National Synchrotron Light Source II

Brookhaven National Laboratory

U.S. DEPARTMENT OF ENERGY

Autonomous X-ray Scattering Experiments at NSLS-II

Masa Fukuto, BNL/NSLS-II

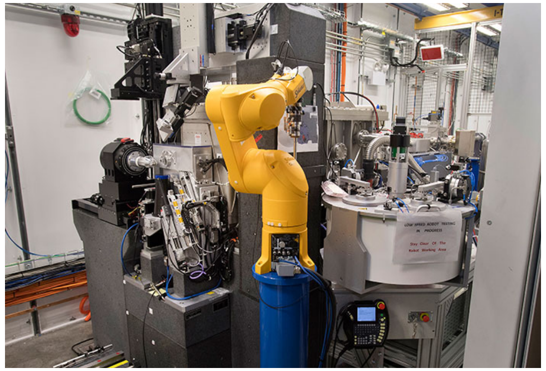
ORNL Second Target Station/Computer Science & Math Workshop, Experiment Automation Breakout, June 23, 2022

Ability to repeat pre-defined tasks precisely & reliably

Ability to learn, decide, and adapt

"Automated"

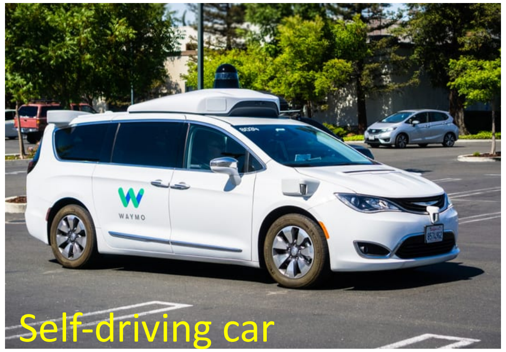
Suited to environment with well-defined tasks



Automated experiments at beamlines, via sample robot, *Bluesky*, analysis pipelines, etc.

"Autonomous"

Suited to dynamic or uncertain environment

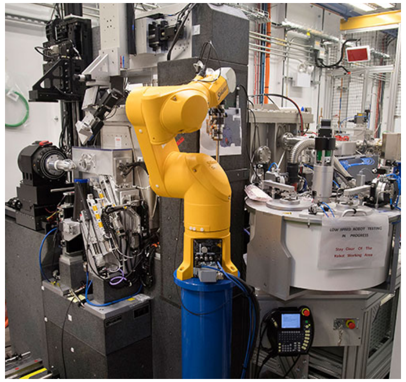


Autonomous experiments at beamlines?

Autonomous experiments: Why?

Bright x-ray source

Fast detectors



Automation

- Data rates at light sources are increasing fast!
  → Humans making experimental decisions is becoming the bottleneck.
  → Machines can analyze data and make decisions about what to do next while the experiment is running.

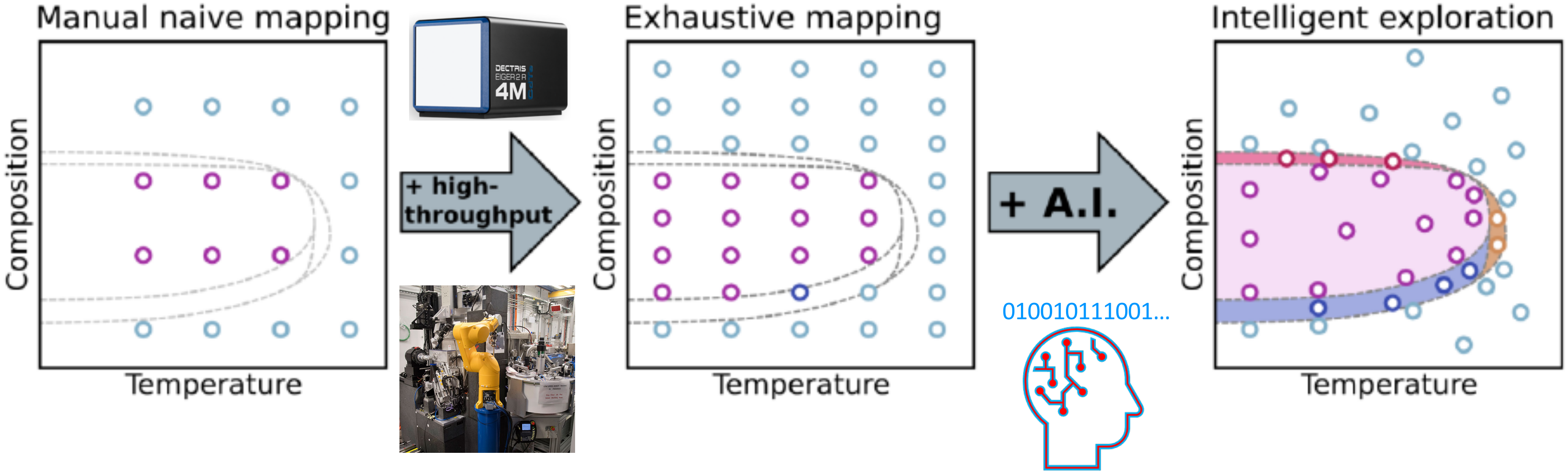
- Machines can efficiently search for the most interesting measurements to make, and make discoveries that humans would have missed or take longer to get to.

From Characterization Instrument to Autonomous Material Discovery Facility

Modern materials are complex

Goal: Beamlines that **autonomously and *intelligently*** explore materials

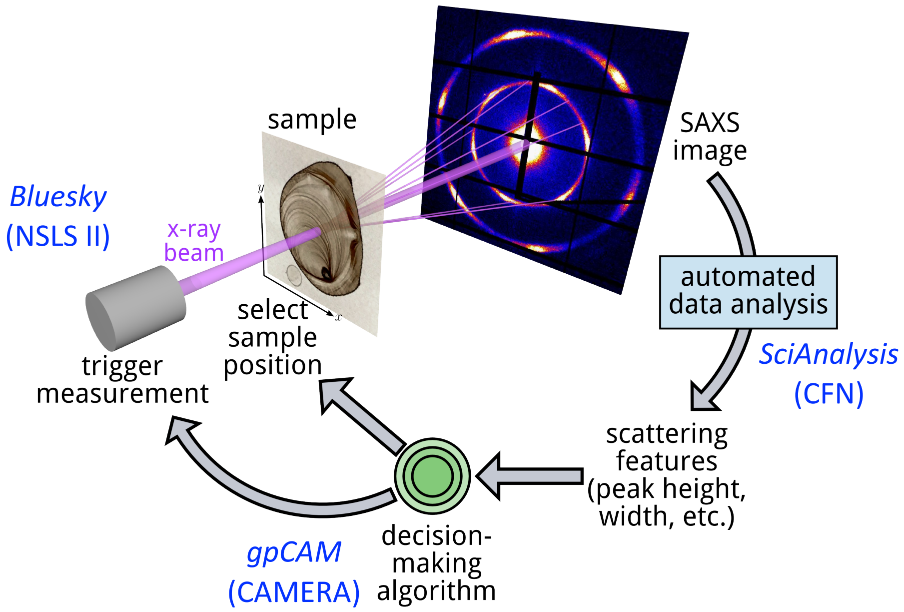
"There is nothing so useless as doing efficiently that which should not be done at all." – Peter Drucker

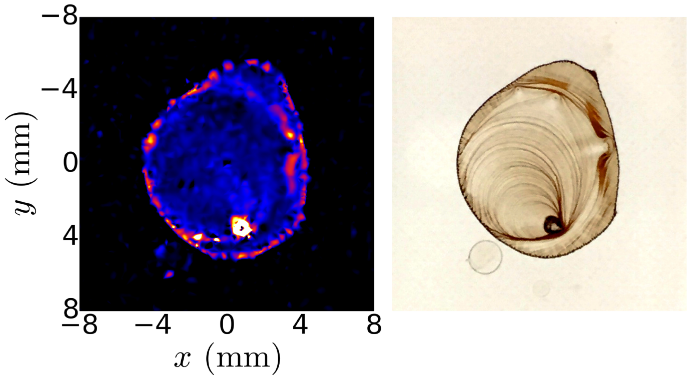


Use artificial intelligence (AI) and machine learning (ML) to run the experiments and make decisions, and leave the experimenter to focus on the big picture!

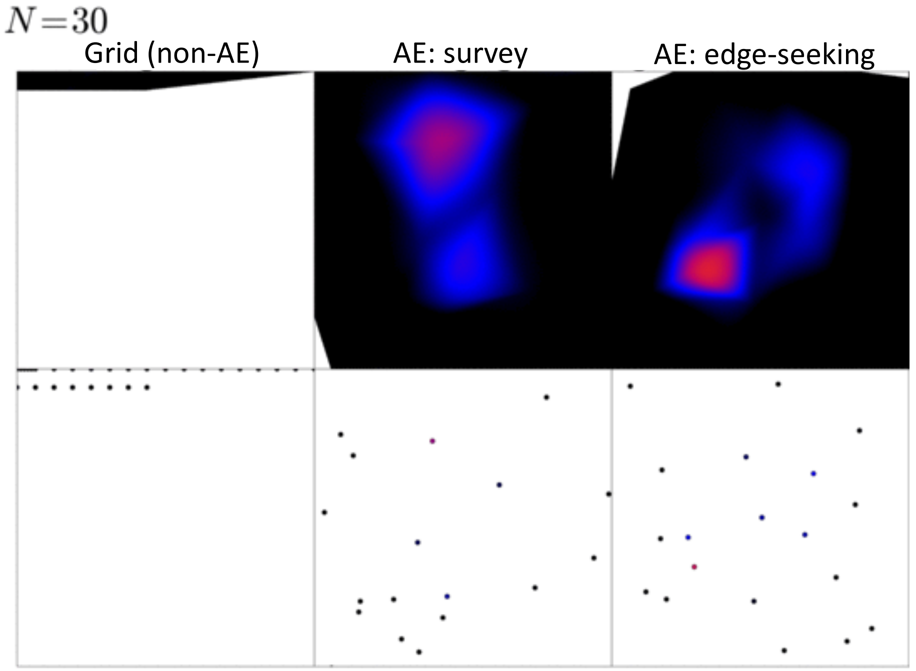
Our First Autonomous X-ray Scattering Experiment

M. Noack et al., *Sci. Rep.* **9**, 11809 (2019); *Sci. Rep.* **10**, 1325 (2020)





Coffee stain-like pattern formed by a drop-cast film of nanoparticle superlattices



(i) Grid scans (non-AE): Pre-programed exhaustive mapping

(ii) Survey (AE): Peak intensity $I(x,y)$ modeled, and locate (x,y) with highest uncertainty for next measurement

→ Suited to quick survey of overall landscape

(iii) Edge-seeking (AE): Uncertainties are weighted by local model gradients

→ Suited to boundary detection and characterization

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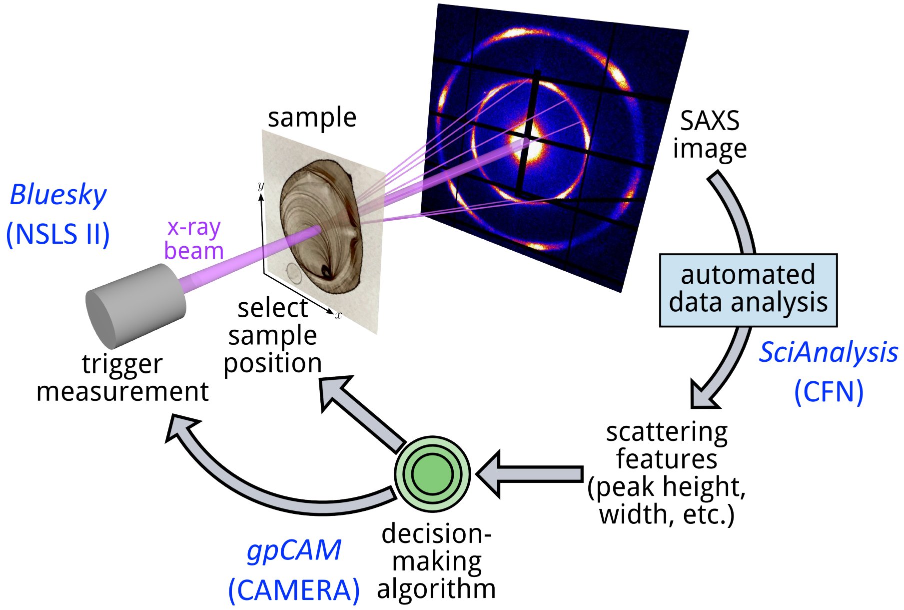

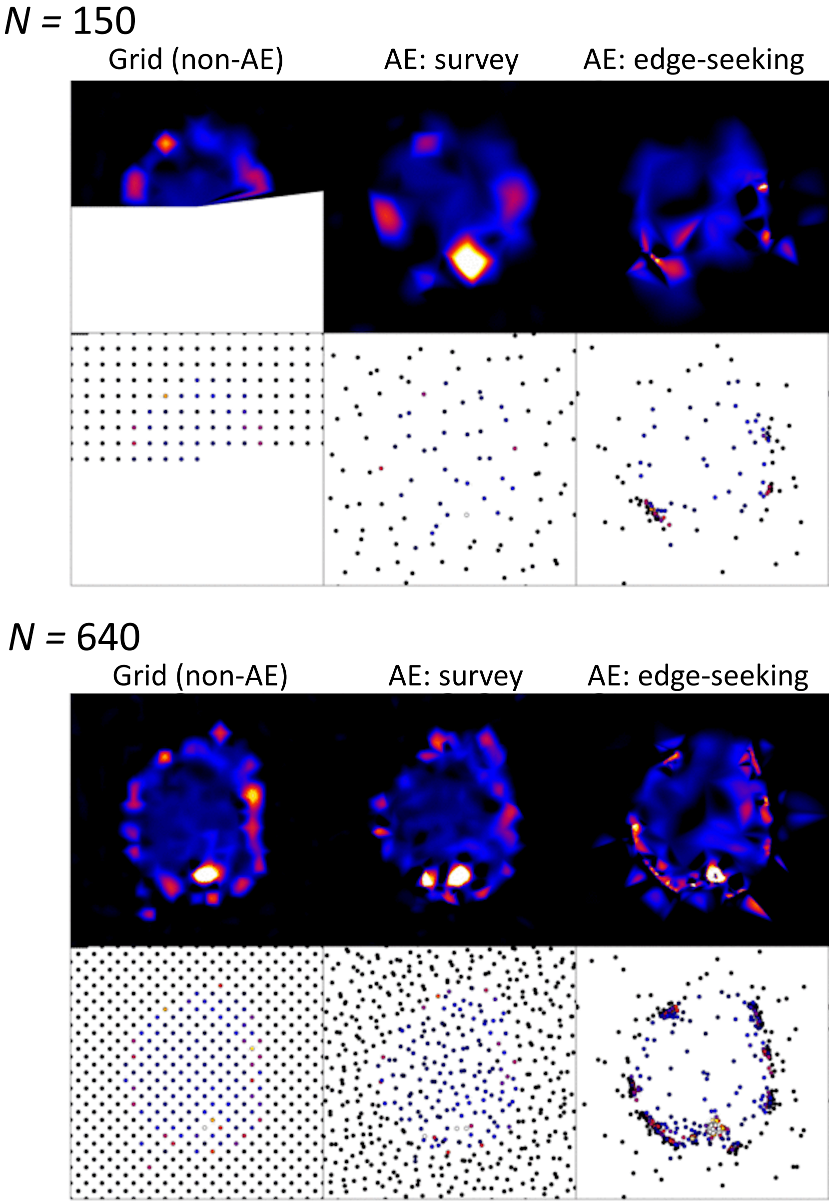

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Autonomous Decision Algorithms (*gpCAM*)

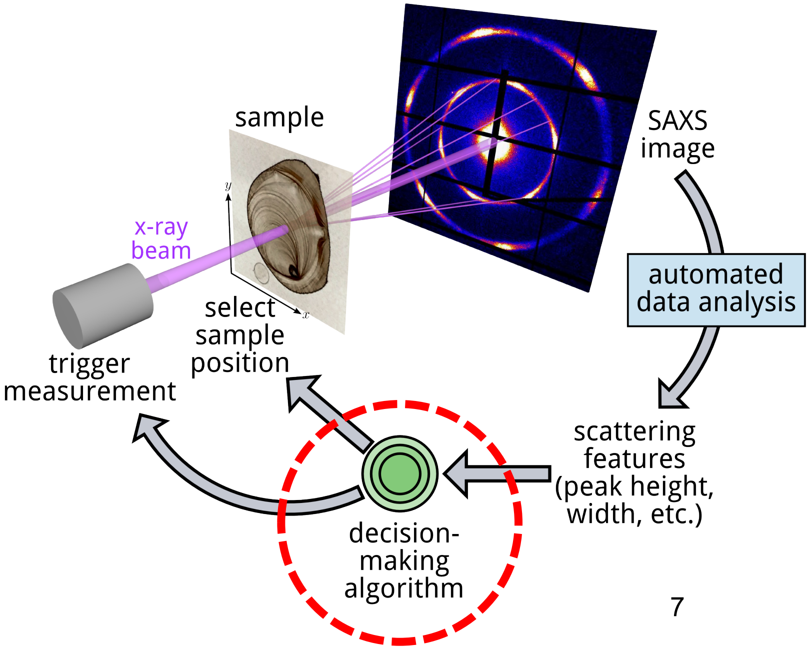
M. Noack et al., *Sci. Rep.* **9**, 11809 (2019); *Sci. Rep.* **10**, 1325 (2020); Sci. Rep. **10**, 17663 (2020)

https://gpcam.lbl.gov/
https://github.com/lbl-camera/gpCAM

- Use **Gaussian Processes** in Bayesian Optimization framework
  - GP to estimate a surrogate model (SM) and uncertainty distribution (UD), based on existing data.
  - Maximize acquisition function based on UD and/or SM to pick the location for next high-value measurements.
- **Decoupled from data analysis** ("dimensional reduction")

  → **independent of experimental techniques**
- In simplest form, **domain knowledge-agnostic → broad applicability**
  - Inputs: Real-time analysis-derived quantities or "signals" (e.g., scattering intensity) as a function of experimentally controlled parameters (e.g., sample position)
  - Outputs: Experimental parameter values for next measurements
- **Multiple input signals** can be used to steer an experiment (one model per signal)

  e.g., peak intensity, grain size, degree and orientation of anisotropy, etc.



Autonomous Decision Algorithms (*gpCAM*)

M. Noack et al., *Sci. Rep.* **9**, 11809 (2019); *Sci. Rep.* **10**, 1325 (2020); Sci. Rep. **10**, 17663 (2020)

- Different steering modes:

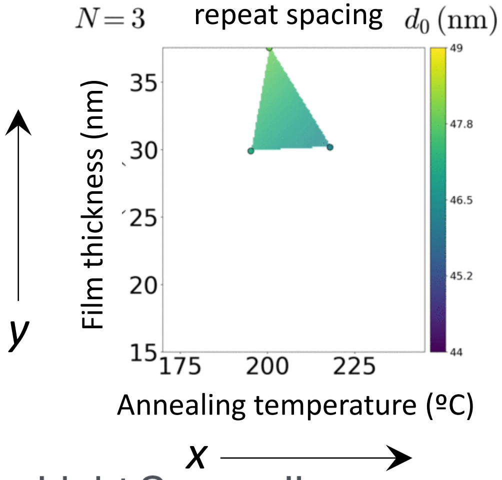
  - **Quick survey**: Acquisition Function (AF) = uncertainty distribution (UD)
  - **Feature optimization**: AF = UD weighted by model values
  - **Boundary detection**: AF = UD weighted by model gradients
  - **Cost optimization**: AF = UD weighted by experimental cost

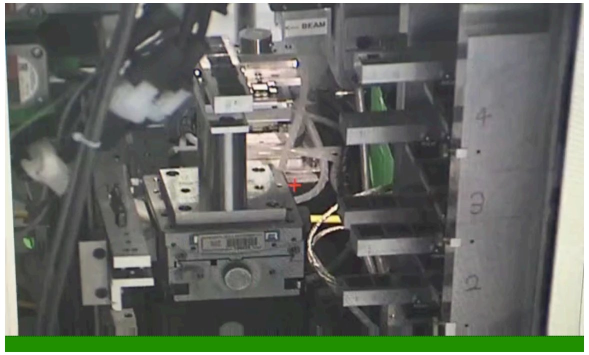
  Sensitive to local data variation (applies to Feature optimization and Boundary detection)

  e.g., time

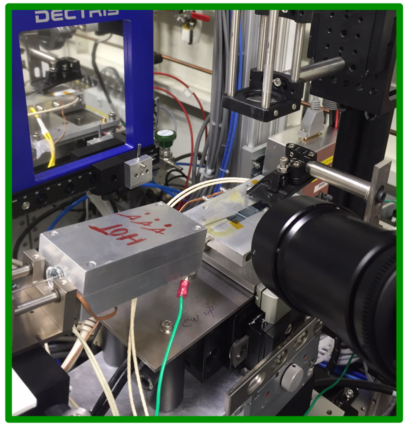
  small < large sample translation < robotic sample exchange < making new samples

Block copolymer combi. film





In-vacuum sample exchanger

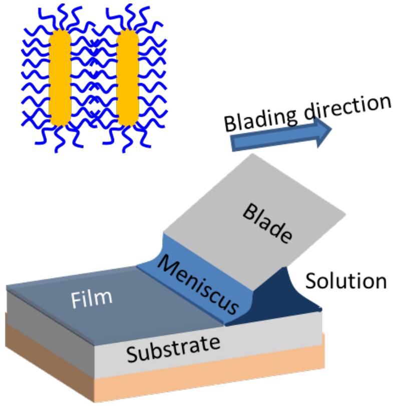


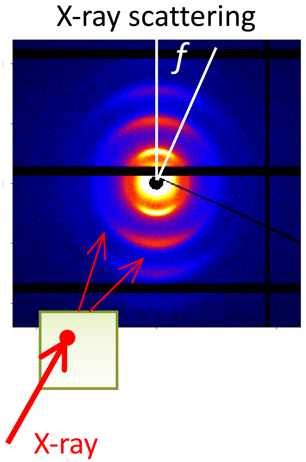
Blade coater for thin film fabrication and annealing

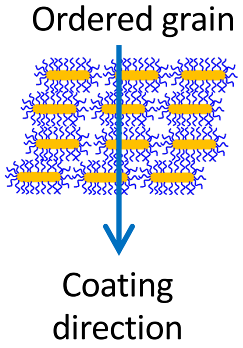
Autonomous Exploration of Material Parameter Space

SAXS exploration of nanoscale ordering in blade-coated polymer-grafted nanorod film User collaboration: J. Streit, R. Vaia (AFRL)

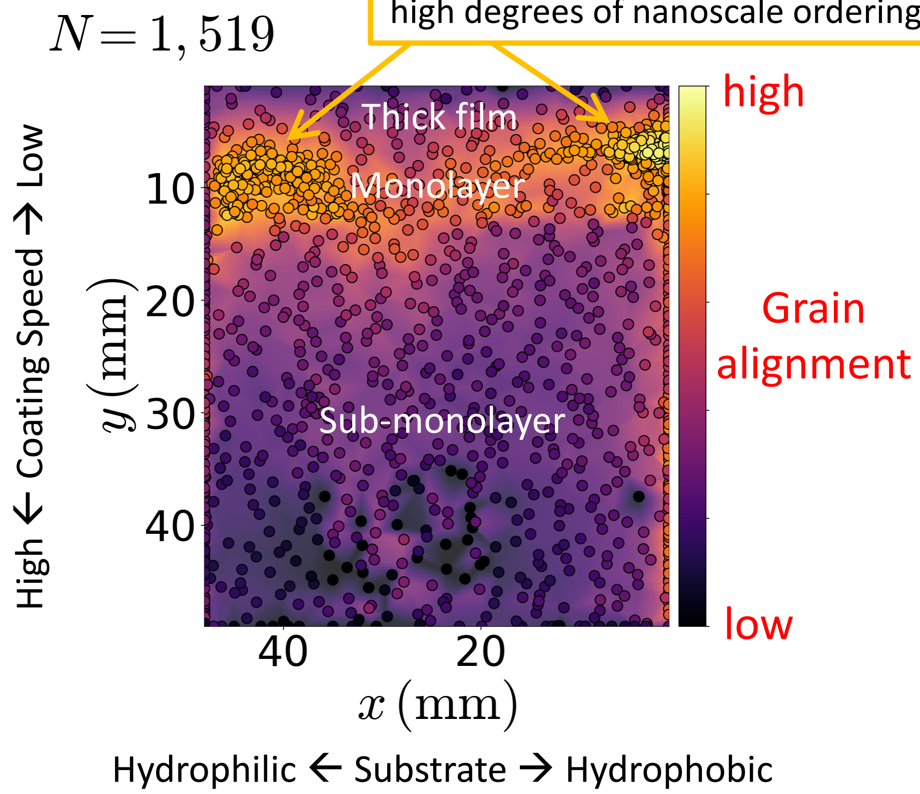
Combinatorial blade-coated film of polymer-grafted nanorods







Optimal coating speed identified for high degrees of nanoscale ordering



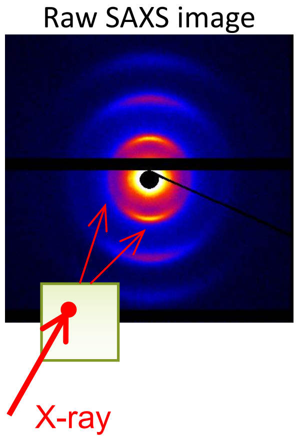
- N < 500 (4 hrs): Survey
- N > 500 (11 hrs): Feature optimization

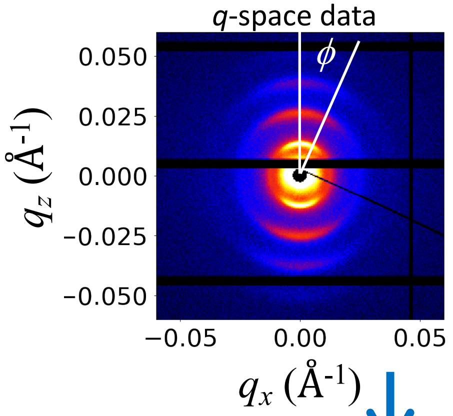
Gridding out at max resolution would have taken 11 days!

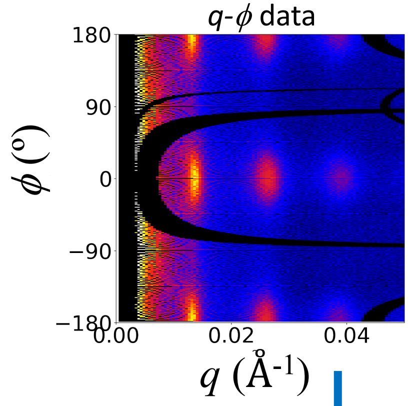
M. Noack et al., *Sci. Rep.* **10**, 17663 (2020); user collaboration: J. Streit, R. Vaia (AFRL)

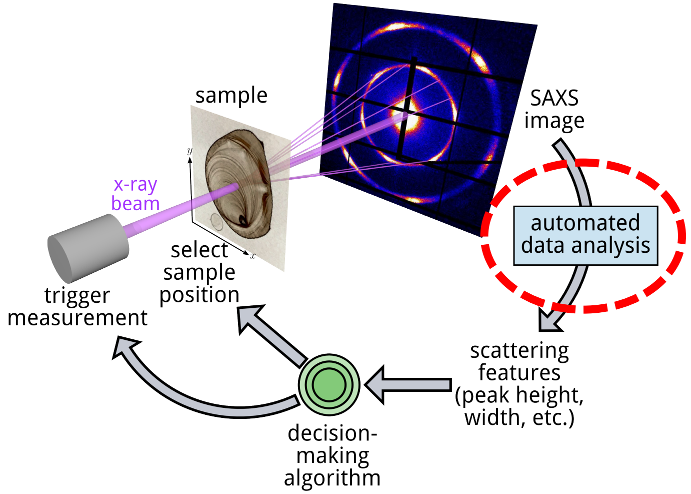
Real-time data processing and analysis

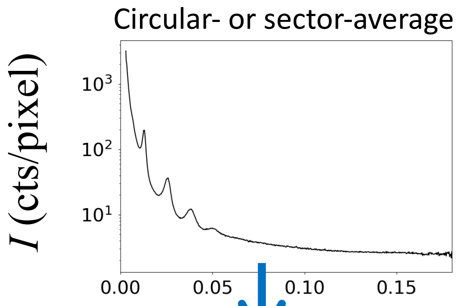
via ***SciAnalysis***: https://github.com/CFN-softbio/SciAnalysis

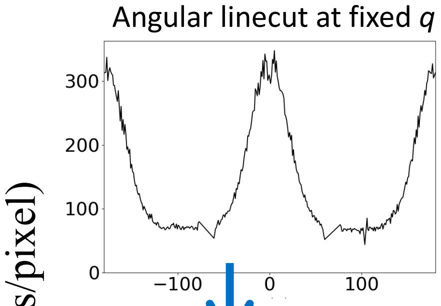


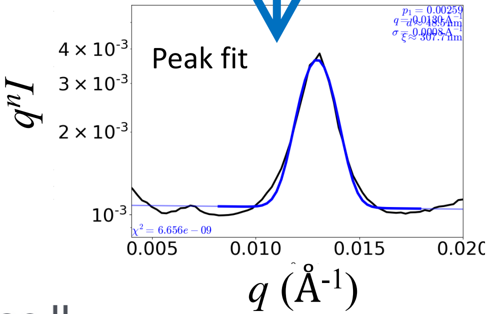


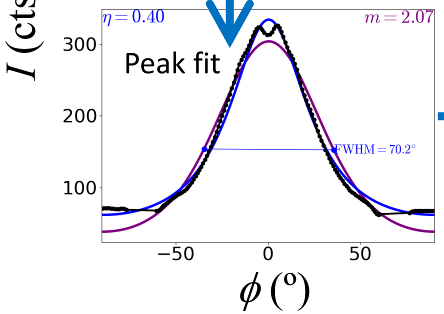


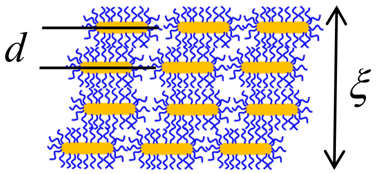




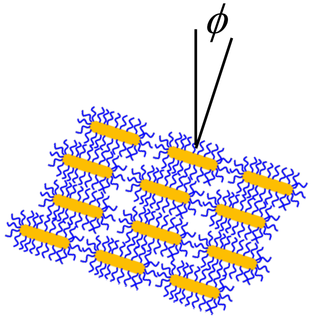








- d-spacing of periodic structure $d = 2\pi/q_{peak}$
- Grain/domain size ξ (from peak width)



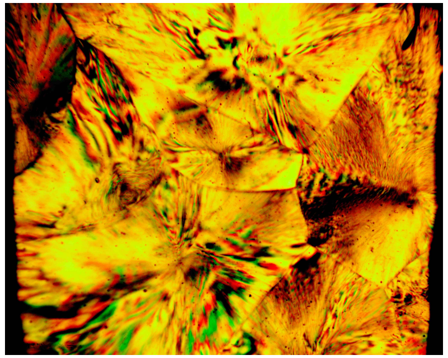
- Orientation of anisotropy ϕ_{peak}
- Degree of anisotropy η ($0 < \eta < 1$, from peak sharpness)

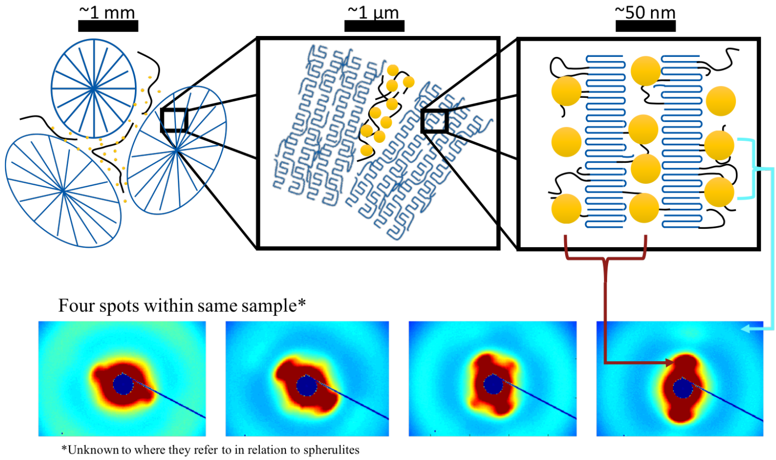
Autonomous Exploration of Real-Space Material Heterogeneity

Microbeam SAXS mapping of polymer/nanoparticle composite films

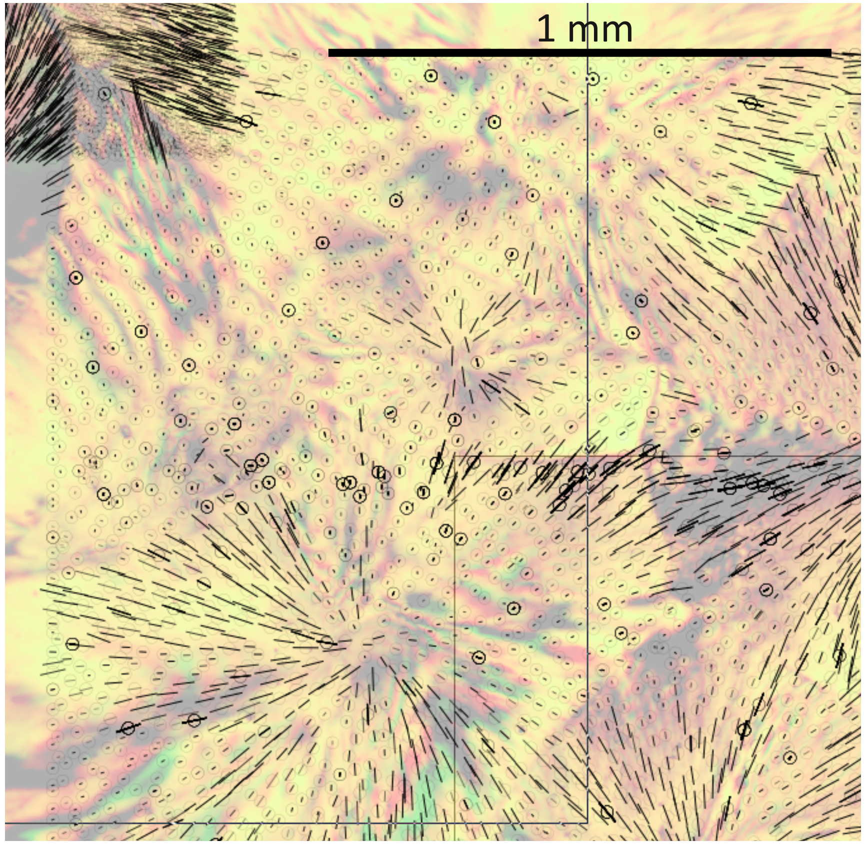
User collaboration: A. Jimenez, A. Krauskopf, S. Kumar (Columbia U.)

POM image of PEO spherulites





14 nm dia. silica NP in PEO



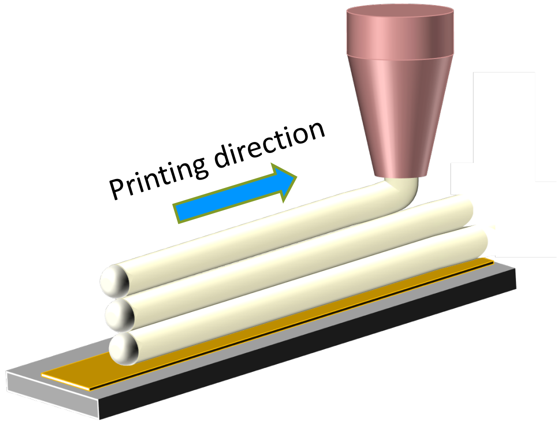
Bar length: degree of anisotropy in NP ordering
Bar direction: parallel to NP layers

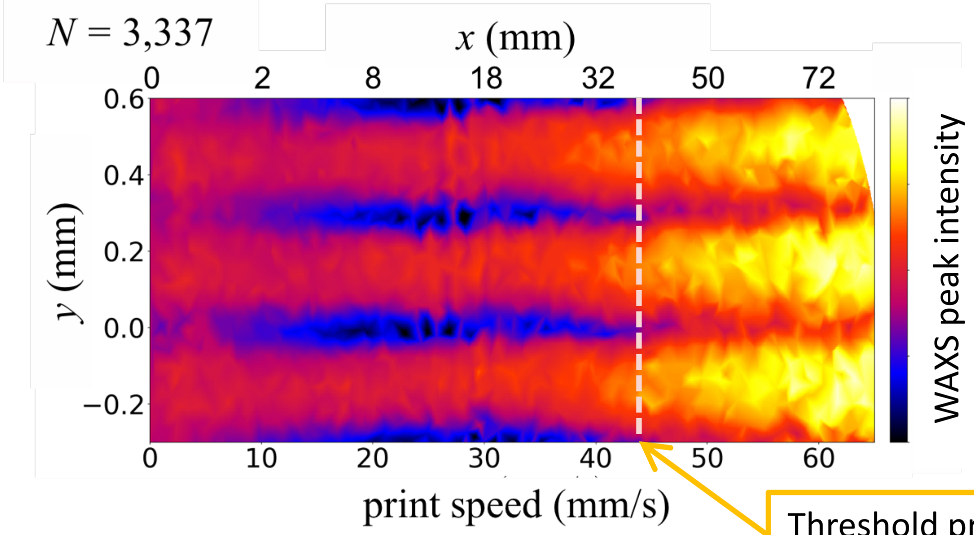
- Polymer crystallization influences NP ordering.
- Direct evidence that NP layers run radially from the nucleation center of each spherulites.

Toward "Domain-Knowledge-Aware" Autonomous Decision Making

Microbeam WAXS mapping of 3D-printed semicrystalline polymer (PLA) filaments

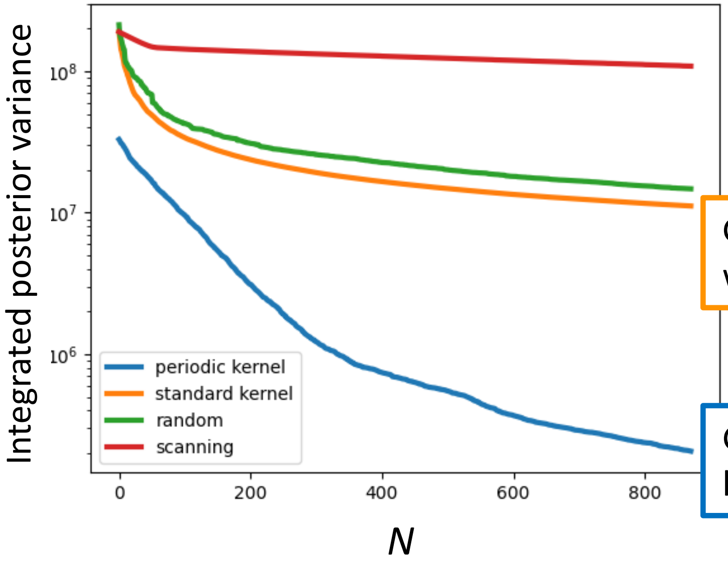
User collaboration: J. Seppala, T. Martin (NIST)





Threshold print speed for high crystallinity

- Hybrid mapping
  - *y*: heterogeneity along stacking direction
  - *x*: gradient in processing parameter (print speed)
- Evidence for shear-induced polymer alignment promoting crystal growth
- **Incorporating knowledge about existence of periodicity (along *y*) yields a better surrogate model faster**

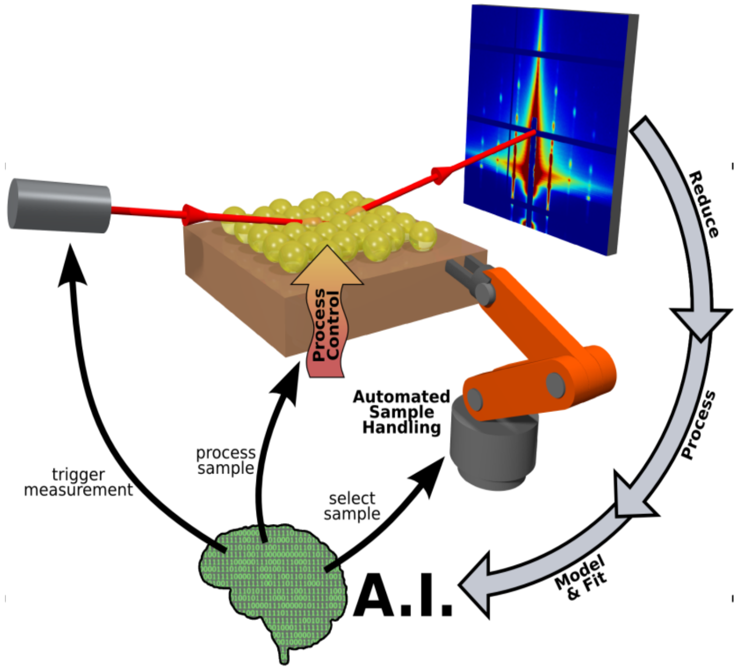


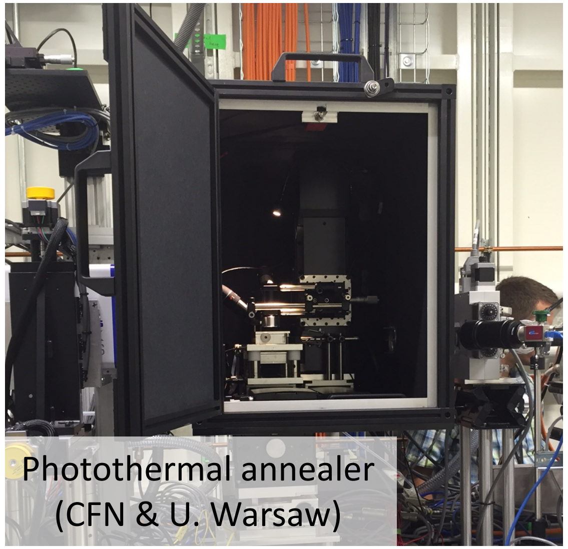
Gaussian Process (GP) with standard kernel

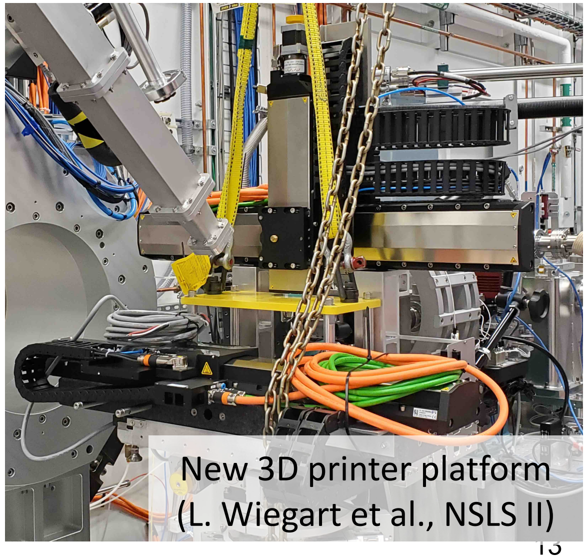
GP with periodic kernel along *y*

Ongoing/Future Work

- Roll out to other beamlines and techniques
- Apply to ***in-situ* material processing** by integrating *in-situ* sample environments and ancillary/proxy non-x-ray probes into autonomous loop
- **ML-accelerated data analysis** (e.g., instant feature recognition and classification of x-ray data to guide downstream analysis)
- **Enhance decision algorithms** to leverage materials knowledge, theory, simulations to guide experiments




Photothermal annealer
(CFN & U. Warsaw)


New 3D printer platform
(L. Wiegart et al., NSLS II)

Additional thoughts

Where autonomous experiments (AE) could be useful?
(Materials science perspective)

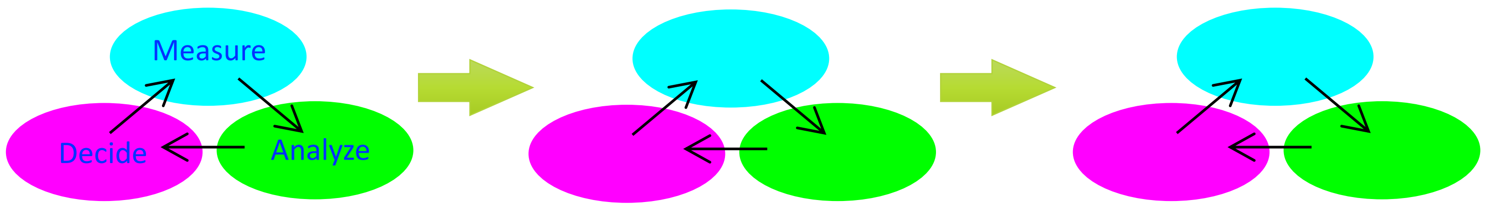
For materials with a lot of existing knowledge or relatively restricted sample space to explore, *automated* experiments may be the better choice than AE.

But AE can be very effective for studying:

- New/novel materials for which a lot is unknown.
- Structure/property of materials involving large and complex material space
  (e.g., many components, many processing variables).
- Materials under dynamic environment where one wants to adaptively explore, understand, and control material processes toward unexpected/optimal structure/functions.
  (“*in-situ/operando*,” “out-of-equilibrium,” “kinetic pathway,” “metastable/transient,” etc.)

Hierarchy of Autonomous Decision Making

e.g., Navigate between different steering modes to suit experimental needs



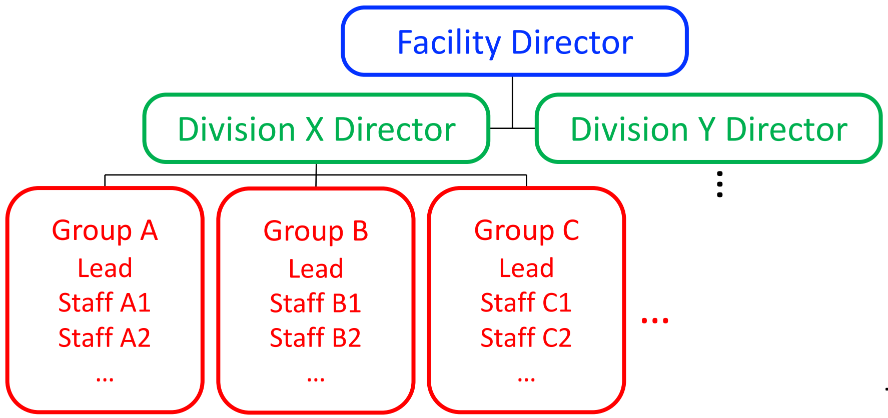
Stage 1: Quick survey to grasp overall landscape

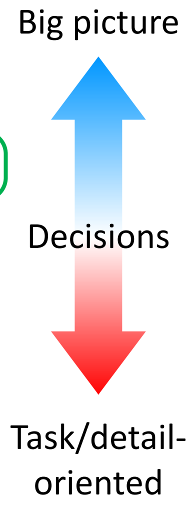
Stage 2: Detect and characterize boundaries (e.g., phase boundaries)

Stage 3: Pinpoint target feature (e.g., maximum grain size)

- How do we leverage human-machine interactions in hierarchical decision-making?
- How should hierarchical decision-making by machines be organized and managed?

e.g., human organization





- Division of labor
- Autonomy and good decisions are important at every level
- Micromanaging hampers momentum: "Hire the best people, get out of the way, let them do what they are good at"

→ Applicable to organizing machine decisions, esp. with advances in HPC?

Role of beamline scientists, users, …

Autonomous Experiments (AE) are meant to **not replace, but liberate and empower** beamline scientists and users.

Automation allowed us to expand the scope of beamline experiments.

Similarly, *AE* will allow beamline scientists and users to:

- Get involved more deeply into scientific aspects of research
- Design more ambitious experiments and manage them at a higher level
- Develop new capabilities toward enhancing our facilities (e.g., addressing data analytics challenges) to attract/pursue impactful research projects/programs

An exhausted user at NSLS (ca. 1999)

AE beam time at NSLS II (Nov. 2019)

In the end, it's the people who make things happen!

"The only thing we know about the future is that it will be different… The best way to predict the future is to create it."
– Peter Drucker

"The path to success … is to form high-performing teams and give them the resources and freedom to do great things."
– Bill Campbell

- **BNL/NSLS-II**
  Ruipeng Li
  Guillaume Freychet
  Misha Zhernenkov
  Lutz Wiegart
  Tom Caswell
  Josh Lynch
  Sanjit Ghose
  Dan Olds
  Phillip Maffettone

- **BNL/CFN**
  Kevin Yager
  Greg Doerk
  Aaron Stein
  Esther Tsai
  Sebastian Russell
  Suwon Bae
  Oleg Gang

- **LBNL/CAMERA**
  Marcus Noack
  Jamie Sethian

- **AFRL**
  Richard Vaia
  Jason Streit

- **U. Warsaw**
  Pawel Majewski
  Arkadiusz Leniart

- **Columbia U.**
  Sanat Kumar
  Andrew Jimenez
  Arejandro Krauskopf
  Sophia Chan

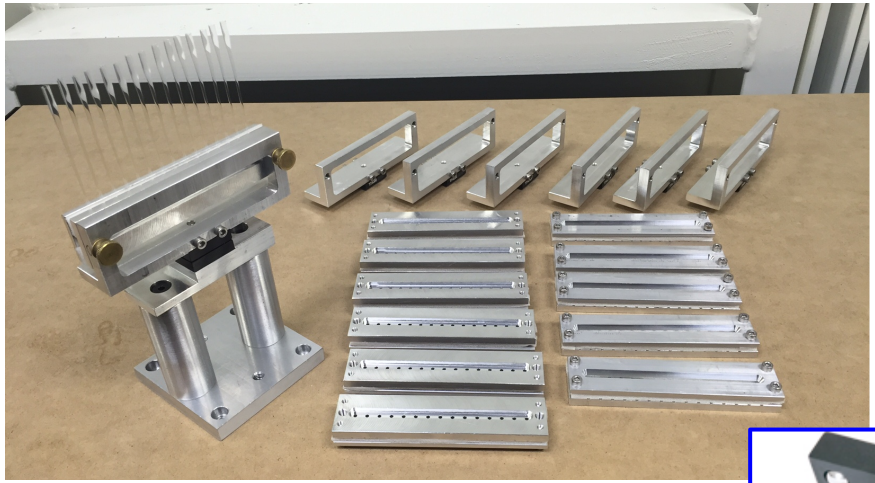
- **U. Penn**
  Christopher Murray
  Katherine Elbert

- **NIST**
  Jon Seppala
  Tyler Martin
  Lee Richter

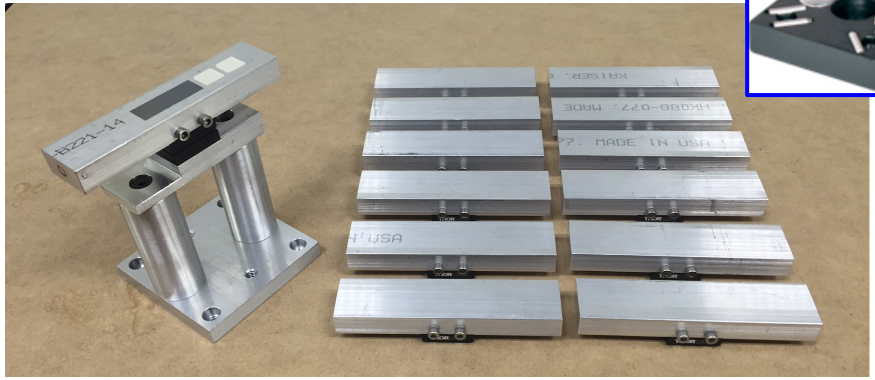
- **SBU**
  Karen Chen-Wiegart
  Chonghang Zhao

Thank you

High Throughput via Robotic Sample Exchange



Sample holders for transmission SAXS/WAXS (15 positions per holder)



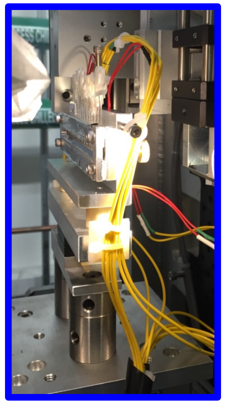
Thin-film substrate holders for GISAXS/GIWAXS (~10 substrates per holder)

Sample exchanger (left) and garage (right) inside sample chamber

Examples of Available In-Situ Setups

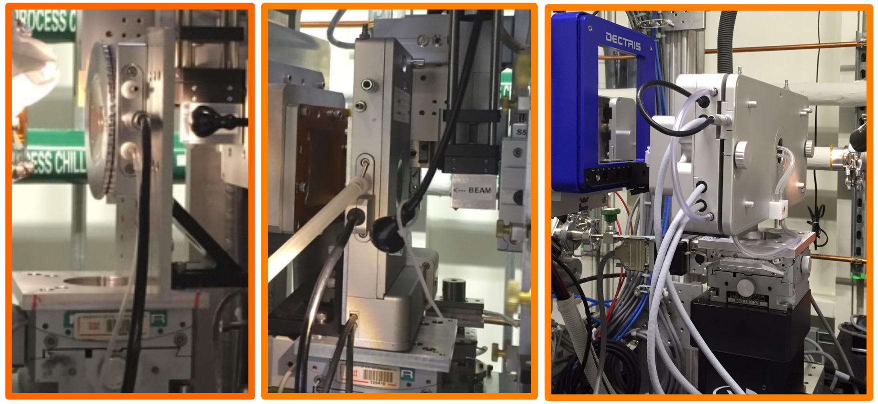
In-house developed/commercial

T-annealing
(GI-SAXS/WAXS)

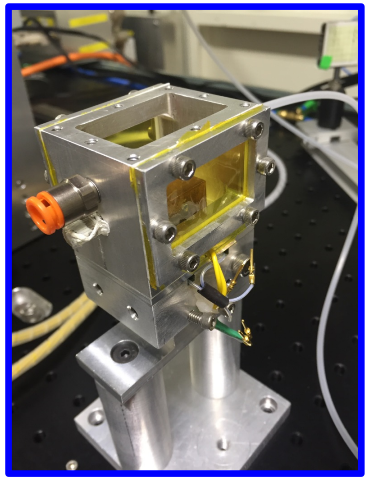


T-annealing
(transmission)

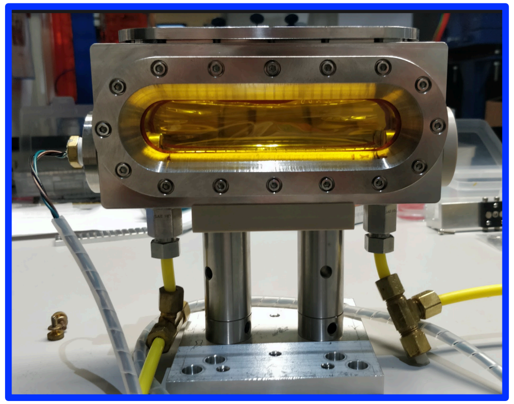
Instec T stage



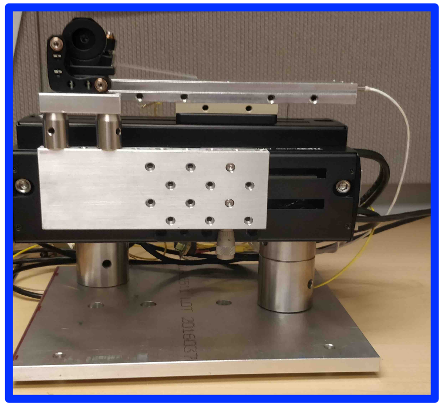
Linkam thermal / tensile / shear stages



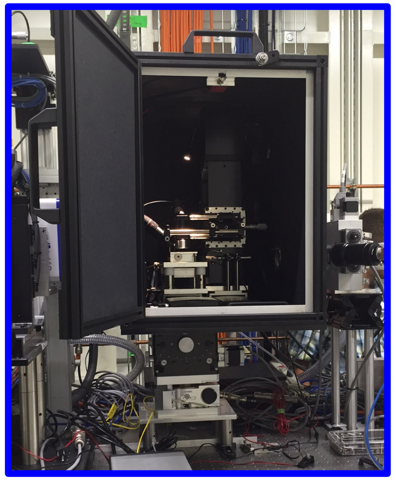
Solvent-vapor
annealing cell



Humidity chamber



Blade coater for solution-
phase thin film fabrication
and T-annealing



CFN IR-laser-based
photothermal annealer