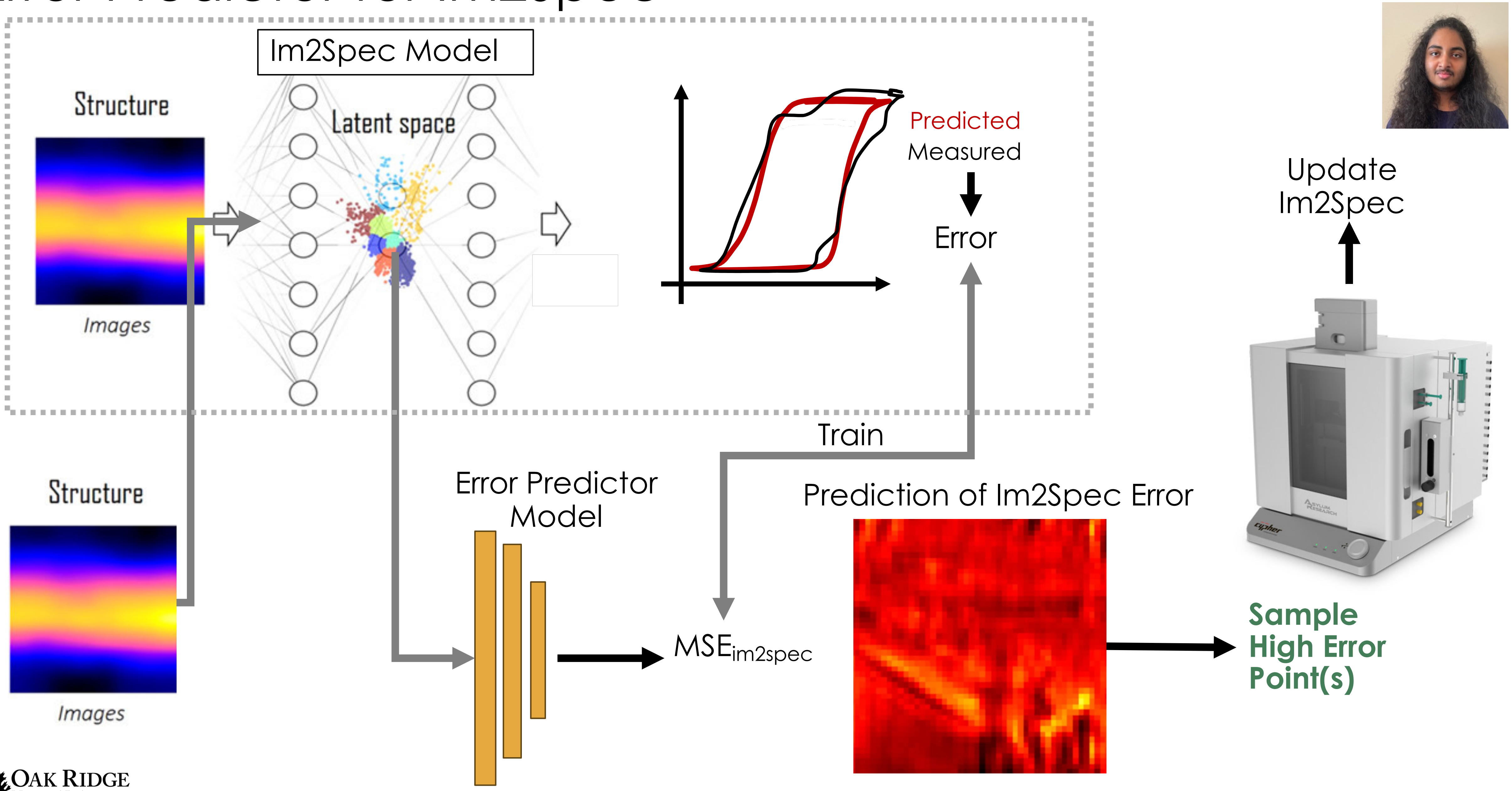
Liu, Yongtao, Kyle P. Kelley, Rama K. Vasudevan, Hiroshi Funakubo, Maxim A. Ziatdinov, and Sergei V. Kalinin. Nature Machine Intelligence 4, 4 (2022): 341-350.

"BO-Lite" Method for Data-driven Model Generation

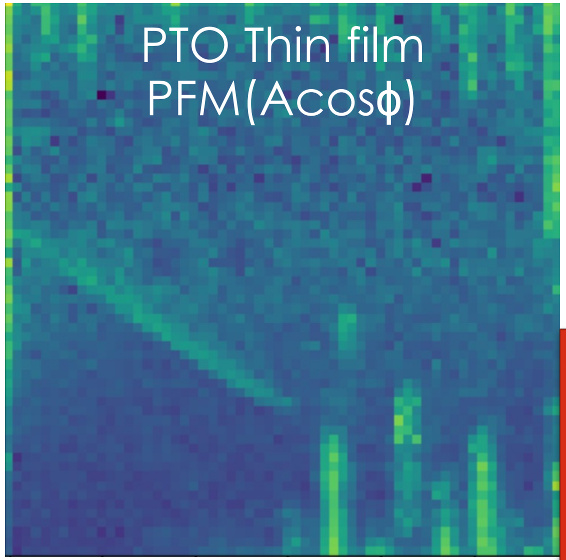
1. BO can work when we have only a few outputs. Large numbers of correlated outputs become computationally expensive
2. The aim of our experiment is to uncover physics. This is accomplished if we can construct a model of images to spectra
3. Ergo, we should sample points that reduce the error in Im2Spec, but the spectra are high dimensional (e.g., 64 dimensional), so standard DKL or BO is not tractable
4. Solution: Use another neural network to predict the error of Im2Spec!

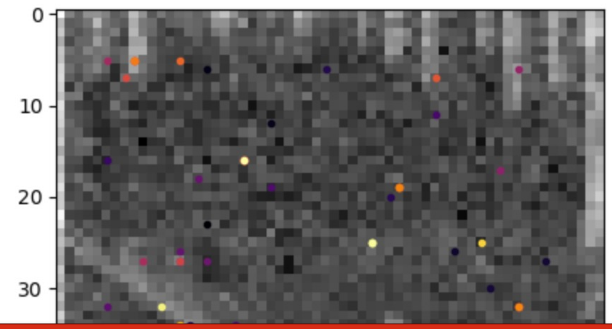
Error Predictor for Im2Spec

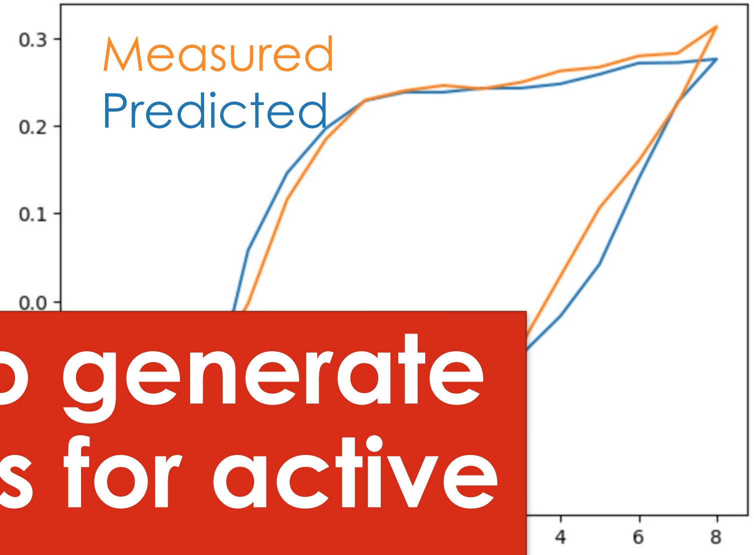
Adi Vatsavai
UNC



Error Prediction Model Results (100 points sampled)

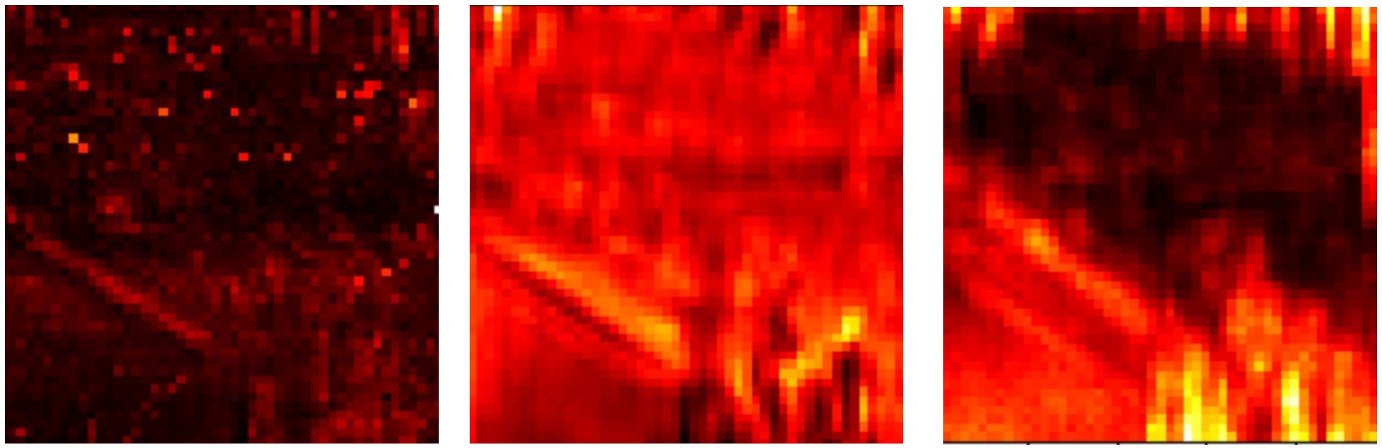


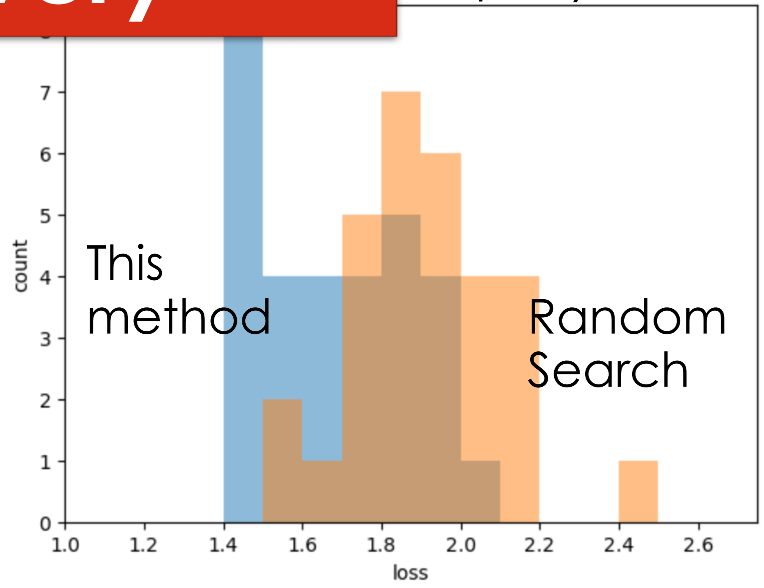




Summary: Try to generate more algorithms for active physics discovery

GT Error





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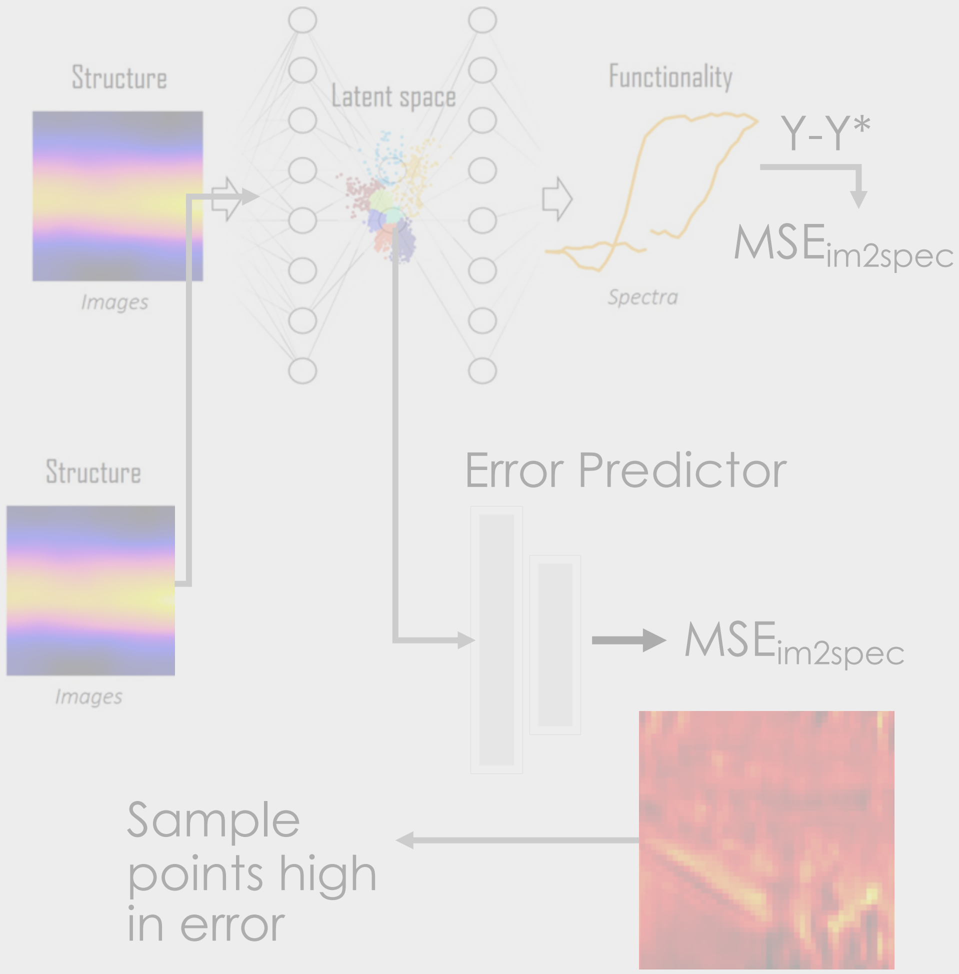
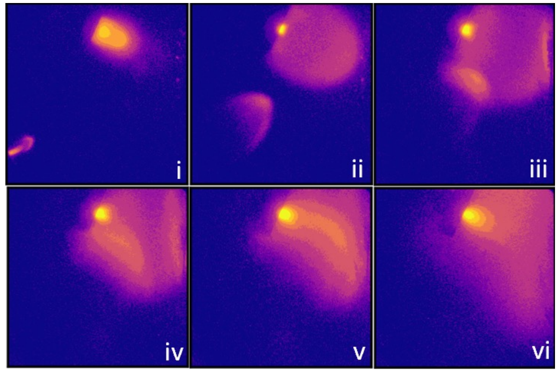


Model-based control for synthesis



To realize control, we need to predict state dynamics

- The "**state process**" is the state of a dynamical system at time instant *n* where *h* is the "state model", θ is a vector of *parameters* and *w* is random noise ~ $\mathcal{N}(0,\sigma)$

$$X_{n+1} = h(X_n, \theta) + w_n$$

- The "**observation process**" is a measurement related to the state X_n where H is some matrix and ξ is random noise ~ $\mathcal{N}(0,\gamma)$

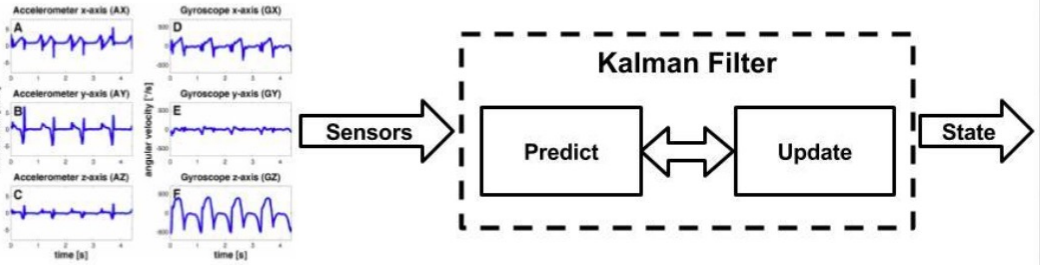
$$Y_{n+1} = HX_{n+1} + \xi_{n+1}$$

Goal: estimate the parameters θ of the state model from $\{Y_n\}$

- Solve this problem with ***particle filters.*** Use the recursive predict-update equations to generate a cloud of particles to approximate the posterior pdfs

https://realitybytes.blog/2017/08/15/state-estimation-kalman-filters/

Traditional method: Kalman Filter



$$\begin{bmatrix} x \\ y \\ z \\ \theta \\ \dot{x} \\ \dot{y} \\ \dot{z} \\ \vdots \end{bmatrix}$$

Simple two-step kinetic model for growth of thin film

Growth of 2D material with PLD can be represented by a simple ordinary differential equation, with two unknown parameters (nucleation and growth coefficients)

Nucleation and Growth

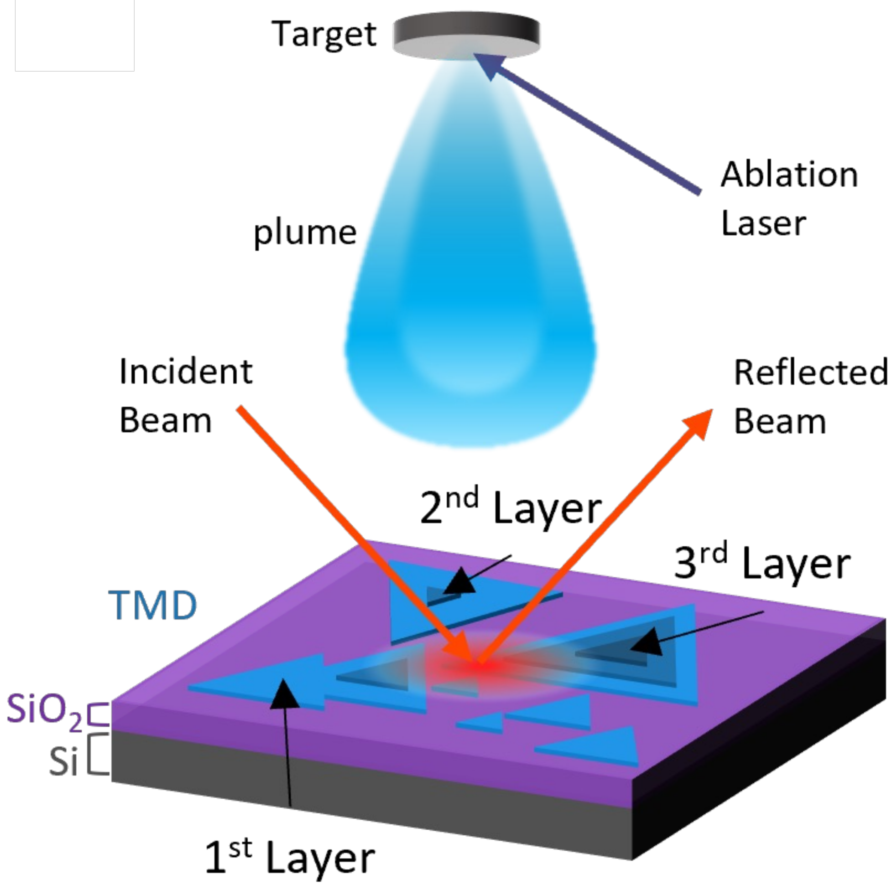
$$A \overset{k_n}{\rightarrow} B, nucleation$$

$$A + B \overset{k_{gr}}{\rightarrow} 2B, growth$$

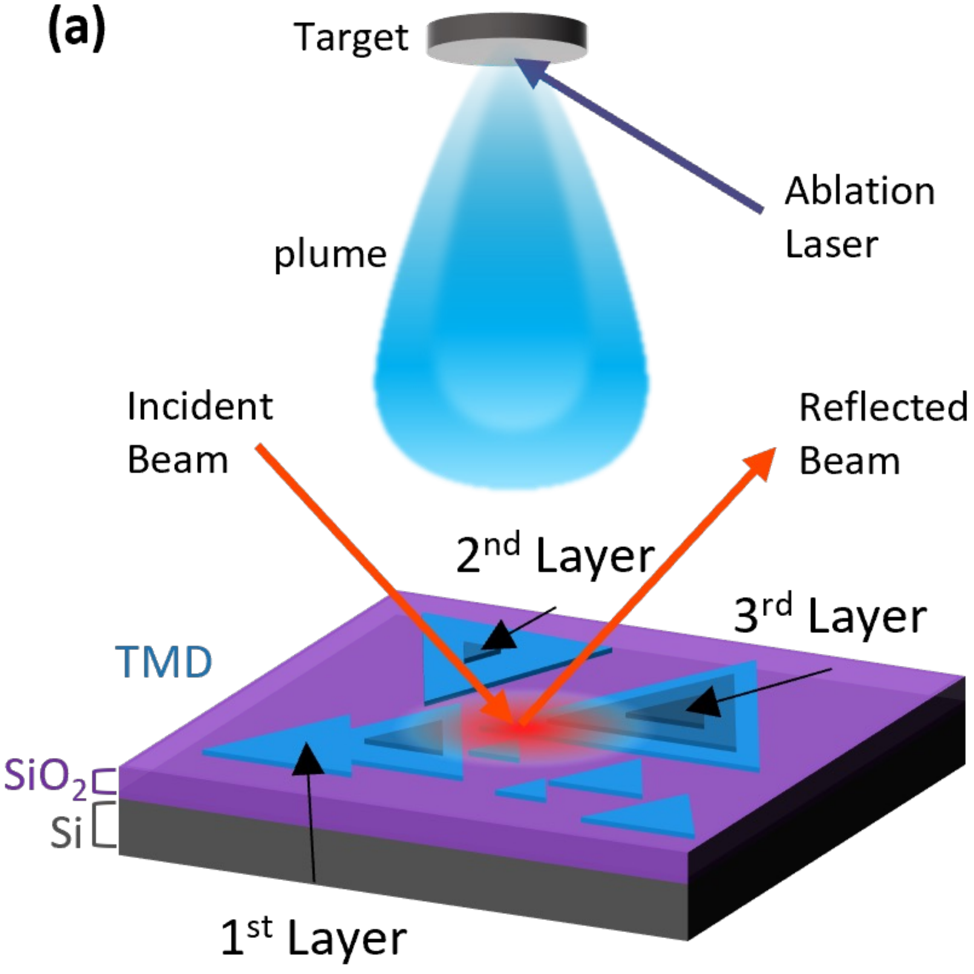
$$\frac{df_i}{dt} = k_{ni}(f_{i-1} - f_i) + k_{gri}(f_{i-1} - f_i)f_i$$

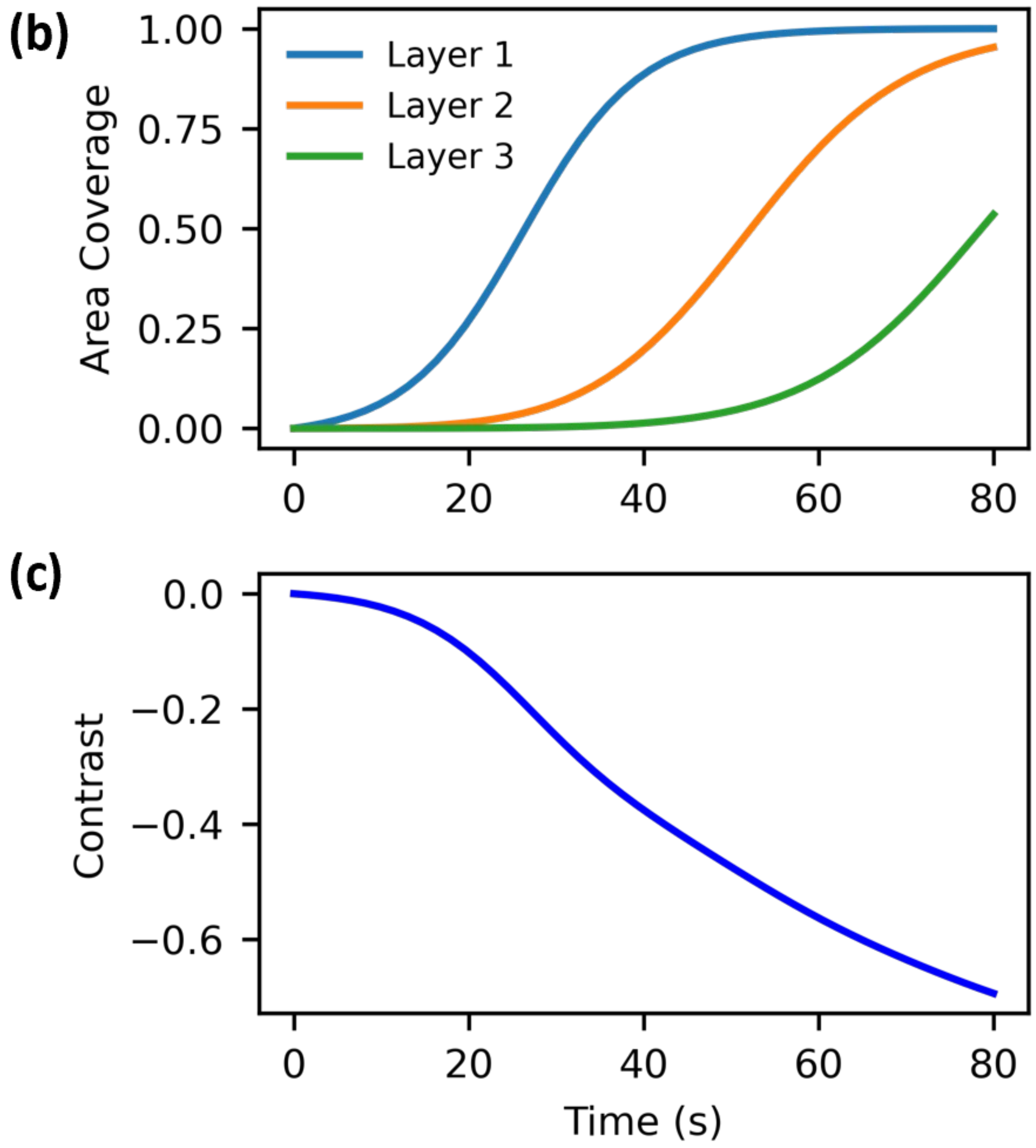
Contrast (observations)

$$c_r(t) = \sum_{i=1}^{N} (c_i - c_{i-1}) f_i(t)$$



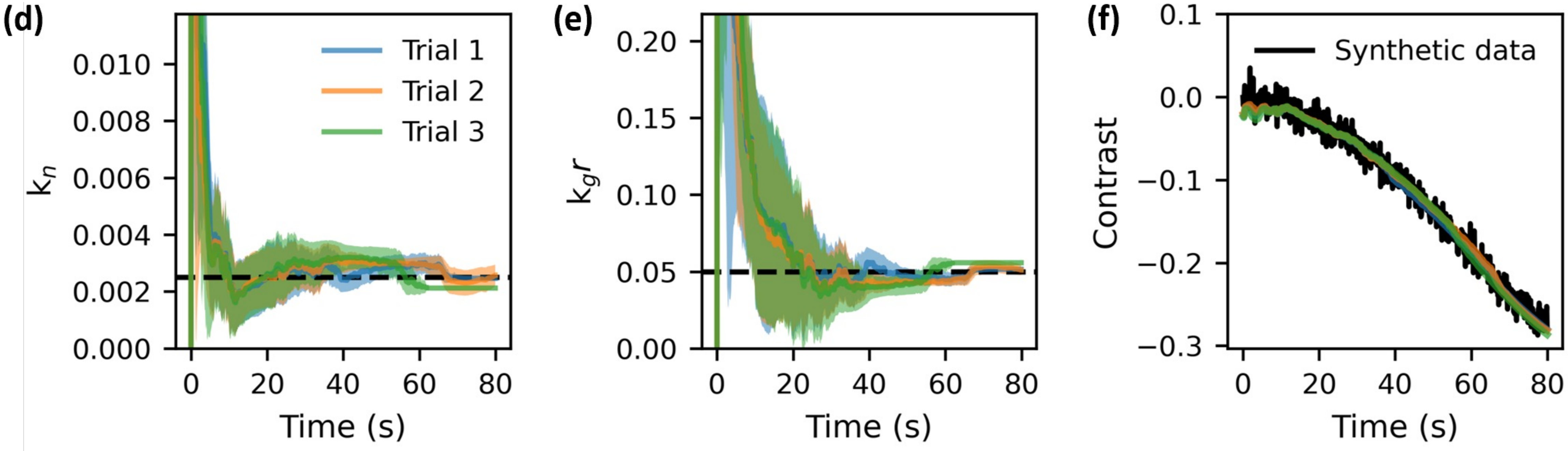
Simple two-step kinetic model for growth of thin film



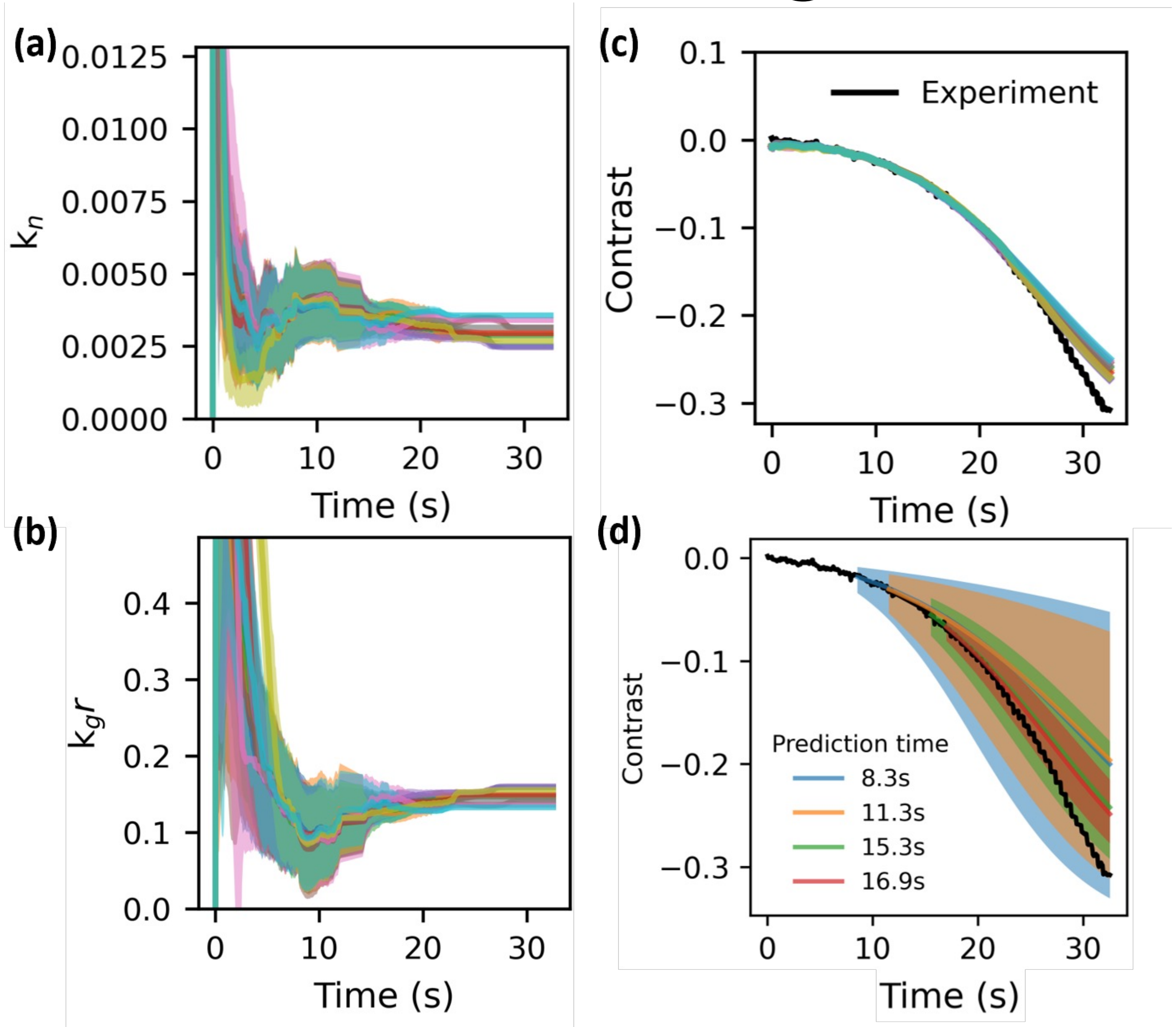


S. Harris et al. (in preparation)

Simulated Data

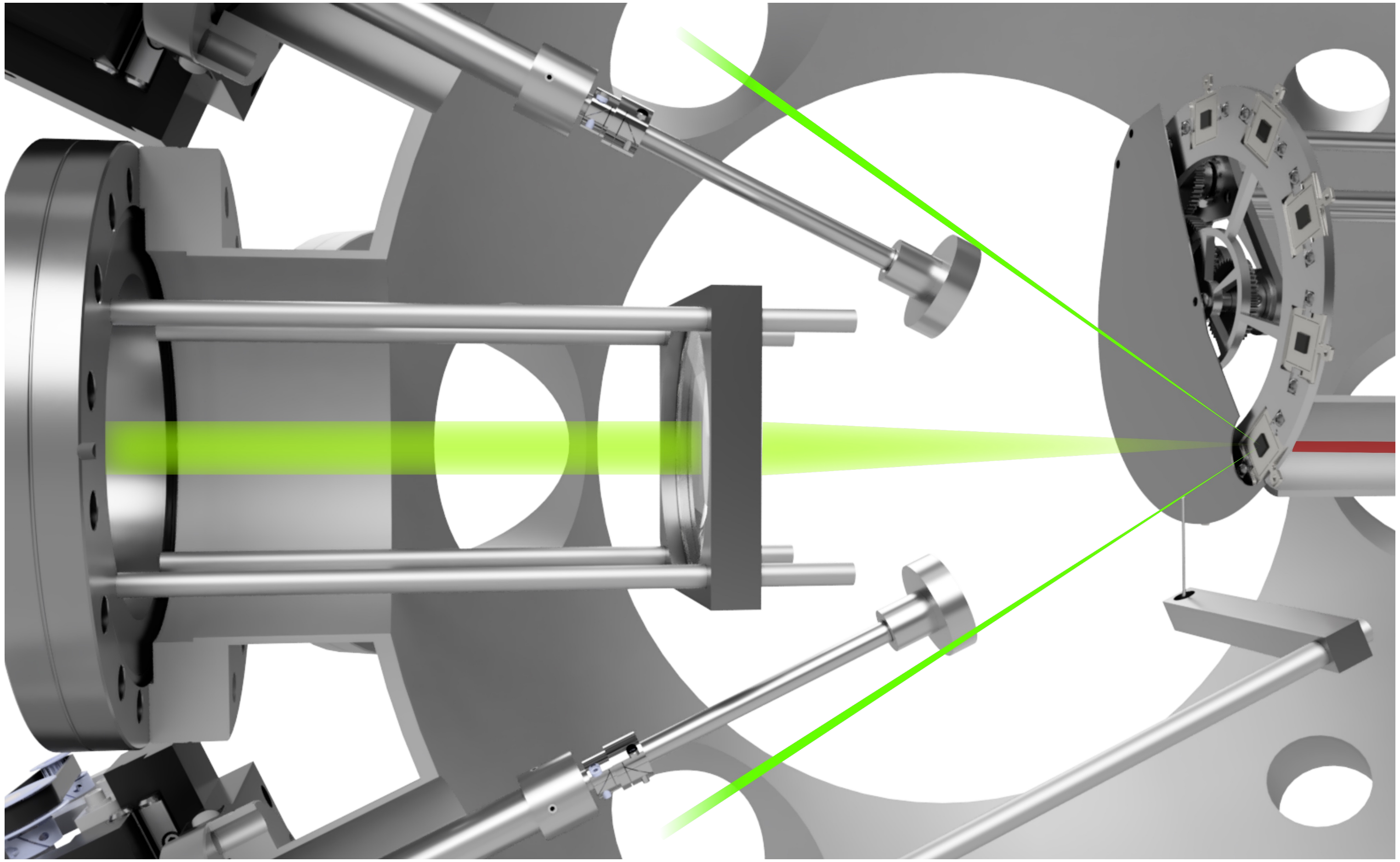


Real Data, Offline Testing



Next-step: use this for real-time control in an online setting

Take Home Message: Simulations + Experiment + ML



- We need to generate algorithms that combine simulations, experiment and ML for producing models with predictive power and high generalizability. This will assist us in physics discovery.

- Model-based control is a promising avenue to realize synthesis of predicted stable and metastable compounds

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vasudevanrk@ornl.gov

Questions