Autonomous X-ray Scattering

Kevin G. Yager (kyager@bnl.gov)
Acknowledgments

- Autonomous implementation
  - **Masa Fukuto** (BNL, NSLS-II)
  - **Marcus Noack** (CAMERA)
  - Jamie Sethian (CAMERA)
  - Ruipeng Li (BNL, NSLS-II)
  - **Esther Tsai** (BNL, CFN)

- Beamlines (BNL, NSLS-II)
  - Mikhail Zhernenkov
  - Guillaume Freychet
  - Lutz Wiegart
  - Sanjit Ghose
  - Dan Olds
  - Phillip Maffettone
  - Joshua Lynch
  - Tom Caswell

- Directed assembly (BNL, CFN)
  - **Greg Doerk**
  - **Aaron Stein**
  - **Sebastian Russell**
  - Suwon Bae

- Electrospray (Yale, U.Penn)
  - Chinedum Osuji
  - Kristof Toth

- Photo-thermal annealing (U. Warsaw)
  - Pawel Majewski
  - Andrzej Sitkiewicz
  - Arkadiusz Leniart

- Polymer composites (Columbia)
  - Sanat Kumar
  - Andrew Jimenez
  - Alejandro Krauskopf

- Nanoparticle superlattices (U. Penn)
  - Chris Murray
  - Katherine Elbert

- Nanorod assembly (AFRL)
  - Richard Vaia
  - Jason Streit

- 3D printing (NIST)
  - Jon Seppala
  - Tyler Martin

- Metal dealloying (SBU)
  - Karen Chen-Wiegart
  - Chonghang Zhao

- Directed assembly (BNL, CFN)
  - **Greg Doerk**
  - **Aaron Stein**
  - **Sebastian Russell**
  - Ruipeng Li (BNL, NSLS-II)

- Beamlines (BNL, NSLS-II)
  - Mikhail Zhernenkov
  - Guillaume Freychet
  - Lutz Wiegart
  - Sanjit Ghose
  - Dan Olds
  - Phillip Maffettone
  - Joshua Lynch
  - Tom Caswell

- Directed assembly (BNL, CFN)
  - **Greg Doerk**
  - **Aaron Stein**
  - **Sebastian Russell**
  - Suwon Bae

- Electrospray (Yale, U.Penn)
  - Chinedum Osuji
  - Kristof Toth

- Photo-thermal annealing (U. Warsaw)
  - Pawel Majewski
  - Andrzej Sitkiewicz
  - Arkadiusz Leniart

- Polymer composites (Columbia)
  - Sanat Kumar
  - Andrew Jimenez
  - Alejandro Krauskopf

- Nanoparticle superlattices (U. Penn)
  - Chris Murray
  - Katherine Elbert

- Nanorod assembly (AFRL)
  - Richard Vaia
  - Jason Streit

- 3D printing (NIST)
  - Jon Seppala
  - Tyler Martin

- Metal dealloying (SBU)
  - Karen Chen-Wiegart
  - Chonghang Zhao
Automate the entire experiment, including decision-making

Liberate human scientist to focus on scientific insight

Accelerate materials discovery
X-ray Scattering

- The goal in x-ray scattering is to determine the structure of a material
Autonomous X-ray Scattering

- Automate entire experiment
  - Instrument
  - Data analysis
  - Decision-making
- Improves efficiency, accelerates discovery
- **Liberates** humans to focus on science

Yager et al. *Nanotechnology* **2023**, 34, 322001
Separate the data analysis from the decision-making
- Dimensional reduction
- Abstraction

Three processes (easy stop/restart)
- **measure** (*Bluesky*)
- **analyze** (*SciAnalysis*)
- **decide** (*gpCAM*)

Communication is “thin”
- \( m \rightarrow a \): Pointer to data
- \( a \rightarrow d \): Result vector
- \( d \rightarrow m \): Parameter-space position
Algorithm: Gaussian Process

- **Construct surrogate model**
  - Fit/interpolate data
  - Select kernel to match physics
  - Estimate hyper-parameters that match data (lengthscales, periodicity, etc.)

- Calculate **uncertainty surface**

- **Construct objective function**
  - Search for maximum in objective
  - Control behavior: gradients, cost, etc.

- As we **iterate**, errors decrease and surrogate model improves

---

Yager et al. *Nanotechnology* **2023**, *34*, 322001
Self-Assembling Materials

- Blends of self-assembling materials become a ‘responsive’ resist
  - Template (pitch, duty cycle) selects morphology

Stein, Wright, Yager, Doerk, Black
*Nature Communications* **2016**, 7, 12366
Autonomy discovered numerous novel morphologies
Autonomy discovered numerous novel morphologies

- skew
- alternating
- ladder

Doerk, Stein, Bae, Noack, Fukuto, Yager
*Science Advances* 2023, 9, eadd3687
parameters: total \[ \geq 12 \] human \[ \rightarrow \] 5
signals: \[ 1,023,183 \] analytics \[ \rightarrow \] 4
points: \[ \infty \] gpCAM \[ \rightarrow \] 26,299

ordered material \( p \) (a.u.)
repeat spacing \( d_0 \) (nm)
grain size \( \xi \) (nm)
novelty \( \chi^2 \) (a.u.)
Layering self-assembling materials can generate new (non-equilibrium) structures.
AI assistants
Domain-specific Chatbot

Large Language Models

- LLMs (c.f. ChatGPT) enable remarkably coherent dialog on any topic
- They sometimes “hallucinate”
- This can be solved by giving the bot domain-specific text
Demo

This is a demo of a domain-specific ChatBot for science. The chatbot uses a large language model (LLM) for generative text, combined with a database of domain-specific text drawn from CFN publications.

User: I want to measure the orientation of a block copolymer morphology in a thin film (cylinders horizontal vs. vertical). Should I use TSAXS, GISAXS, or GTSAAXS?

CFNBot: To measure the orientation of a block copolymer morphology in a thin film (cylinders horizontal vs. vertical), you can use either GISAXS (Grazing-Incidence Small-Angle X-ray Scattering) or GTSAAXS (Grazing-Incidence Transmission Small-Angle X-ray Scattering). Both techniques provide valuable information about the structure of thin films.

GISAXS allows for the observation of monolayers of nanomaterials and depth profiling of thin films. It can provide information about the unit cell, morphology, repeat spacing, orientation distribution, and grain size in soft materials. However, analyzing the data obtained from GISAXS can be challenging due to refracted effects at the ambient-film interface causing distorted images of reciprocal-space.

On the other hand, GTSAAXS is an alternative to GISAXS that can obtain undistorted data without requiring lengthy reconstruction. It involves aligning the incident x-ray beam on the downstream edge of a sample/substrate, rather than directing the beam towards the center of the sample as typically done in GISAXS. GTSAAXS is simpler and can record an image without refraction-distortions or multiple-scattering complications. However, it requires arranging the sample of interest to be positioned near the edge of the substrate.
Virtual Scientific Companion

- NLP allows scientists to plan experiments in natural language
- LLM helps refine the experiment, generates Python code to execute at the beamline
- Dialogue with models and analysis pipelines

Esther Tsai
Early Career Award

Potemkin, Soto, Li, Yager, Tsai *arXiv* 2024, 2312.17180
Future: AI Assistants

Nanoscience Chatbot
- Find/discuss relevant information
- Brainstorming

Data Navigator
- Organize data

Experimental Assistant
- Plan/control experiments (including AE)

Future: Swarm of AI Agents
- Specialized agents for experiments, data, publications
- Agents talk to each other; surface only important topics to human
- Swarm effectively expands the human’s memory, planning, and thinking
Autonomous experiments can explore arbitrary science problems
- Imaging/mapping
- Combinatorial
- Real-time synthesis/processing

Language models can automate diverse science tasks
Acknowledgments

- Autonomous implementation
  - Masa Fukuto (BNL, NSLS-II)
  - Marcus Noack (CAMERA)
  - Jamie Sethian (CAMERA)
  - Ruipeng Li (BNL, NSLS-II)
  - Esther Tsai (BNL, CFN)

- Beamlines (BNL, NSLS-II)
  - Mikhail Zhernenkov
  - Guillaume Freychet
  - Lutz Wiegart
  - Sanjit Ghose
  - Dan Olds
  - Phillip Maffettone
  - Joshua Lynch
  - Tom Caswell

- Directed assembly (BNL, CFN)
  - Greg Doerk
  - Aaron Stein
  - Sebastian Russell
  - Suwon Bae

- Electrospray (Yale, U.Penn)
  - Chinedum Osuji
  - Kristof Toth

- Photo-thermal annealing (U. Warsaw)
  - Pawel Majewski
  - Andrzej Sitkiewicz
  - Arkadiusz Leniart

- Polymer composites (Columbia)
  - Sanat Kumar
  - Andrew Jimenez
  - Alejandro Krauskopf

- Nanoparticle superlattices (U. Penn)
  - Chris Murray
  - Katherine Elbert

- Nanorod assembly (AFRL)
  - Richard Vaia
  - Jason Streit

- 3D printing (NIST)
  - Jon Seppala
  - Tyler Martin

- Metal dealloying (SBU)
  - Karen Chen-Wiegart
  - Chonghang Zhao