NREL
Transforming ENERGY

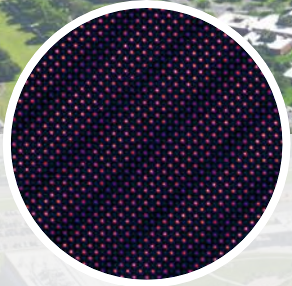
From Chaos to Clarity
Autonomous Materials Discovery for Extreme Environments

Steven R. Spurgeon
Colorado School of Mines
November 14, 2024

Carnegie Mellon → Drexel → PNNL / UW → NREL / CU

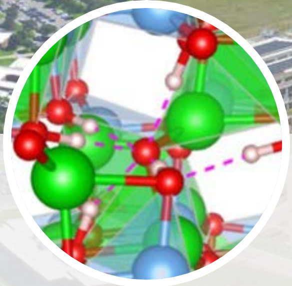
Thin Film Synthesis

Enabling devices for electronics, computing, and energy



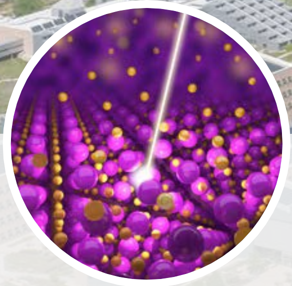
Atomic-Scale Microscopy

Understanding the fundamental building blocks of matter



Defect Engineering

Quantifying and harnessing property-defining defects



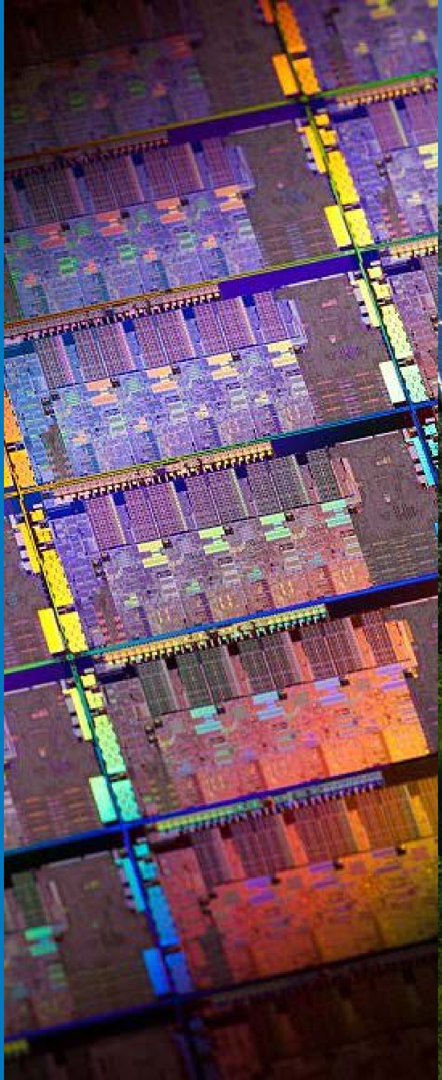
Degradation Pathways

Charting evolution in real-world extreme environments

Autonomous Science

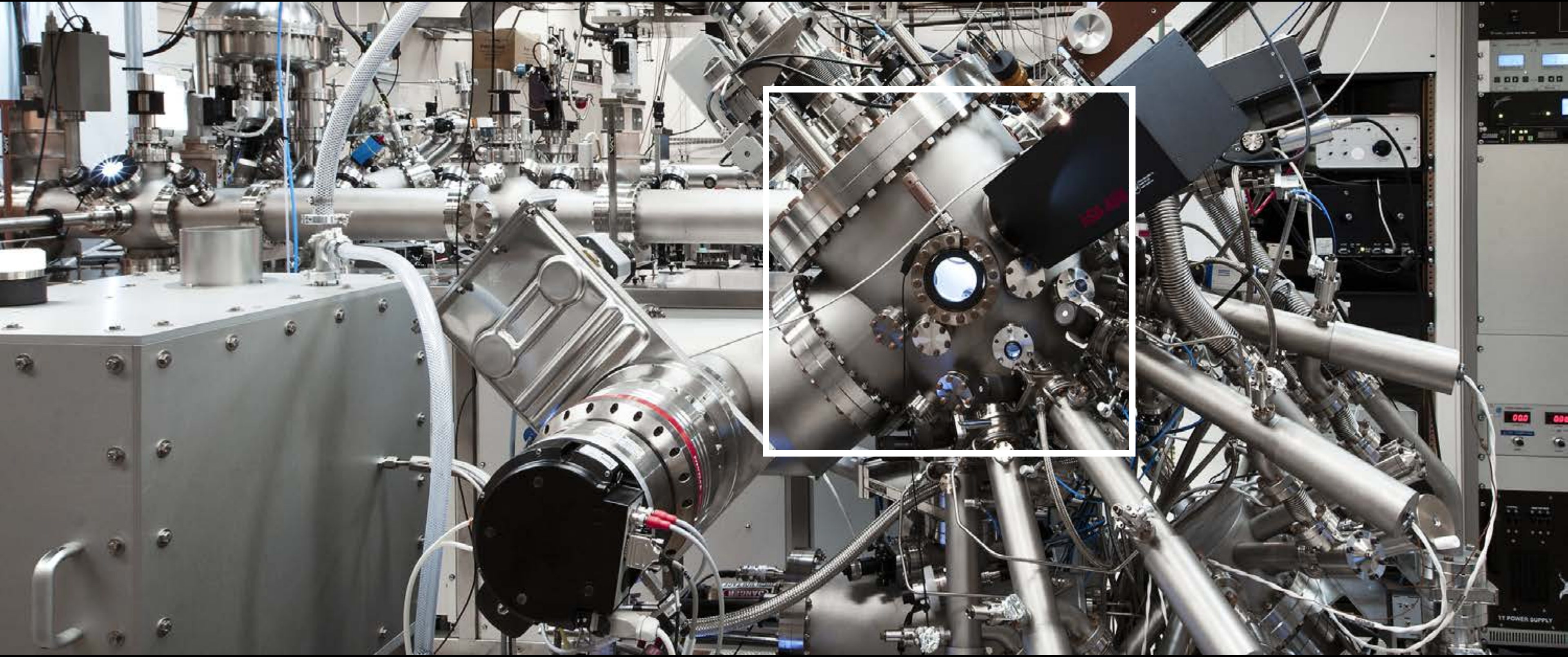
AI-accelerated materials discovery and design

Everyday technologies are critically reliant on mastering materials lifecycles



Wafer: Intel Company
PV: NREL
Battery: Battery Technology Online

Mastery depends on understanding of complex synthesis pathways…



Oxide Epitaxy Lab @ PNNL

Mastery depends on understanding of complex synthesis pathways…

Kaganer. Paul Drude Institute.

…and degradation behavior in real-world extreme environments.

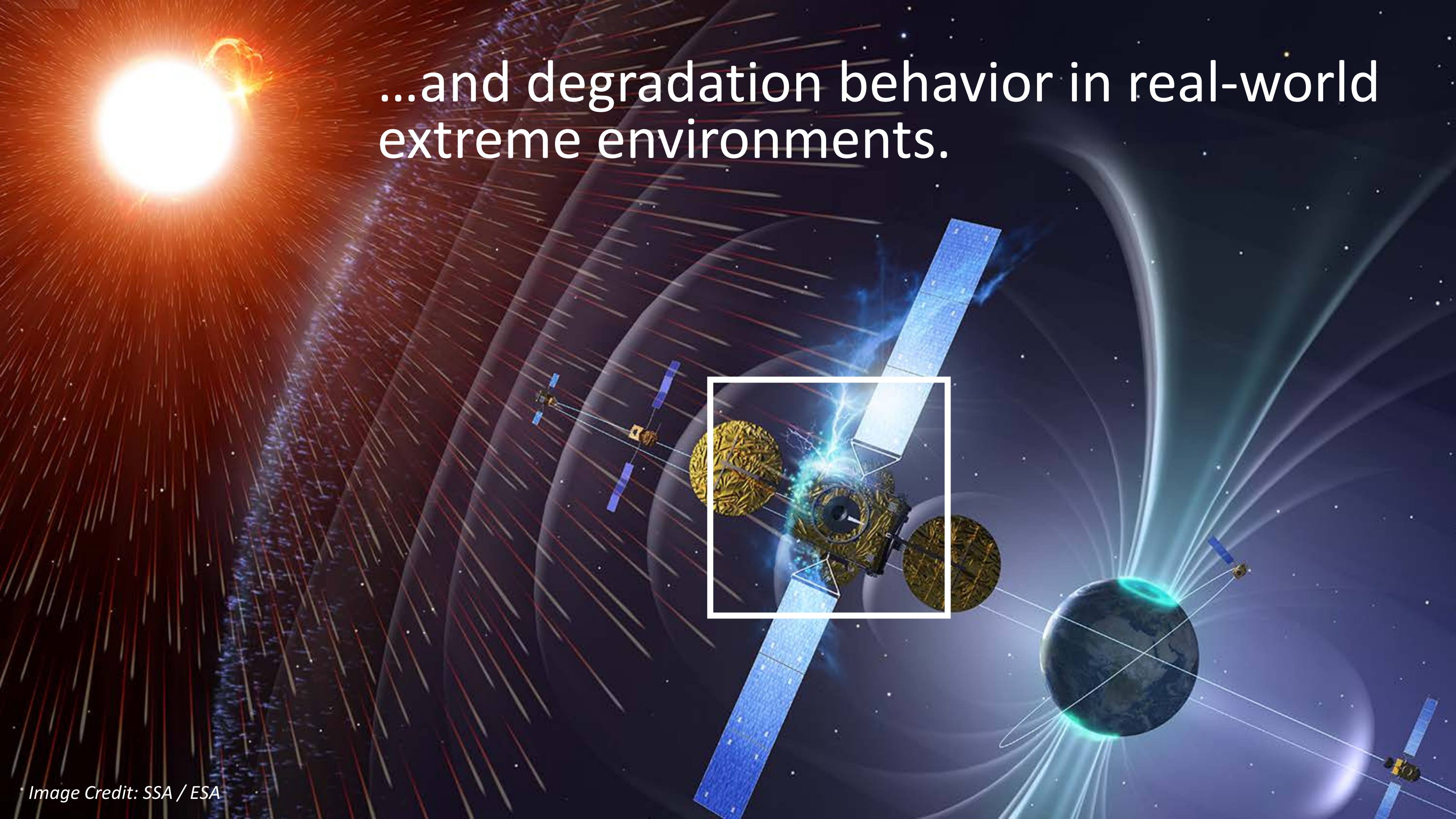
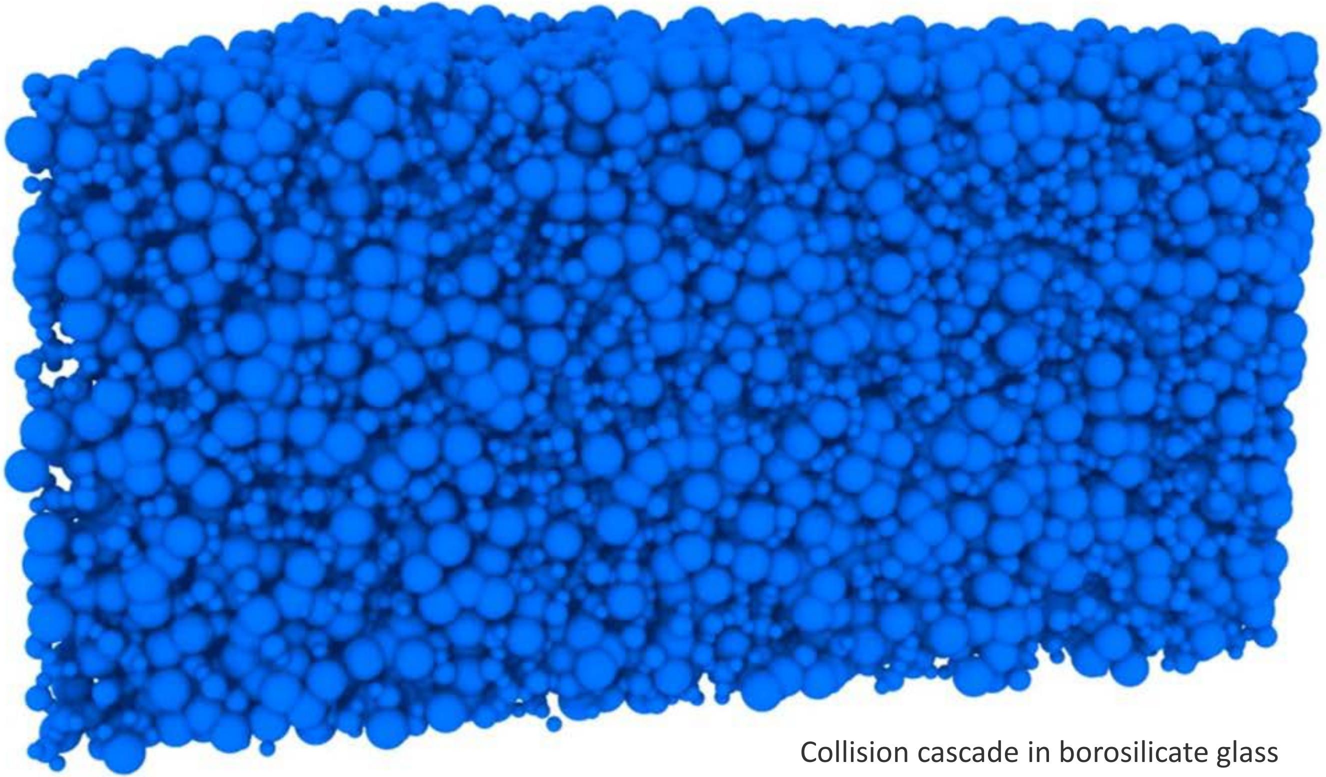


Image Credit: SSA / ESA

…and degradation behavior in real-world extreme environments.

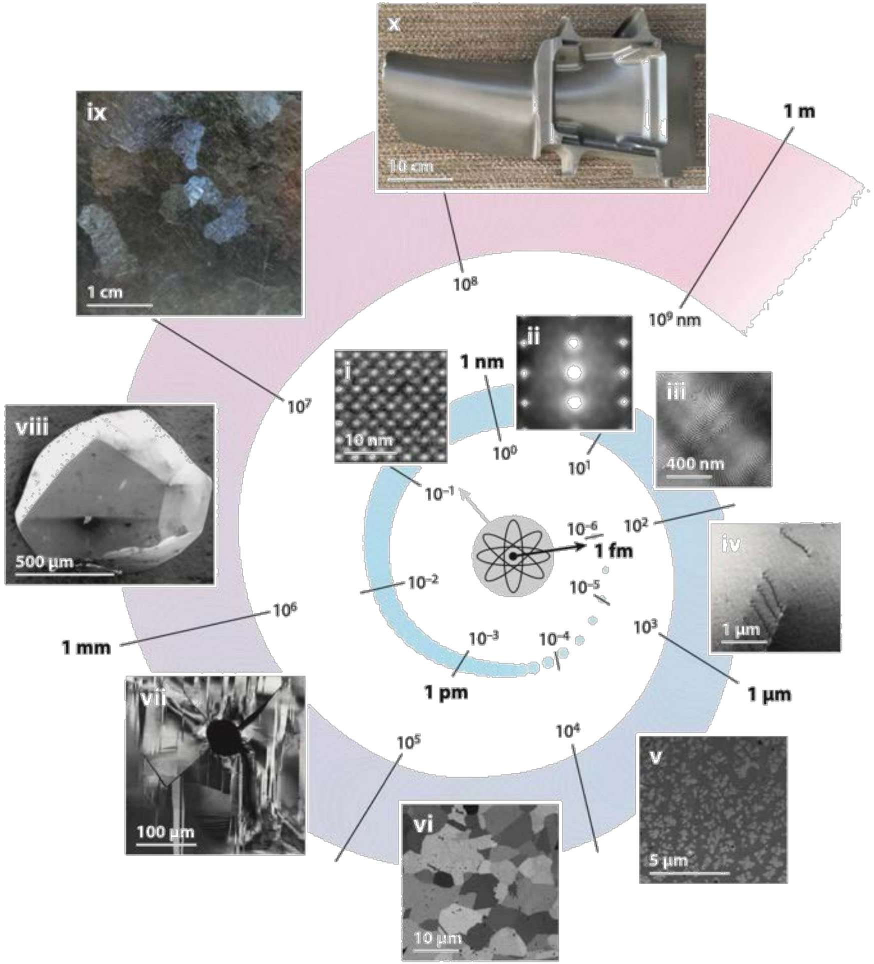


Collision cascade in borosilicate glass

D.A. Kilymis et al. J Non-Cryst Solids. 432, 354-360 (2016).

My research aim is an ontology of the materials lifecycle:

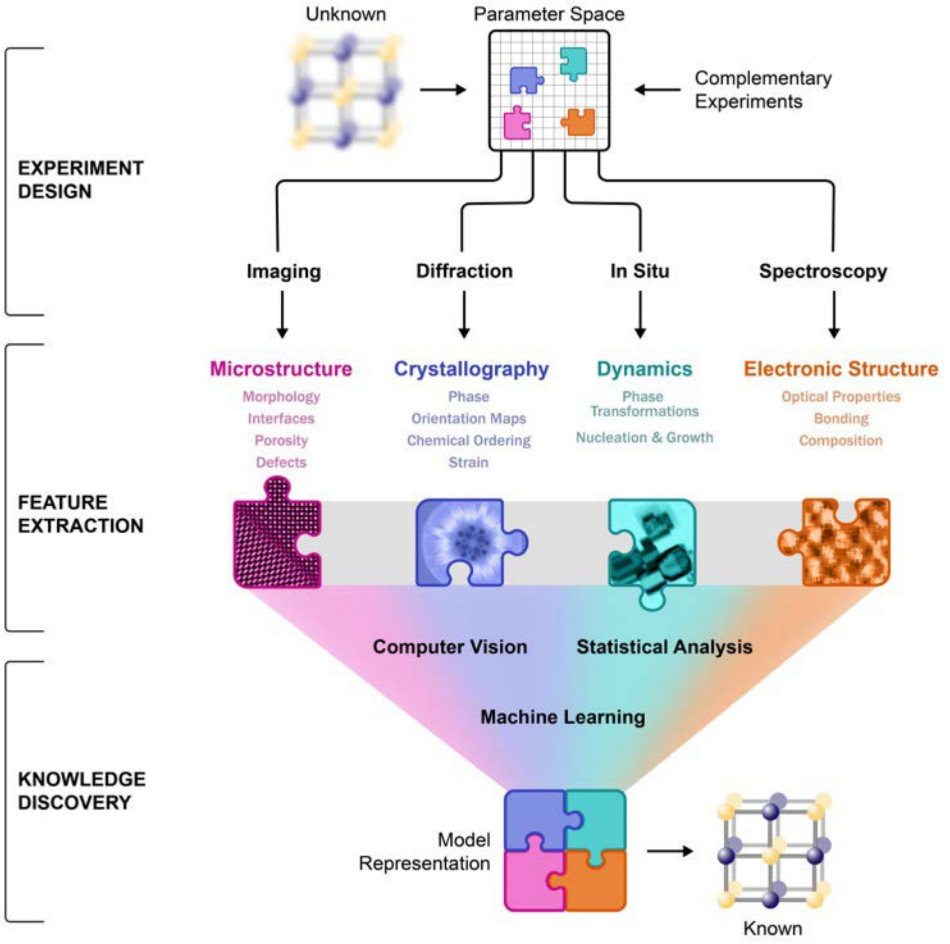
“A systematic mapping of data to meaningful semantic concepts…” ***across spatial and temporal scales***



Kalidindi et al. Ann. Rev. Mater. Res. 45: 171-193. (2015).

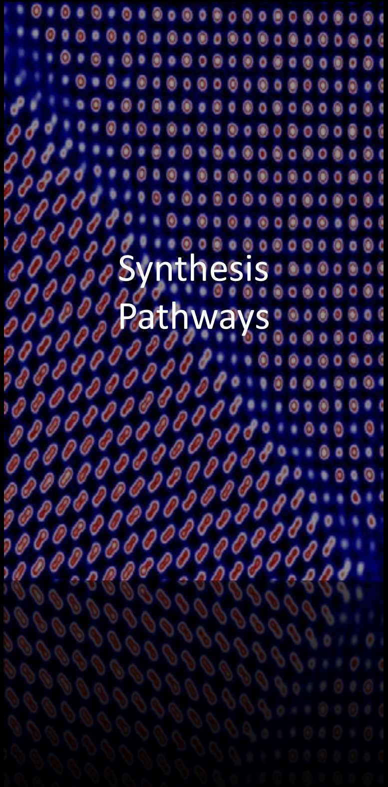
Quote adapted from: https://blog.palantir.com/ontology-finding-meaning-in-data-palantir-rfx-blog-series-1-399bd1a5971b

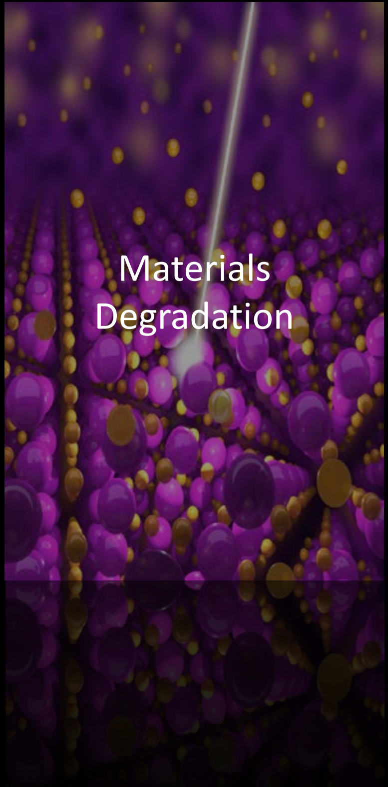
Autonomous science plays an important role in developing and harnessing this ontology

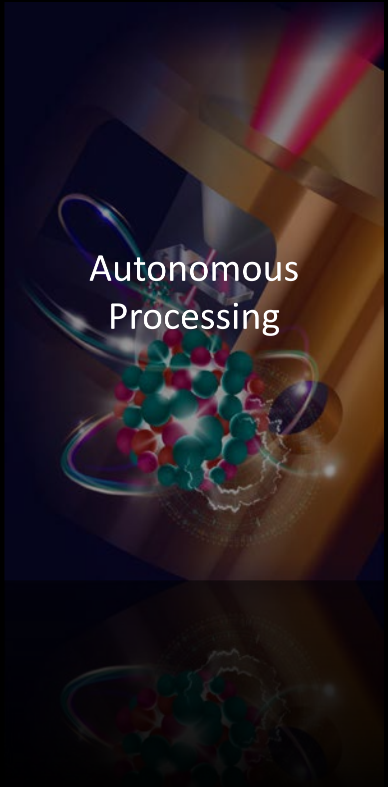


Spurgeon et al. Nature Mater, 20(3), 274–279. (2021).

Today I'll discuss accelerating materials design through autonomous science in three areas







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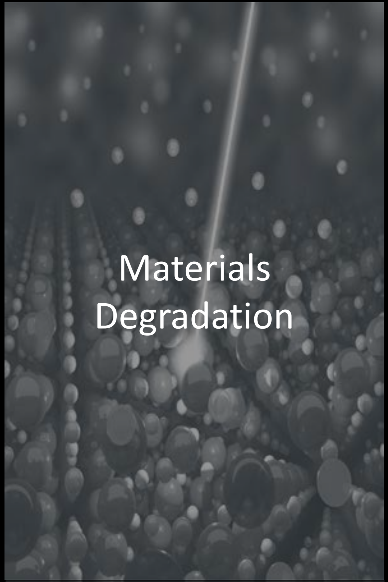
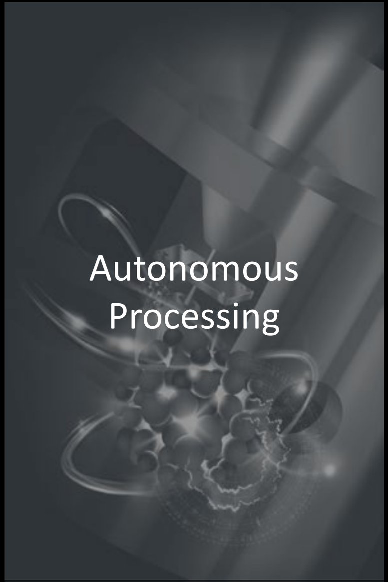
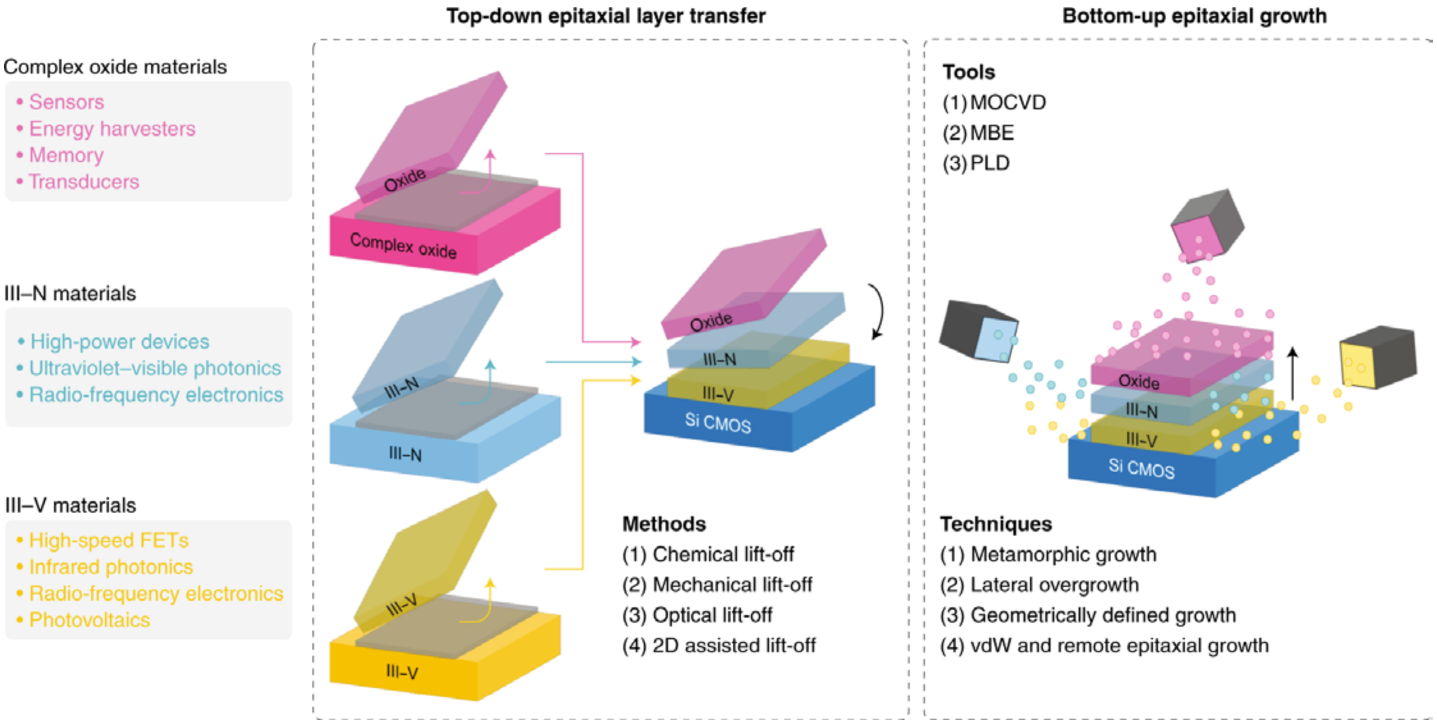




Epitaxial integration of semiconductors and oxides is a critical enabler for emerging devices

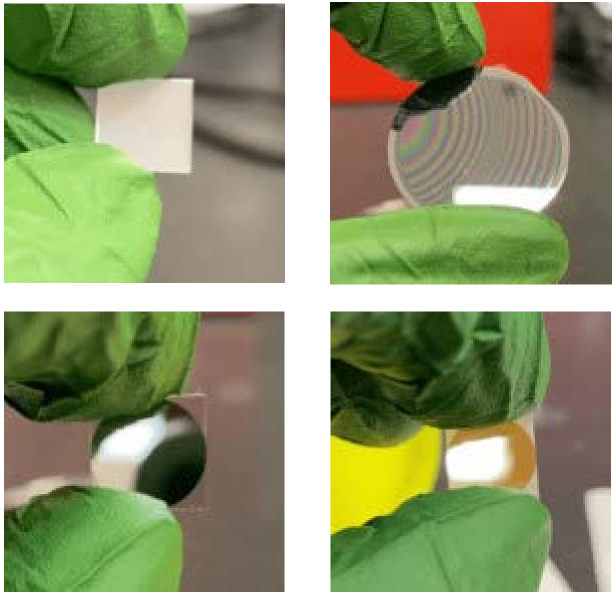


How do we define and control these interfaces?

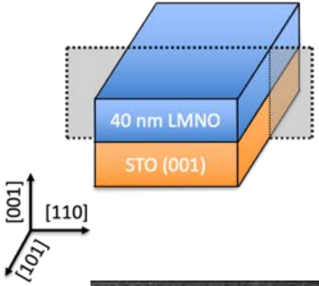
Kum et al. Nature Electron. 2, 439–450 (2019).

Tailored materials design requires direct local probes of structure and chemistry

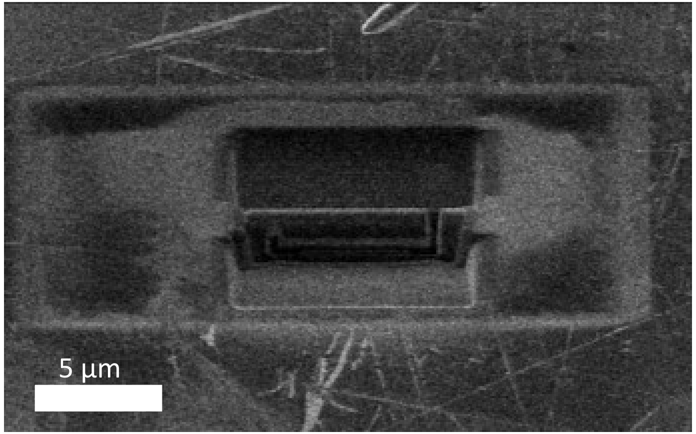
Functional Thin Films for Energy Applications



~1 cm



Site Specific Metrology



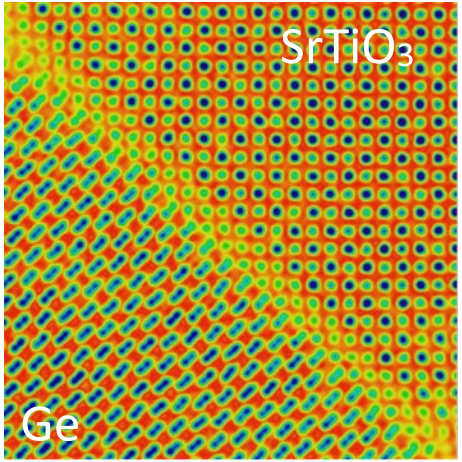
Focused Ion Beam

STEM

Spurgeon et al. Chem Mater, 28.11. 3814–3822. (2016).

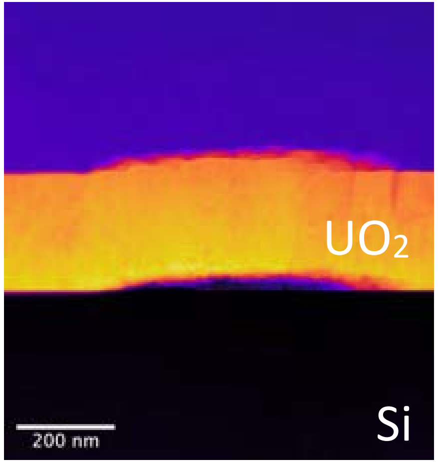
Electron microscopy can richly inform lifecycle models to achieve predictive control

Structure → **Chemistry**



Imaging

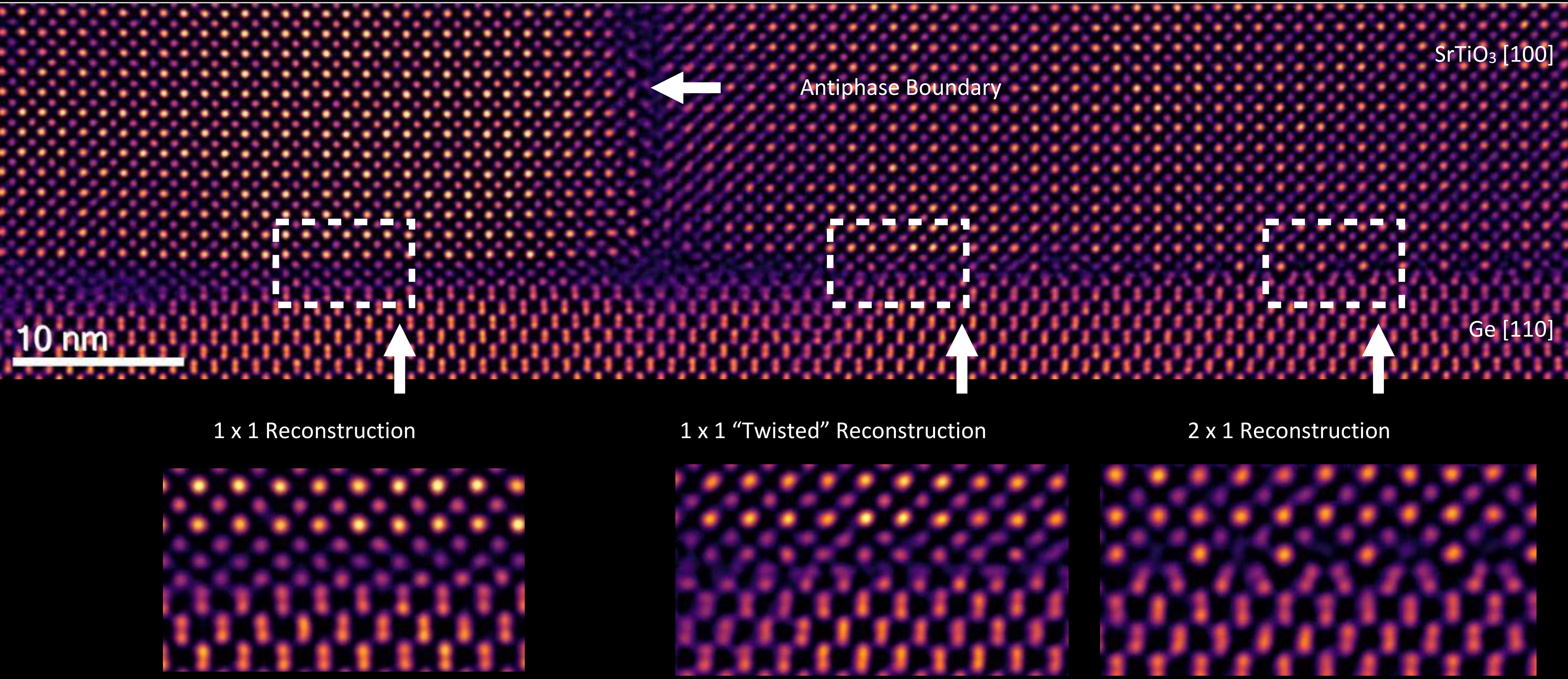
Diffraction



Spectroscopy

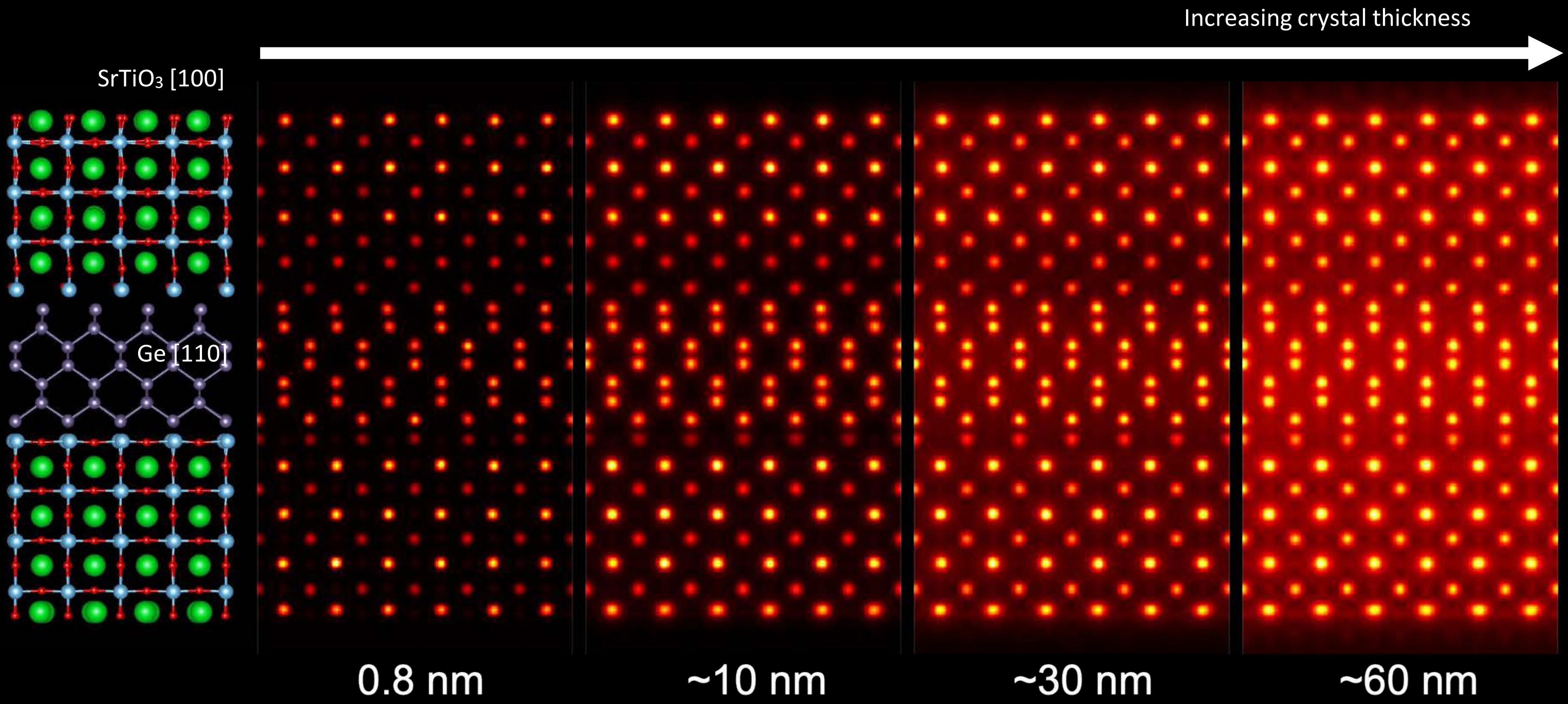
Du et al. Phys Rev B 2, 094602. (2018). | Popel et al. ACS Appl Mater Int. 12, 39781. (2020). | Spurgeon et al. PNAS 116, 17181. (2019).

We have examined interfacial reconstructions in STO / Ge grown via low oxygen pressure MBE



Du et al. Phys Rev Mater. 2. 094602. (2018).

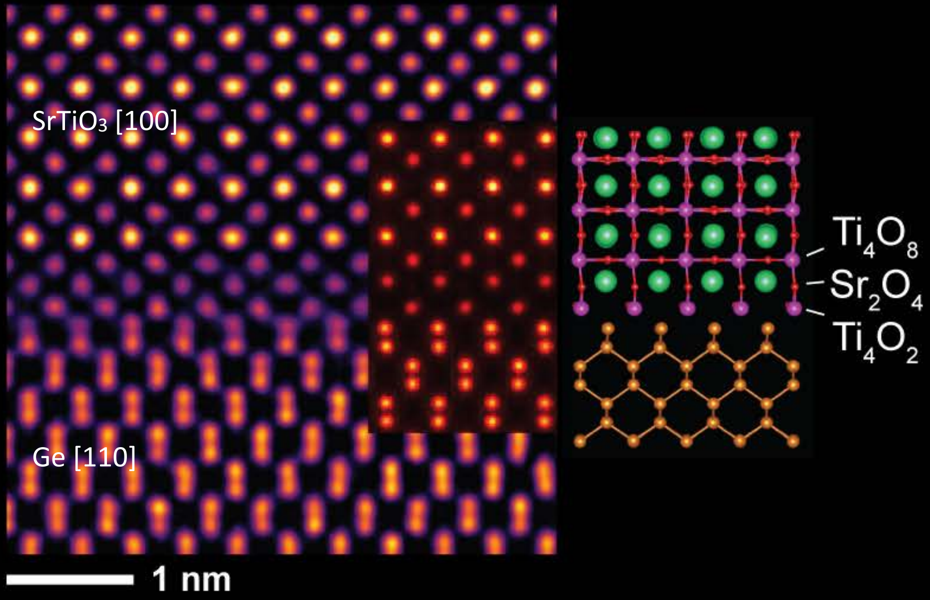
We can directly integrate DFT calculations of interface structures with microscope data



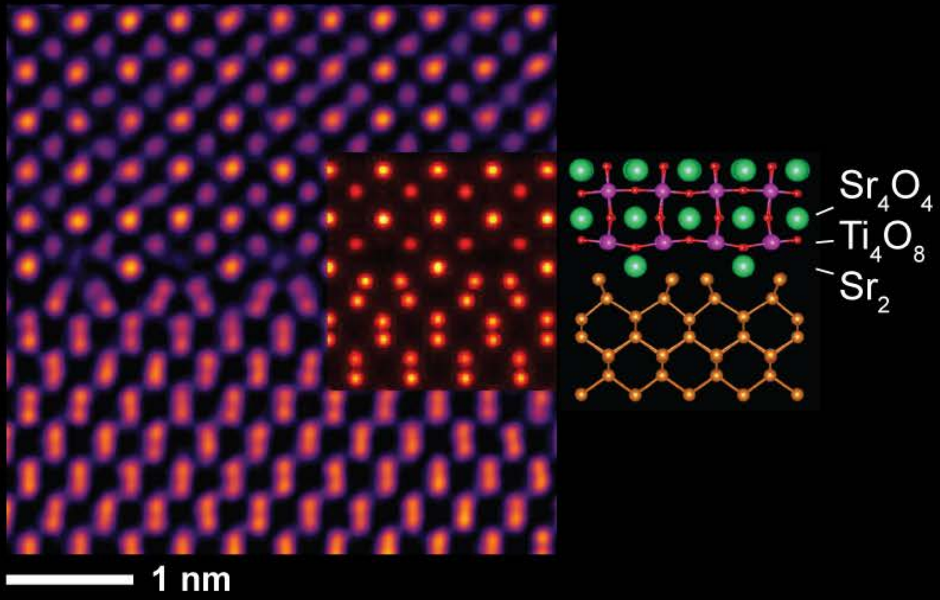
Du et al. Phys Rev Mater. 2. 094602. (2018).

This allows us to determine solutions for interface reconstruction and resulting band structure

1 x 1 Reconstruction

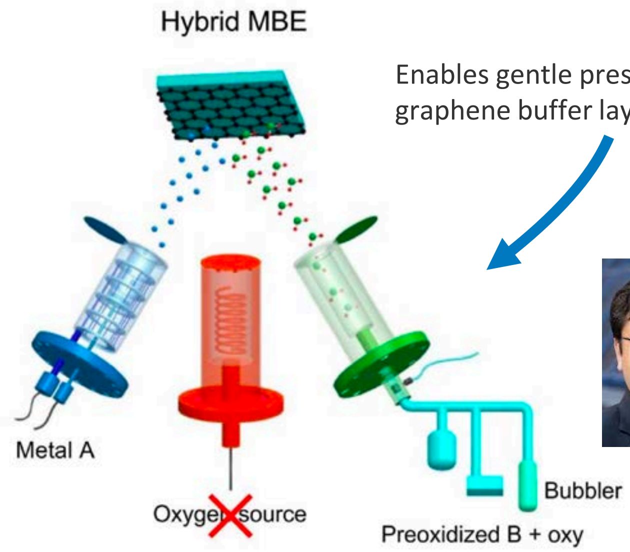


2 x 1 Reconstruction



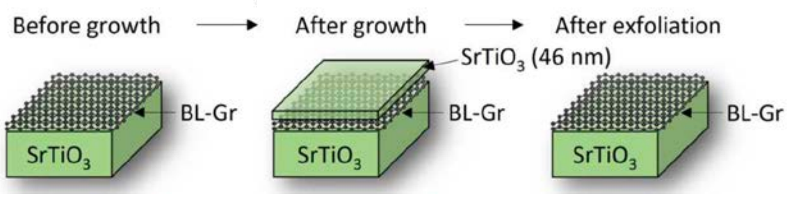
Du et al. Phys Rev Mater. 2. 094602. (2018).

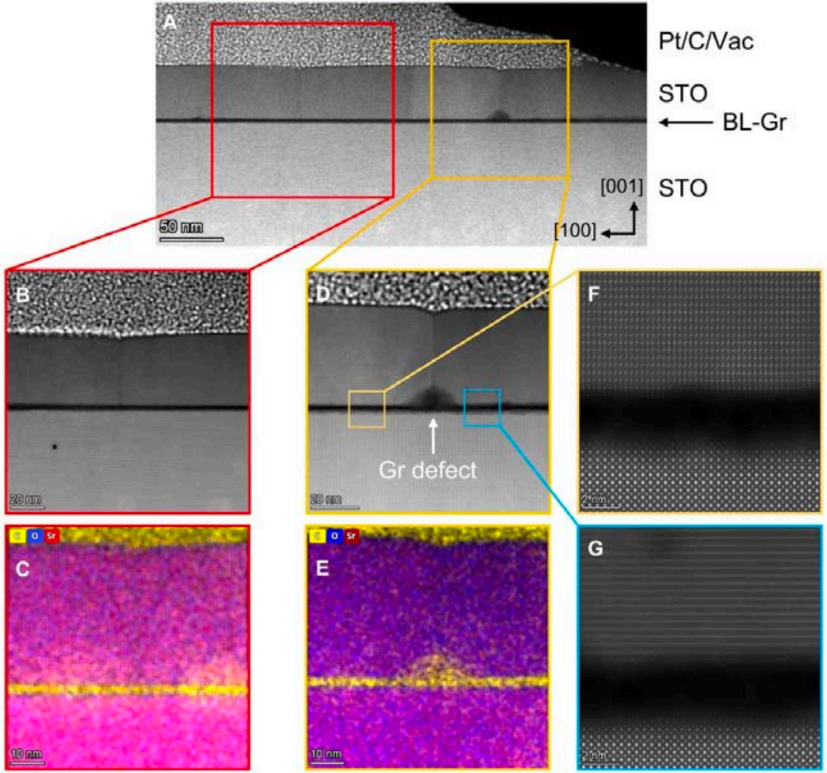
Using hybrid MBE growth, we have demonstrated successful STO lift off from buffered graphene



Enables gentle preservation of graphene buffer layers

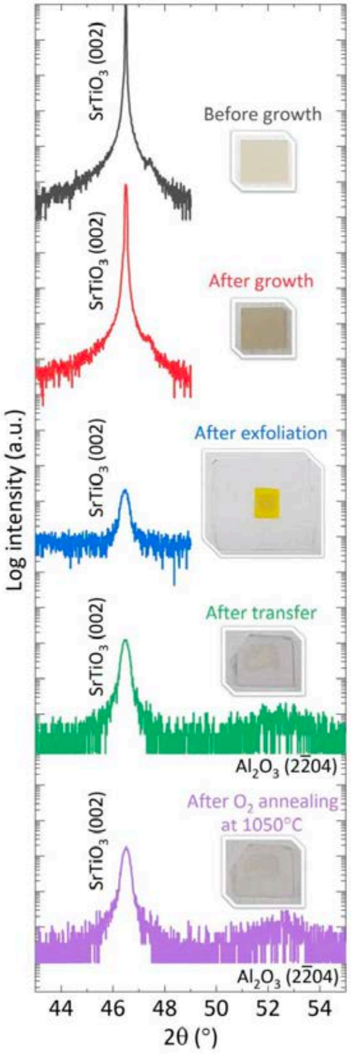
Bharat Jalan
U. Minnesota

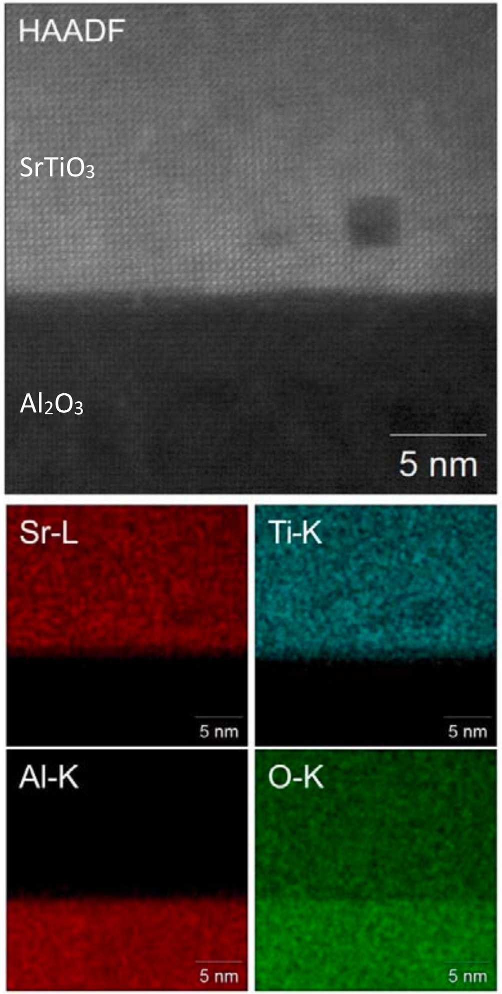




Yoon et al. Sci Adv 8, eadd5328 (2022).

We can transfer crystalline films to foreign substrates, unlocking completely new architectures

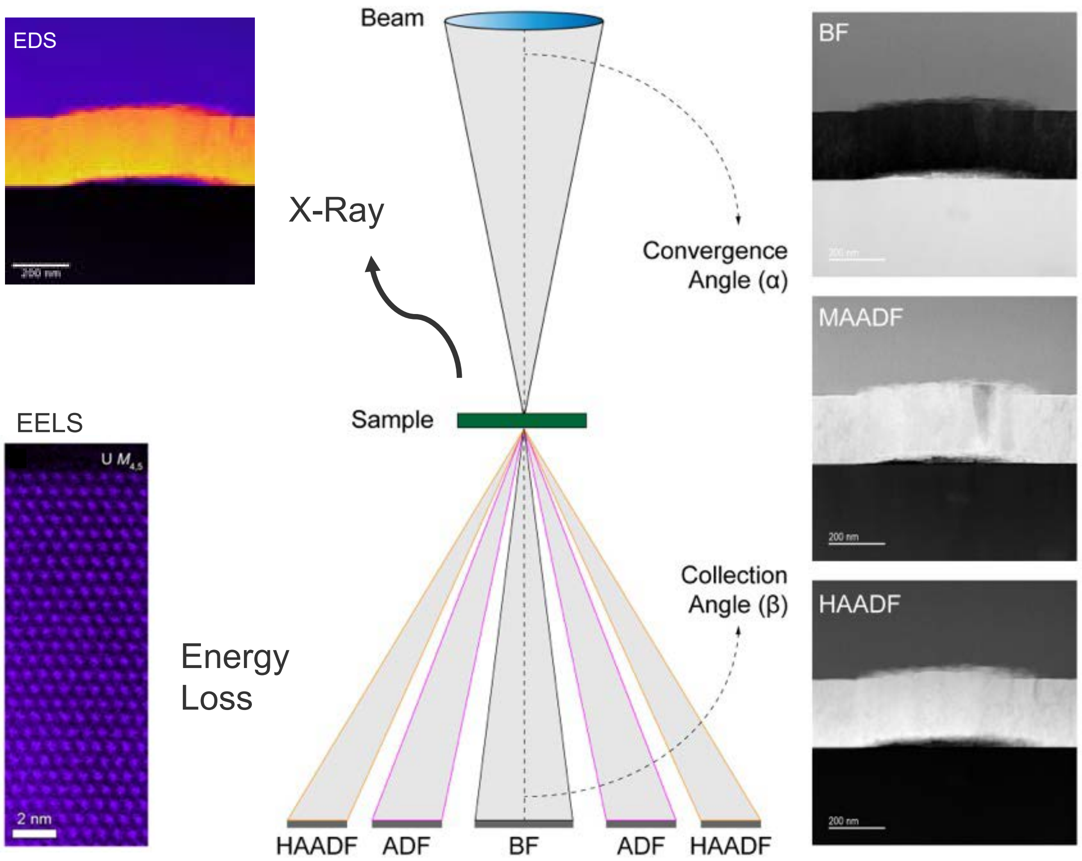




Yoon et al. Sci Adv 8, eadd5328 (2022).

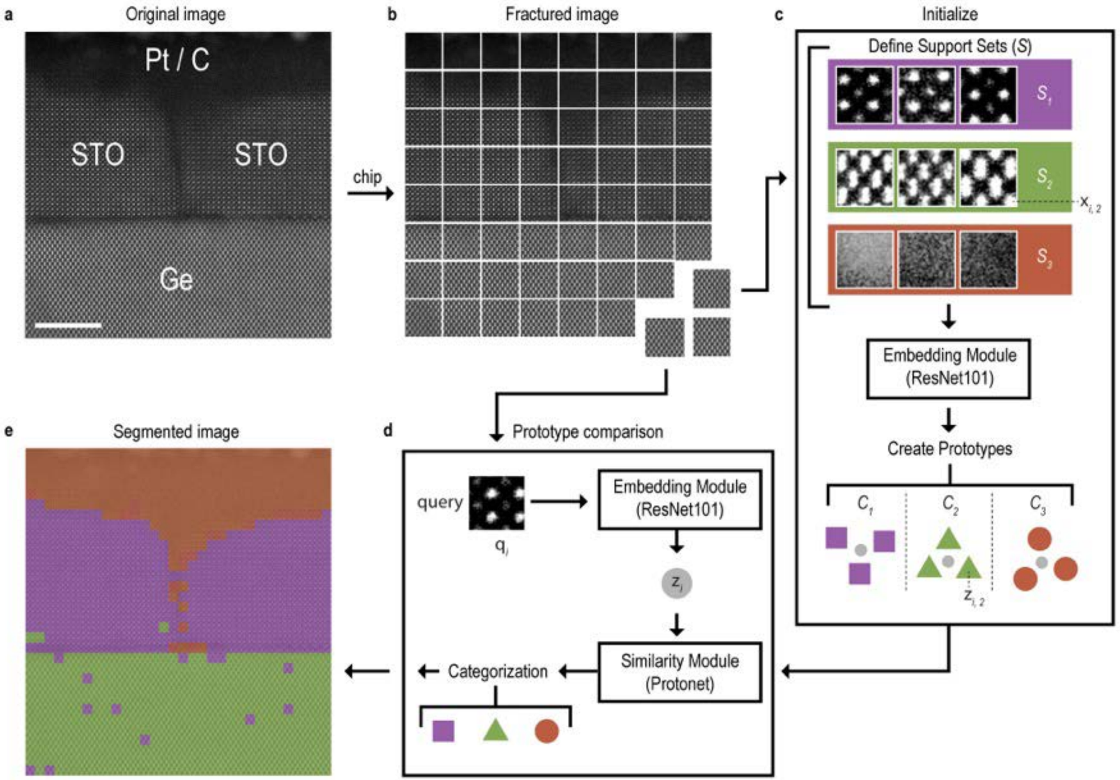
The Challenge

How do we move from laborious one-off “hero” experiments to truly statistical models?



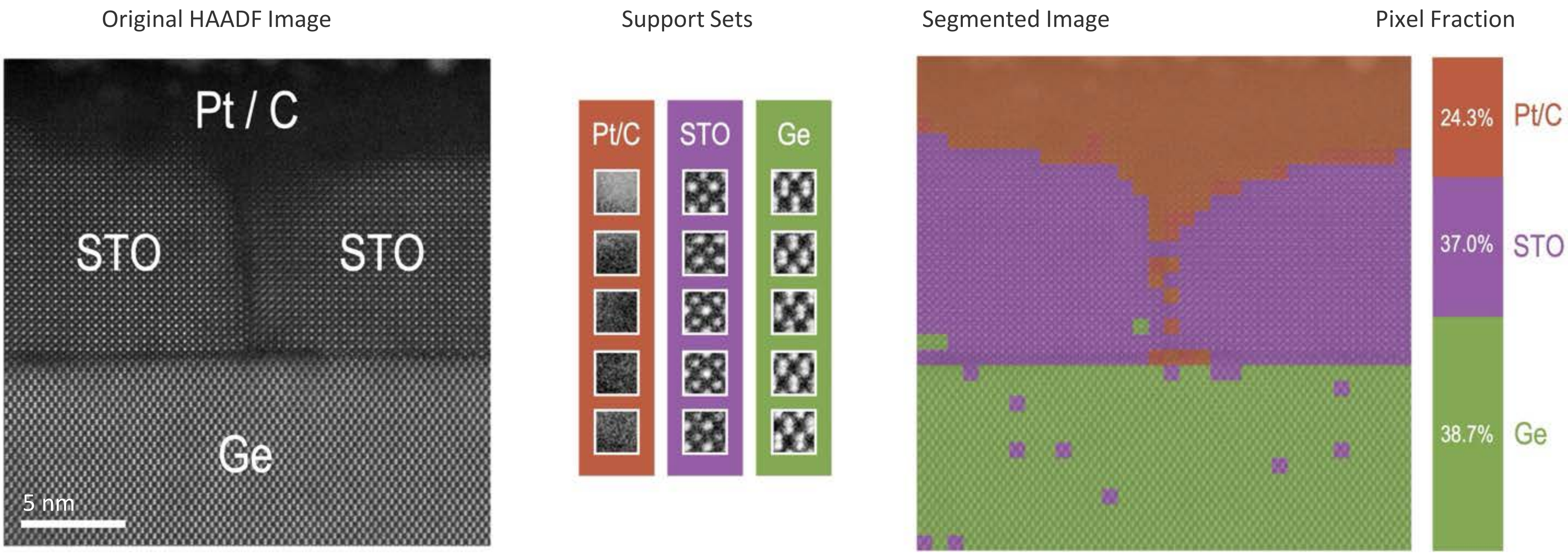
Spurgeon. (2020). DOI:10.48550/arXiv.2001.00947
Spurgeon et al. PNAS 116, 17181. (2019).

Few shot ML uses limited prior knowledge to classify features in discovery scenarios



Akers et al. npj Comp. Mater., 7(1), 187. (2021).

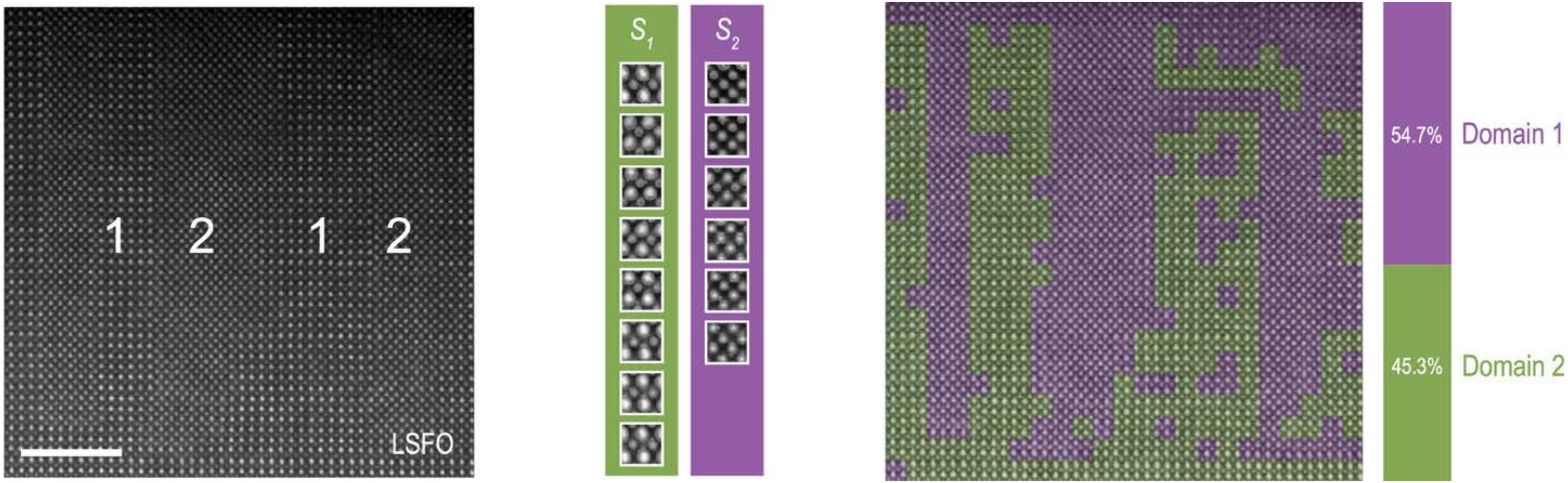
We can rapidly classify atomic motifs in data to understand phase distributions in spintronics



Akers et al. npj Comp. Mater., 7(1), 187. (2021).

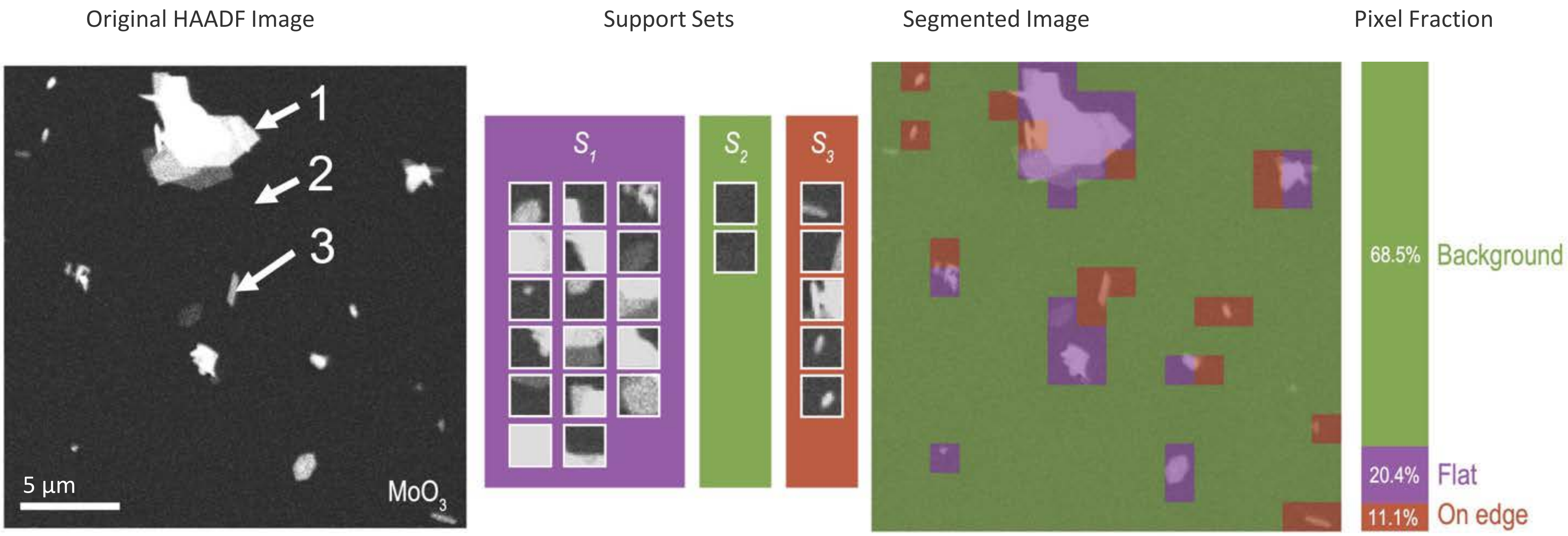
We can also identify defective domains within oxides for memristors

Original HAADF Image | Support Sets | Segmented Image | Pixel Fraction



Akers et al. npj Comp. Mater., 7(1), 187. (2021).

Such a model can easily be applied to different synthesis tasks such as precursors for PVs

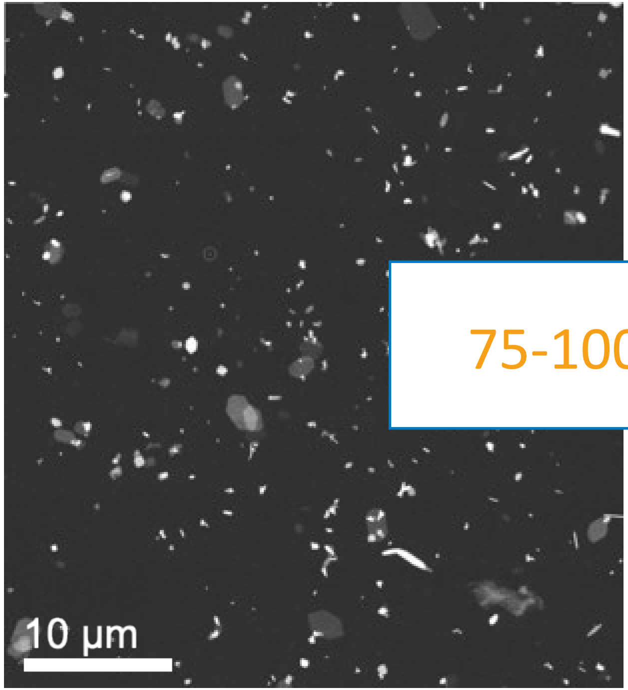


Akers et al. npj Comp. Mater., 7(1), 187. (2021).

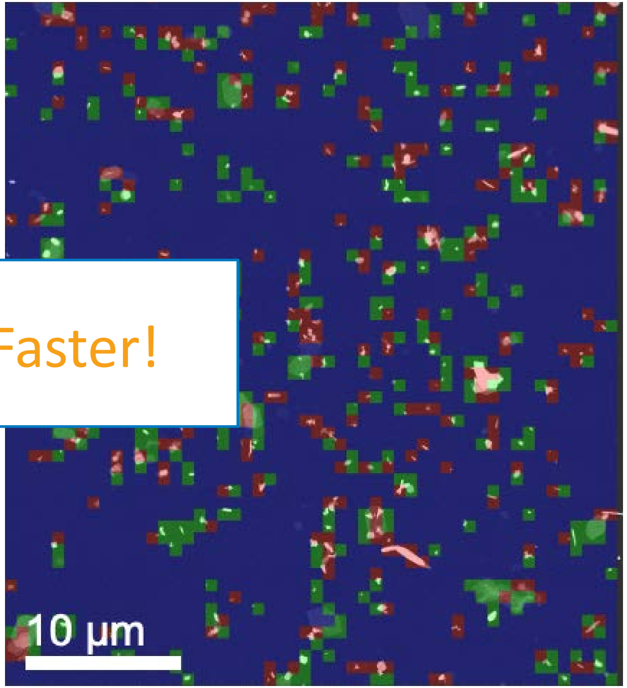
We can ultimately extract materials descriptors in a faster and more reproducible manner

MoO_3

Manual Analysis
10 minutes



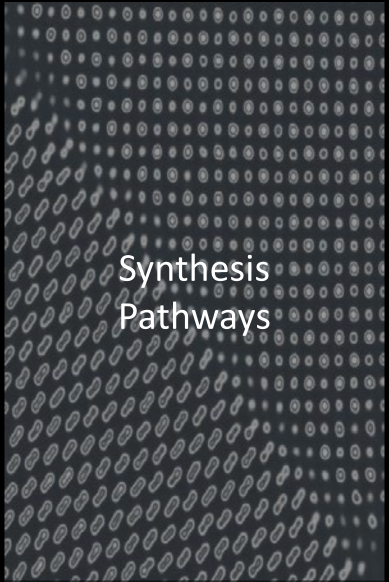
75-100x Faster!

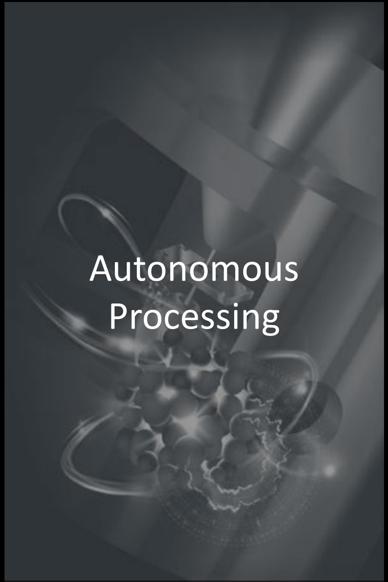


Few-Shot
Task 2
8 seconds

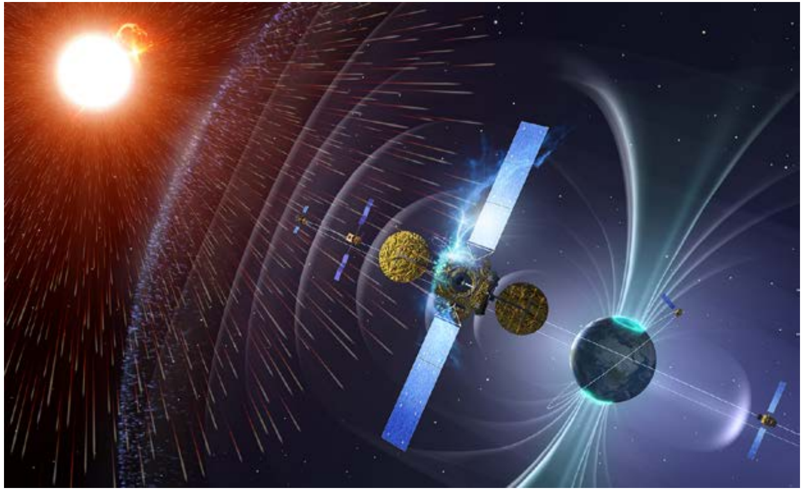
Akers et al. npj Comp. Mater., 7(1), 187. (2021).

Today I'll discuss accelerating materials design through autonomous science in three areas

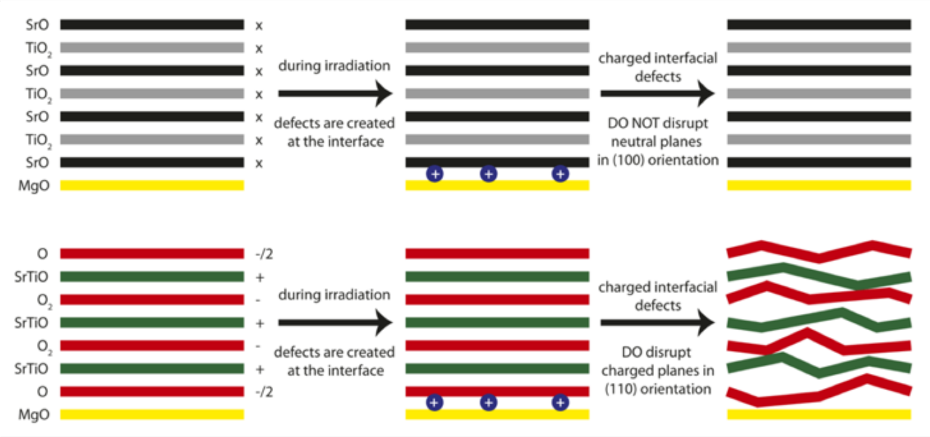




Controlling materials degradation is critical for electronics and sensors in extremes

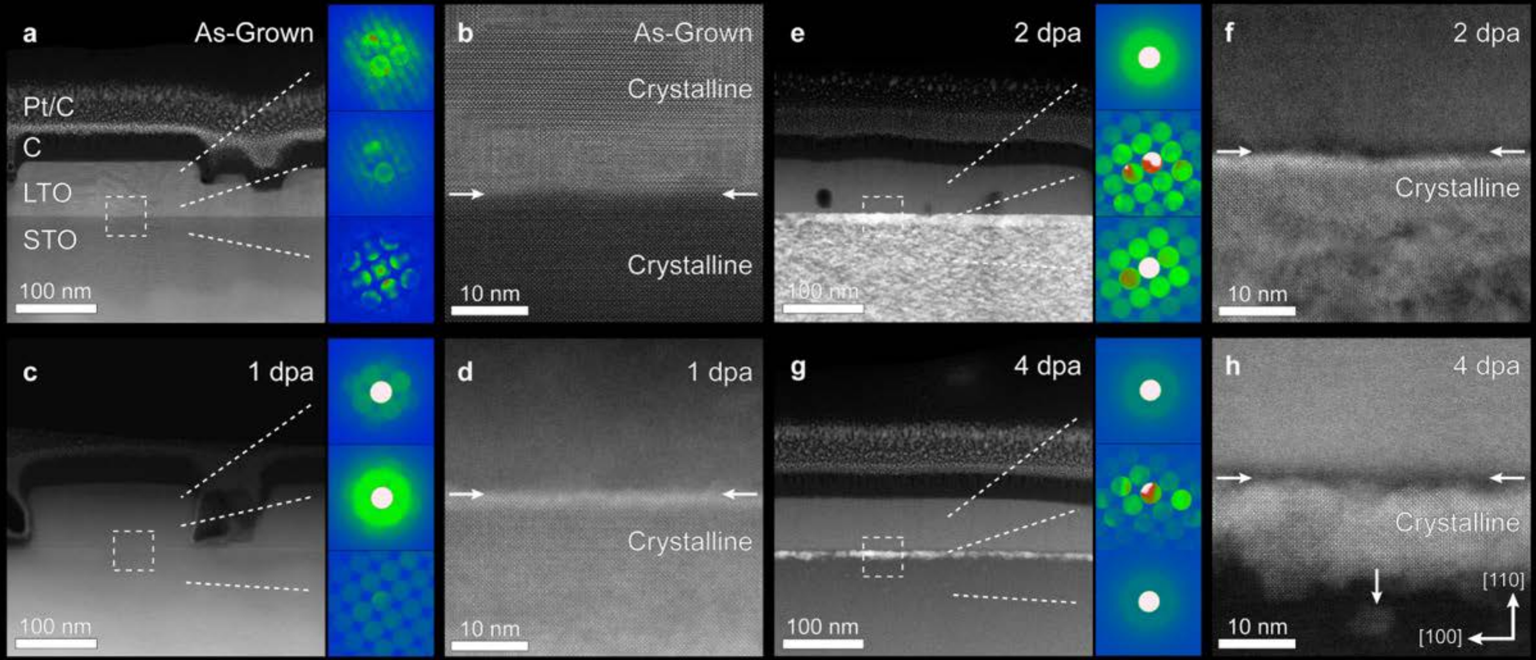


How does interface configuration affect radiation-induced disorder in devices?



Aguiar et al. J. Mater. Res. 29(16), 1699–1710 (2014). | Spurgeon. Curr. Opin. Solid. State Mater. Sci. 24(6), 100870 (2020). | Image: SSA / ESA

We can visualize damage buildup at these interfaces at stages of irradiation



Spurgeon et al. Adv. Mater. Interfaces. 7(8). 1901901944. (2020).

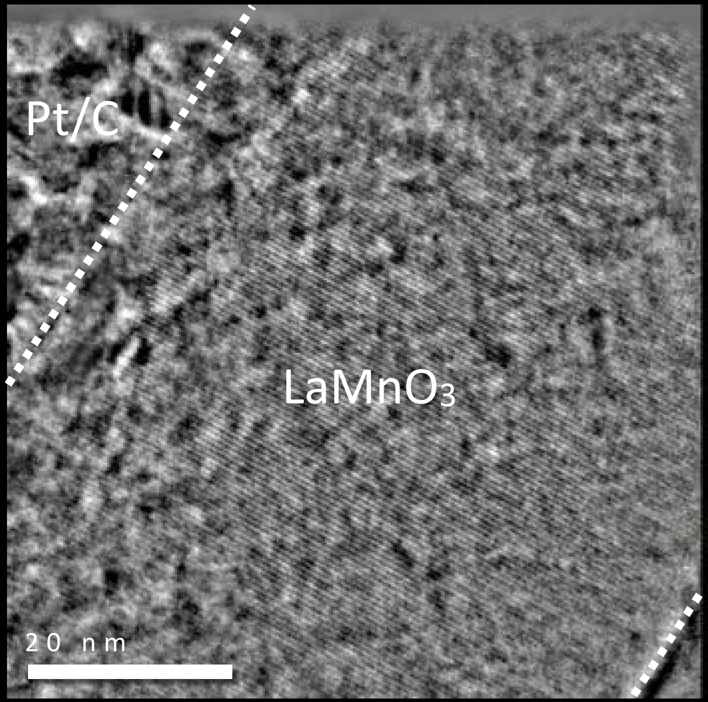
At CINT we took this one step further, visualizing materials breakdown in situ using the I^3TEM

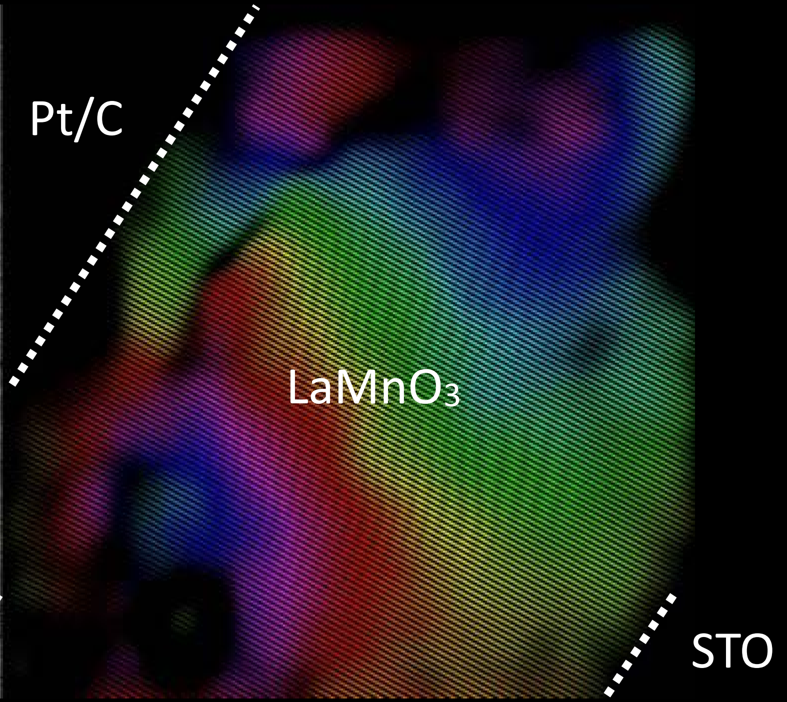
Khalid Hattar (UTK)

Chris Barr

https://tours.sandia.gov/ibl_info.html

In situ TEM shows that disorder percolates through the material in a non-uniform manner

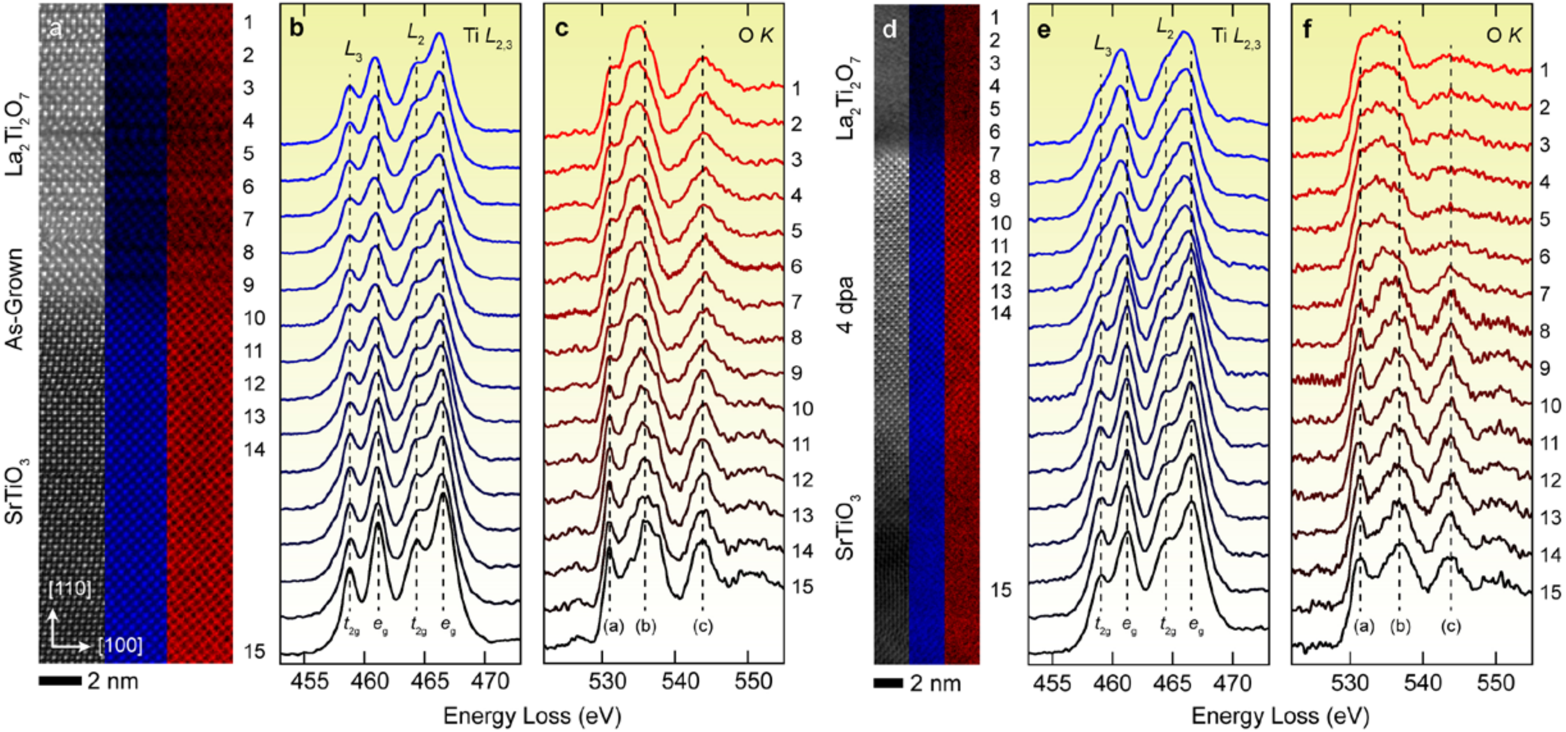




1 Hr Irradiation

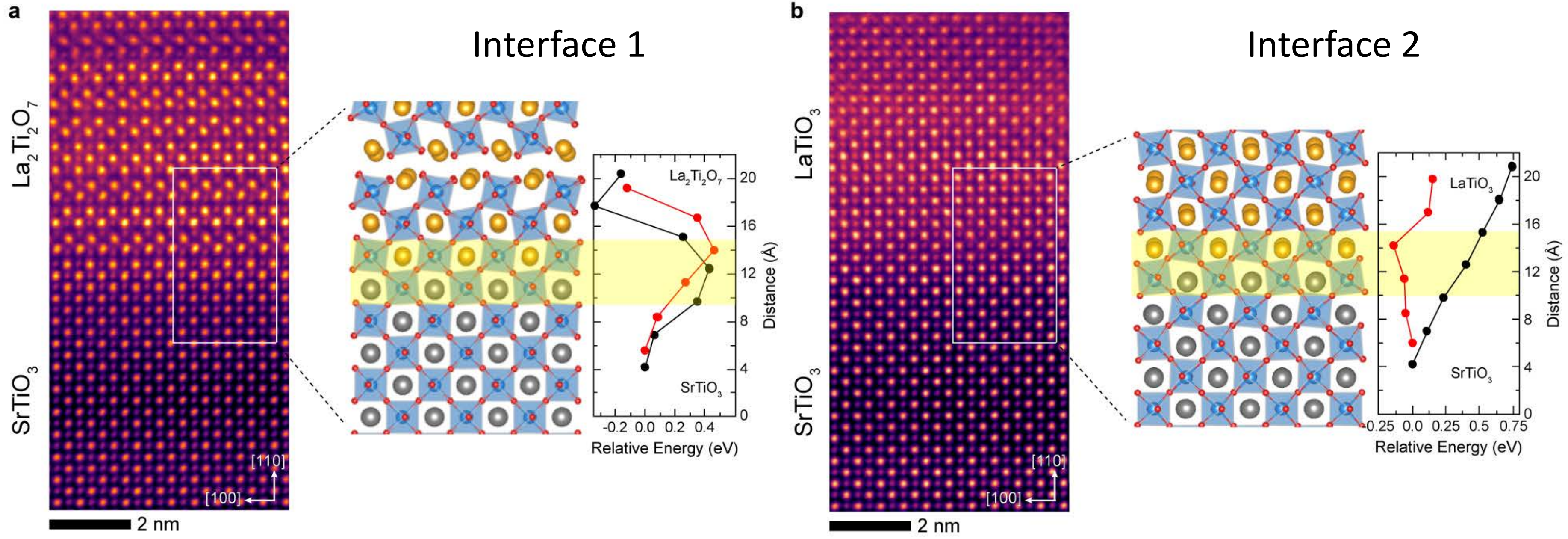
Matthews et al. Nano Letters, 21(12), 5353–5359. (2021).

Atomic-scale spectroscopy reveals changes in oxygen vacancies upon irradiation



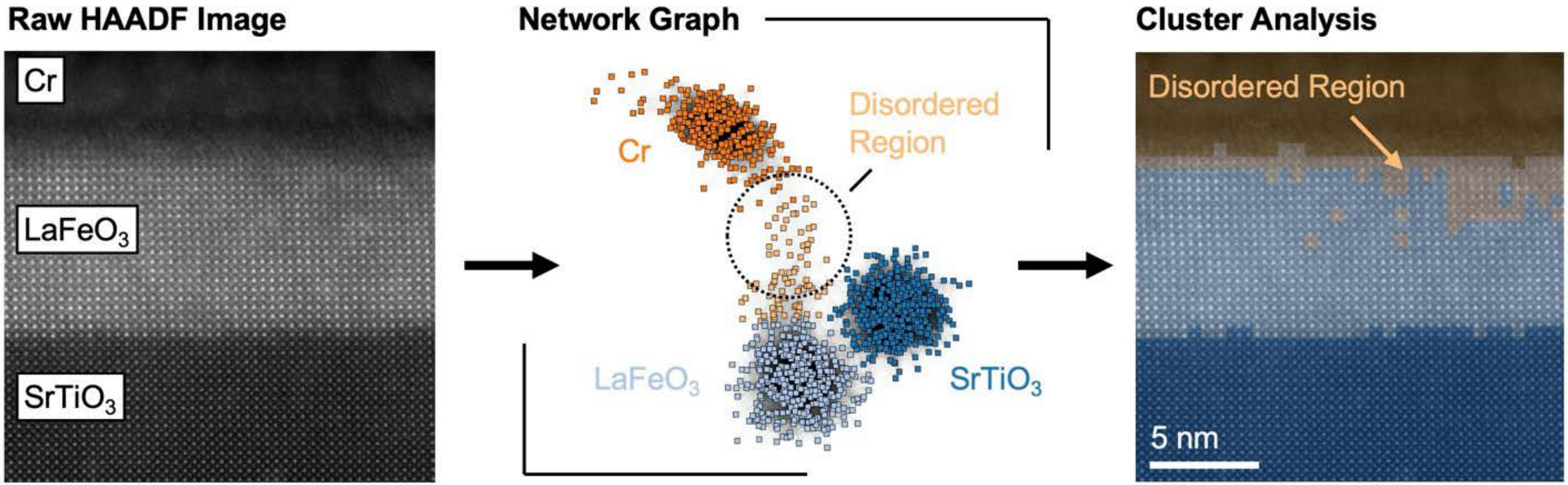
Spurgeon et al. Adv. Mater. Int. 7(8). 1901901944. (2020).

This informs DFT calculations, which show differences in oxygen vacancy formation energy



Spurgeon et al. Adv. Mater. Int. 7(8). 1901901944. (2020).

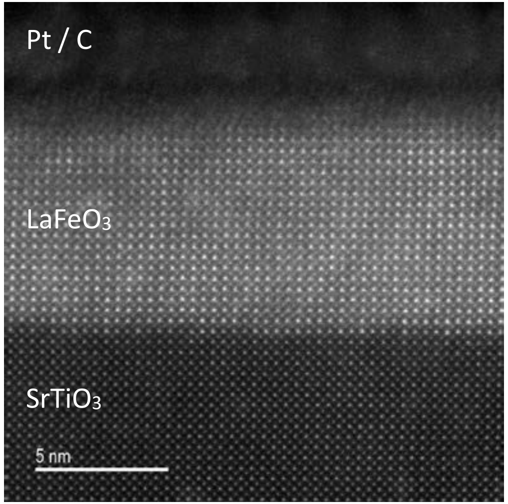
But how do we begin to move toward more statistical models of defects?



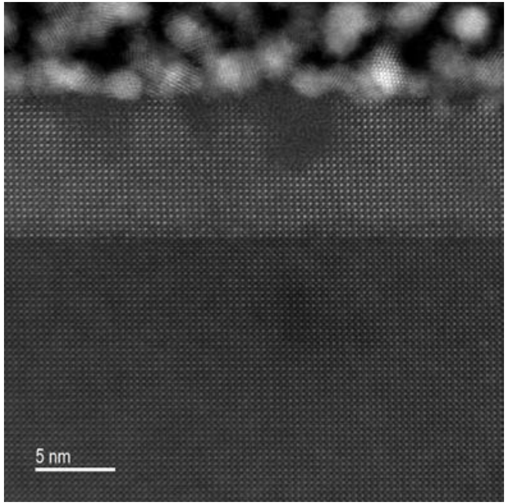
Ter-Petrosyan et al. Proc. Thirty-Seventh Conf. on Neural Information Processing Systems (NeurIPS). (2023). DOI:10.48550/arXiv.2311.08585

These descriptors help us evaluate degradation during irradiation in an unsupervised manner

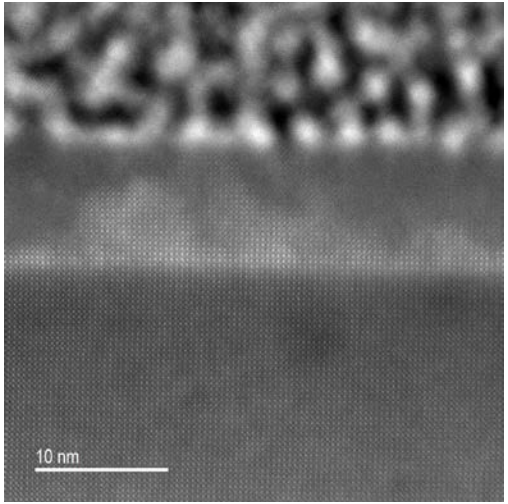
0 dpa



0.1 dpa



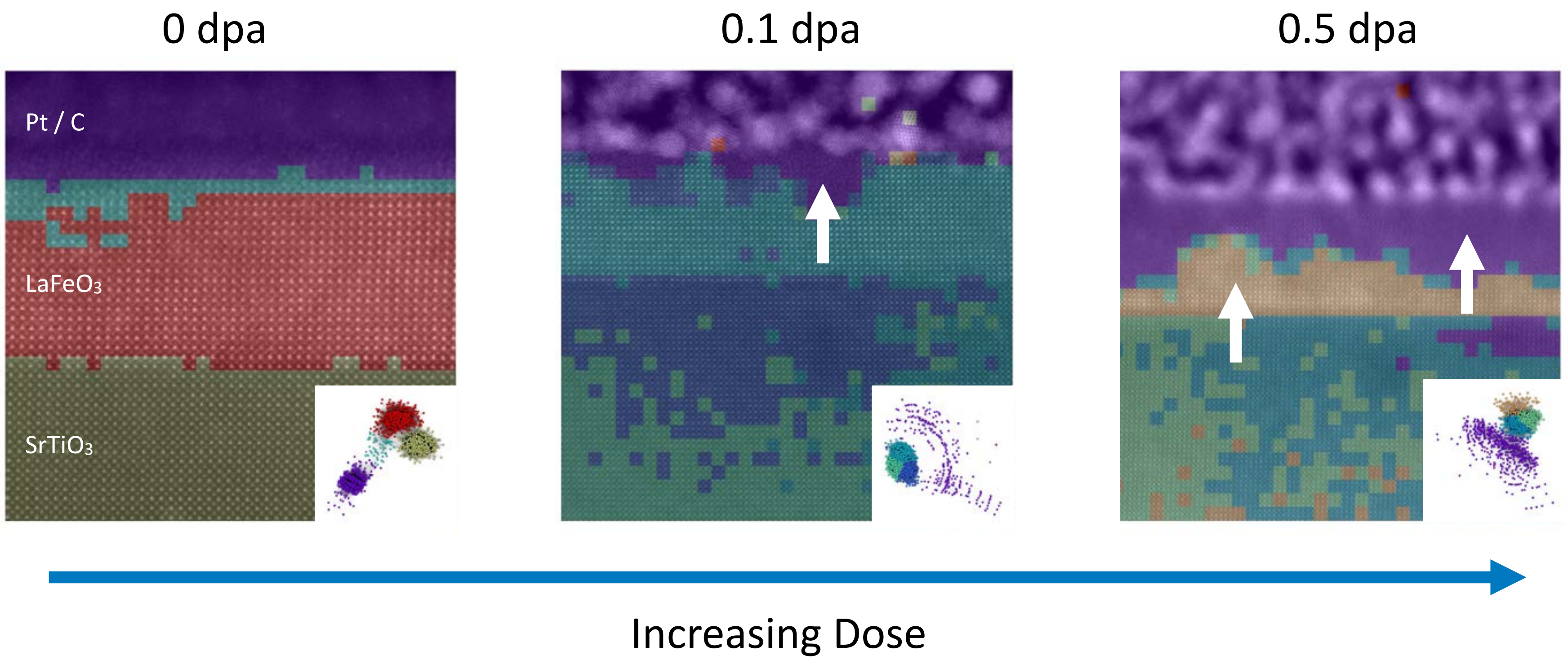
0.5 dpa



Increasing Dose

Ter-Petrosyan et al. Proc. Thirty-Seventh Conf. on Neural Information Processing Systems (NeurIPS). (2023). DOI:10.48550/arXiv.2311.08585

The signature of radiation damage can be tracked and used to guide next steps of synthesis



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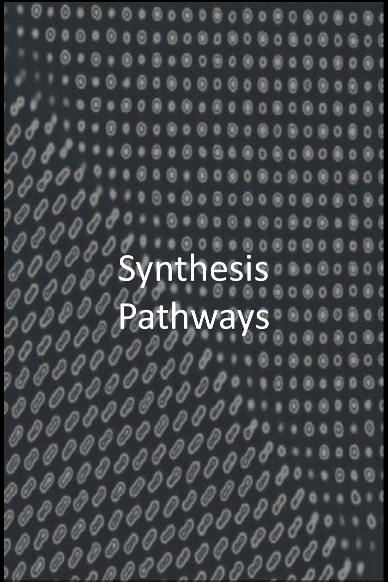
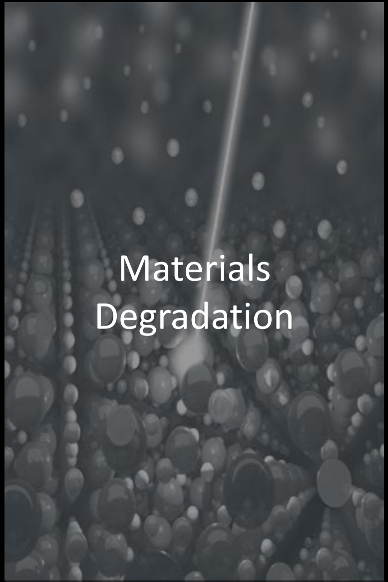
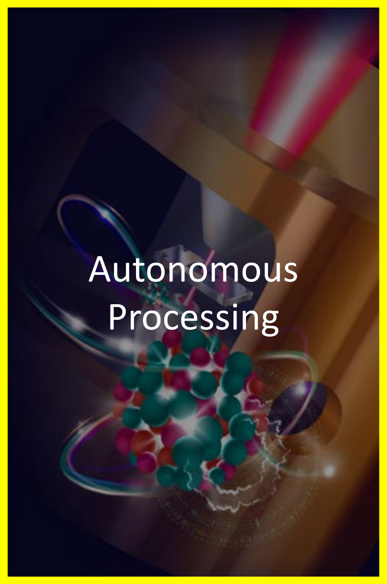
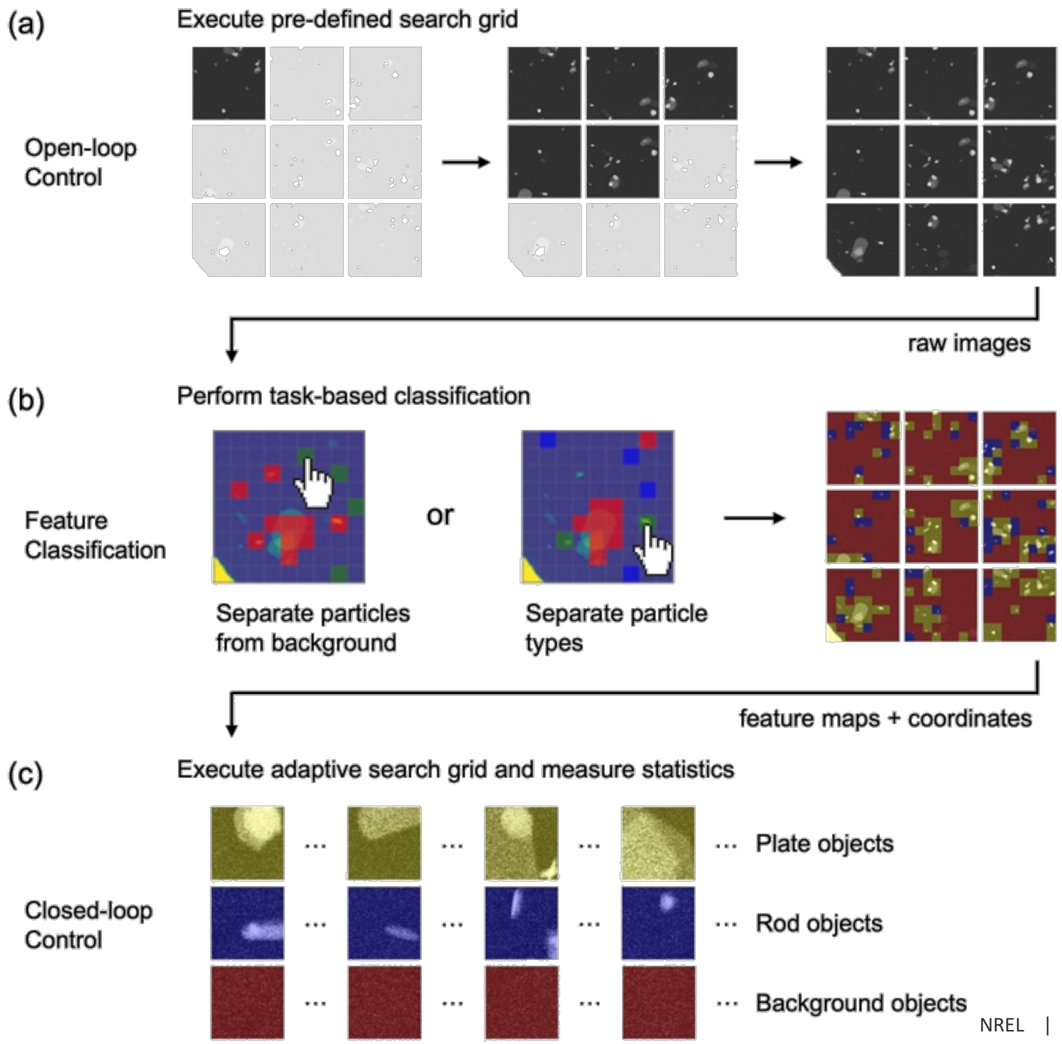






This platform enables intelligent closed-loop experiments and statistical analyses

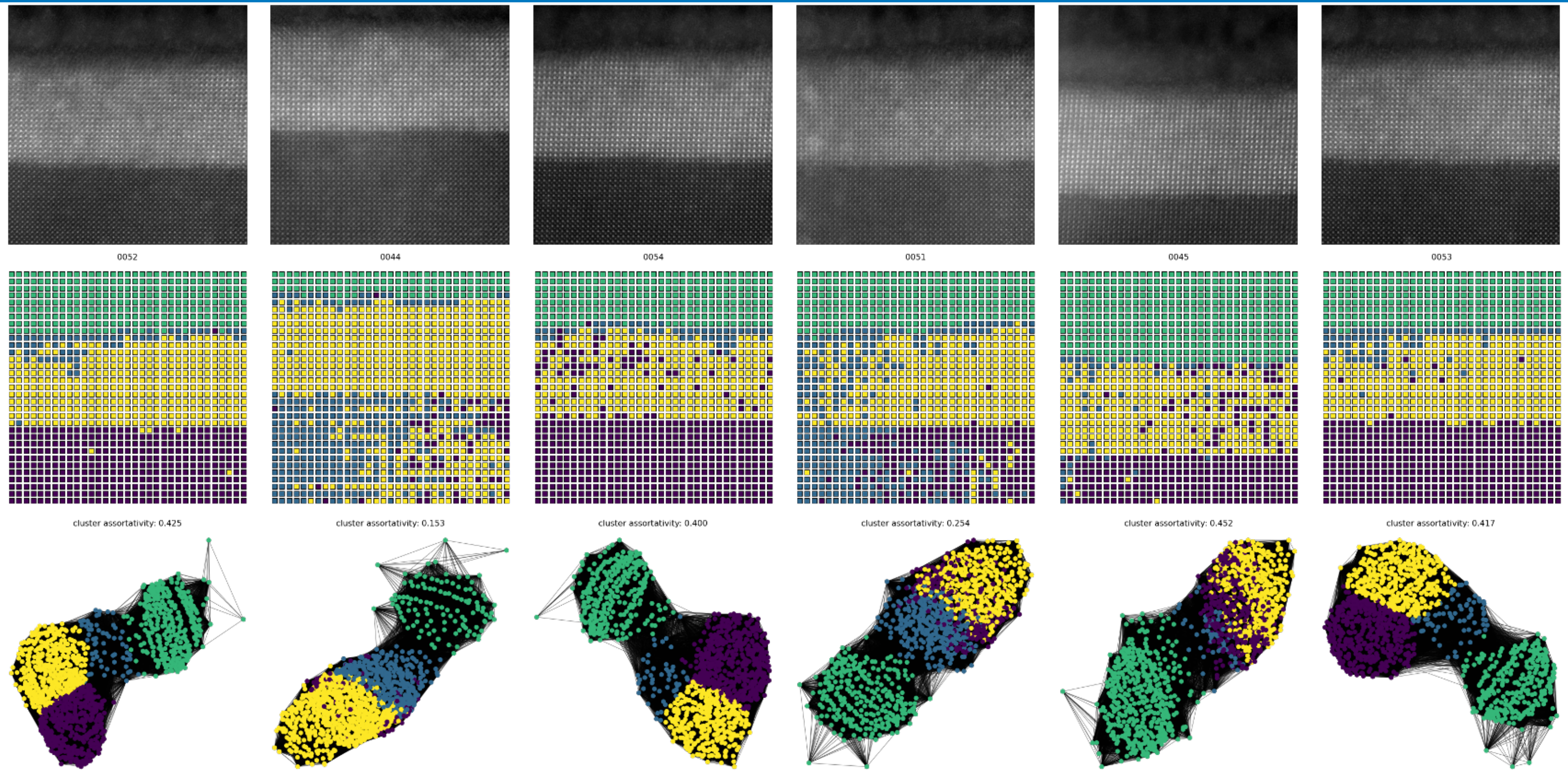
Olszta et al. Micro. Microanal., 28 (5), 1611-1621. (2022).



2024 R&D 100 WINNER

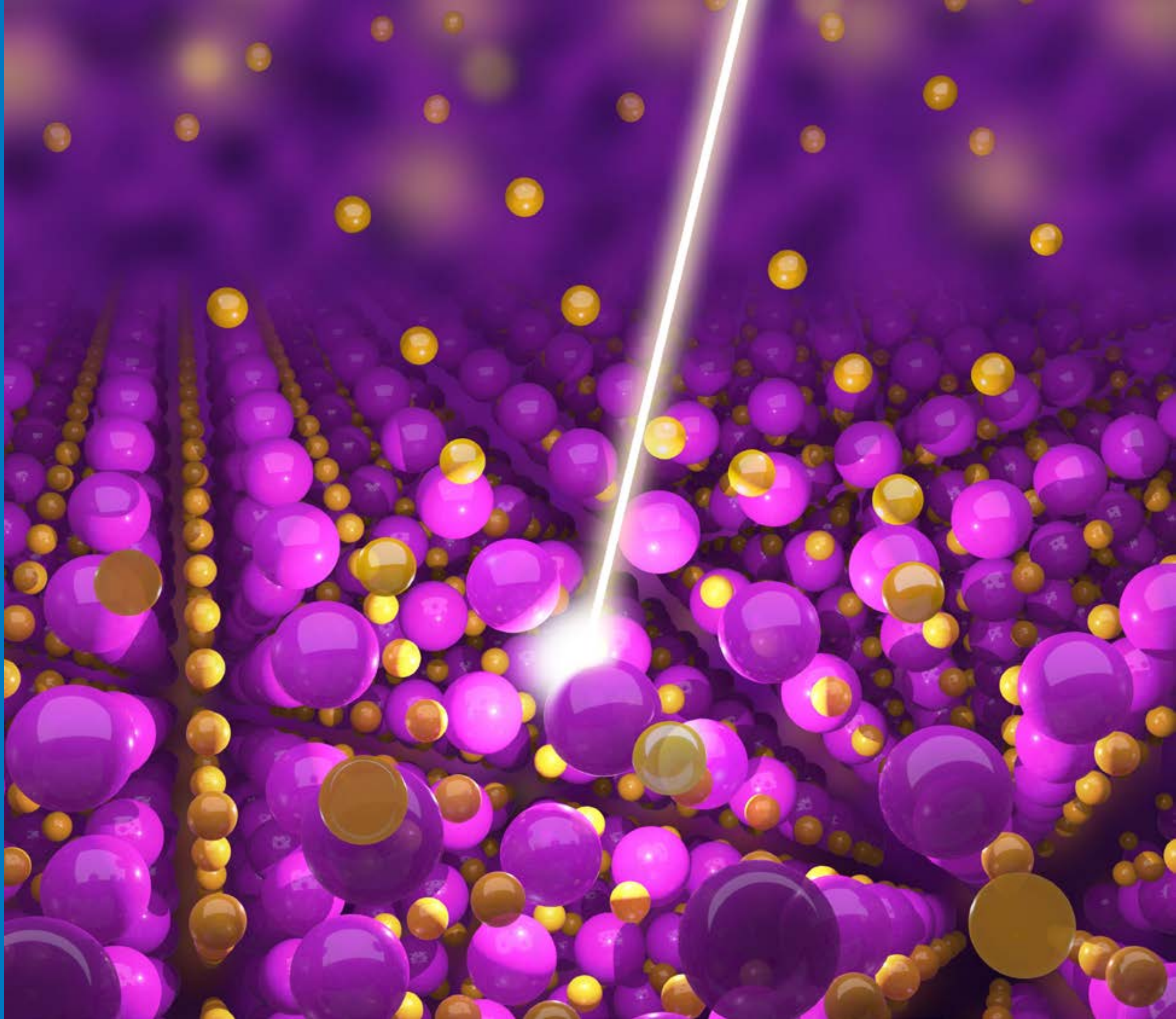
PATENT PENDING
HTTPS://YOUTU.BE/XKYJ1UAE6JE

We can build statistical libraries of synthesis and degradation products



Ter-Petrosyan et al. Proc. Thirty-Seventh Conf. on Neural Information Processing Systems (NeurIPS). (2023). DOI:10.48550/arXiv.2311.08585

Taking this a step further, can we autonomously process materials?



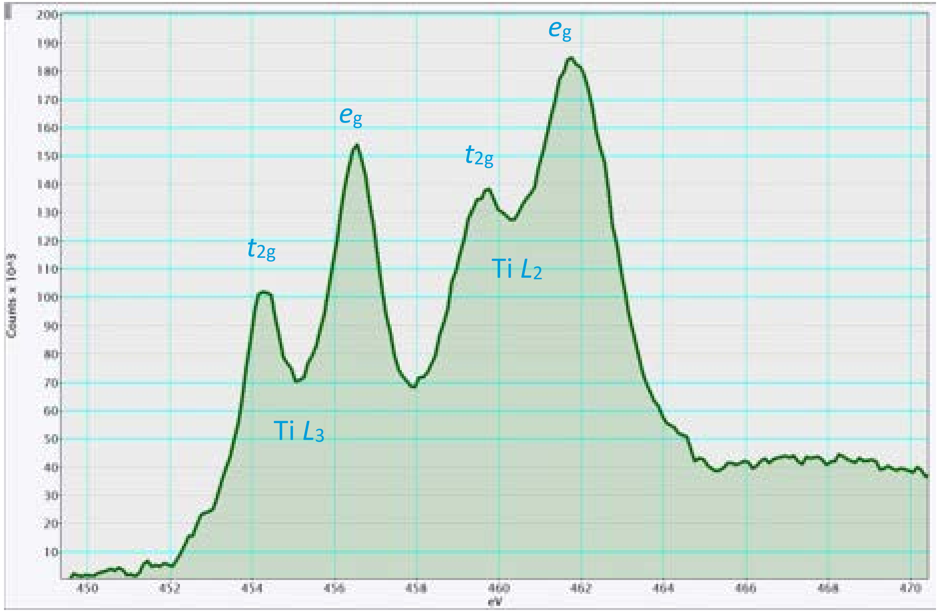
We can controllably run reactions in our microscope to impart specific defect configurations

In Situ Reduction of STO

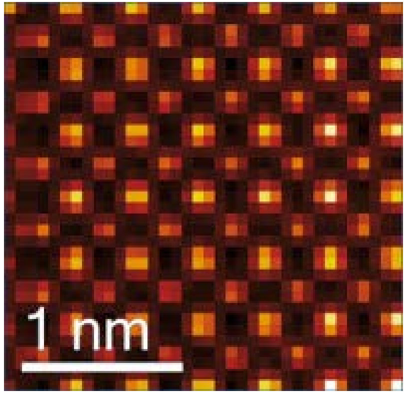
Dose Rate ~10^9 e / (Å^2 • s)

Temp ~20 ºC

EELS

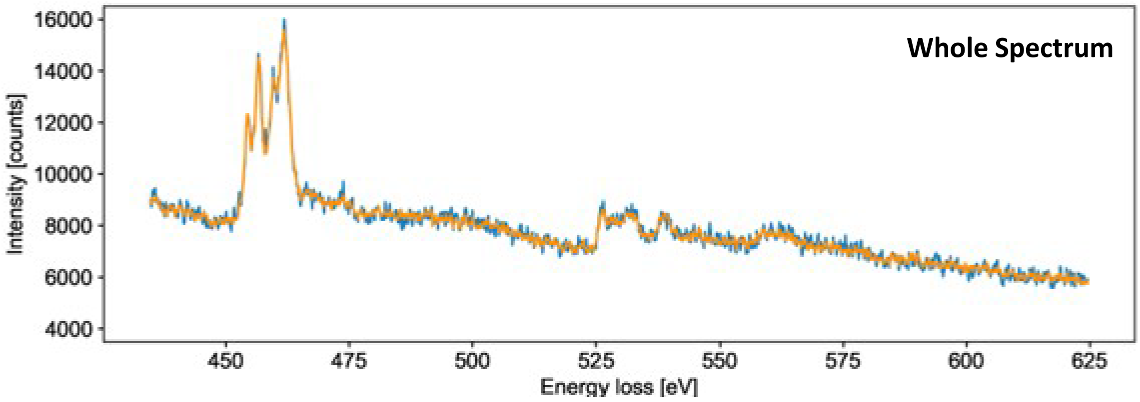


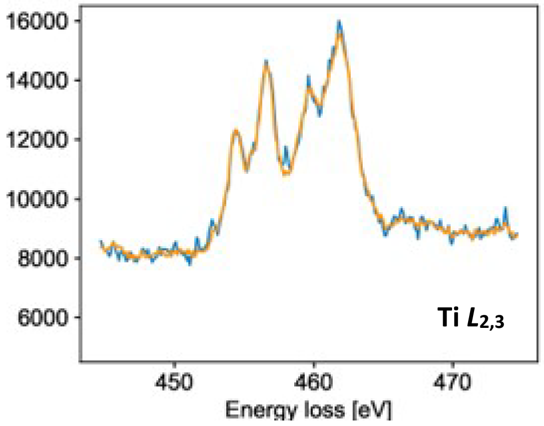
HAADF

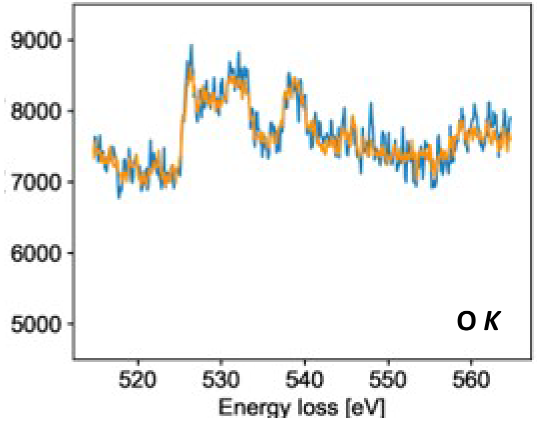


Lewis et al. npj Comp. Mater., 8. 252. (2022).

Recurrent neural networks allow us to predict defect evolution to intelligently direct them







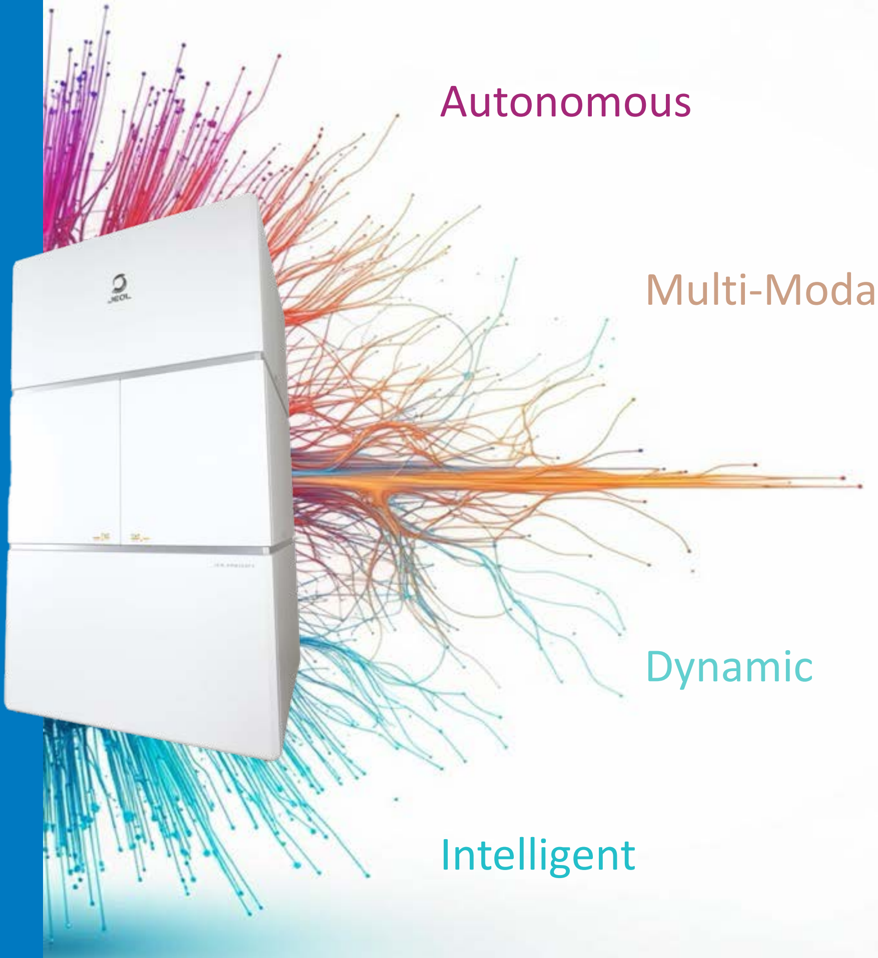
Lewis et al. npj Comp. Mater., 8. 252. (2022).

What is next?

NREL is leading a $14M recapitalization of our electron microscopy center, with a focus on in situ and autonomous science

What is next?

NREL will be home to a new autonomous electron microscope platform built around dynamic and adaptive experiments

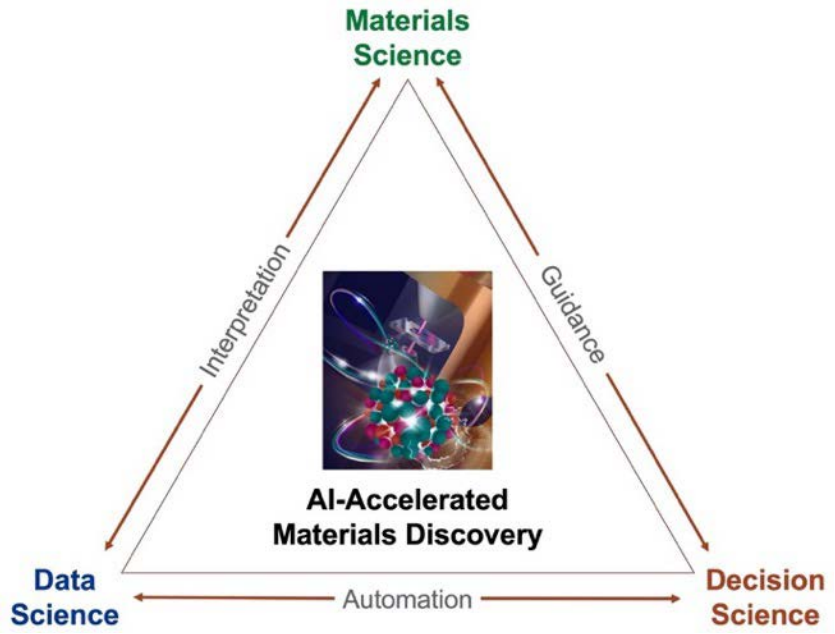


AM^3

The Autonomous Multi-Modal Microscope

Autonomous science is revealing hidden materials lifecycles and transforming the design of clean energy systems.

For more information visit:
https://tinyurl.com/z8ryk4y3



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- Internship Dates: Summer 2025
- Application Due: January 8, 2025
- Apply: https://science.osti.gov/wdts/suli

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SCAN ME

Contact me!

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UVa SULI Student Grace Guinan (Left) and NREL Postdoc Michelle Smeaton (Right)