THE UNIVERSITY of TENNESSEE KNOXVILLE

Virtual Summer School:

Machine Learning in Electron Microscopy

Sergei V. Kalinin and Gerd Duscher,
University of Tennessee, Knoxville

Maxim Ziatdinov and Rama Vasudevan,
Oak Ridge National Laboratory

Maxim Ziatdinov

Rama Vasudevan

Gerd Duscher

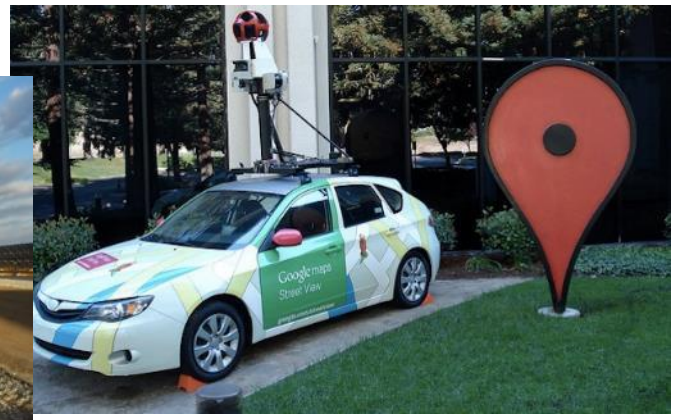
Ayana Ghosh

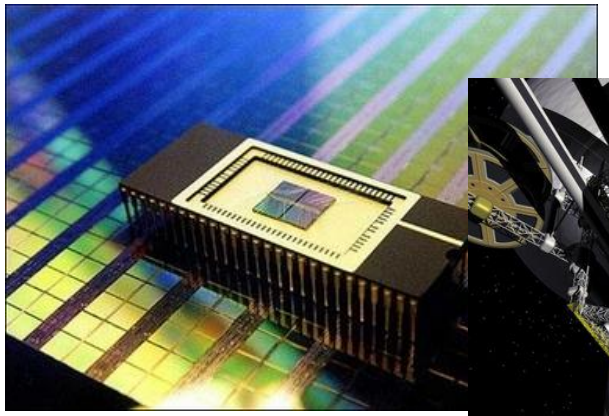
Tommy Wong

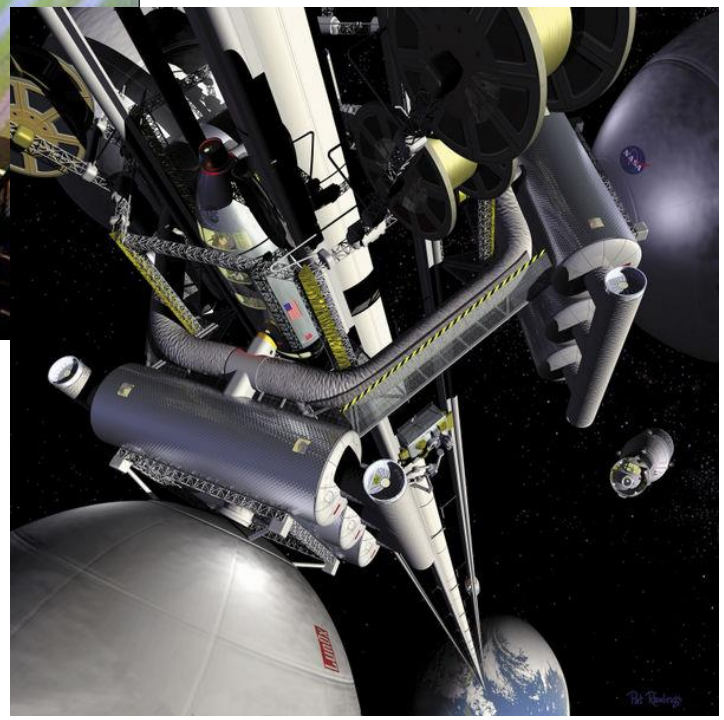
Kevin Roccapriore

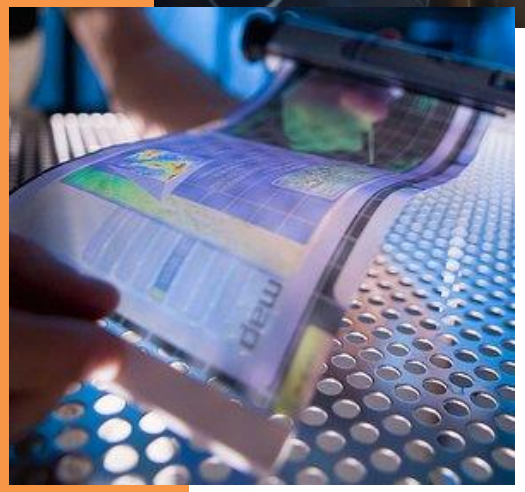
Thorfinn Roccapriore

The World is Material Opportunity









Predicting crystal structure by merging data mining with quantum mechanics

CHRISTOPHER C. FISCHER[1], KEVIN J. TIBBETTS[1], DANE MORGAN[2] AND GERBRAND CEDER[1]*

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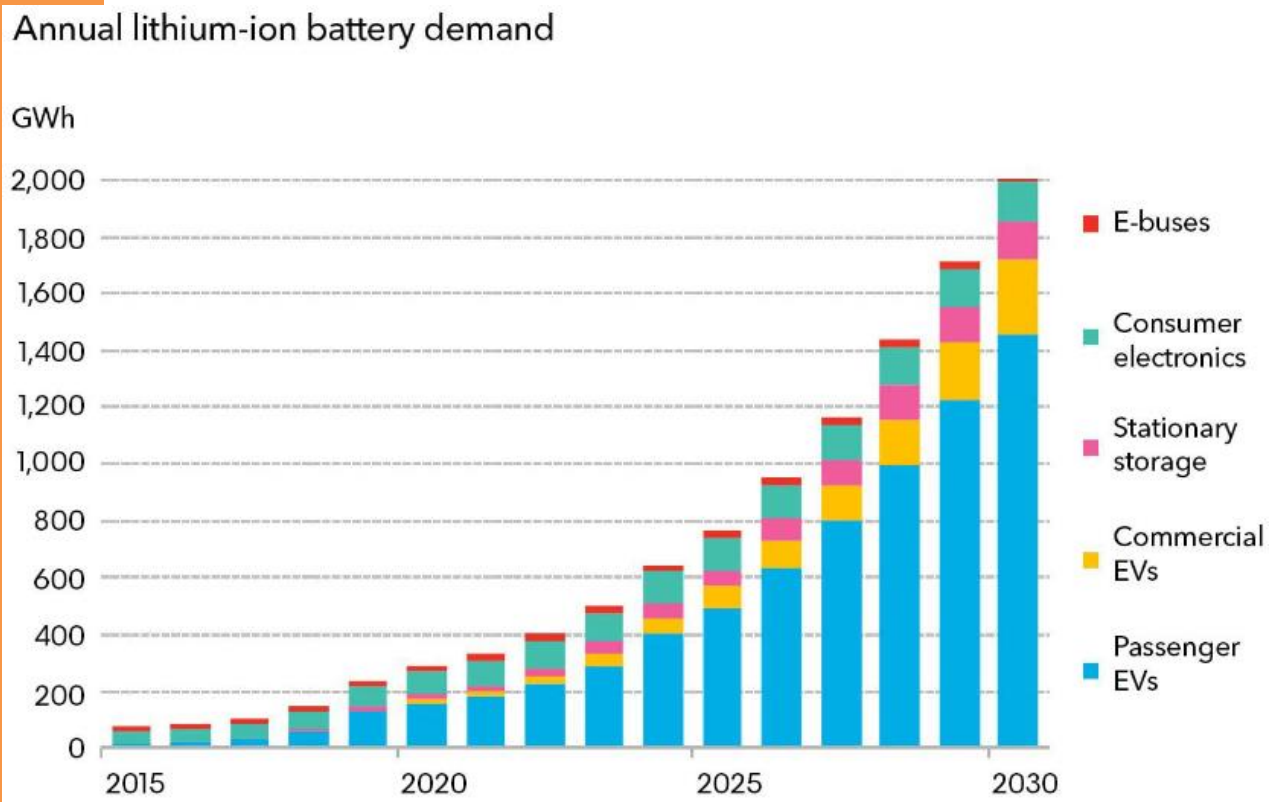
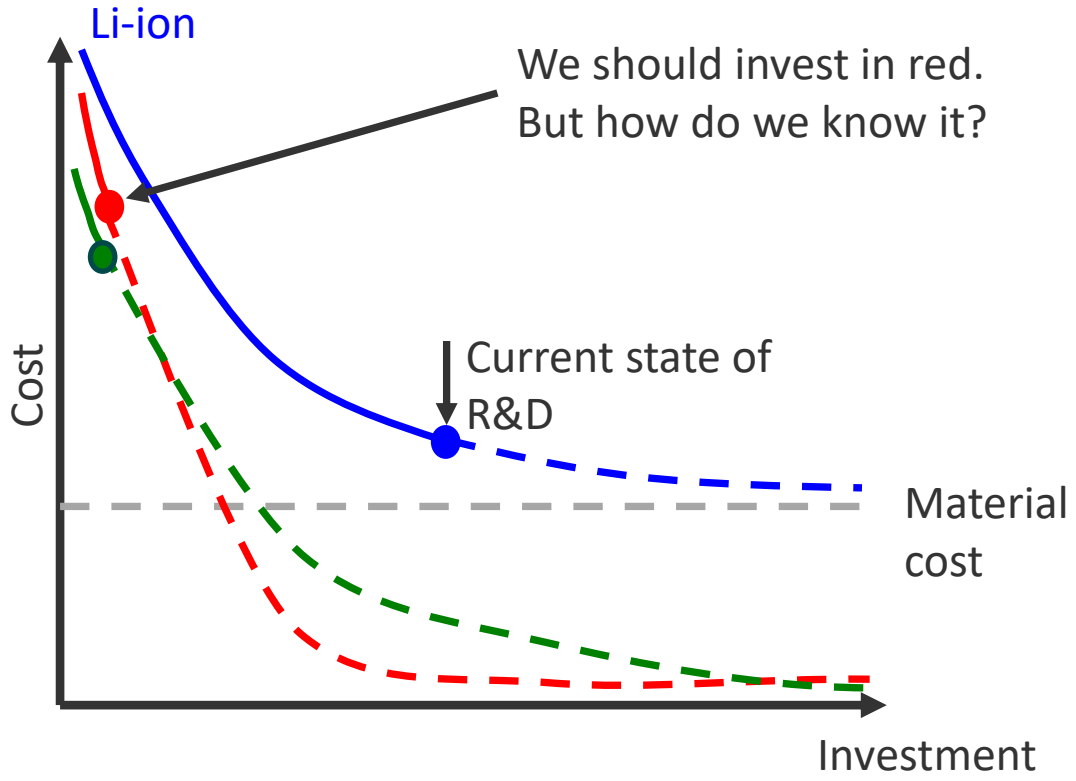
- **"Improve":** Renewable energy, self-driving cars, transparent displays, new memory technologies
- **"Discover":** Room temperature superconductivity, high mechanical stress materials
- **"Engineer":** Quantum computing, single-atom catalysts, biomolecules

Functionality, manufacturability, cost

Batteries: Li-ion and Beyond

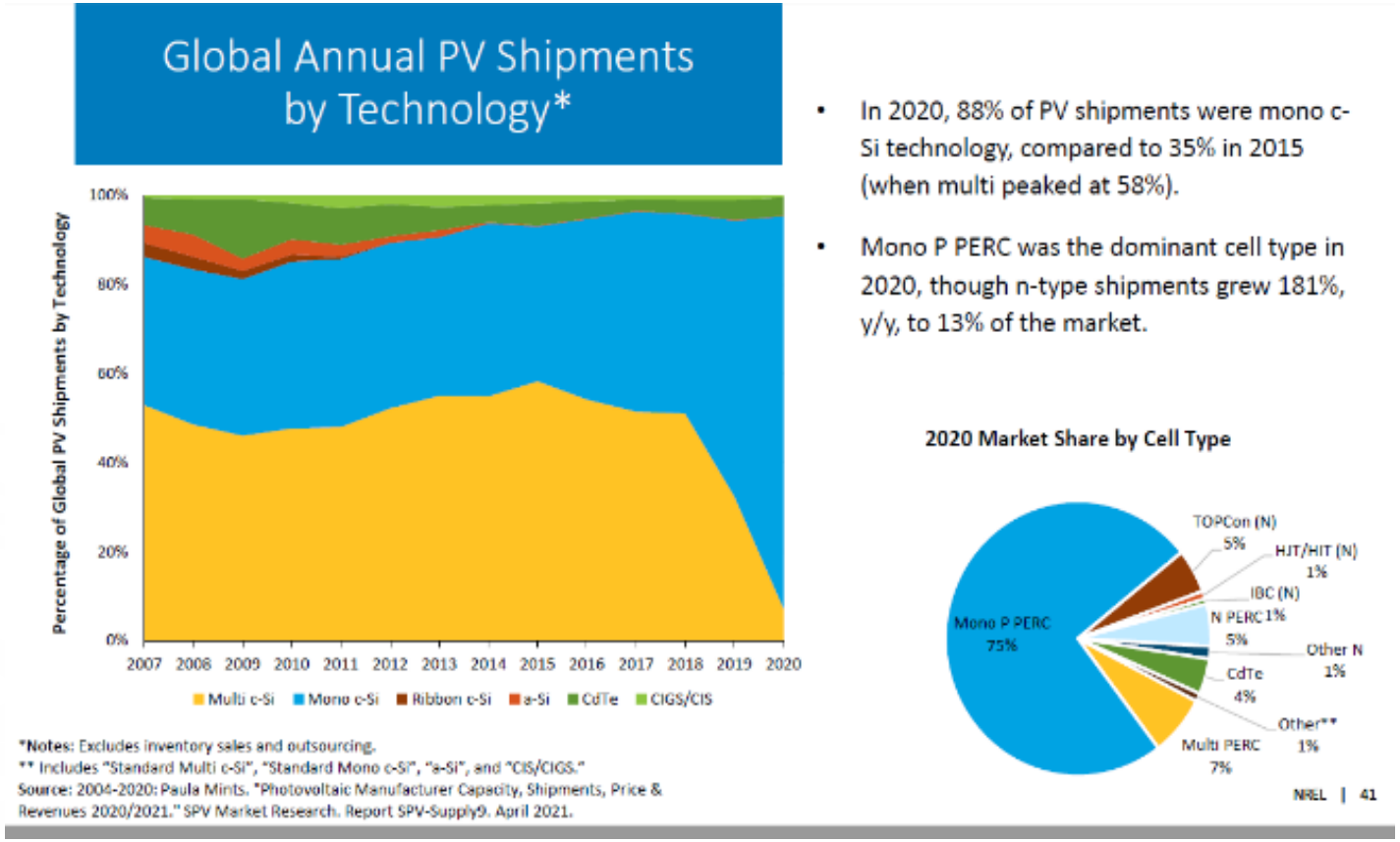




- Batteries are required element of energy transition (EVs, ESS, mobile devices)
- Currently Li-ion is the primary technology
- Optimization of Li-ion batteries takes years (even with same process on new Gigafactory)
- However, it is far from Goldilock zone for ESS or energy transport

- How can we optimize usage and safety for Li-ion batteries in EVs?
- How do we select beyond Li technologies for ESS?

Solar Energy: Will Silicon Ever Reign?



- In 2020, 88% of PV shipments were mono c-Si technology, compared to 35% in 2015 (when multi peaked at 58%).
- Mono P PERC was the dominant cell type in 2020, though n-type shipments grew 181%, y/y, to 13% of the market.

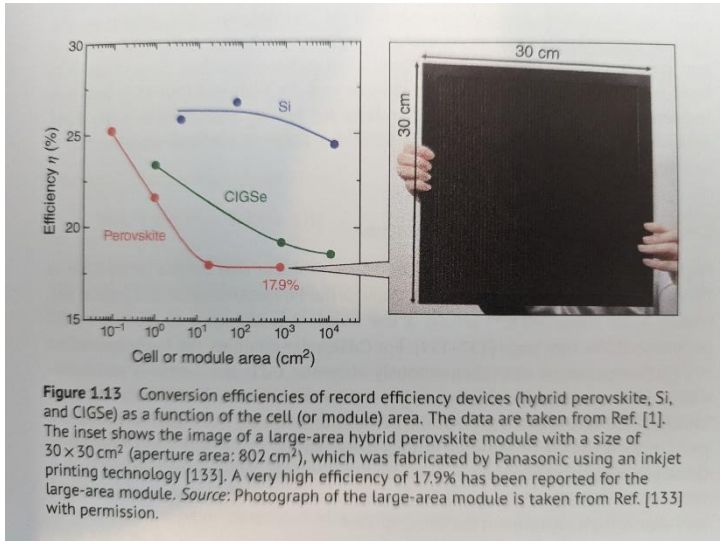
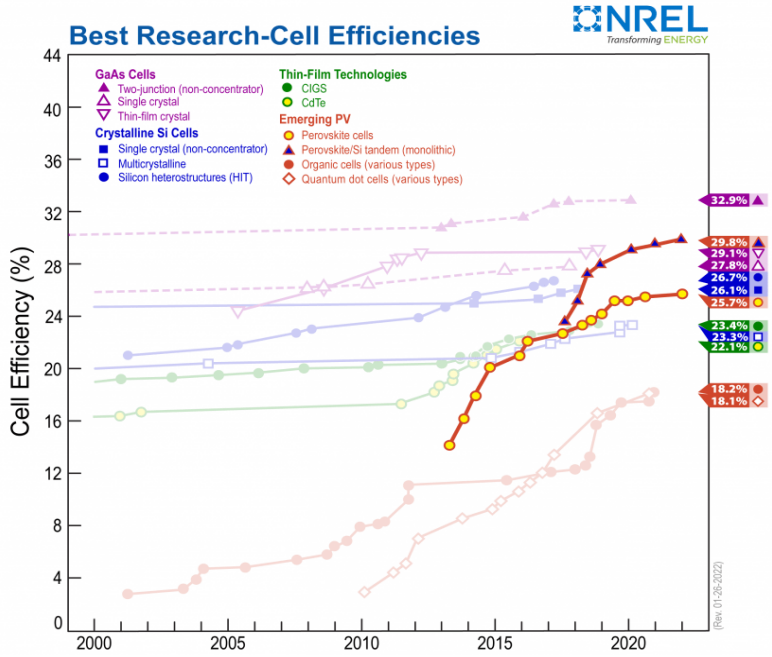
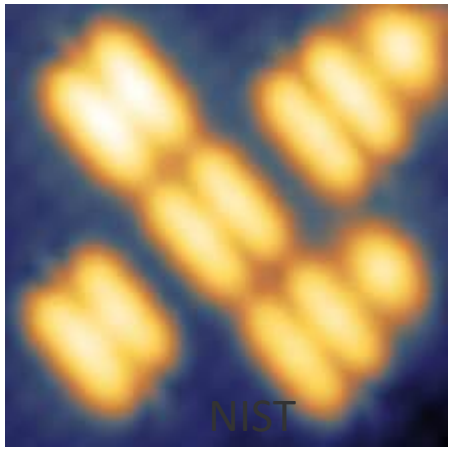


Figure 1.13 Conversion efficiencies of record efficiency devices (hybrid perovskite, Si, and CIGSe) as a function of the cell (or module) area. The data are taken from Ref. [1]. The inset shows the image of a large-area hybrid perovskite module with a size of $30 \times 30\ cm^2$ (aperture area: $802\ cm^2$), which was fabricated by Panasonic using an inkjet printing technology [133]. A very high efficiency of 17.9% has been reported for the large-area module. *Source*: Photograph of the large-area module is taken from Ref. [133] with permission.

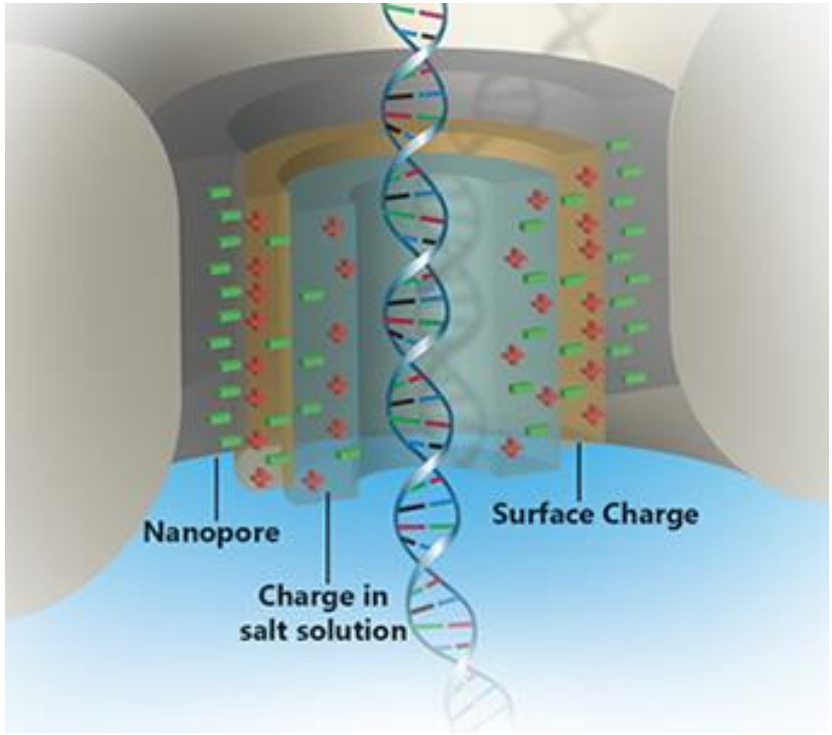


- Solar energy is the fastest growing energy sector
- Si is now reining material – however, it is really not the optimal material for PV (heavy, expensive)!
- Hybrid perovskites can be used as ideal PV materials – if we can make them stable and scale manufacturing!

Quantum Computing and Single Molecule Bio



Zyvex Labs



Oxford Nanopore

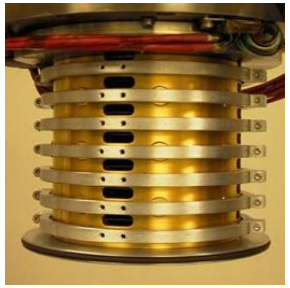
- Direct atomic fabrication: quantum communications and quantum computing, environmental sensing
- Single-molecule biological devices

- Success story 1: cryo-electron microscopy
- Success story 2: nanoelectron diffraction

We need to go small to go big!

The Lab on the Beam

Image atomic columns

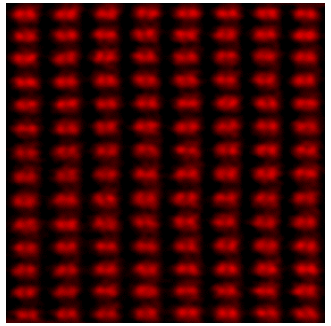


First AC (Nion)
1997

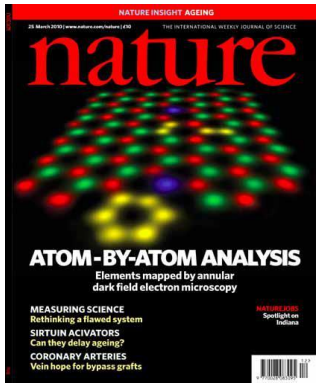
Spectra from single atoms

Image light atoms and sensitive materials

0.61 A resolution

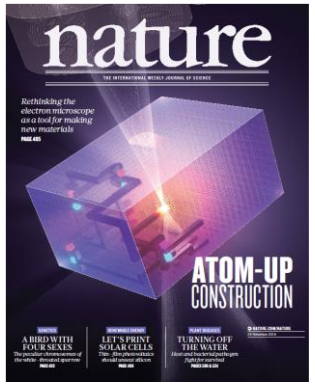


Diffraction from subatomic volumes



Beams with orbital momentum

Beam manipulation



- Vortex beams
- High-res EELS
- In-situ holders
- 4D STEM
- Physics extraction
- Atomic manipulation

Sample
Source
Detector
Data
Feedback

Images from A. Lupini

2002 Prototype correctors

2006 TEAM project

2012 Broad adoption of AC STEM

2012 Segmented detectors

2014 4D STEM

2016 Vortex beams

2018

2012 DCNNs

2014 GANs, VAEs

AlphaGo

2016 Attention Transformers

2018 Invariant VAEs

2020 muZero

AlphaFold

2022 LLMs

However

We run the instruments the same way as decades ago:

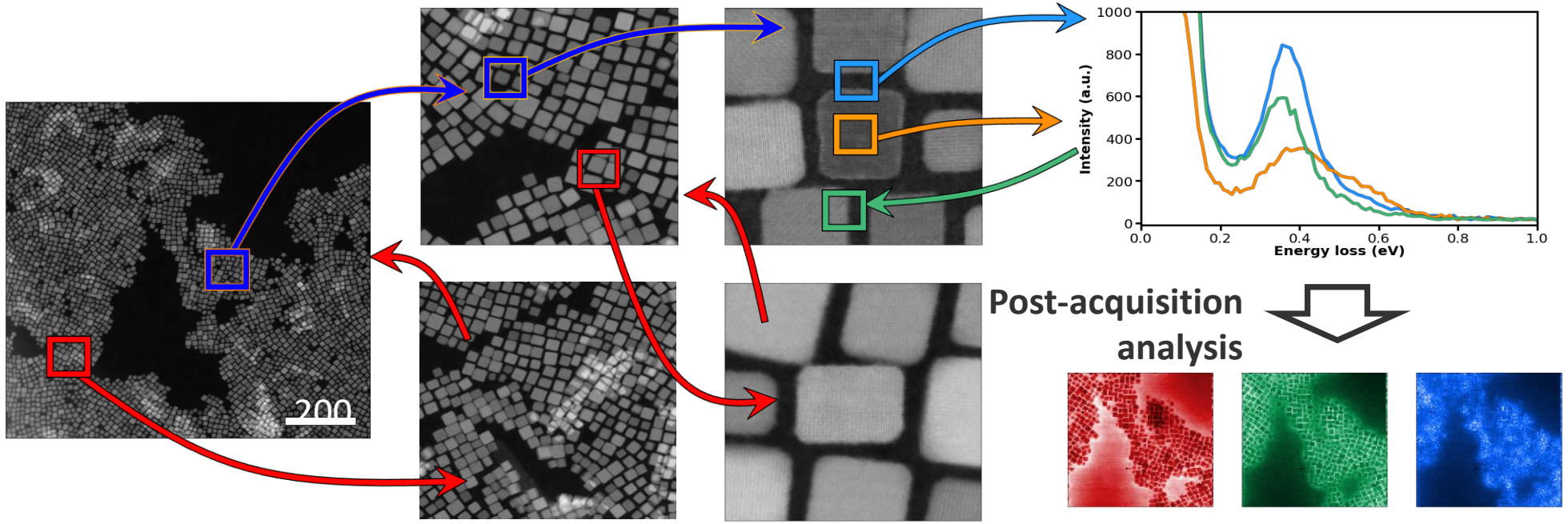
- Rectangular scans
- Point spectroscopy
- Hyperspectral imaging (spectroscopy on the grid)

Opportunities:

- Very high sampling images (>2k)
- Multidimensional spectroscopies
- Objects of interest are non-uniform in image plane

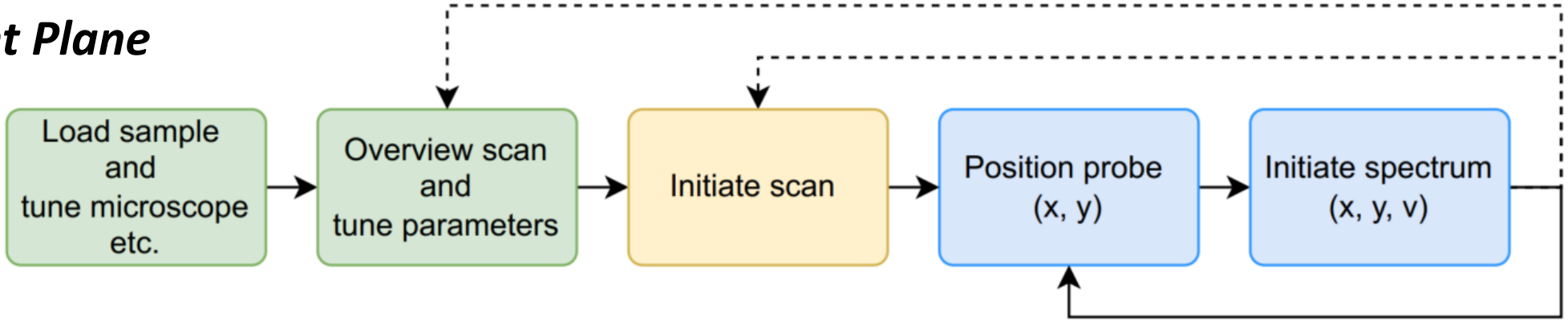
- Why microscopy?

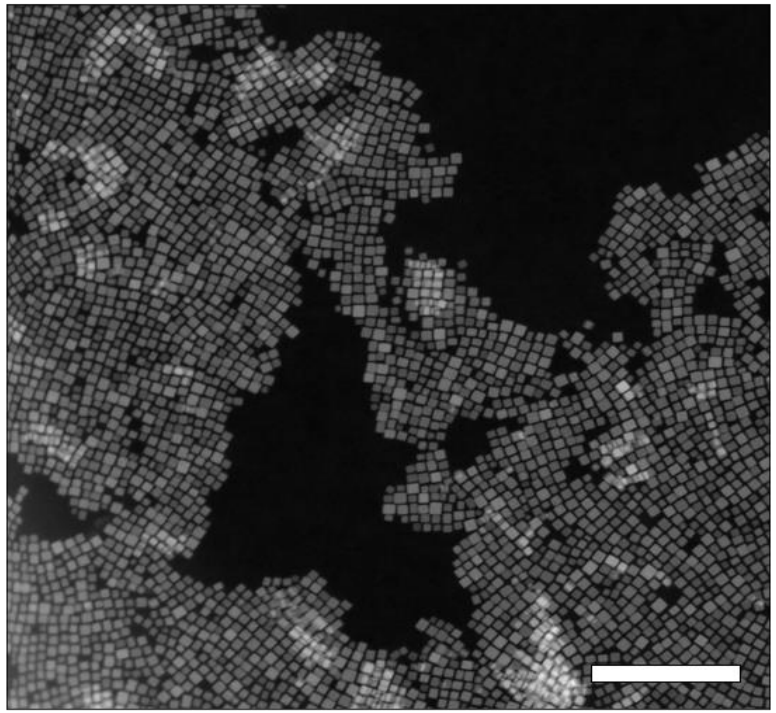
Workflows in STEM

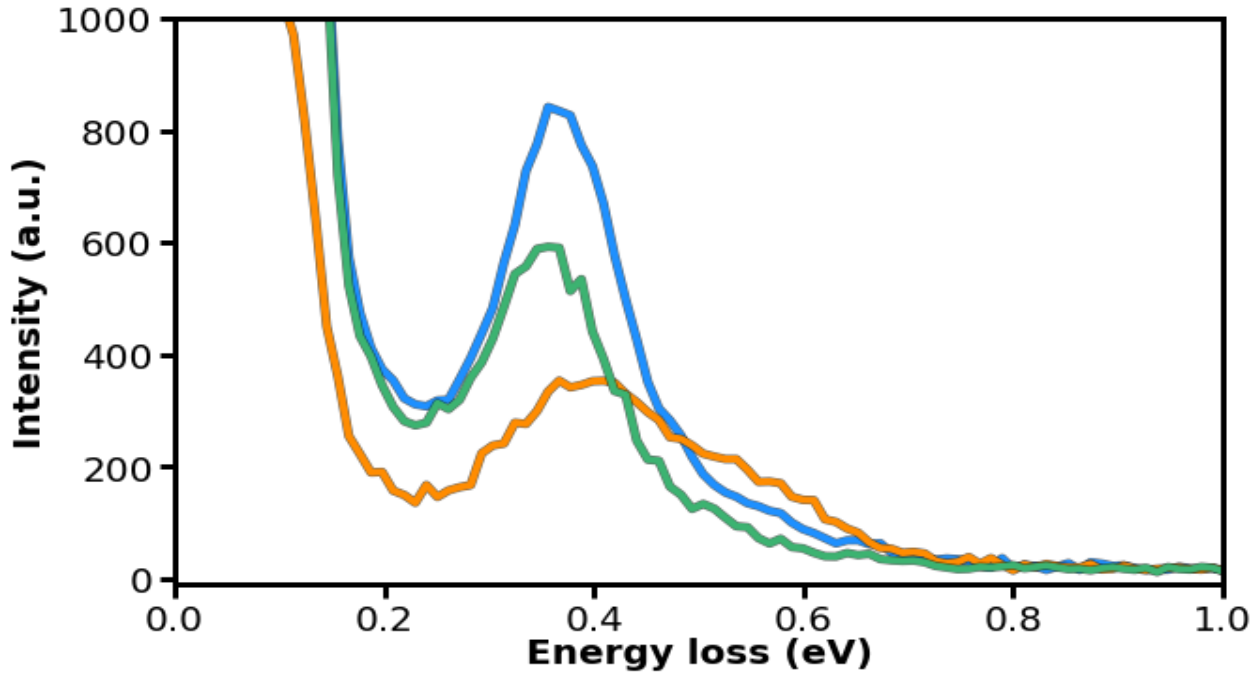


Minimal instruction set control language

Instrument Plane







- 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**

This workshop:

Learn how to transition from image quantification to ML-enabled workflows

Tentative Program

1. Outline and structure of the course - June 6

2. Imaging in Scanning Transmission Electron Microscopy - June 9
3. Spectroscopy in STEM - June 13

4. Linear methods and dimensionality reduction for spectral data - June 16

5. High-resolution and Z-Contrast Imaging -June 20
6. Image registration methods - June 23
7. Linear methods and dimensionality reduction for imaging data - June 27
8. Diffraction and 4D STEM - June 30
9. Bringing Cloud and Edge to STEM: from tool to ecosystem - July 4
10. Image simulations - July 7

11. Deep convolutional networks - July 11
12. DCNN for image data - July 14
13. DCNN case studies - July 18

14. Gaussian processes and Bayesian Optimization - July 21
15. Bayesian Inference, Structured GP, and Hypothesis Learning - July 25

16. Variational Autoencoders – 1 - July 28
17. Variational Autoencoders – 2 - August 1
18. Encoders-decoders and structure-property relationships - August 4
19. Special topic: VAE for any tasks - August 8

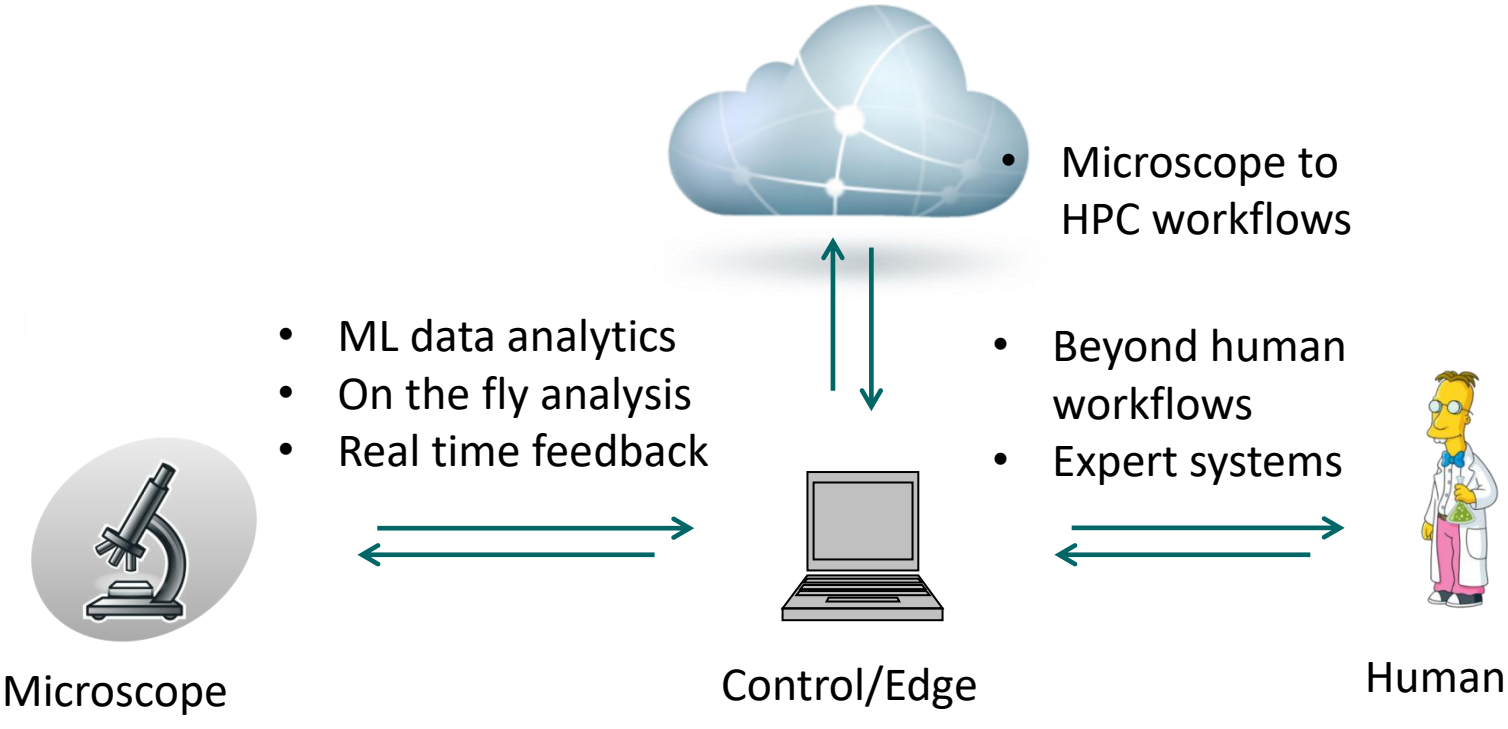
20. Deep kernel learning: EELS and 4D STEM – August 11
21. DKL forensics and human in the loop - August 15

22. Special topics: Reinforcement learning - August 18
23. Special topics: Learning physics form images - August 22
24. Special topic: Causality - August 25

1. Over 15-year experience in applying ML to experimental physical problems
2. Over 5-year experience in automated and autonomous microscopy via edge computing
3. Experience in ML for materials synthesis and synthesis/characterization workflow design
4. Leading teams developing data infrastructure infrastructure for scanning probe and electron microscopy

Sergei V. Kalinin

- Ph.D. 2002: U. Penn
- 2002-2022: Oak Ridge
- 2022-2023: Amazon, GC
- 2023: U. Tennessee, Knoxville



Rama Vasudevan

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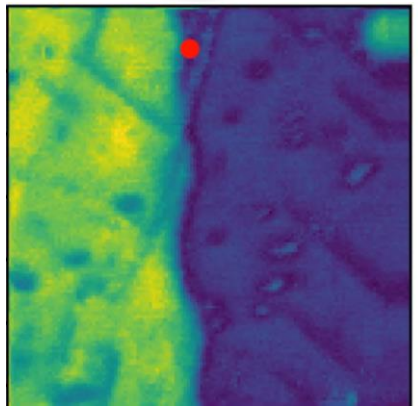

- Science Interests: Autonomous labs, microscopy, open-source scientific software, machine learning and reinforcement learning
- Currently group leader of Data NanoAnalytics at CNMS/ORNL

- Running/Cricket/Gardening. Former opening bowler/now relegated to part time pie thrower
- Avid Gardener. Wannabe Farmer.

Maxim Ziatdinov

Dr. Maxim Ziatdinov, who has devoted his life's work to the masterful synthesis of machine learning, experimental practices, and theoretical principles, all aimed to expedite discoveries in the grand tapestry of physical sciences.

He has meticulously woven together science-informed machine learning algorithms, as detailed and intricate as the astrolabes of the Citadel, each informed and refined by prior realms of knowledge. His scholarly endeavors have also led to the creation of crucial pathways, akin to ancient Valyrian roads, connecting the latest instrumental platforms with the sheer might of high-performance computing facilities.

A master craftsman of the modern age, he has forged numerous software artifacts, each imbued with a unique purpose, much like the famed blacksmiths of Qohor. Among these are **AtomAI**, a tool as keen as Valyrian steel in the realm of deep learning applications for microscopy, **pyroVED**, a mystical sigil for invariant autoencoders in image and spectral analysis, and **GPax**, akin to the magical weirwood trees, it empowers physics-based active learning and Bayesian optimization in automated experiments. Each tool is shared freely in the community, like a lord sharing his feast with his people, contributing to a broader and deeper understanding of our world.

In the dominions of knowledge and discovery, Dr. Ziatdinov is not a solitary wanderer, but a cherished member of two esteemed brotherhoods. He holds vital roles in the noble houses of the CSED and CNMS at the towering fortress of ORNL, both renowned for their dedication to unraveling the mysteries of our universe.

Gerd Duscher

Research

- Electrical activity at interfaces and crystal defects
- Atomic and electronic structure at interfaces
- Atomic resolution Z-contrast imaging
- Atomic resolution electron energy-loss spectroscopy
- Atomic structure calculations with density functional theory

Dr. Duscher earned his master's degree (Diplom) in Physics from the University of Regensburg, Germany in 1990, and his Dr. Sci. in Materials Science and Engineering from the University of Stuttgart in 1996.

His dissertation research was performed at the Max-Planck Institute for Metals Research, Stuttgart, where he held a Fellowship. He used analytic TEM methods at both his masters and his doctoral research to investigate the relationships between the atomic structure and macroscopic properties. In his dissertation research he studied the influence of oxide precipitates on the electrical activity of grain boundaries in silicon.

In 1997, Dr. Duscher joined the group of Dr. Steve Pennycook in the Solid State Division at the Oak Ridge National Laboratory as a Postdoctoral Research Associate where he studied grain boundaries in oxides. In 1998, Dr. Duscher was a staff member of the Max-Planck Institute and in 1999 he accepted a position as a Research Assistant Professor for Theoretical Physics with Dr. Sokrates Pantelides at Vanderbilt University.

In 2001 Dr. Duscher joined the faculty at the North Carolina State University, Raleigh, and in 2006 he became the director of the Silicon Wafer Engineering and Defect Science a NSF IUCRC center. In 2008 Dr. Duscher joined the faculty at The University of Tennessee . He has authored or co-authored more than 100 technical papers and has contributed to over 50 (25 invited) technical presentations at national and international conferences.

Ayana Ghosh

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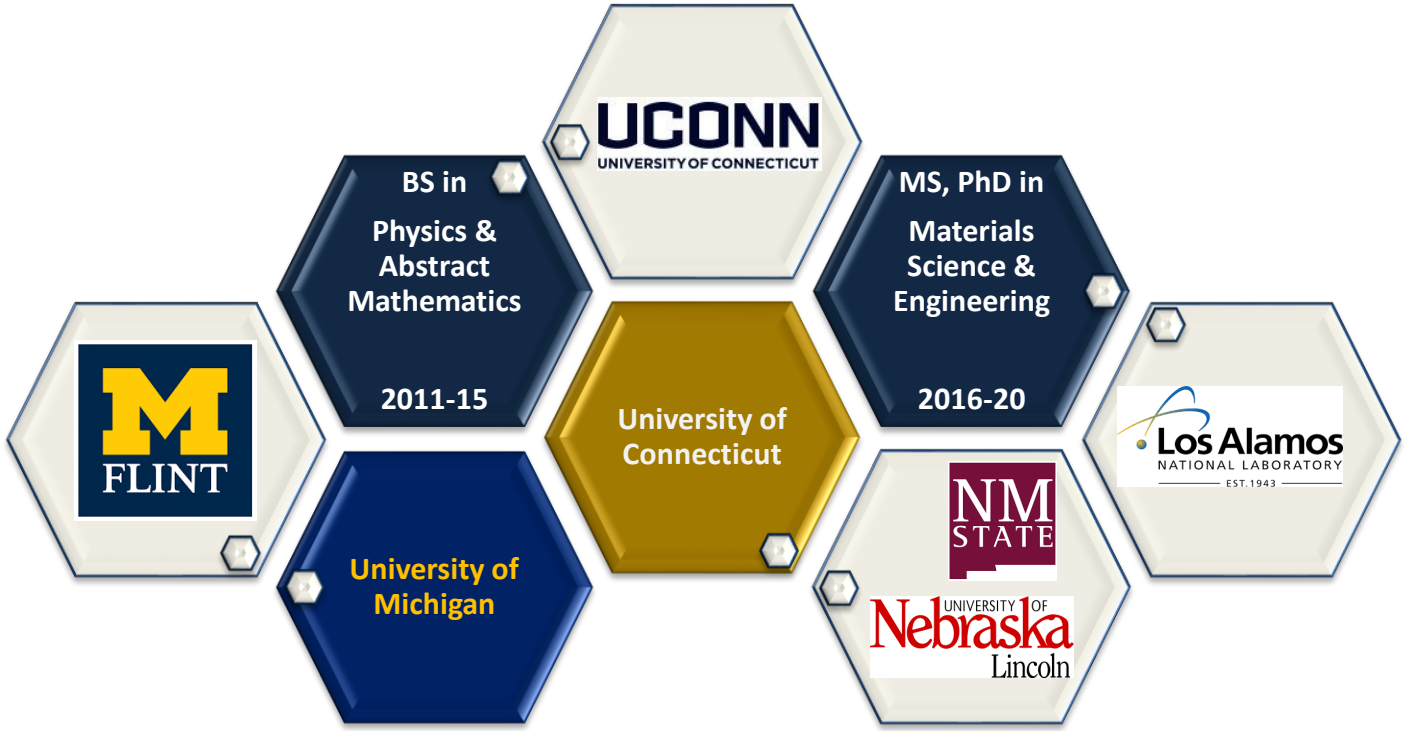
- Materials
- Physical Models
- Machine Learning & Causal analysis
- Combine Theory & Experiments



OAK RIDGE National Laboratory

Postdoc at ORNL
(Sept. 2020 – Feb. 2023)

Research Scientist at ORNL
(March 2023 -- Present)

Kevin Roccapriore:

- Staff scientist at Oak Ridge National Laboratory's Center for Nanophase Materials Sciences (CNMS).
- PhD in Physics from the University of North Texas in 2018, which was nanofabrication for nanophotonics applications.
- Post-doc at ORNL (2018-2021), focusing on machine learning in the scanning transmission electron microscope (STEM) toward automated experiments, including STEM-EELS and 4D-STEM.
- Currently, focuses on ML on-the-fly in the STEM for precise atomic fabrication by site-specific atom targeting with the electron beam, automated experiments learning and utilizing structure-property relationships, and automated multidimensional data acquisition and analysis.

Tommy (Chun Yin) Wong

- Research interests: deep learning & computer vision, electron microscopy, radiation damage
- 3rd year Ph.D. student in Energy Science & Engineering, Bredesen Center, University of Tennessee
- M.S. in Materials Science & Engineering '22, University of Tennessee
- B.S.E. in Nuclear Engineering & Radiological Sciences '20, University of Michigan

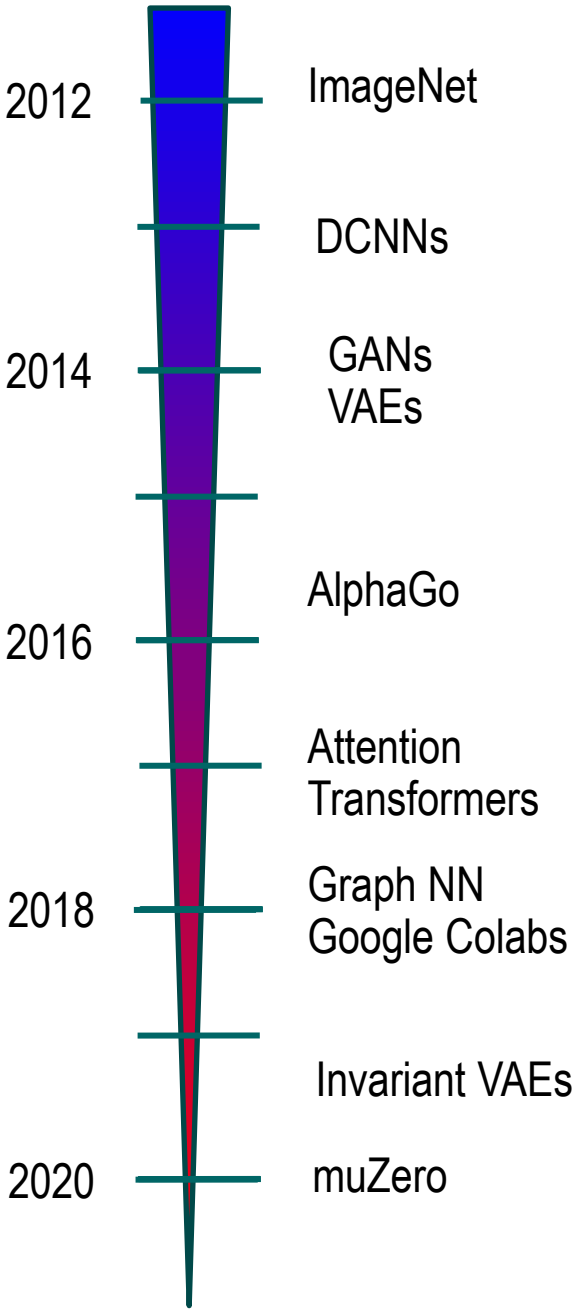
Why Machine Learning?

- Last decade has experienced an explosive growth of machine learning and artificial intelligence applications
- These developments have spanned areas from computer vision to medicine to autonomous systems and games
- However, the progress and impact as applied to experimental physical sciences has been minimal….

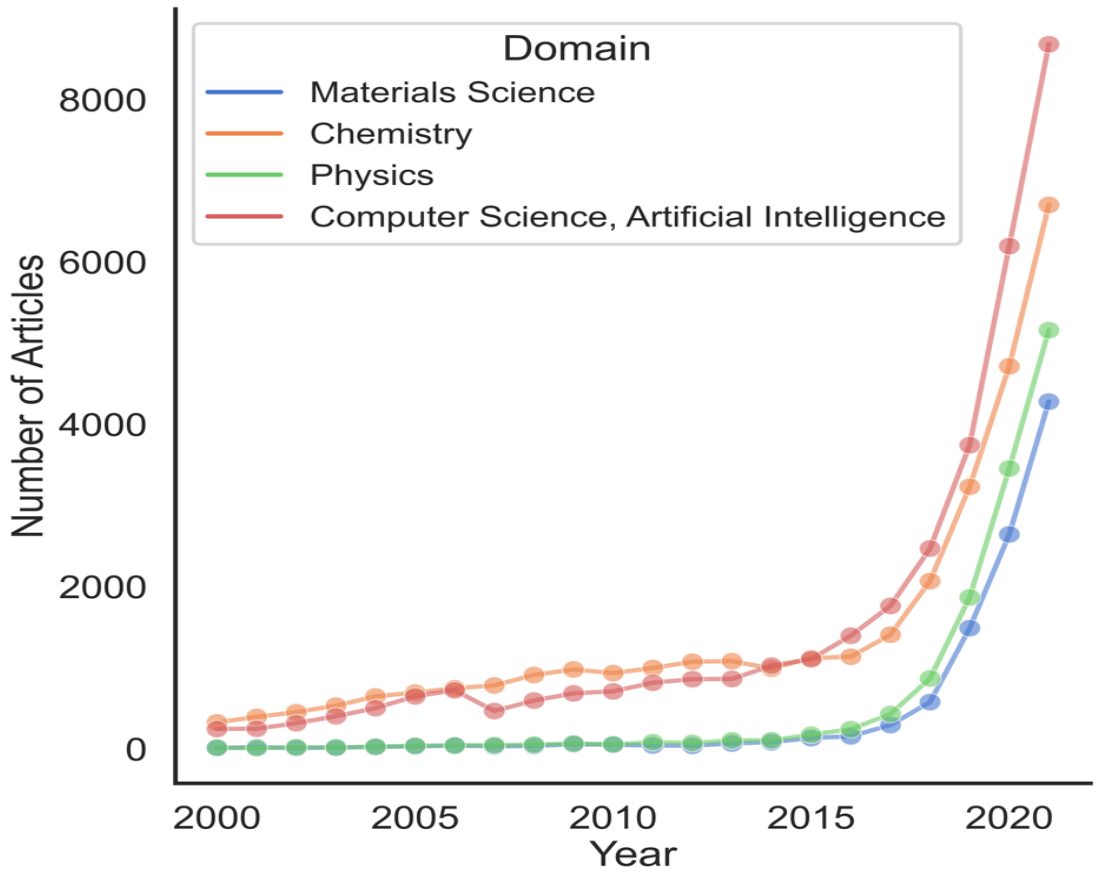
Why is it difficult?

- Requires domain expertise and domain-specific goals
- Deeply causal and hypothesis drive nature of domain sciences
- No single answer: culture, not a method
- Infrastructure, open code, open data
- **Most important:** active nature of scientific process

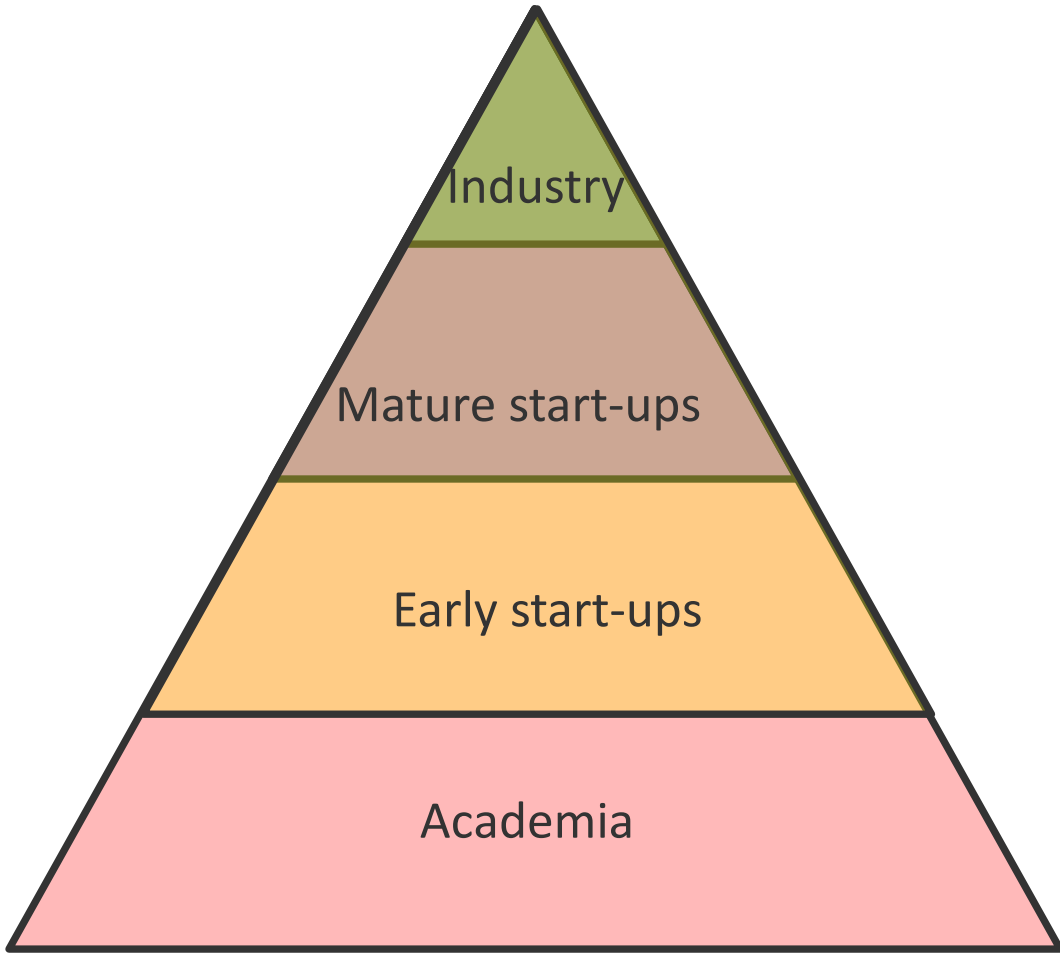
Microsoft: GitHub
Meta: Open Catalyst,
Meta: Papers with Code
Toyota: TRI
Google: AlphaFold
NVIDIA: protein folding



ML in Domain Sciences



Analysis by B. Blaiszik, Argonne



- The rapid adoption of ML in domain sciences and industrial R&D is a very recent trend
- Technologies and workforce emerge from academia into industry
- We can estimate potential growth rates comparing to cloud computing 15 - 20 years ago

“Eras” of ML in Industry

- **Before 2000:** It’s all about IT (dotcoms, Amazon, etc)
- **2000 - 2010:** It’s all about collecting and searching data (Facebook, Google, Uber, etc.)
- **2010 – 2020:** What do we learn from data (correlative era)
- **2020 – now:** Physics is the new data

arXiv:2204.05095

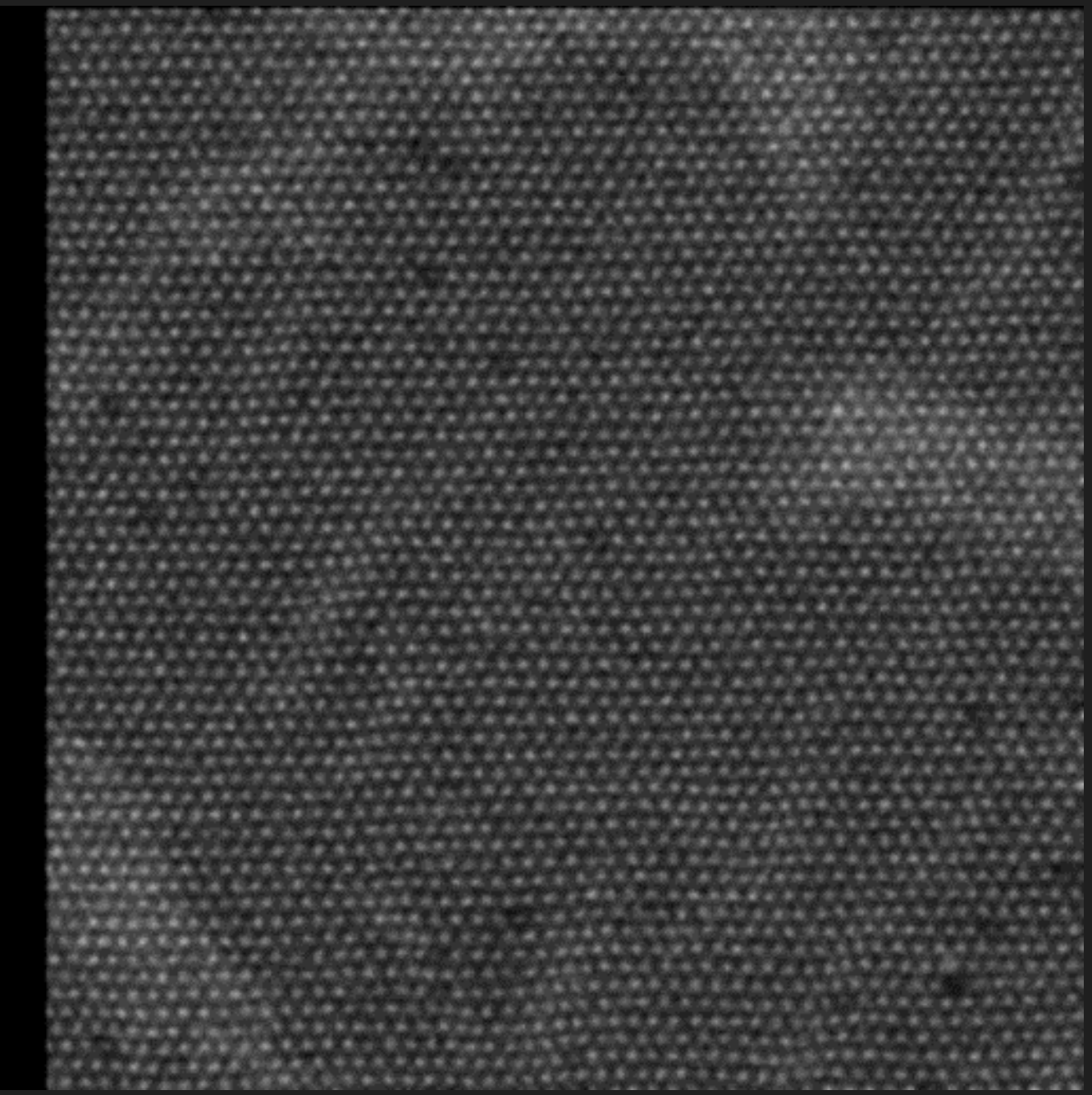
- Classical machine learning is underpinned by the existence of the large static data sets – from MNIST to emerging medical, bio, faces, etc.
- Real world problems are associated with the large distribution shifts, often small data sets, and presence of uncontrollable exogenous factors
- Also, real world problems are often active learning: we interrogate the data generation process and provide feedback, not deal with static data sets
- However, we often have extensive prior knowledge of past data, physical laws generalizing them, and strong set of inferential biases

arXiv:2005.01557

ML for real-world applications is different!

Can Machine Learning help us analyze the acquired data?

Dynamic Atomic Changes



Observations of atomic dynamics induced by beam (or temperature, field, etc.) gives information on multiple atomic configurations as they form and evolve. Can we:

1. Minimize beam damage:
    a. Low dose imaging
    b. Low beam energy
    c. Tailored measurement grid?

2. Can we learn the transformation mechanisms from atomic dynamics?

3. Can we learn force fields between the atoms?

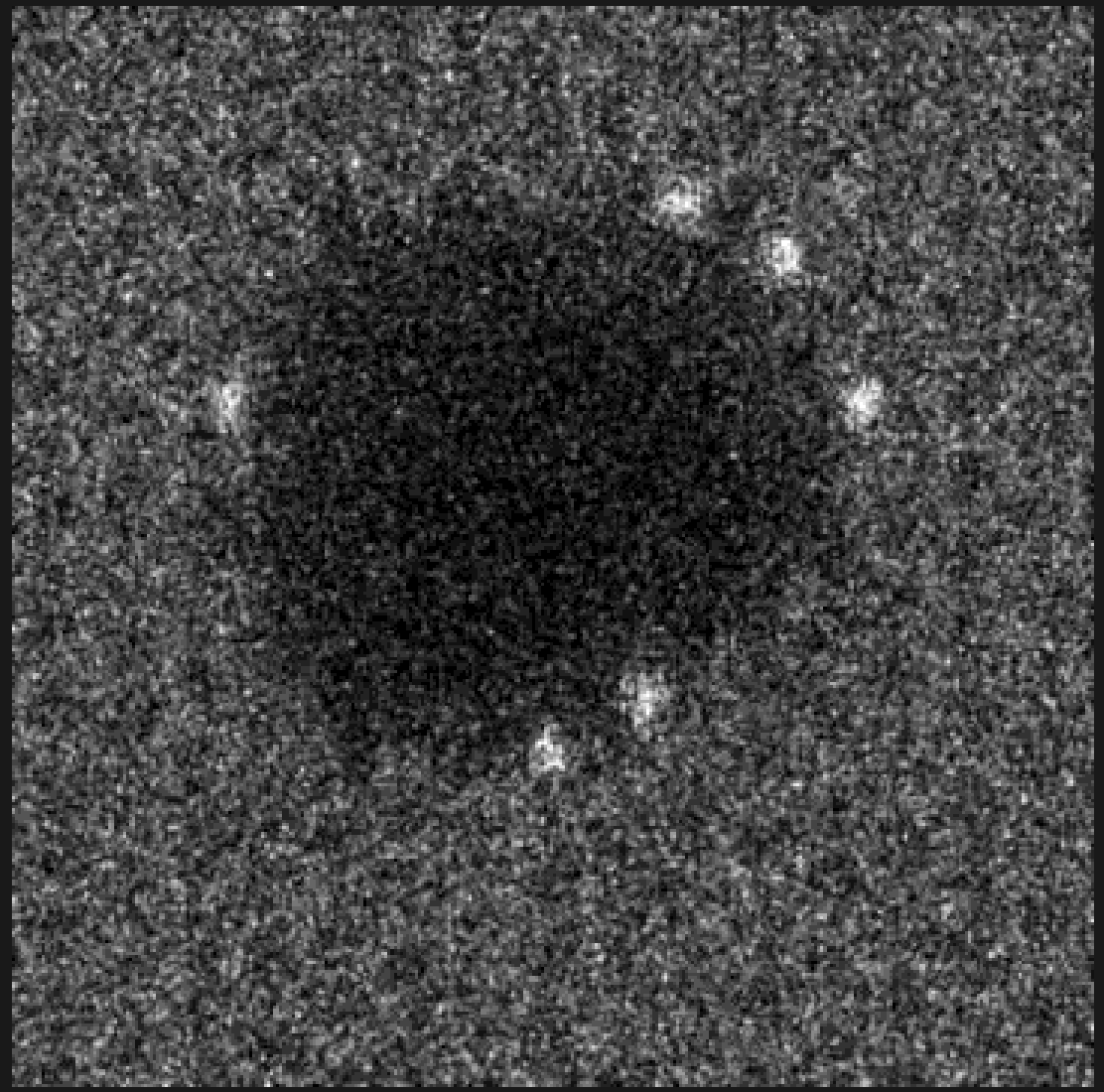
4. Do we really have to take EELS and 4D STEM measurements everywhere?

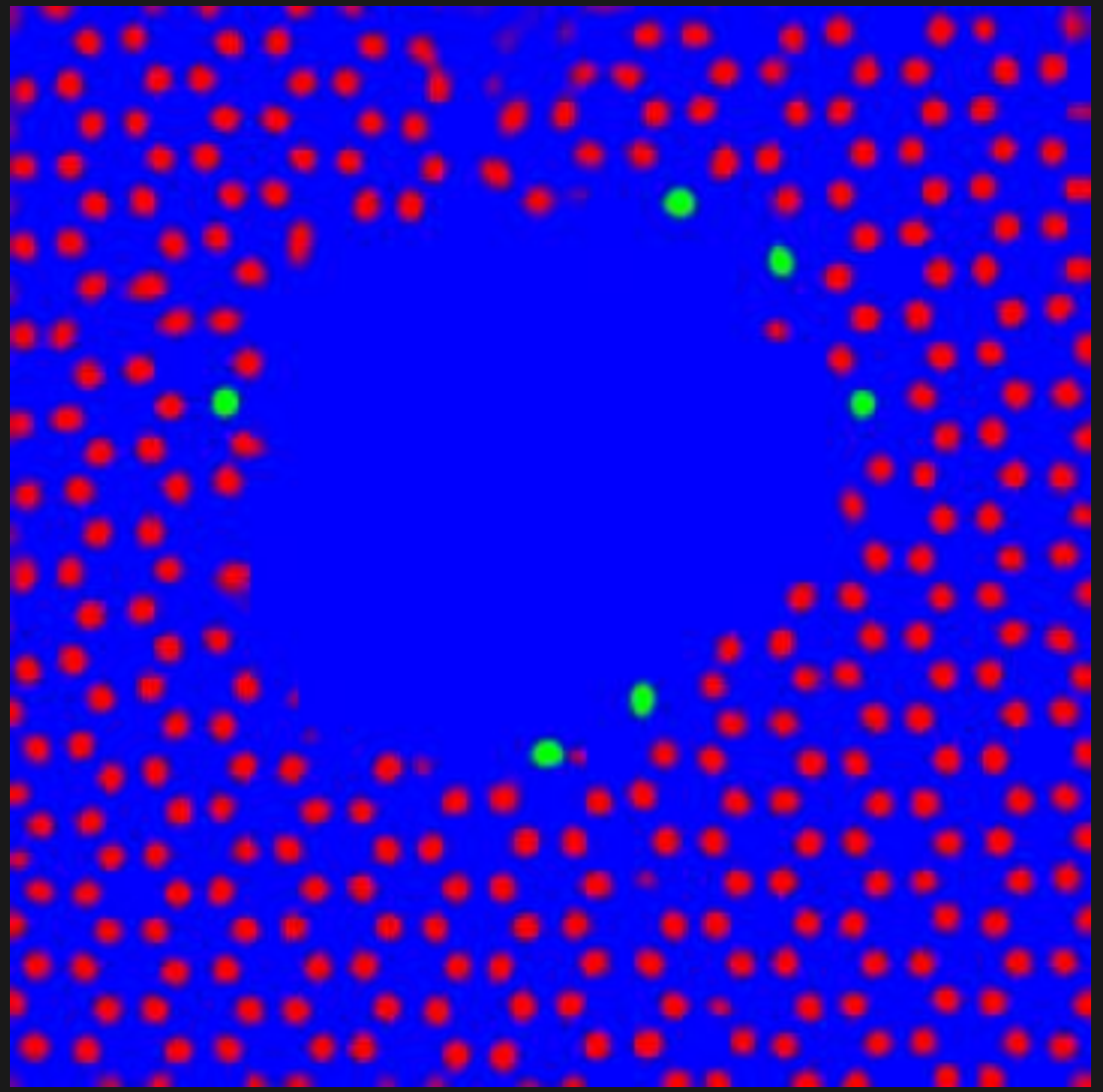
5. Can we learn how to **direct** beam induced transformations atom-by-atom?

Deep learning works like a charm for:

- Drift correction
- Denoising
- Data processing/dimensionality reduction
- Feature finding (physics is in the training set)

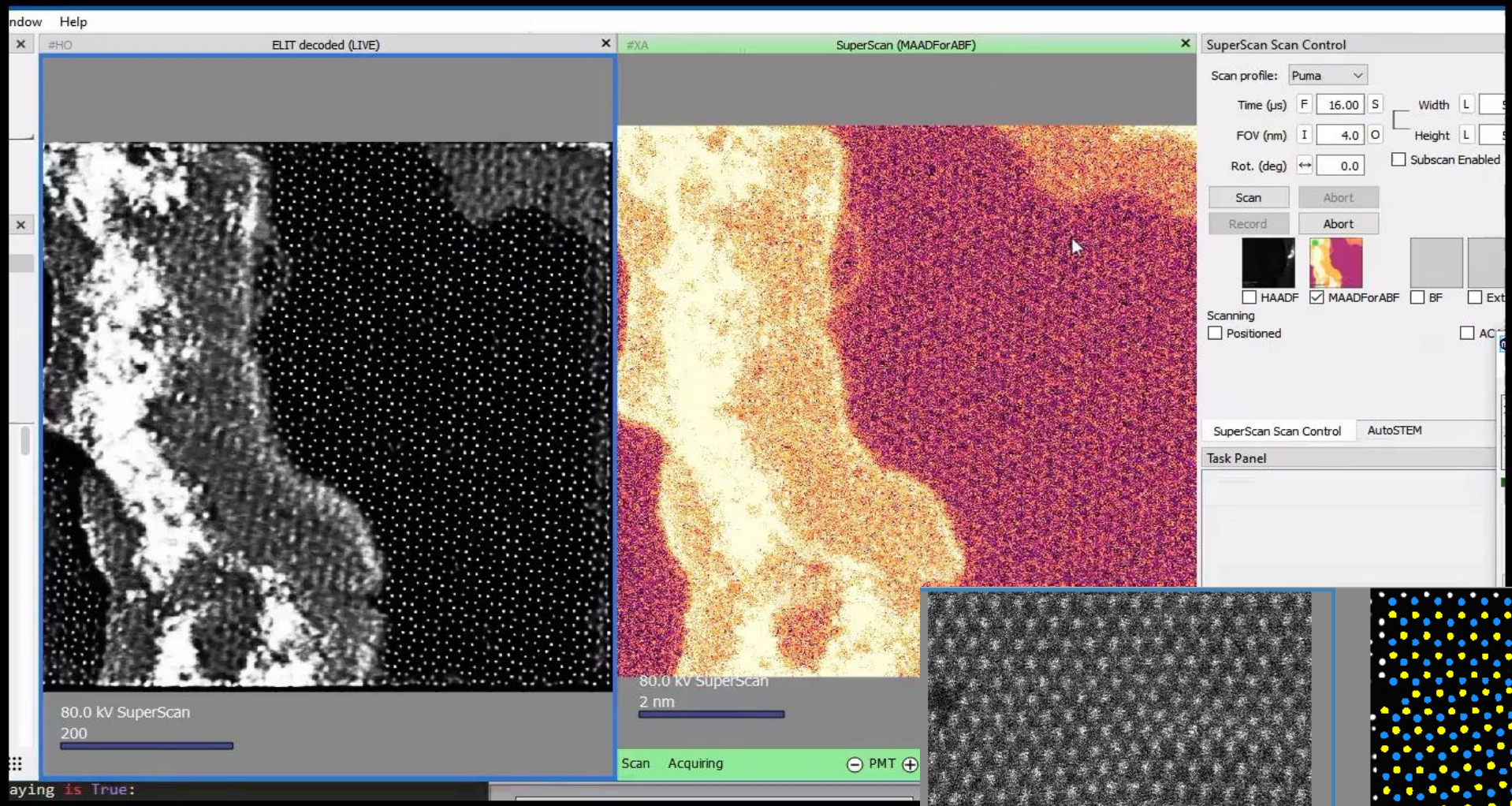
M. Ziatdinov et al, ACS Nano 2017





Automated Experiment:

almost easy…. If you know what you are looking for



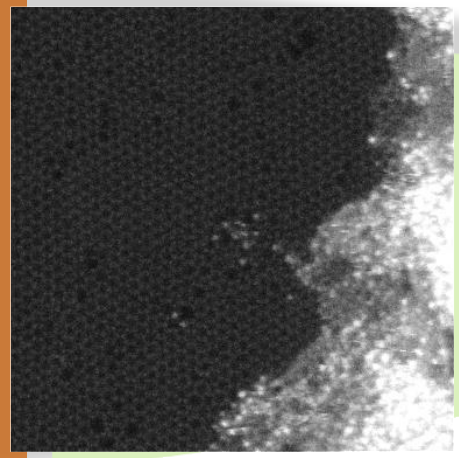
Automated control

Implementation: Kevin Roccapriore, Ayana Ghosh, Sergei V. Kalinin & Maxim Ziatdinov, 2021

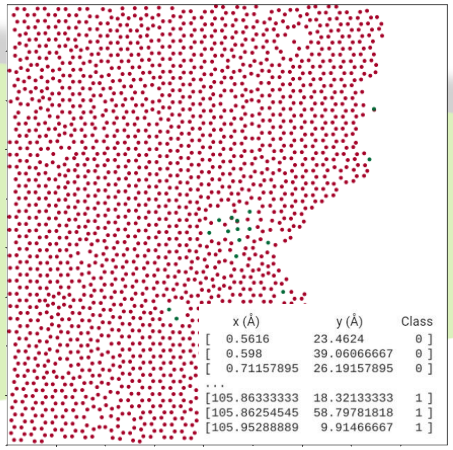
From Microscope to Simulation Environment

Stage 1 | Stage 2 | Stage 3

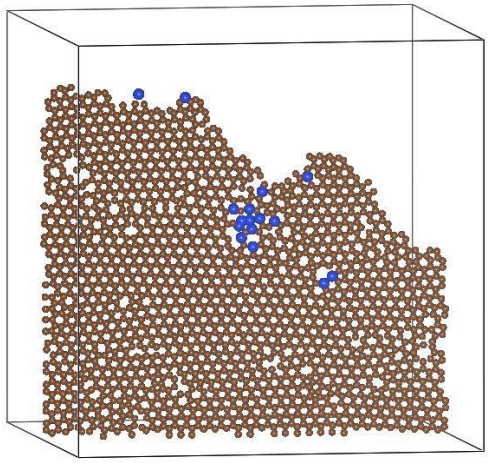
Microscopic images → **Atomic Types & Coordinates** → **Atomistic Simulations** → **Physical Phenomena**



DCNN



Transform

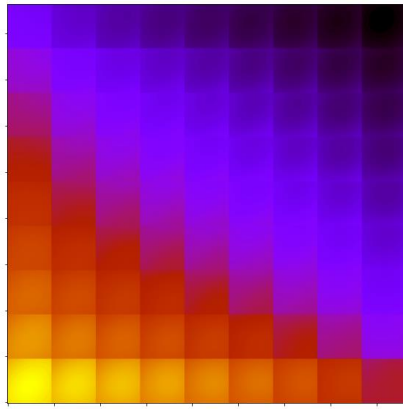


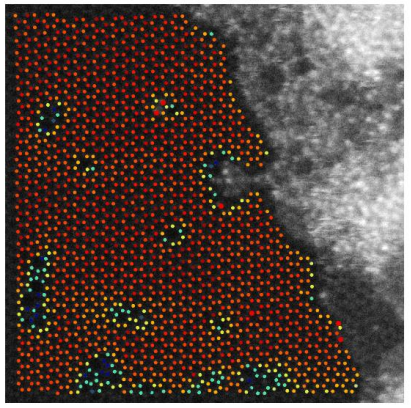
Materials Properties

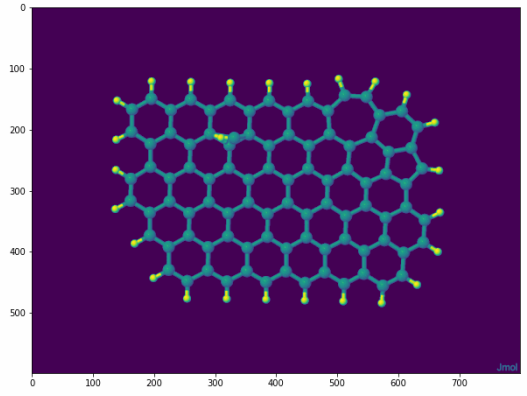
Feature Finding & Physics-informed learning

Data **Machine Learning** **Human-in-the-Loop** **Learn Physics**

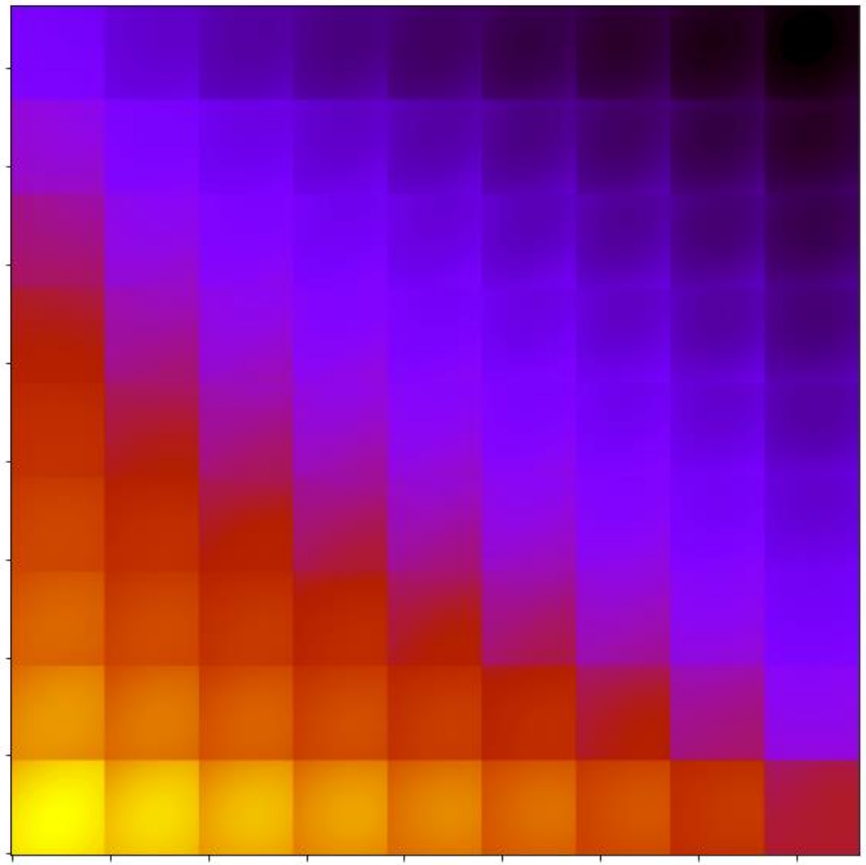
- DCNNs for atom finding
- Invariant VAEs for physics discovery
- Conditional VAEs
- GP reconstructions
- Piping data in DFT/MD environment

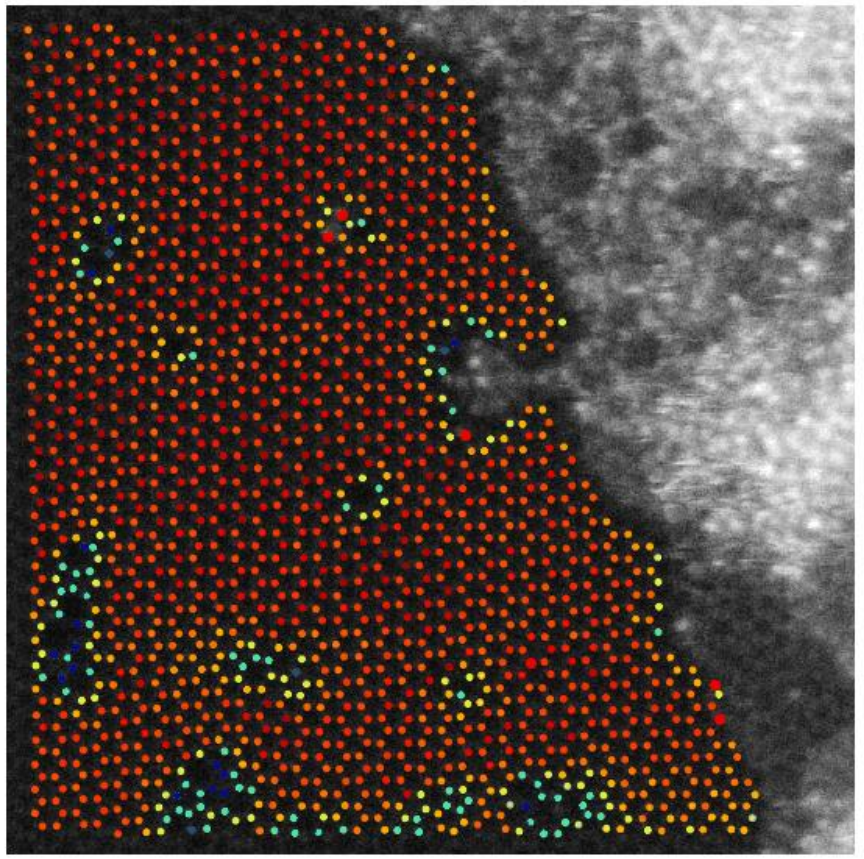






Discovering Building Blocks





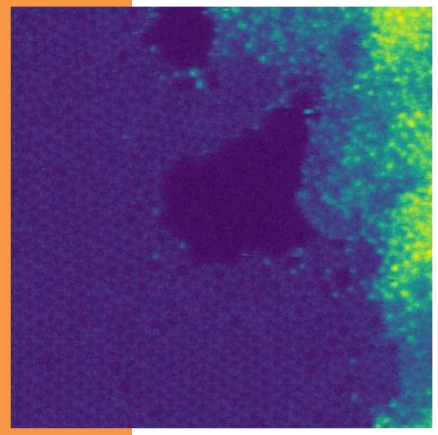
- Classical physical descriptions can be defined locally only in Bayesian sense
- We can argue that local descriptors are simple, if not necessarily known
- And the rules that guide their emergence are also simple, if not known
- rVAE discovers trends and (joint rVAE) building blocks irrespectively of orientation or shift in the image plane

Discovering Building Blocks

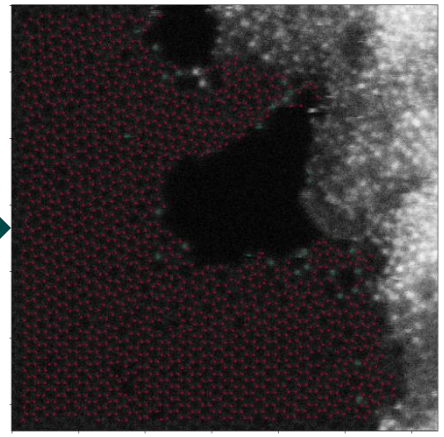
Atom exists, are discrete, and have known shape

Structures are parsimonious in nature

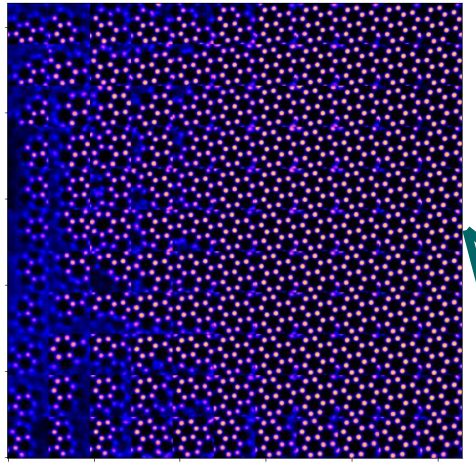
Raw observation



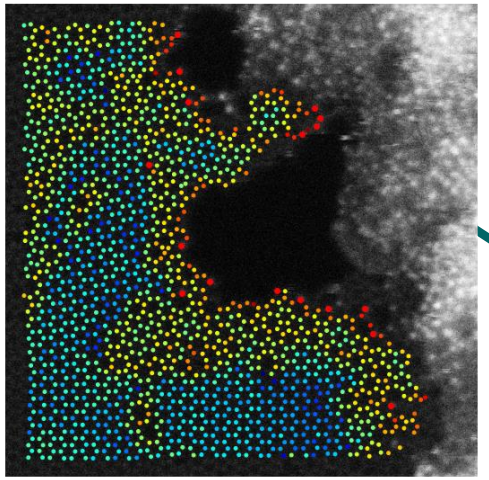
DCNN decoded



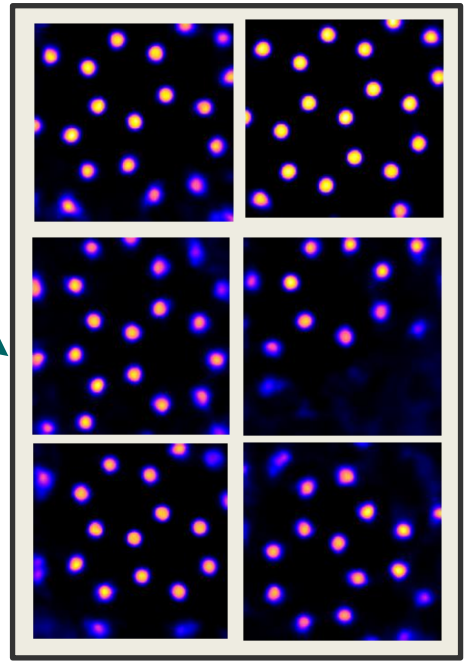
Latent space of srVAE



Latent labels



I. Molecules

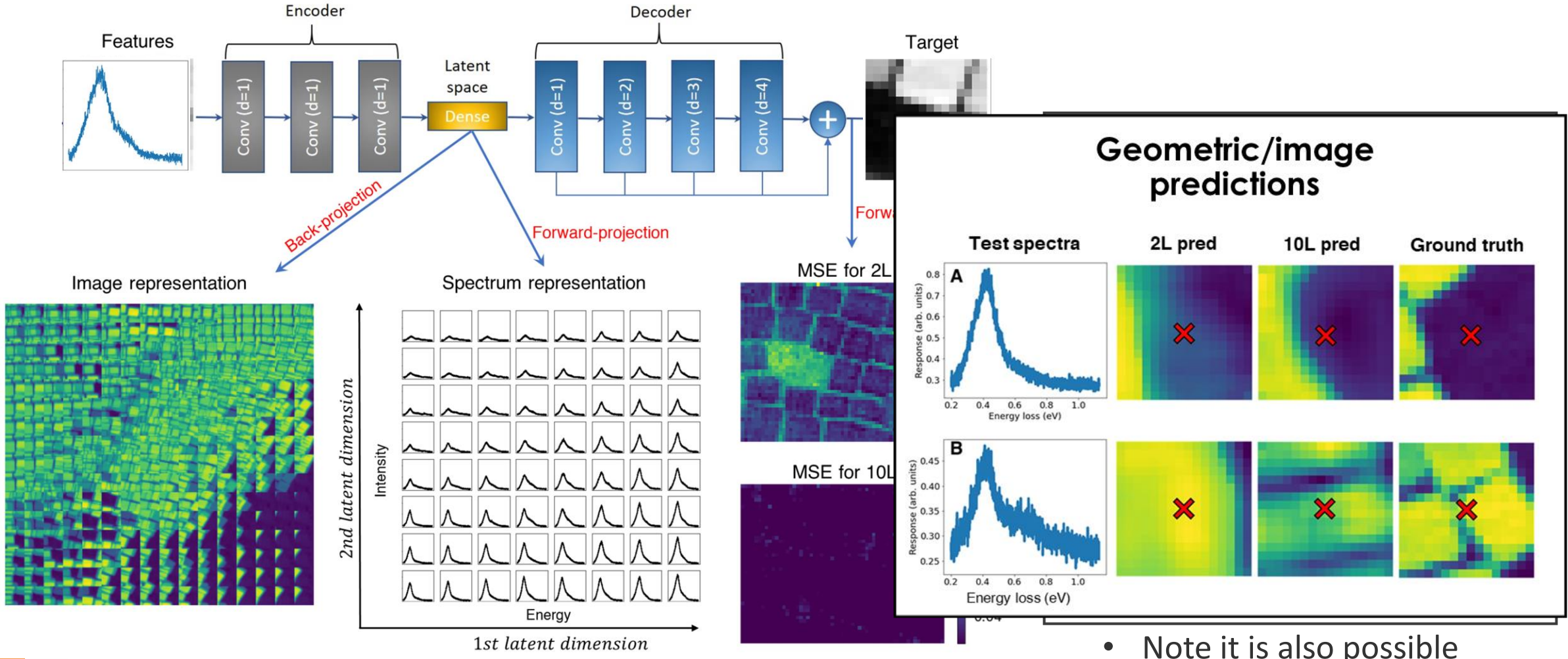


II. Reaction networks

A -> B -> C

These look like textbook organic molecular fragments…
Except that unsupervised DCNNs do not know organic chemistry.

Predicting responses



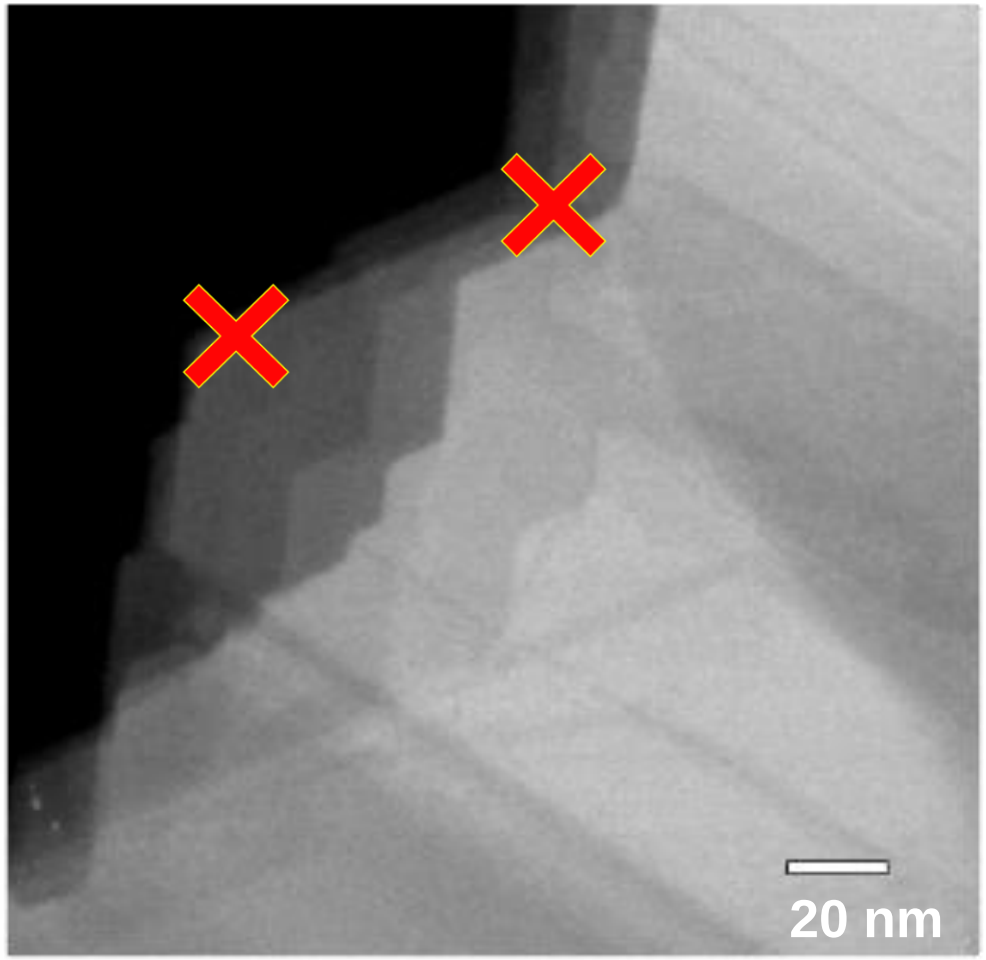
Define **# of latent dimensions** to represent the data in a low dimensional space.

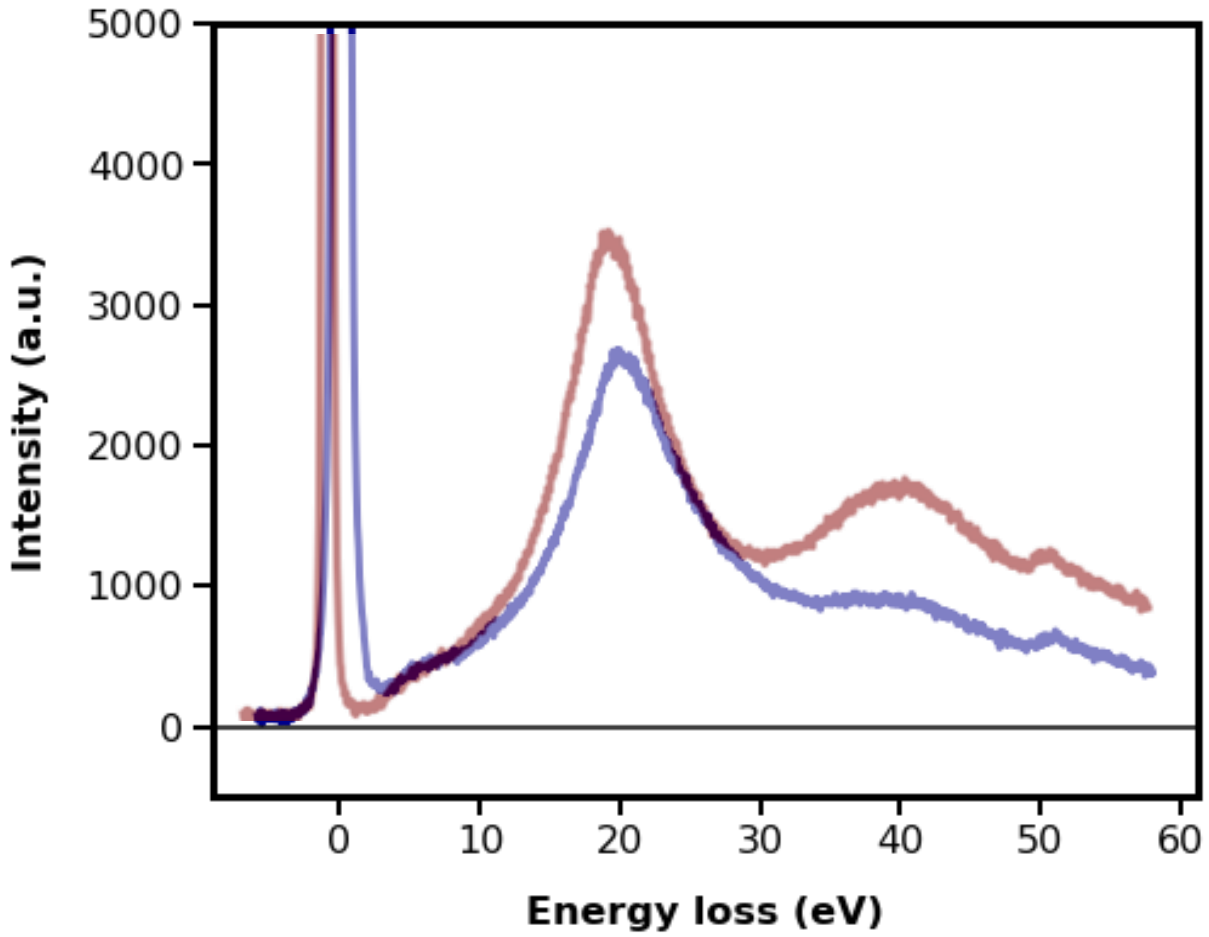
- Note it is also possible to do image prediction from spectrum (**reverse**)

Automated Experiment:

… as a scientist…

Can ML run experiment as a scientist?





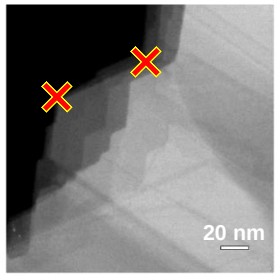
1. What if we have full access to structural information
2. And want to choose locations for (EELS, 4D STEM, CL, EDX) measurements
3. So as to **learn** relationship between structure and spectrum fastest
4. Or **discover** which microstructural elements give rise to specific **desired** spectral features?

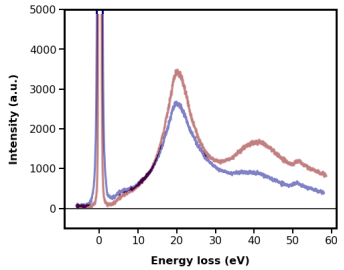
Physics-based feature engineering:

Deep kernel learning – Bayesian optimization

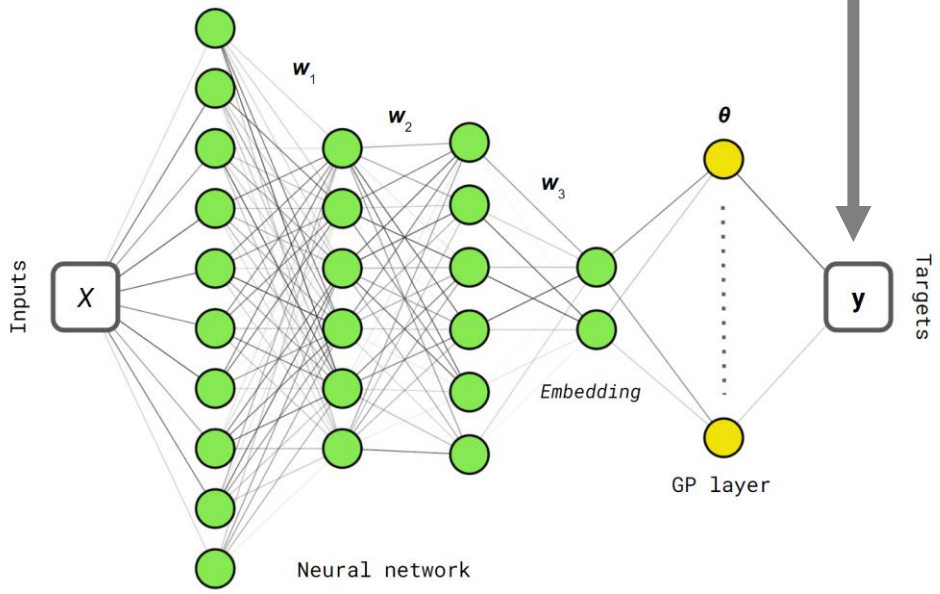
Specify physics criteria

Active learning





Scalarizer



Acquire **structural data**

Measure a **spectrum**

Train DKL model with new data

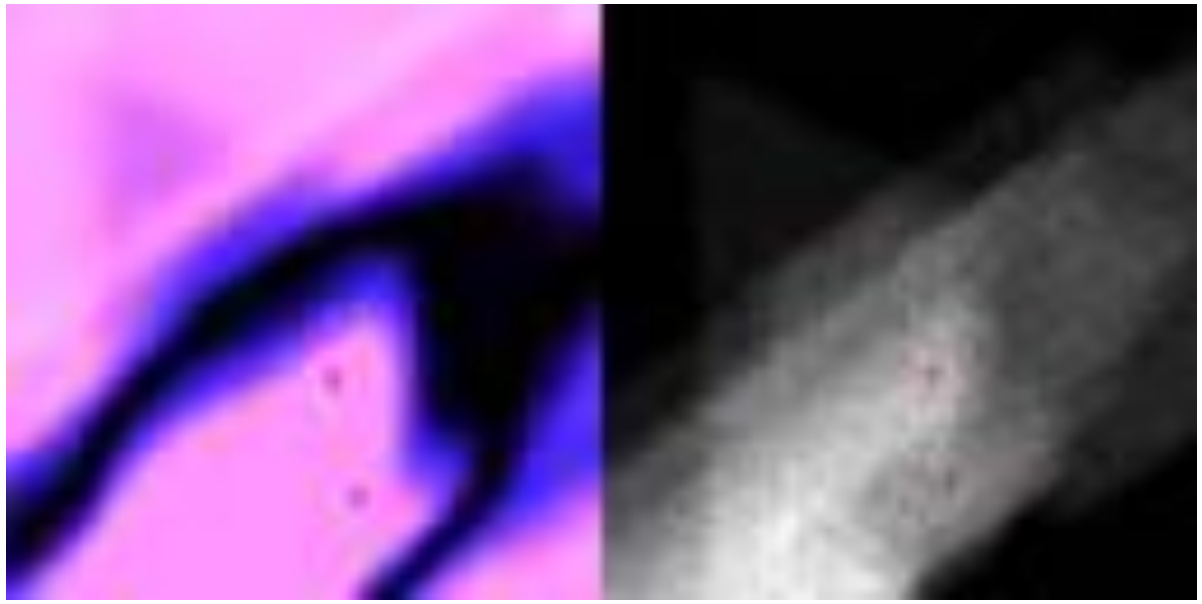
Decide next position (optimize physics criteria)

Allows navigation of the system to search for physics

Discovering Regions with Interesting Physics

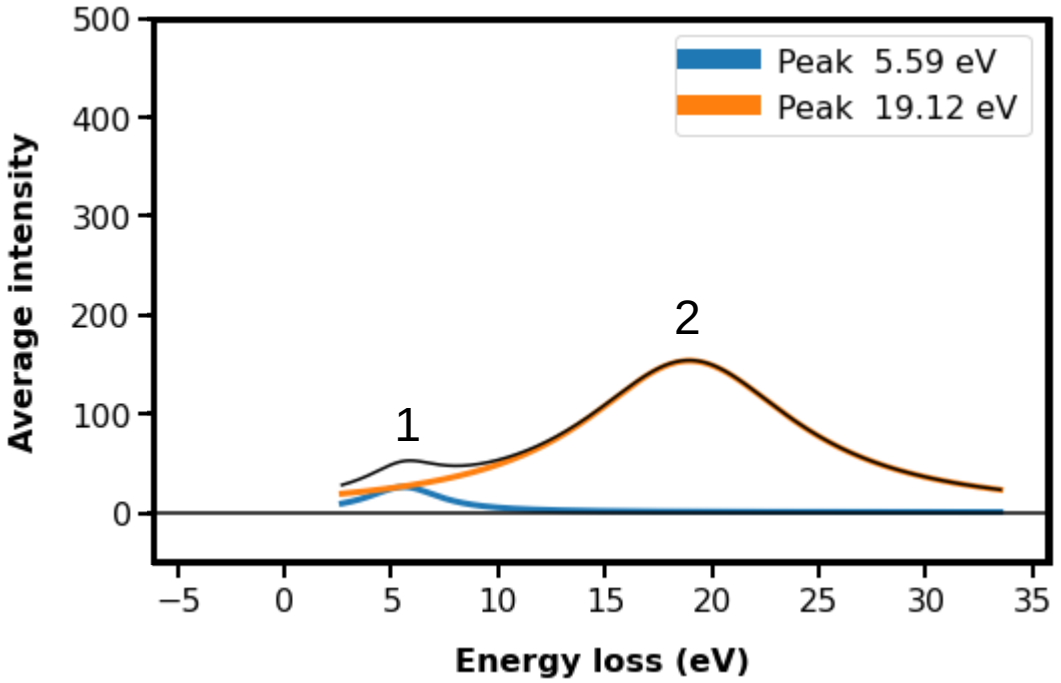
- **Discovering physics in a "new" material $MnPS_3$**
- **Curve fitting** to help enforce physical processes

"Acquisition function" HAADF-STEM



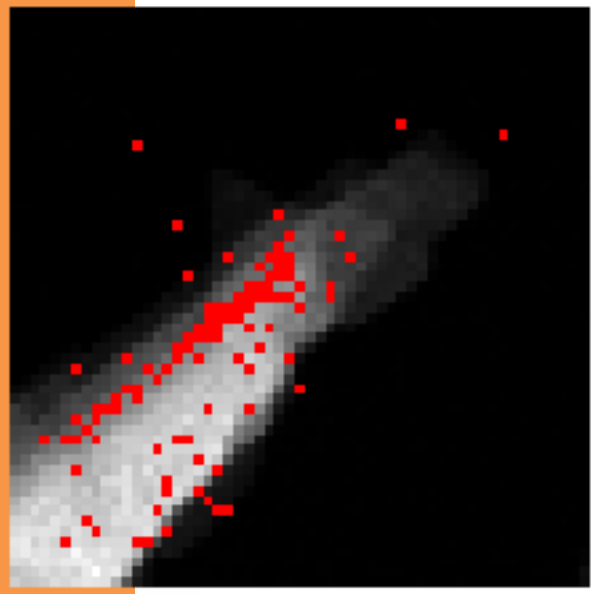
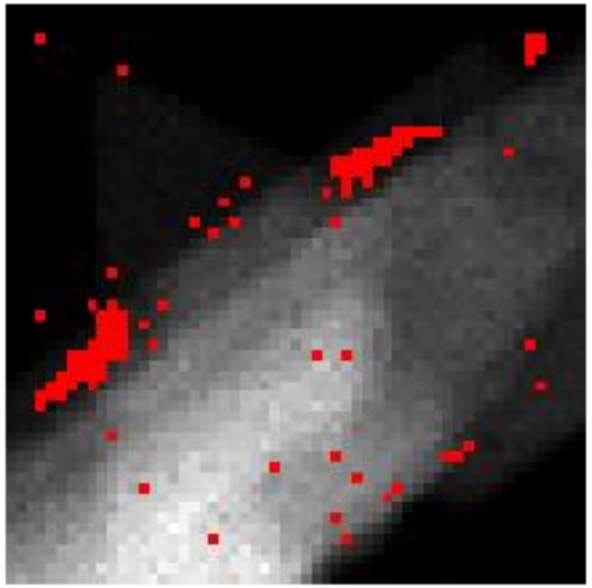
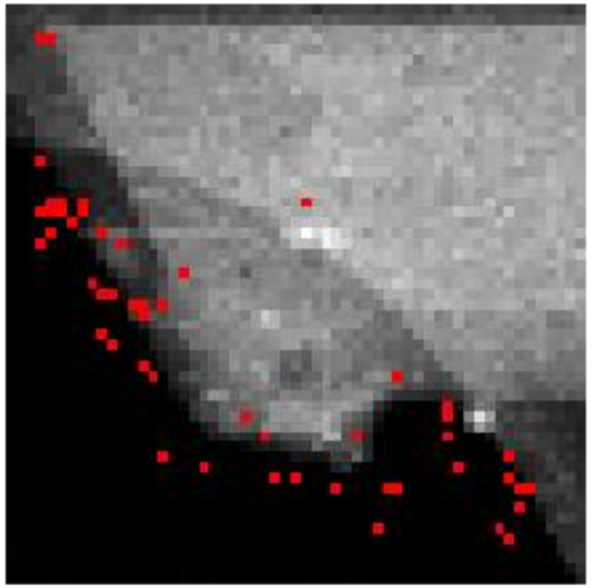
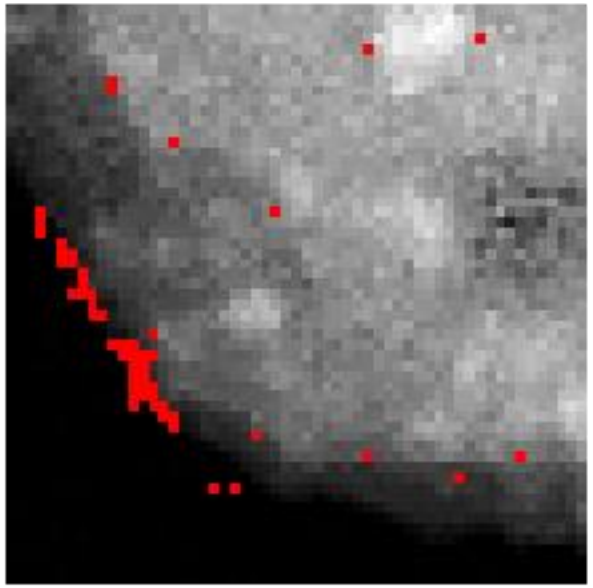
Physics search criteria:

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



More Examples of Physics Discovery

- Very similar behavior when searching for the same criteria!
- Success!

Discovery pathway depends on the reward structure (scalarizer that defines signature of physics we want to discover)!

Changing the Criterion

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

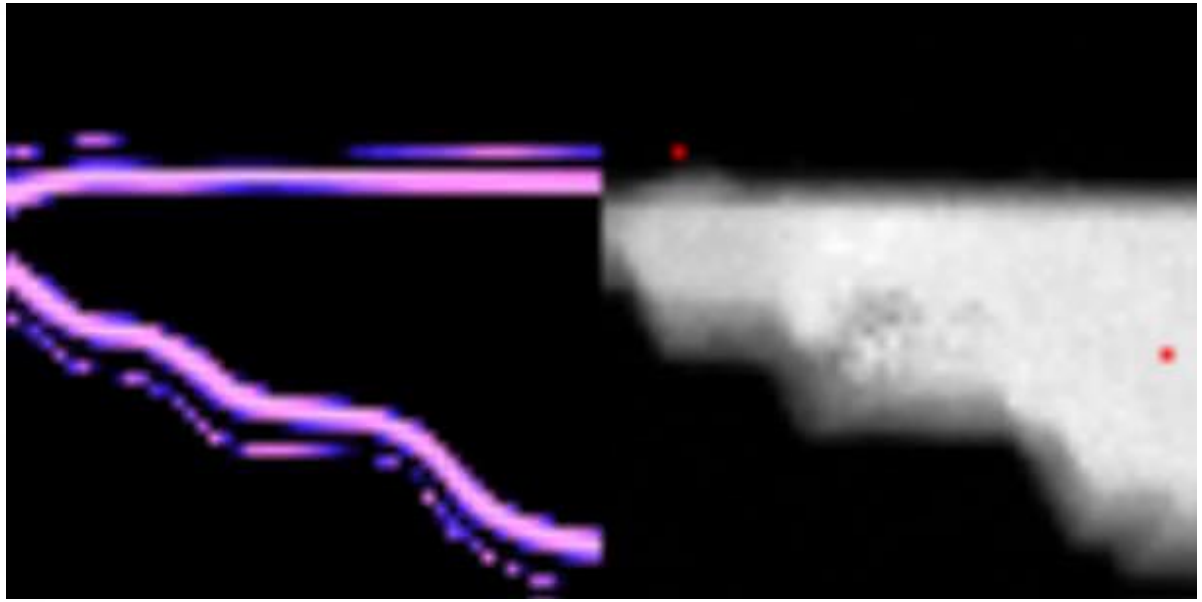
Physics search criteria:

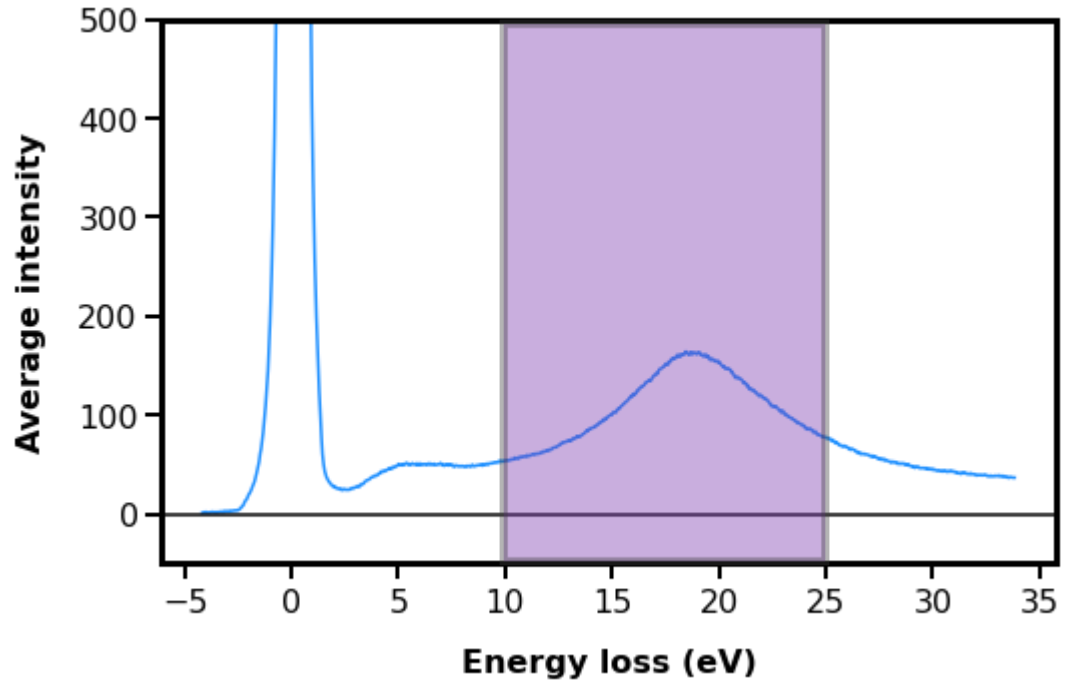
$$\boldsymbol{Maximize(f)}$$

(Specific peak intensity)

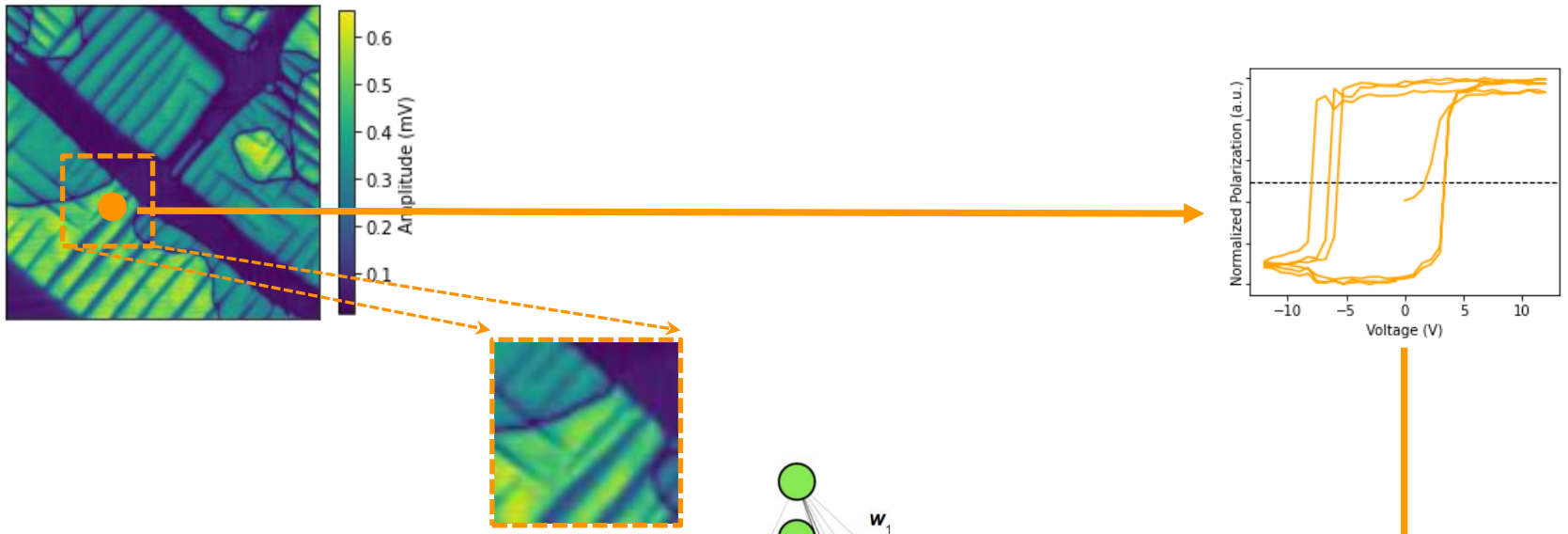
"Acquisition function"

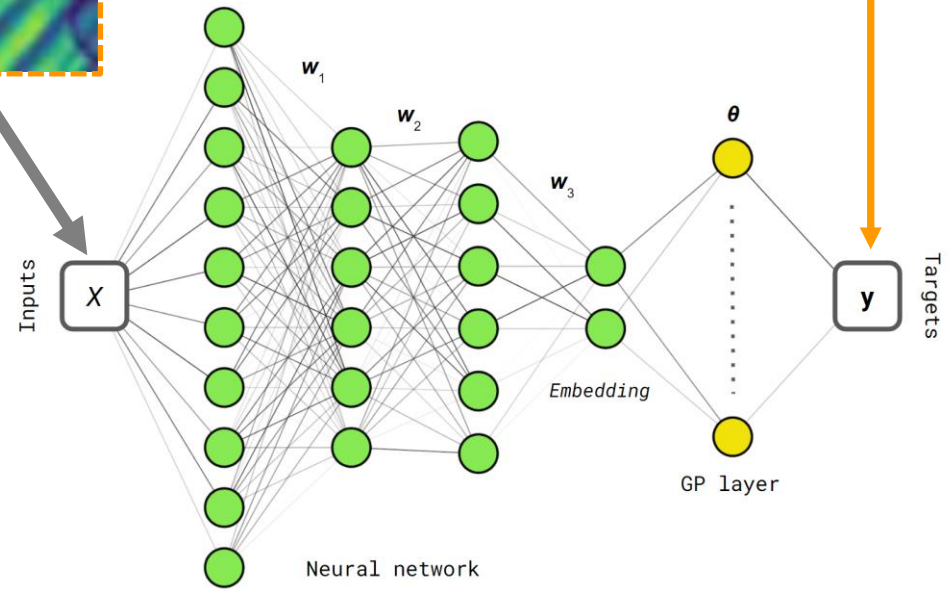
HAADF-STEM
+ points visited





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

Specify physics criteria

Acquire **structural data**

↓

Measure a **spectrum**

↓

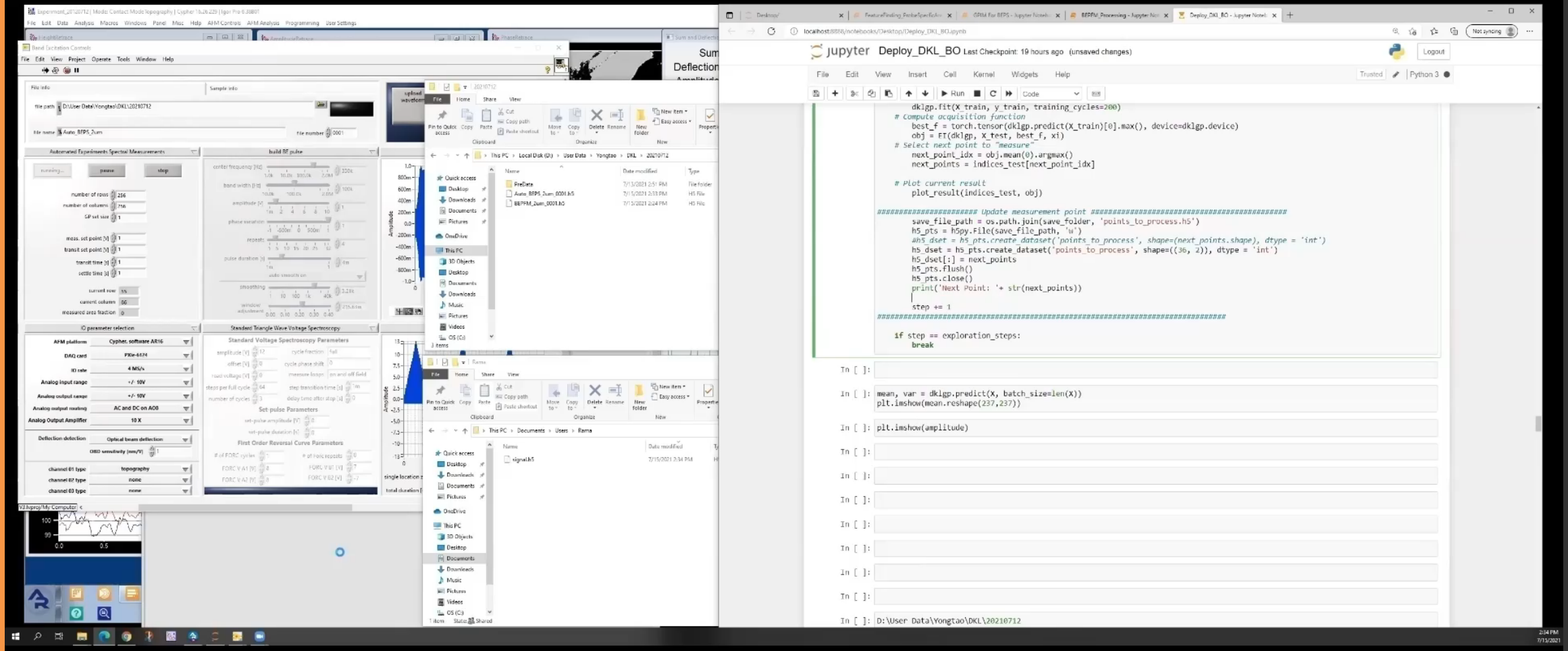
Train DKL model with new data

↓

Decide next position (optimize physics criteria)

Allows navigation of the system to search for physics

Deep Kernel Learning AE

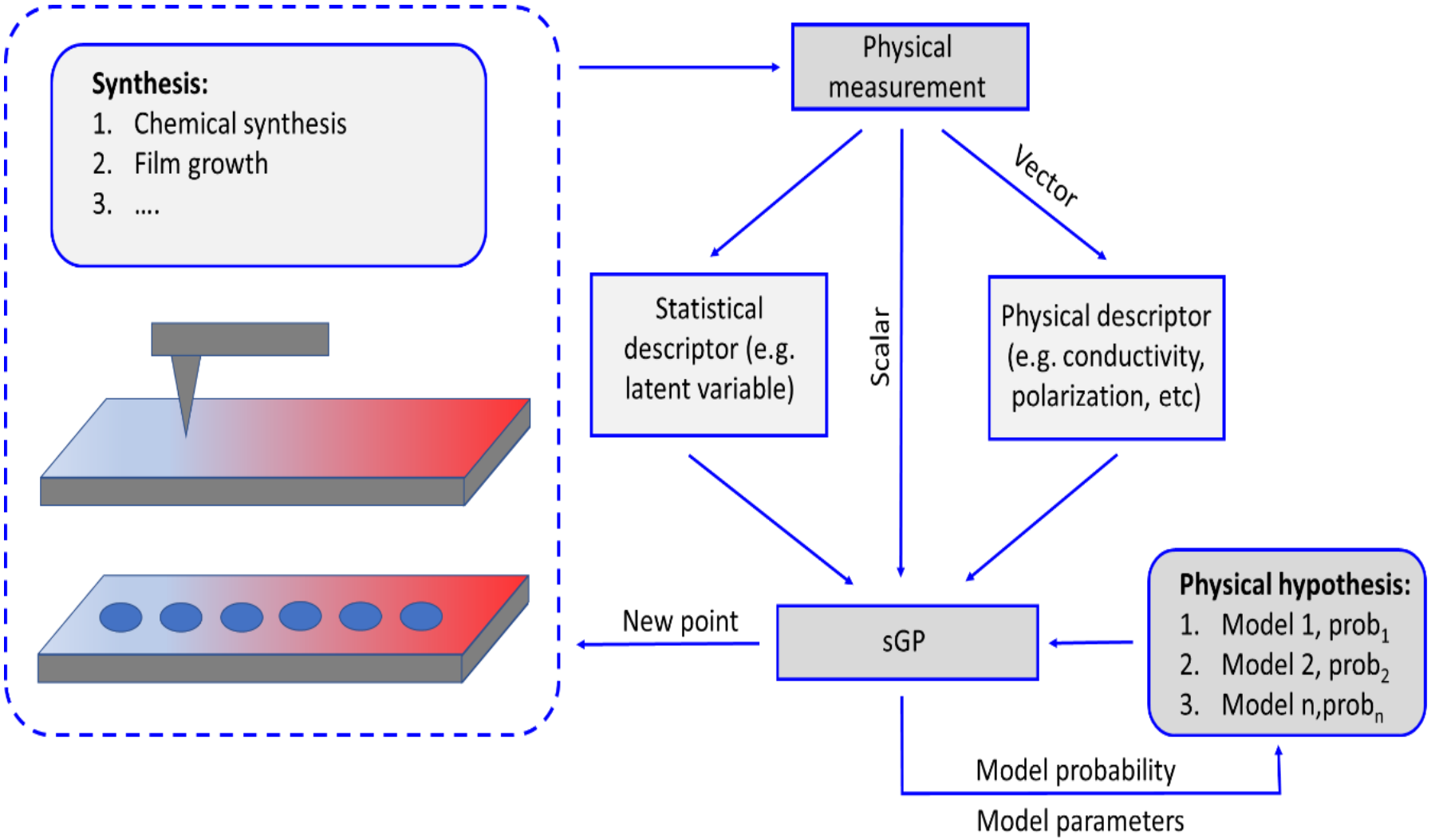


Automated Experiment:
… as a scientist…

Bayesian optimization:

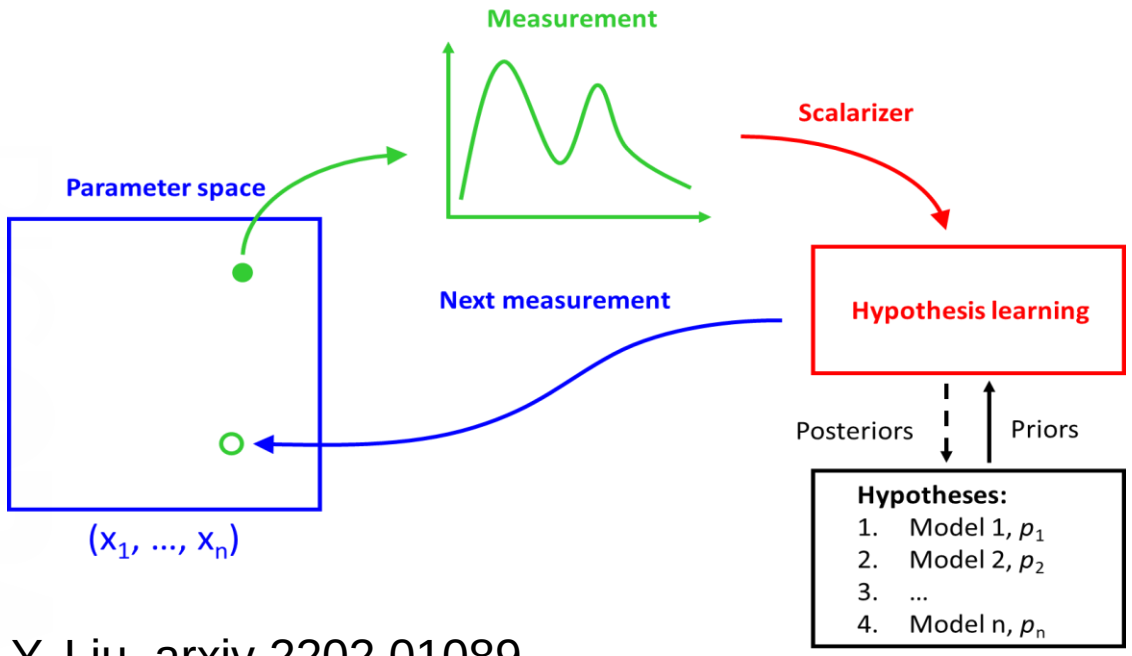
1. Works only in low-dimensional spaces
2. The correlations are defined by the kernel function (very limiting)
3. We do not use any knowledge about physics of the system
4. We do not use cheap information available during the experiment (proxies)

Hypothesis Active Learning

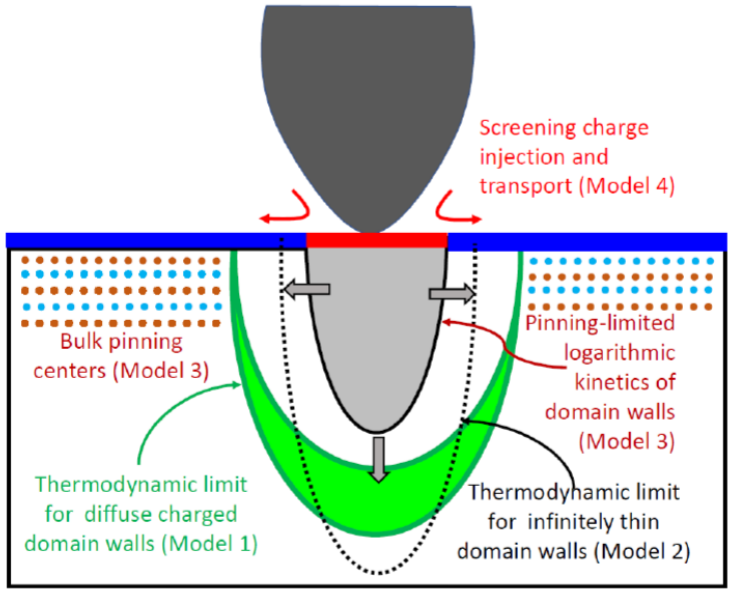


Hypothesis Learning

- Can ML algorithm think like a scientist?
- Yes – automated experiment can pursue hypothesis-driven science!



Y. Liu, arxiv 2202.01089
Y. Liu, arxiv 2112.06649



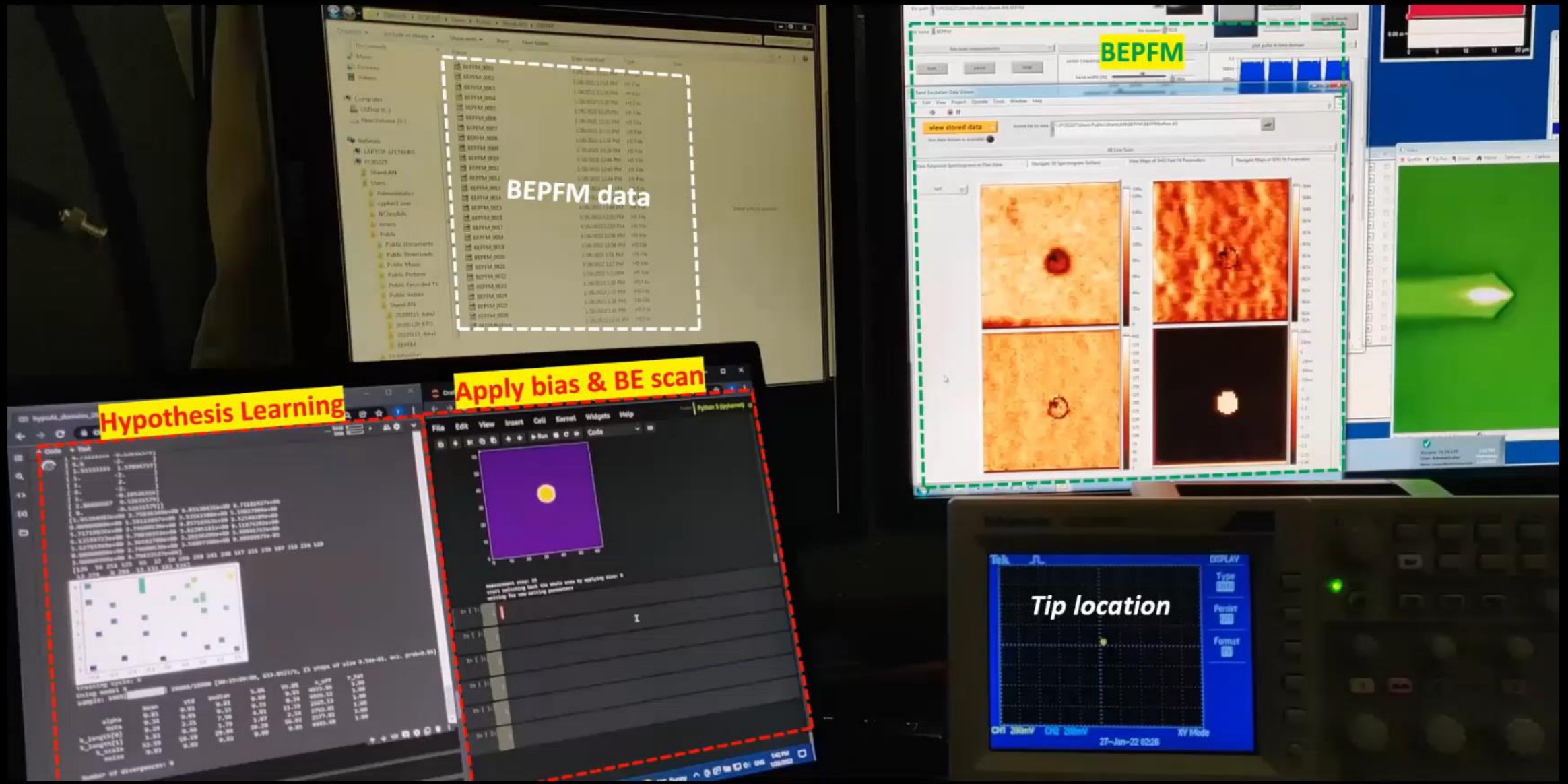
Thermodynamic 1

Thermodynamic 2

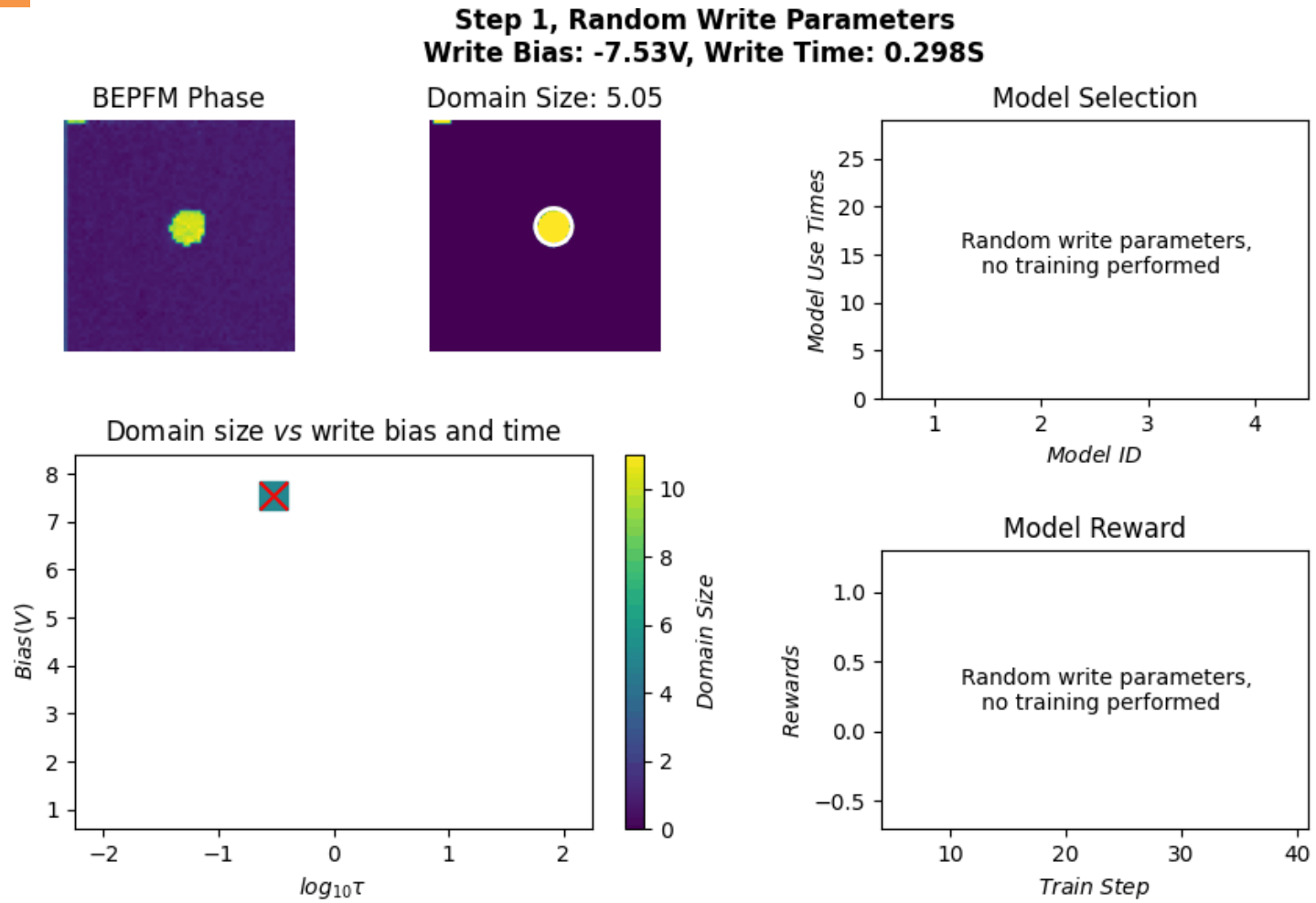
Wall pinning

Charge injection

	Model Equation
Model I	$r(V) = r_{cr} + r_0\sqrt{\left(\frac{V}{V_c}\right)^{2/3} - 1}$
Model II	$r(V) = r_{cr} + r_0\sqrt[3]{\left(\frac{V}{V_c}\right)^{2} - 1}$
Model III	$r(V,t) = V^{\alpha}\log\tau$
Model IV	$r(V,t) = V^{\alpha}\tau^{\beta}$


BEPFM data
BEPFM
Hypothesis Learning
Apply bias & BE scan
Tip location

Hypothesis learning in action

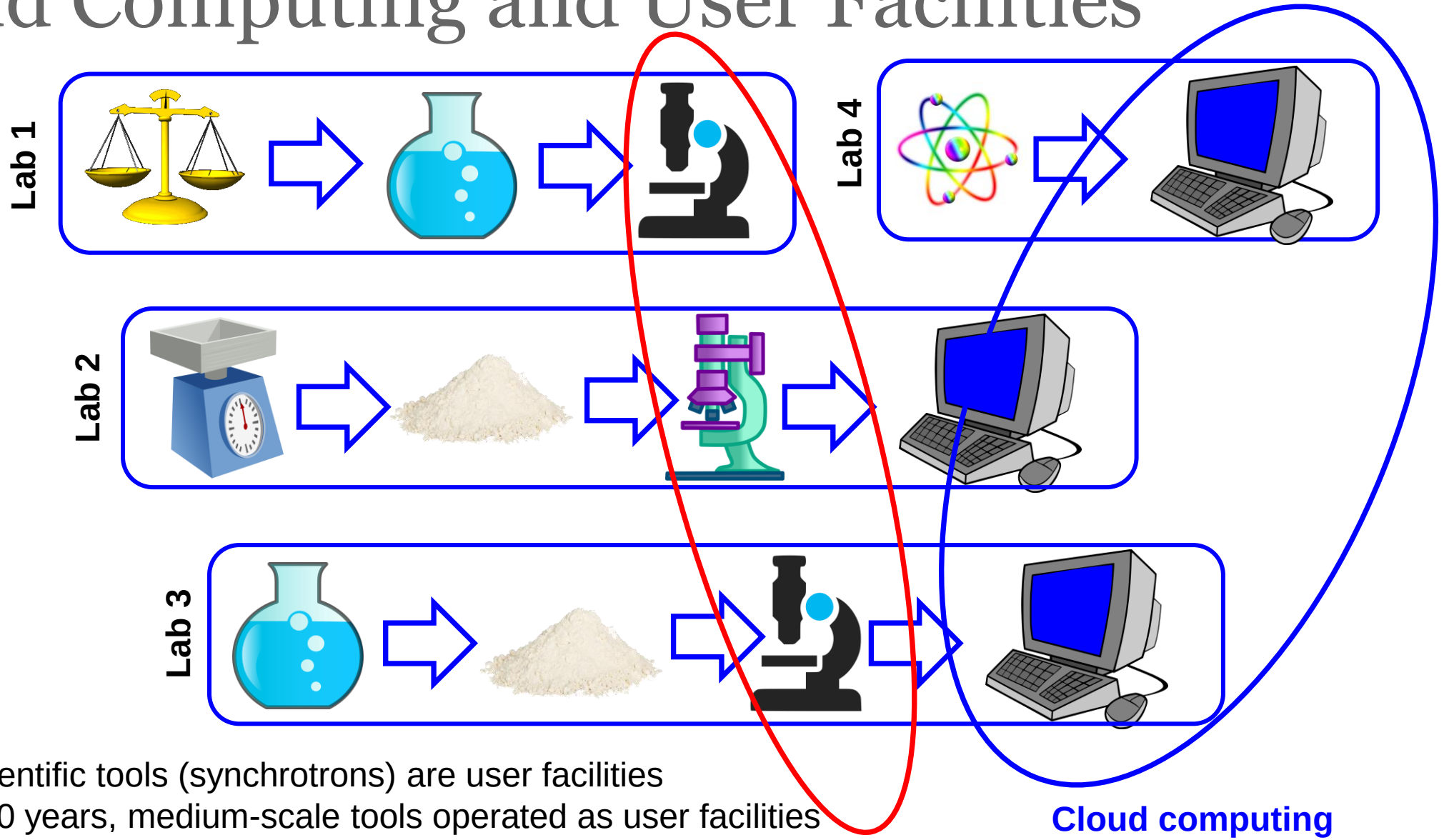


- ML algorithm has 4 competing hypothesis on domain switching mechanisms
- These hypothesis represent full set of possibilities for this system
- The microscope chooses experimental parameters in such a way as to establish which hypothesis is correct fastest
- Important: the same approach can be implemented in synthesis and electrical characterization
- Machine learning meets hypothesis-driven scientific discovery!

Y. Liu, arxiv 2202.01089
Y. Liu, arxiv 2112.06649

Future of Instrumentation

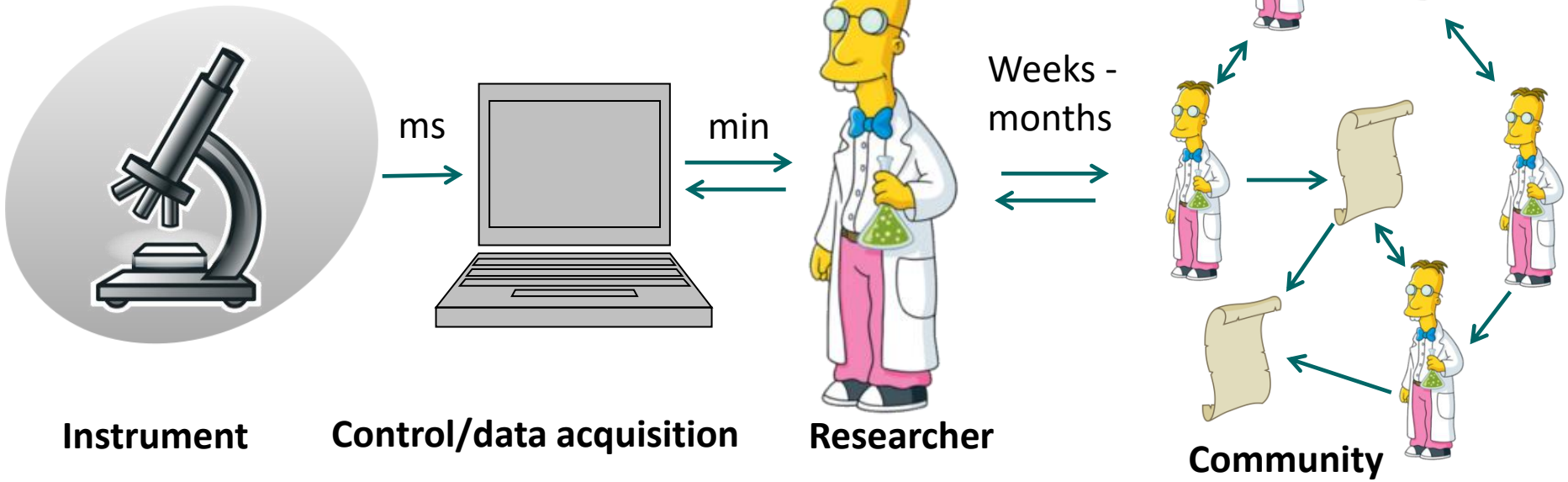
Cloud Computing and User Facilities



- Big scientific tools (synchrotrons) are user facilities
- For ~20 years, medium-scale tools operated as user facilities
- Enterprise computing -> cloud computing
- Over last 5 years, cloud labs are emerging
- What about the workflows?

Classical Instrumental Research

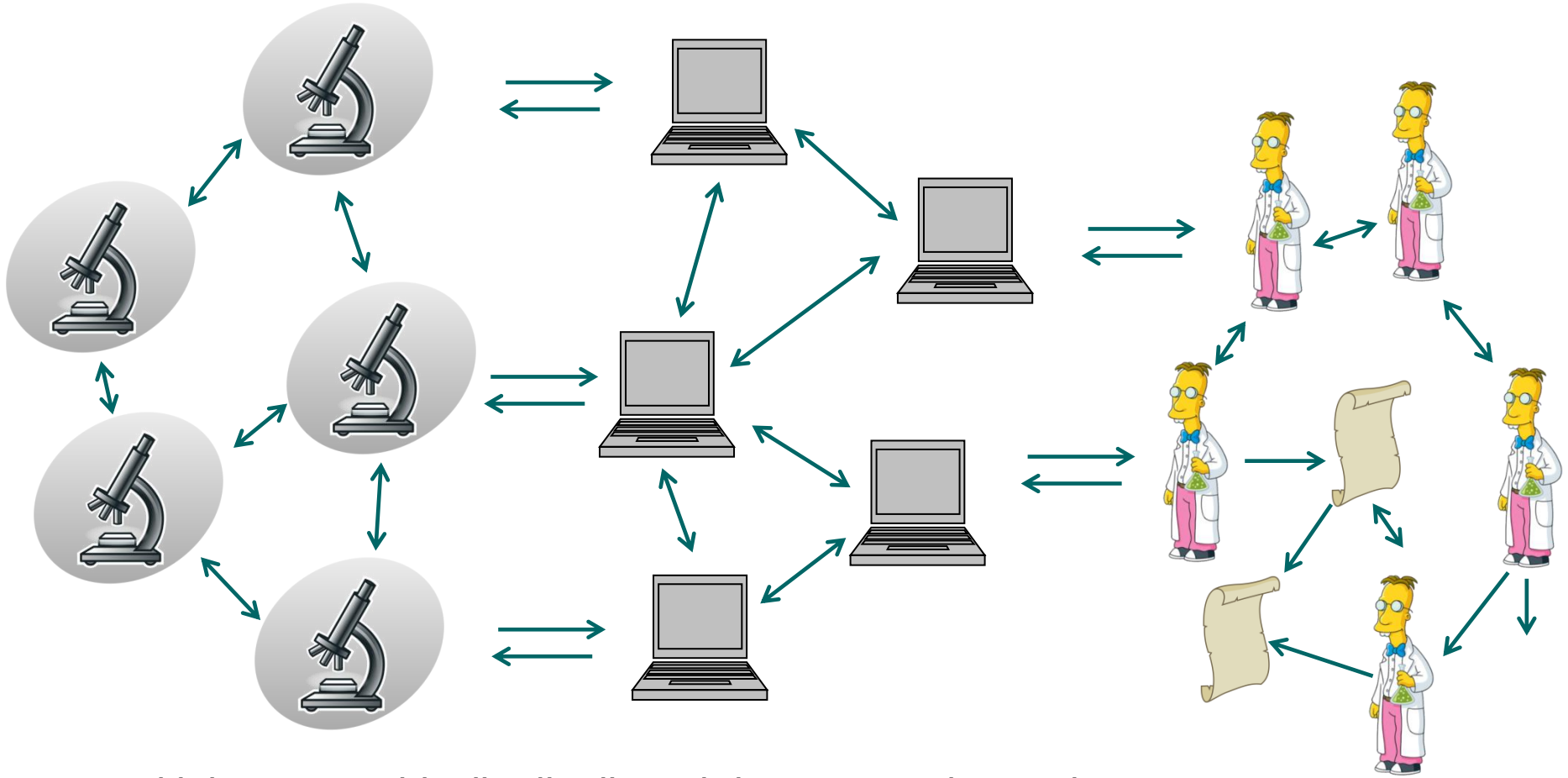
SPM: 30,000+ platforms worldwide:
Large weakly connected instrumental network
(S)TEM: ~100s top level machines,
much stronger integrated community



- Social networking/education
- Publications/citations

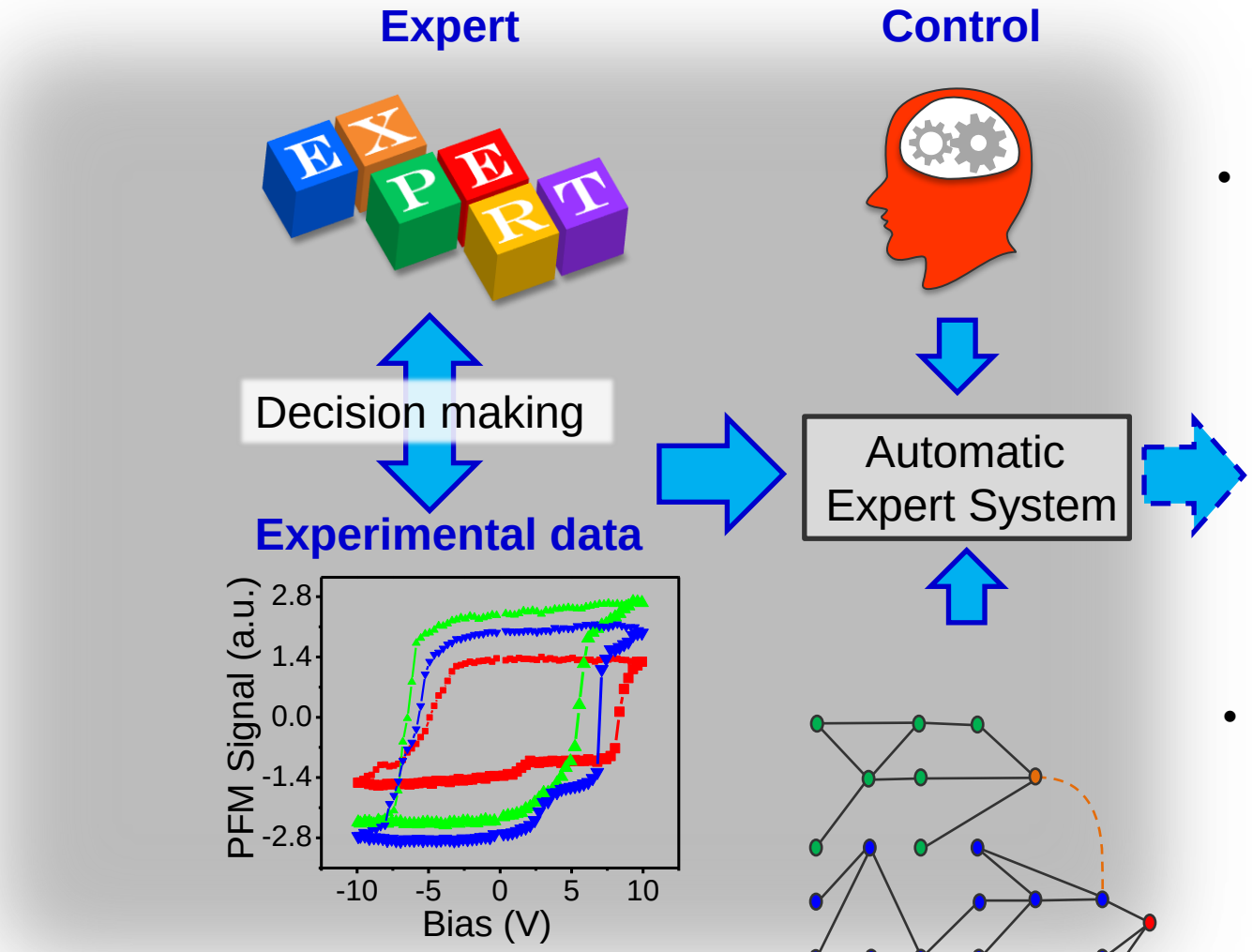
1. Only small fraction of data stream from the instrumentation is captured
2. Only small fraction of captured data is analyzed, interpreted, and put in the context
3. Human-machine interaction during acquisition is often slow and can be non-optimal
4. Human interpretation of data is limited: bias and ignoring serendipity
5. Information propagation and concept evolution in scientific community is slow and affected by non-scientific factors

Step 1: Cloud Integration



1. Multiple geographically-distributed data generation node
2. Full capture of instrumental data stream /compression/curation
3. Coordination of protocols and data/metadata across the cloud
4. Cloud-based processing and dimensionality reduction
5. Community-wide analytics

Step 2: Cloud Analytics

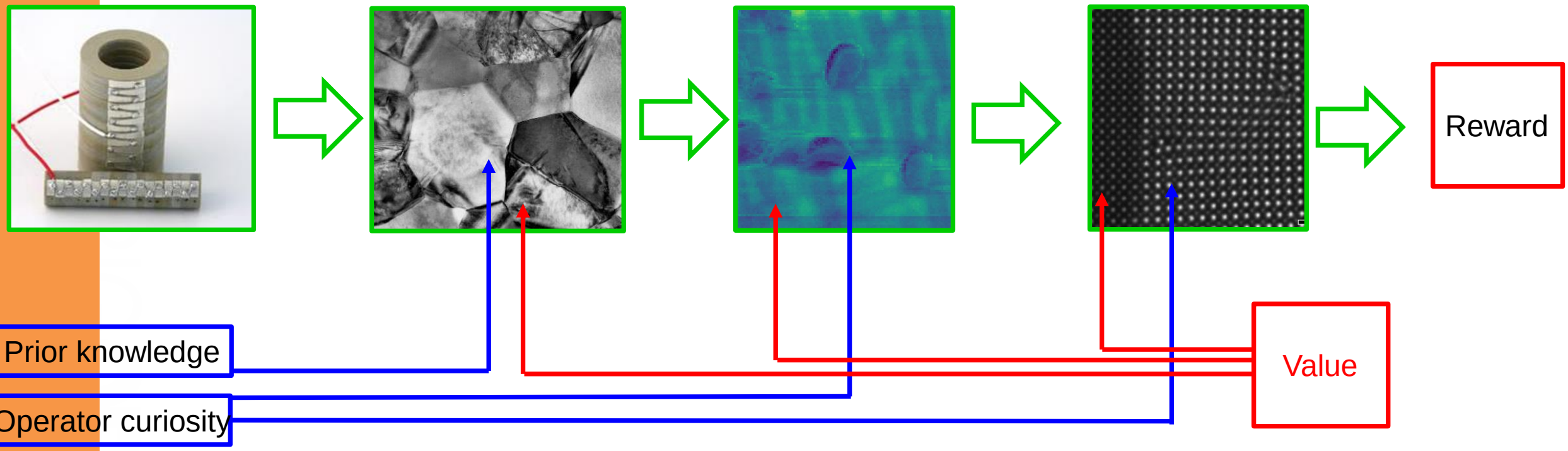


- **Synthesis of expertise:** factor in human expert knowledge
- **Context search:** published results data mining/social networks

Future:

- Automated analysis of routine data
- Identification of anomalies
- Initial training of new practitioners
- Data centers: information based on knowledge

Step 3: Workflow Design



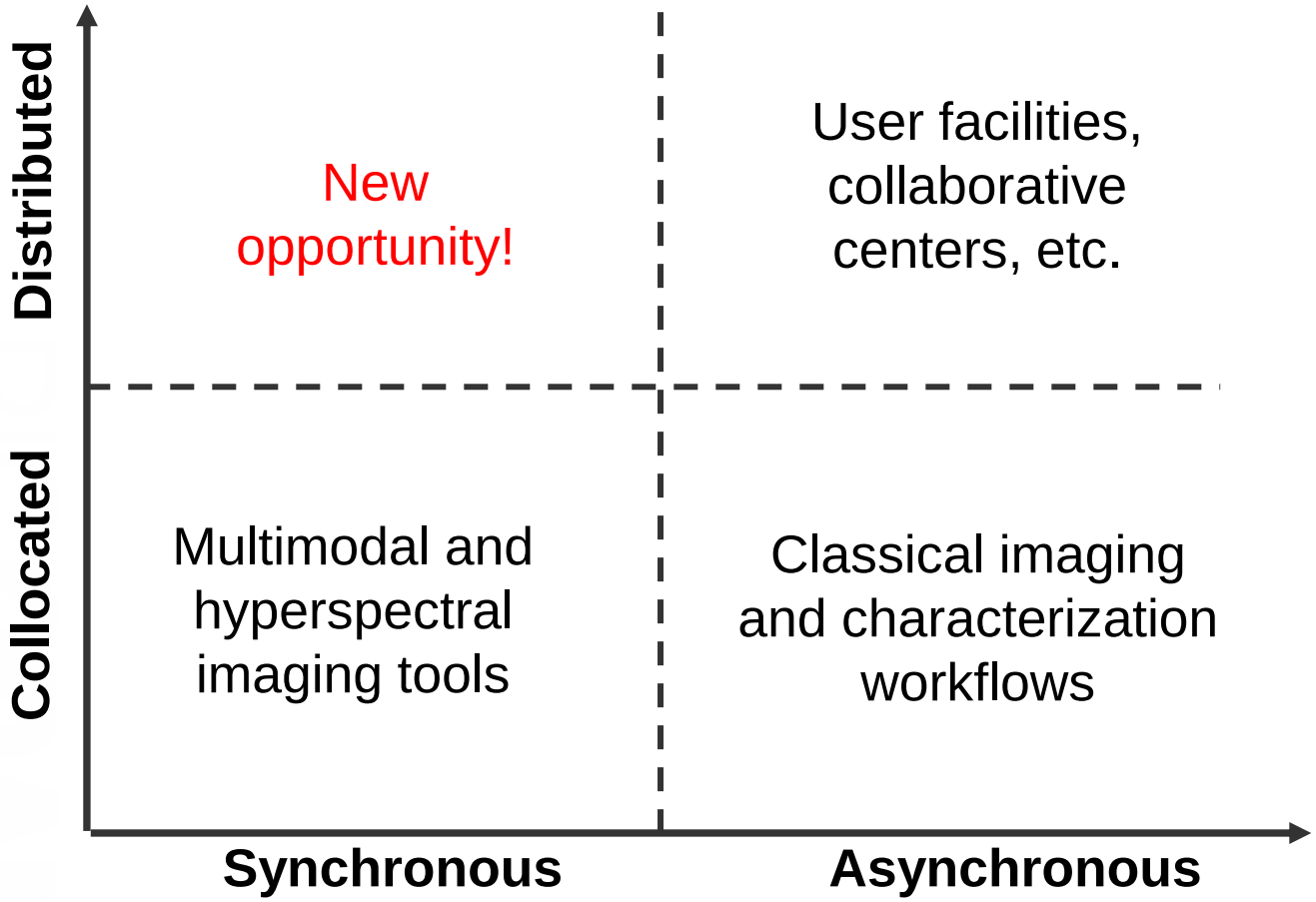
Traditional experiment:

1. Always based on workflows
2. Ideated, orchestrated, and implemented by humans
3. The “gain of value” during the workflow implementation is uncertain

Value of the step is key element:

- Either based on prior knowledge
- Or defined in a sense of the reinforcement learning Q-function

Cloud Labs: Facilities of the Future



Emerald Cloud Lab, SF and CMU

1. Combined human-machine workflow implementation
2. Computer orchestrating agent
3. How would beyond human workflows be ideated?

And technology

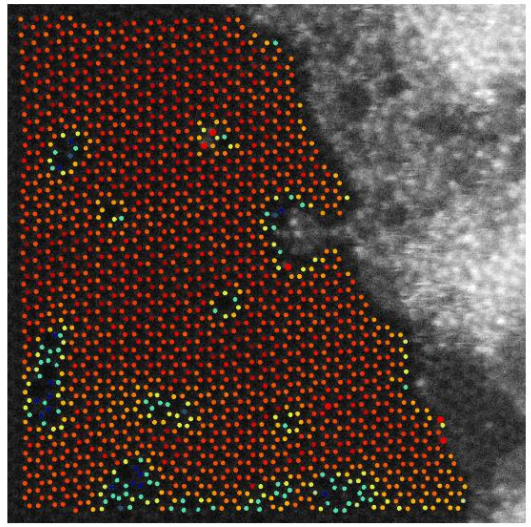
"New directions in science are launched by new tools much more often than by new concepts. The effect of a concept-driven revolution is to explain old things in new ways. The effect of a tool-driven revolution is to discover new things that have to be explained."

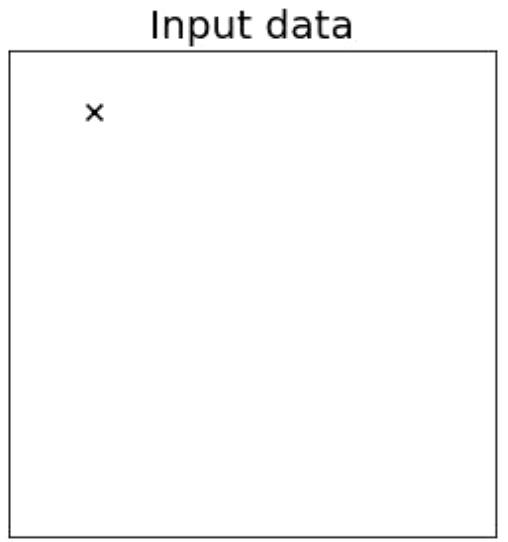
Freeman Dyson

- Cloud connection and data infrastructure is necessary, but not enough
- Can we define workflow design, building, and optimization?
- Potential to change way R&D is performed: academia, start-ups, industry

DIY ML

- **AtomAI:** comprehensive toolbox for DCNN-basec supervised exploration of STEM and SPM Data:
- **PyroVED:** building structure-property correlations and unsupervised and semi-supervised physical discovery
- **GPim:** Gaussian processing toolbox for image analytics and automated experiment
- **gpax:** hypothesis-driven structured Gaussian Processes
- **PyCroscopy:** General data formats, workflows, and image analytics





Welcome to Microscopy Project!