M3 LEARNING
Multifunctional | Materials | Machine

Automatic Feature Extraction from Hyperspectral Imagery using Deep Recurrent Neural Networks

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Department of Materials Science and Engineering, Lehigh University

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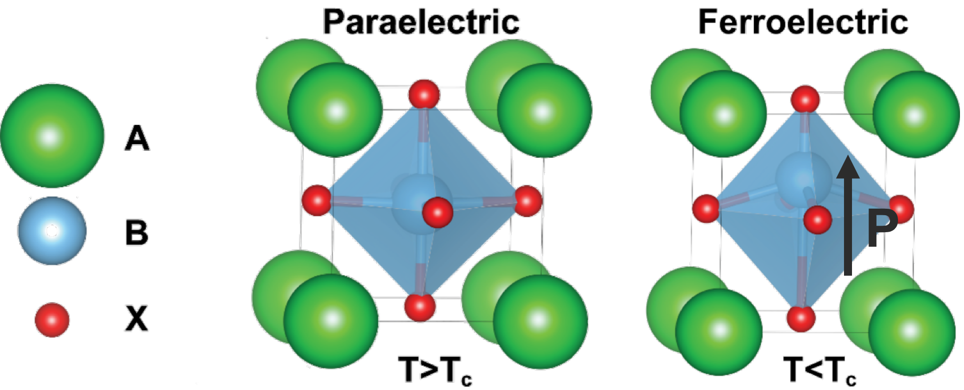
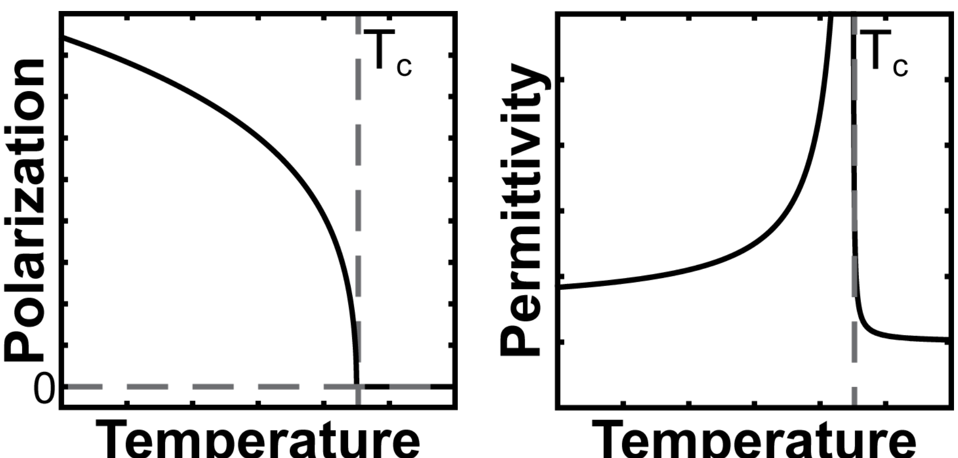
Ferroelectricity 101

Perovskite Ferroelectrics

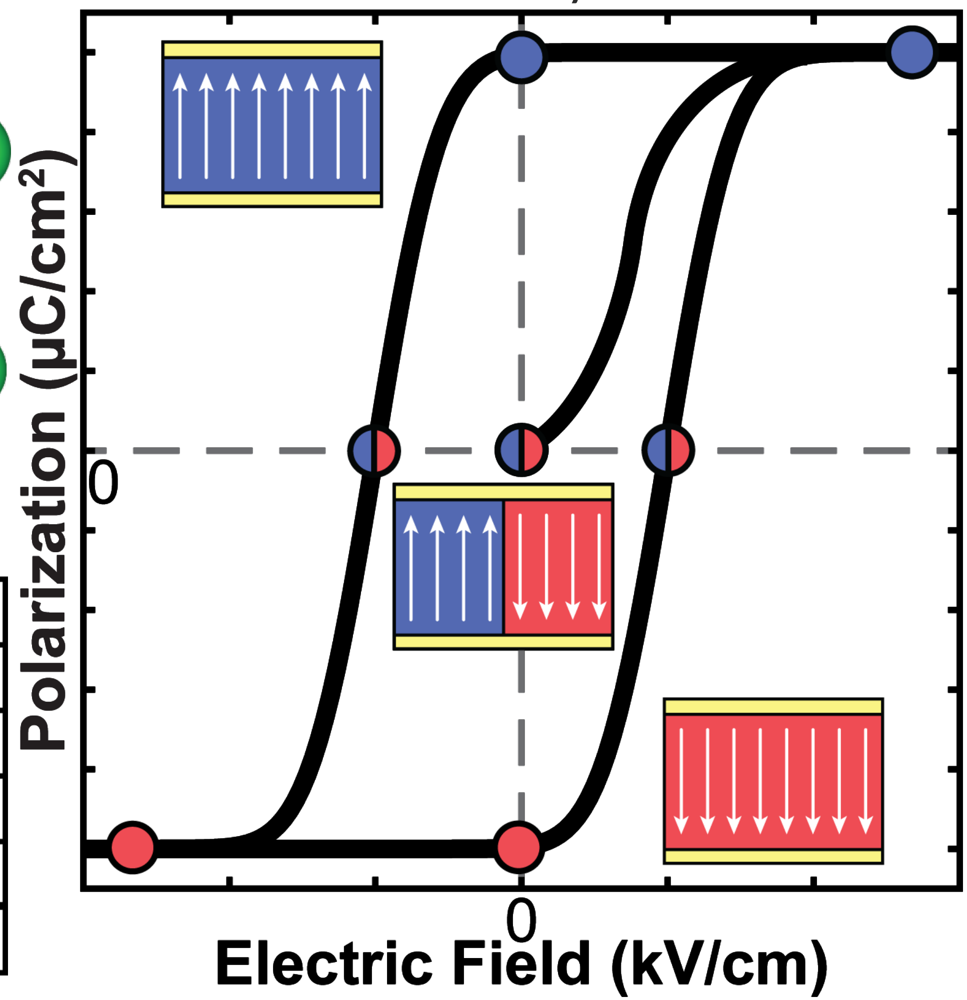


Ferroelectric Transition



- Susceptibility maximized near materials phase transitions

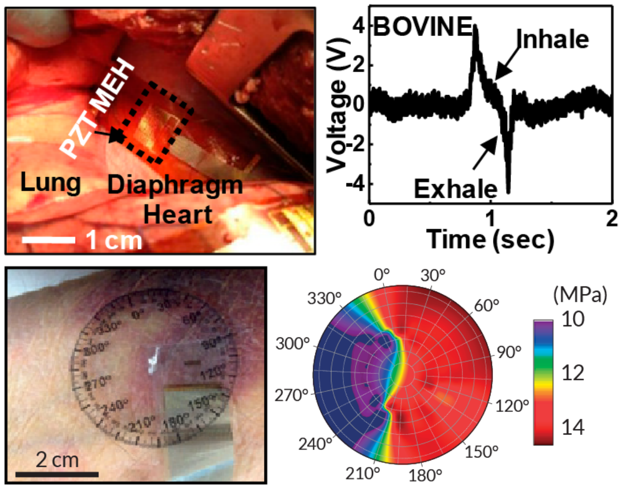
Ferroelectric Hysteresis



- Field switchable spontaneous polarization

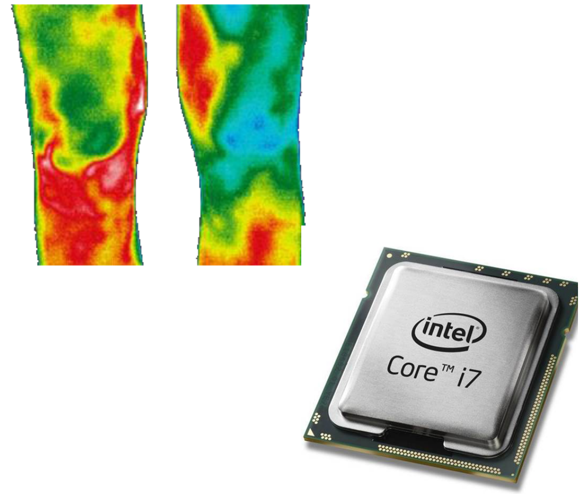
Multifunctional Ferroics

Piezoelectric



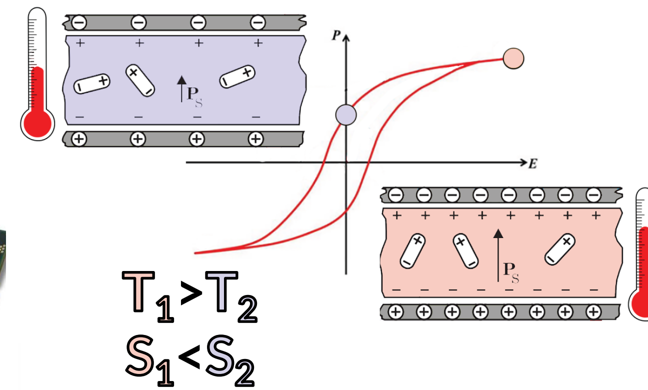
Dagdevirena *et al.*, *PNAS*. **111**, 5, 1927 (2013)
Dagdevirena *et al.*, *Nat. Mater.* **14**, 728 (2015)

Pyroelectric

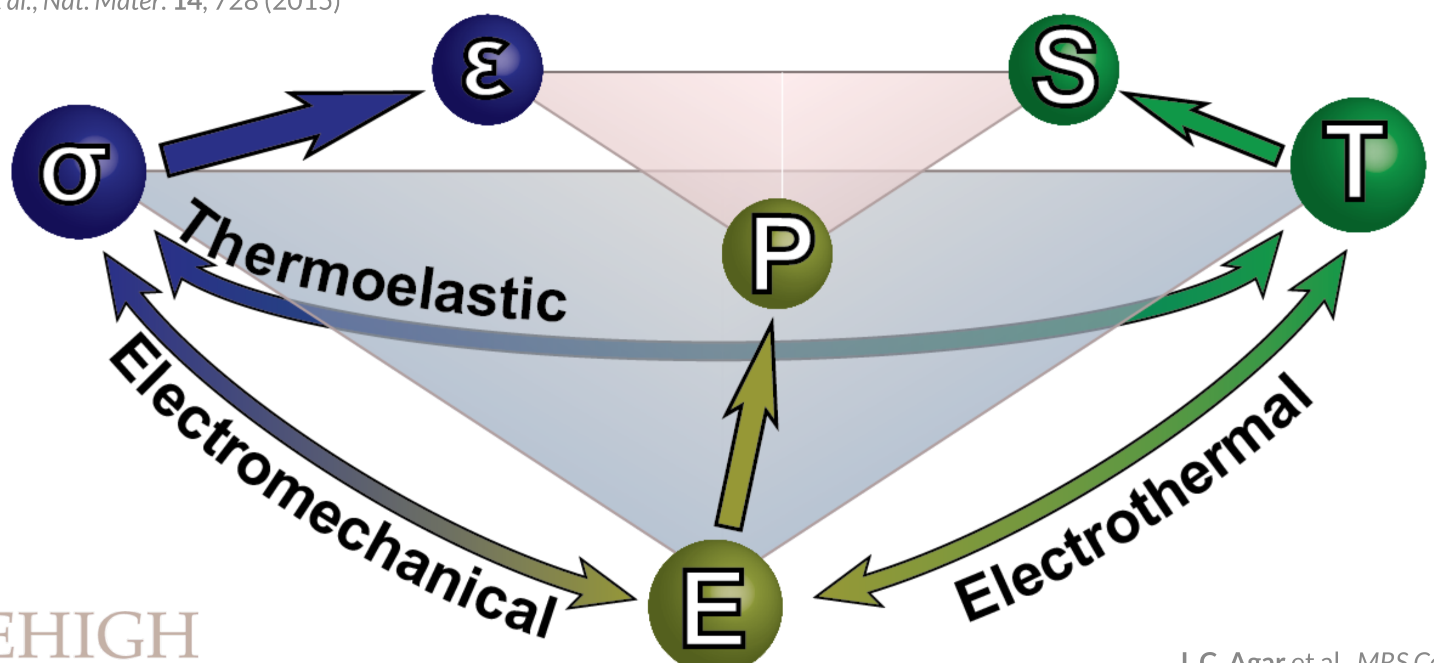


C. Hildebrandt *et al.*, *Sensors*. **10**, 4700 (2015)

Electrocaloric



S.B. Lang, *Phys. Today* **58**, 31 (2005)



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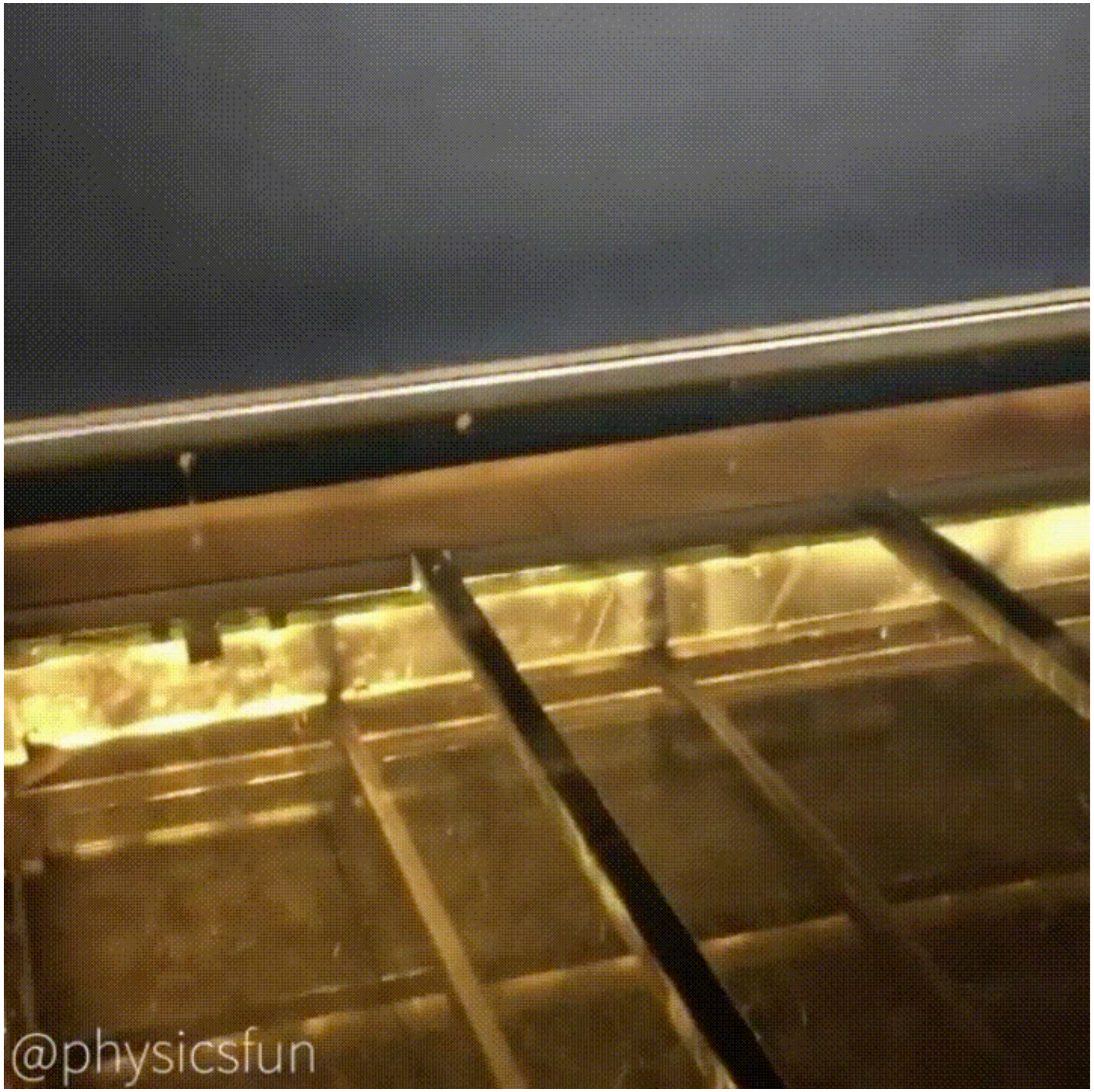
J. C. Agar et al., *MRS Commun.* **6**, (2016)

Need for Ferroelectrics



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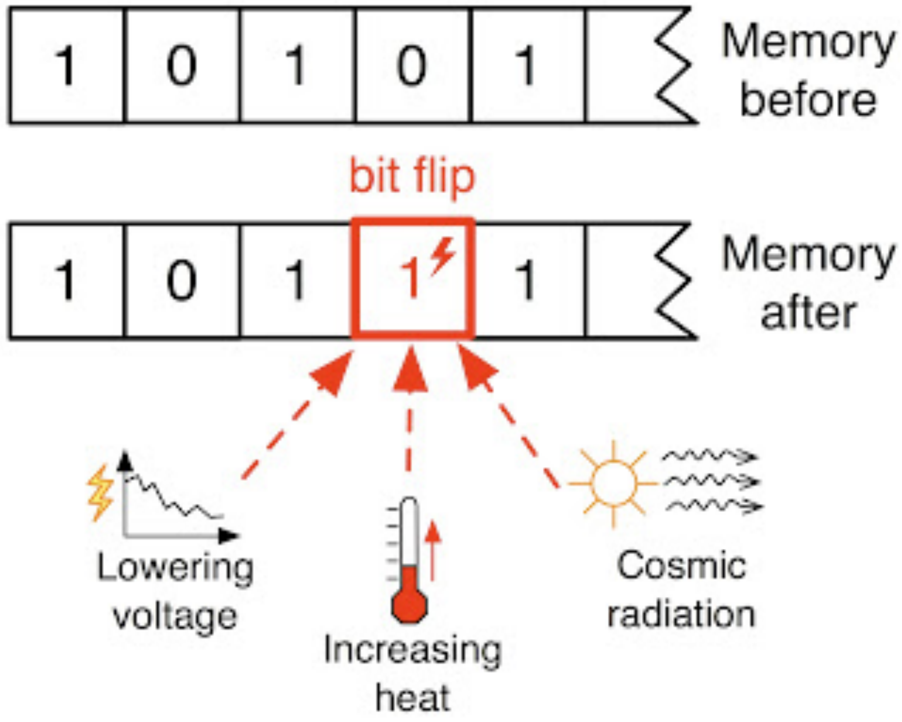
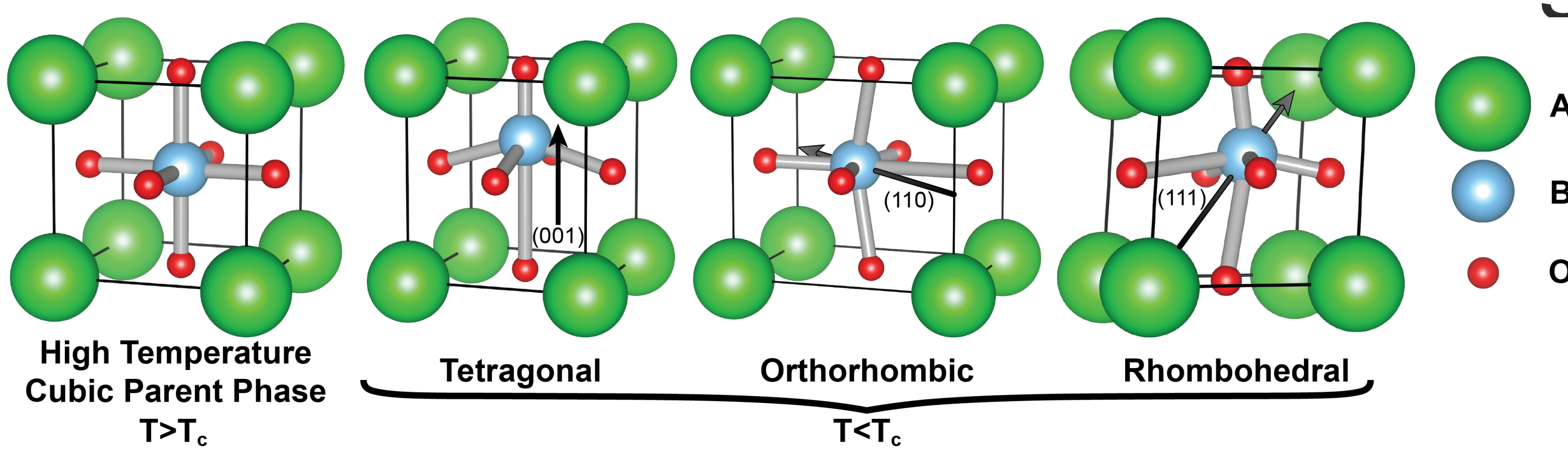
J. C. **Agar** et al., *MRS Commun.* **6**, (2016)

Need for Ferroelectrics

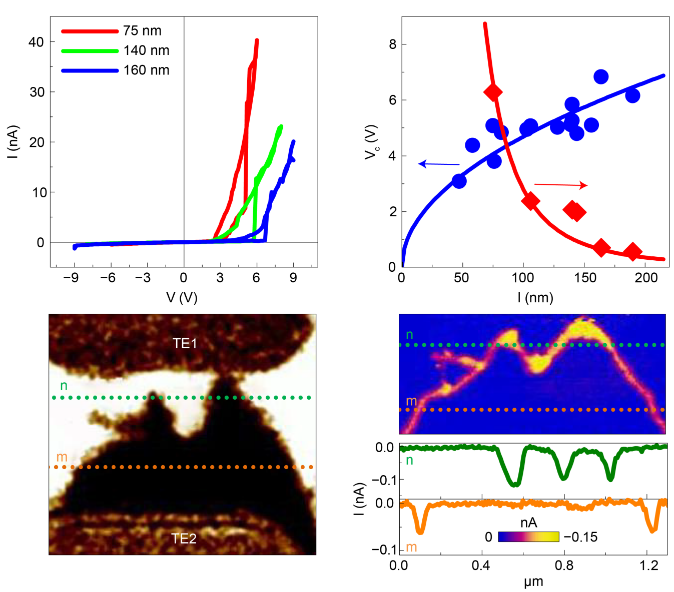


Controlled Ferroelectric Switching



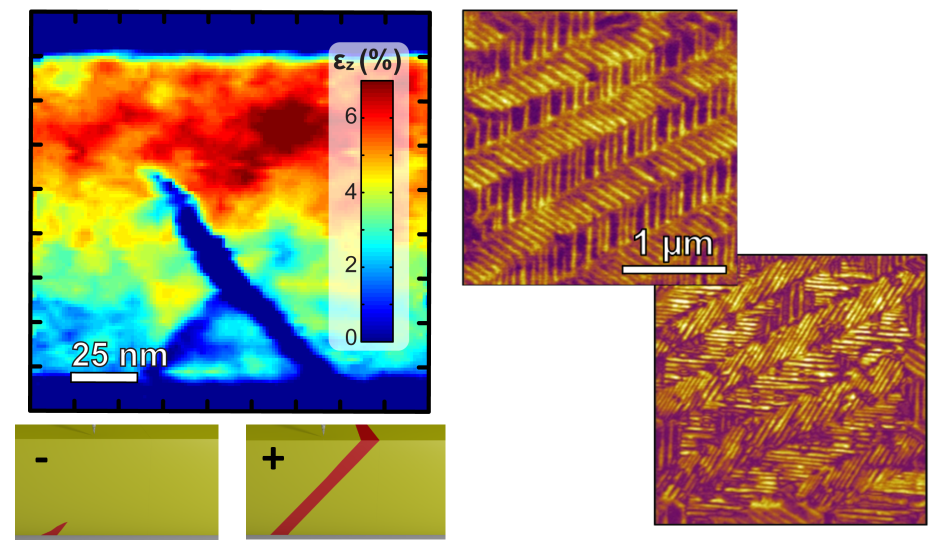
Controlled ferroelectric phase transformations → electrical conductivity, dielectric constant, elastic modulus, piezoresponse, etc.

Domain Wall Devices



J. Jiang *et. al. Nat. Mater.* **17**, 49, 2017

Complex Switching



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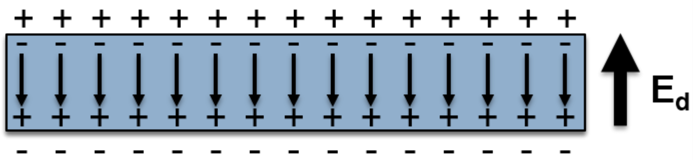
Ruijuan Xu

Ferroelectric Domain Structures

Designing domain structures → Manipulating energies

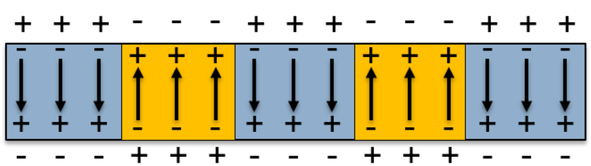
$$E_{Total} = E_{Landau} + E_{Depolarization} + E_{Strain} + E_{Domain\ Wall}$$

Landau/Bulk Energy ($E_{Lan.}$)



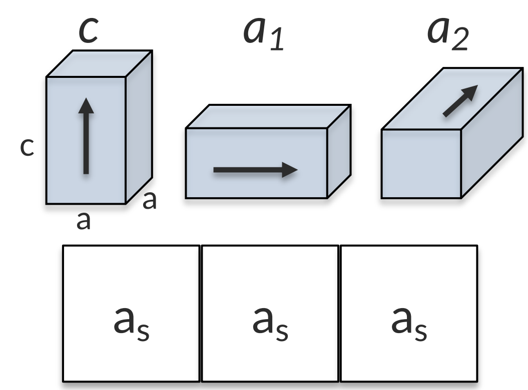
Orbital interaction → Polar distortion

Depolarization Energy (E_D)



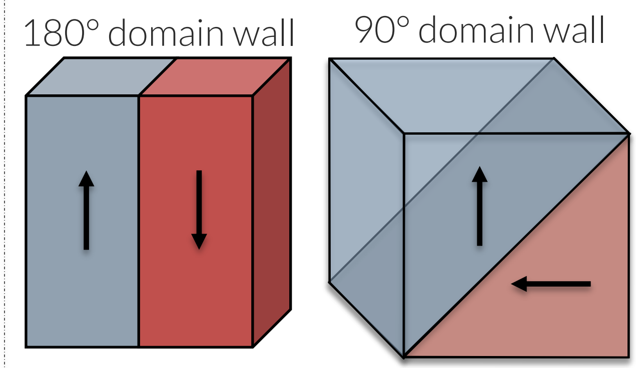
Minimize charge asymmetry

Strain Energy (E_S)



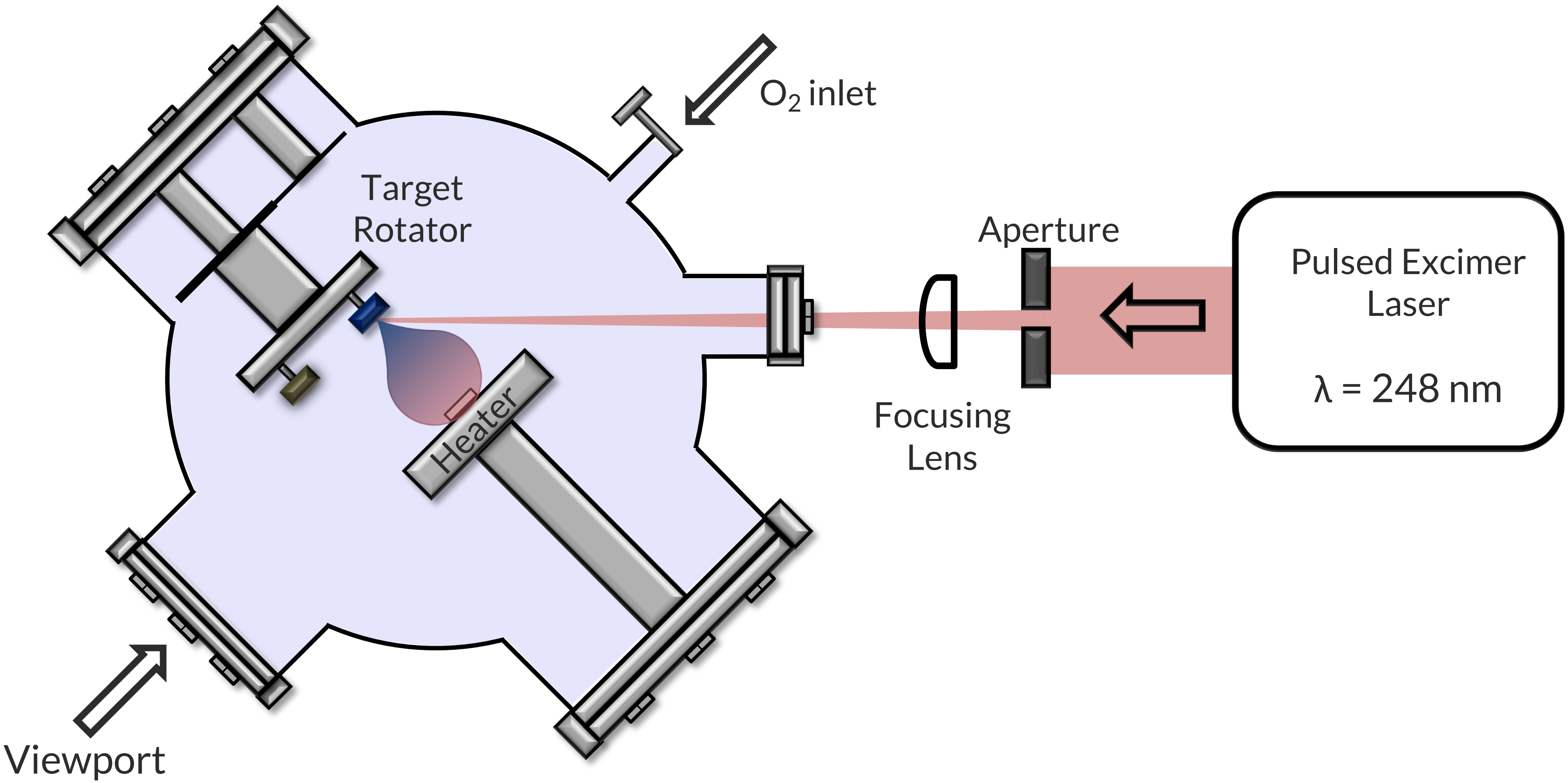
Minimize average lattice mismatch
→ Varying unit-cell orientation

Domain Wall Energy (E_{DW})



Minimize ∇P → Reduce # of DW +
only head-to-tail DW

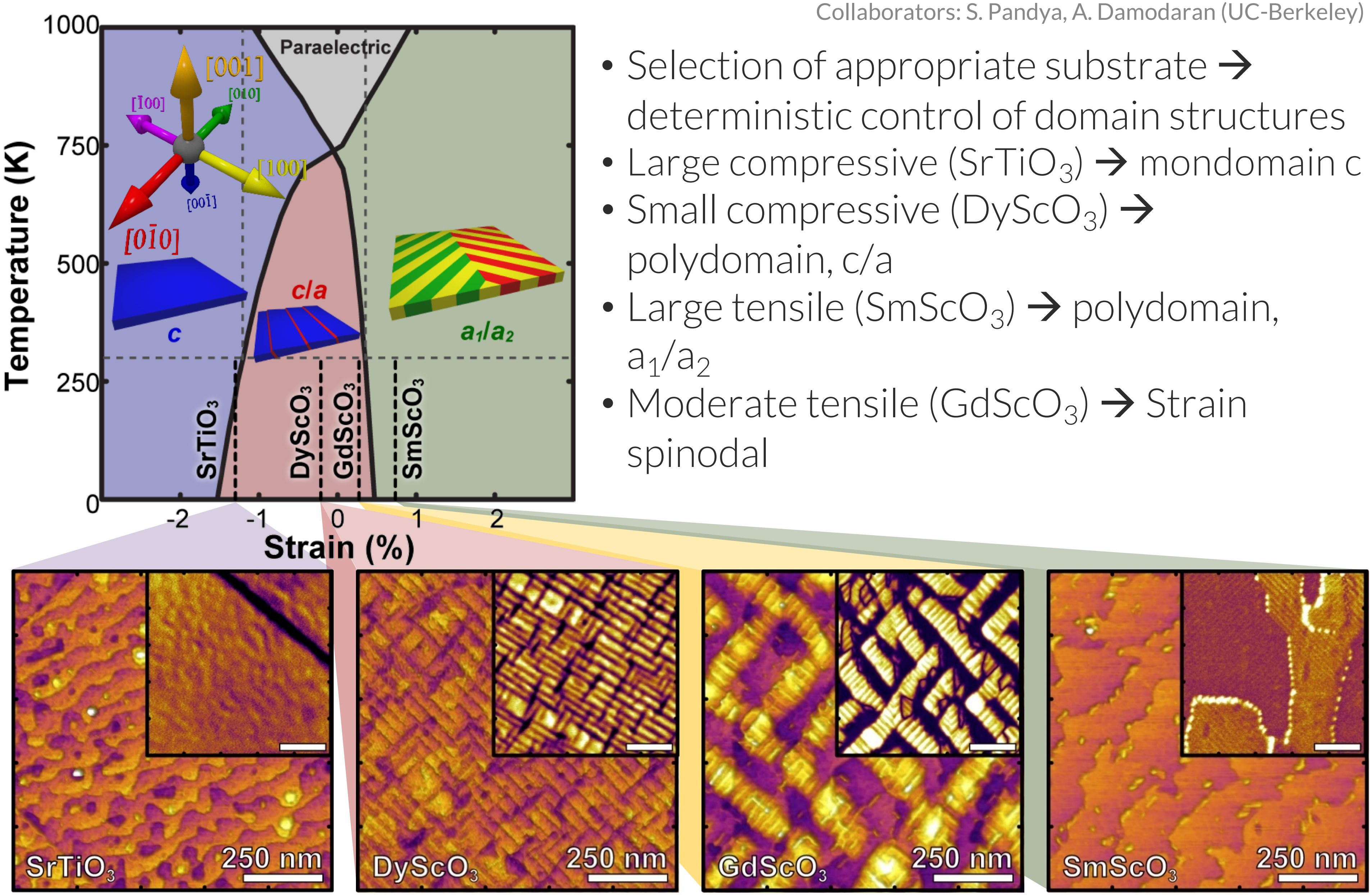
Thin-Film Epitaxy Pulsed-Laser Deposition



- Epitaxial Heterostructures with unit-cell-level control
- Growth of a wide variety of complex materials systems
- Stoichiometric and defect control during growth

Growth of Tensile Strained $PbTiO_3$

Collaborators: S. Pandya, A. Damodaran (UC-Berkeley)



- Selection of appropriate substrate → deterministic control of domain structures
- Large compressive ($SrTiO_3$) → mondomain c
- Small compressive ($DyScO_3$) → polydomain, c/a
- Large tensile ($SmScO_3$) → polydomain, a_1/a_2
- Moderate tensile ($GdScO_3$) → Strain spinodal

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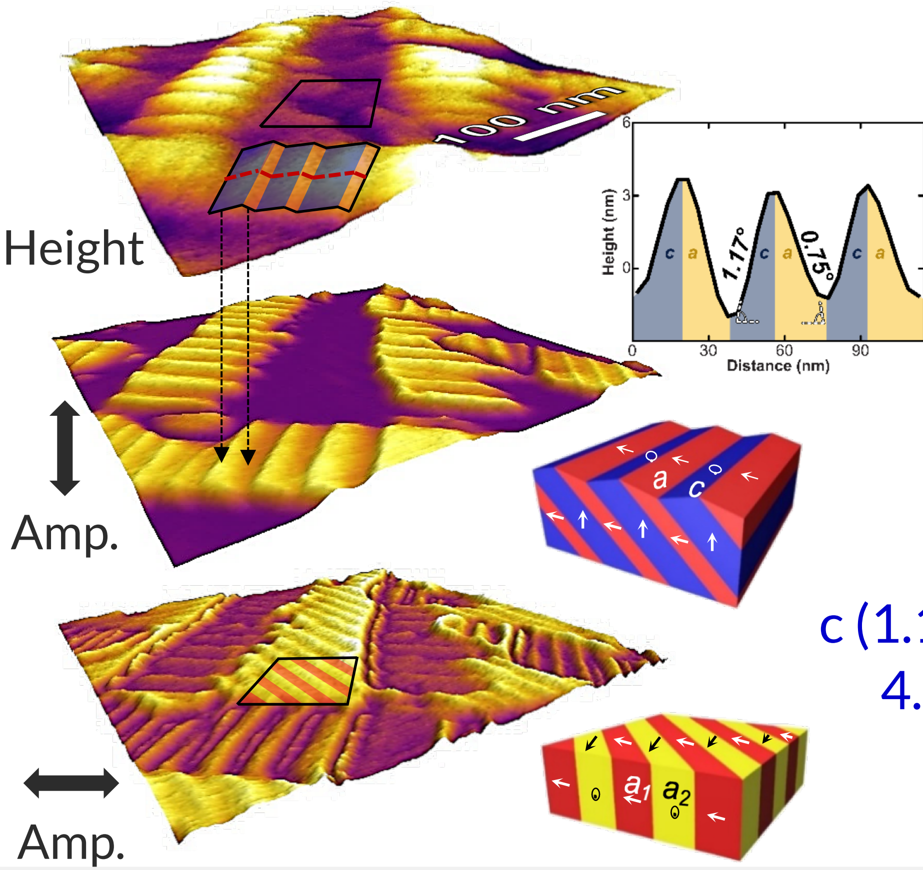
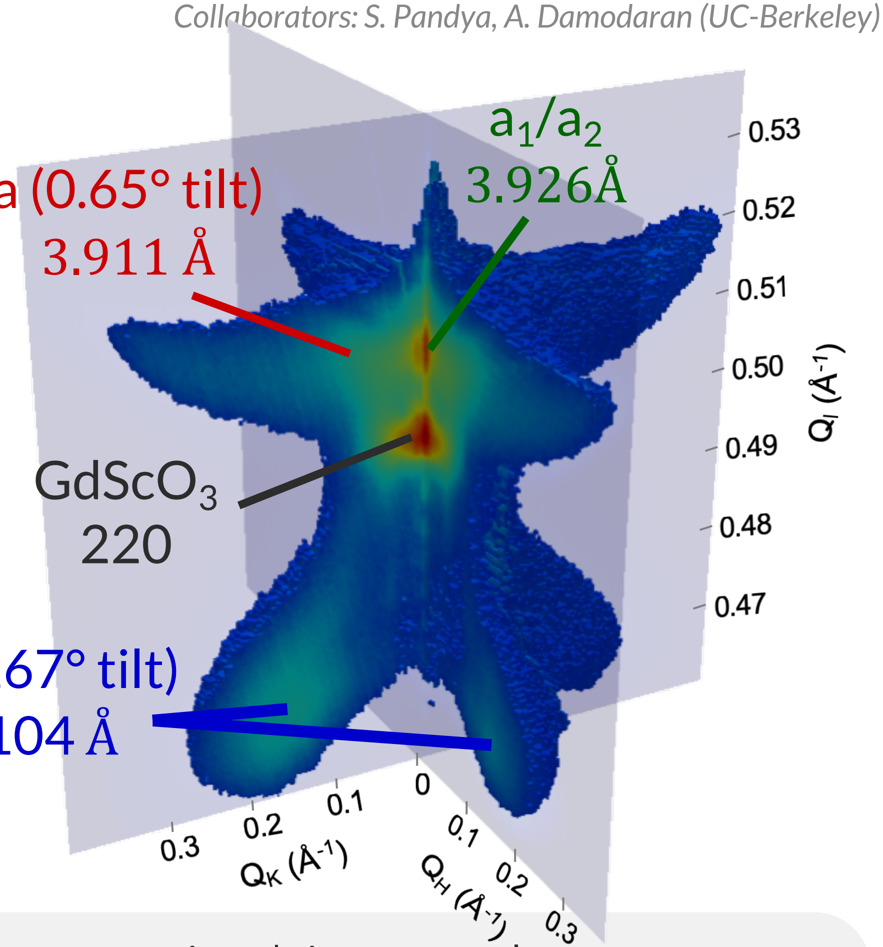
Understanding Hierarchical Domain Structures

Collaborators: S. Pandya, A. Damodaran (UC-Berkeley)





1. How does ferroelectric switching occur in this complex structure?
2. What are its implications for the dielectric, piezoelectric, and pyroelectric susceptibilities?

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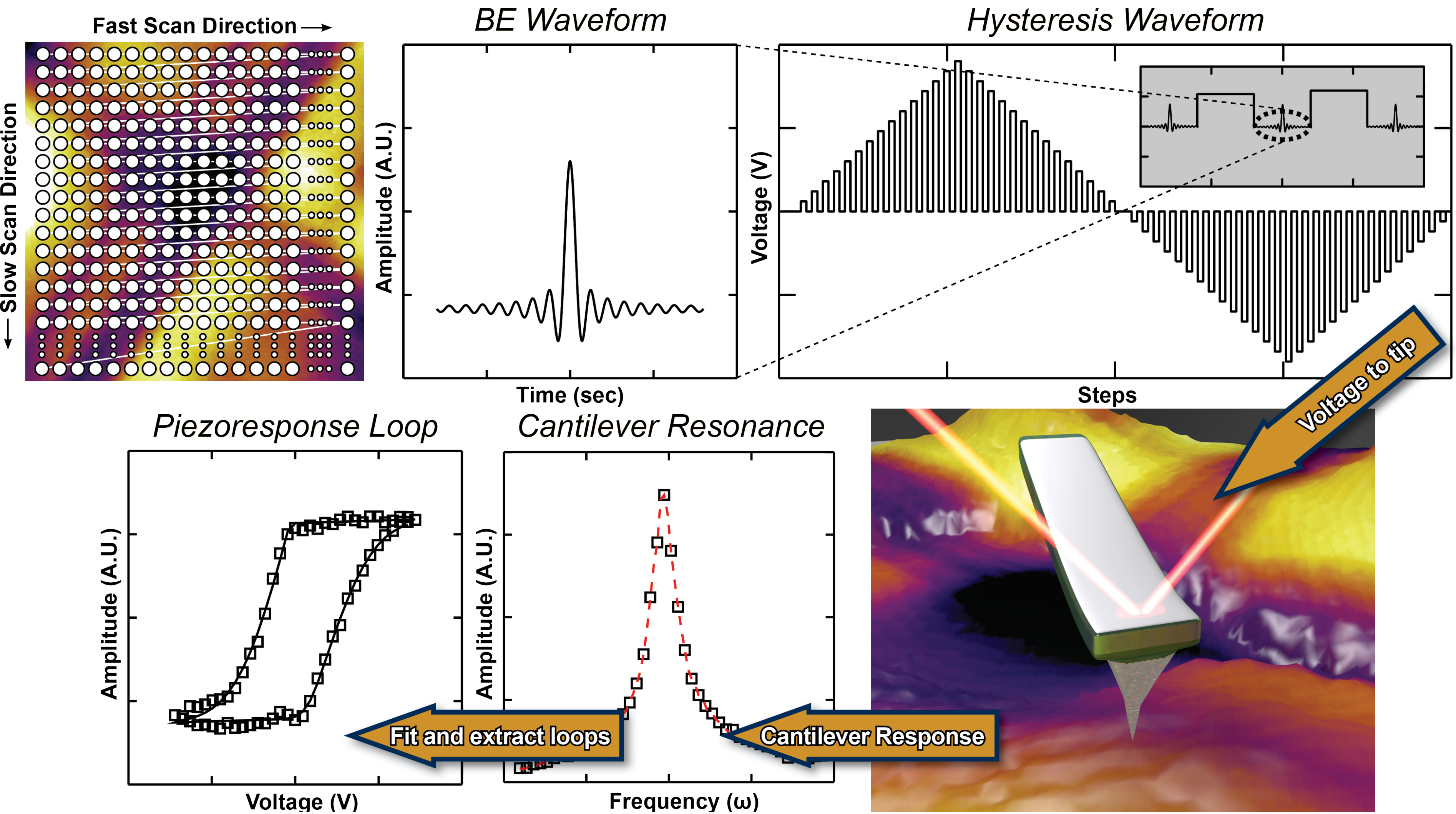
Band Excitation Piezoresponse Spectroscopy

Collaborators: R. Vasudevan, S. Jesse, N. Balke and S. Kalinin (Oak Ridge National Laboratory)

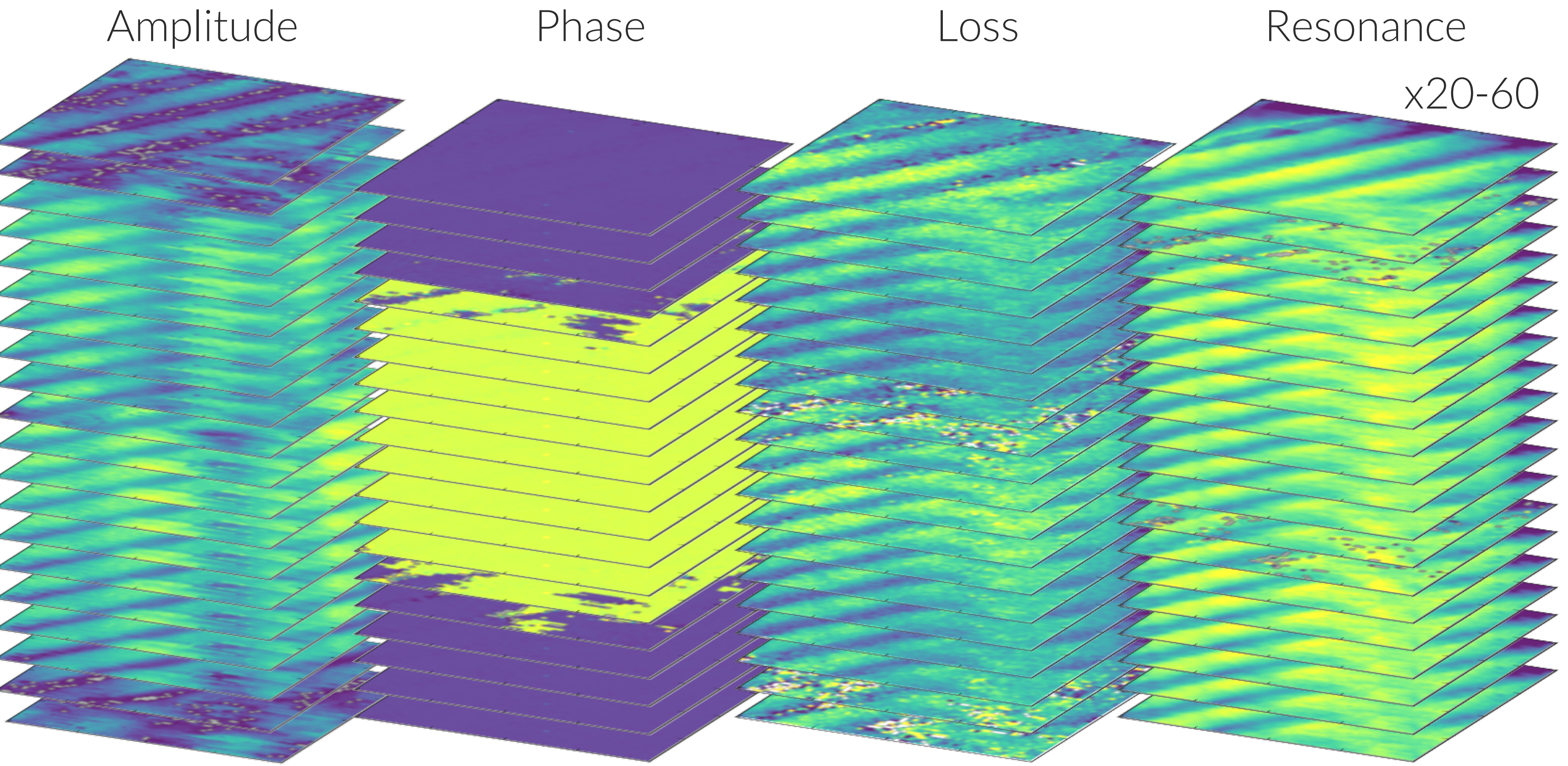


Band excitation (BE) PFM allows the spatially resolved measure of piezoresponse, modulus, and electromechanical dissipation

Visualizing Ferroelastic Switching

BE-PFM → contains the information → untenable to analyze

Piezoresponse = A cos(ϕ); (x = 40-100, y = 40-100, V = 50-700, cycle = 1-6)



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Visualizing Ferroelastic Switching

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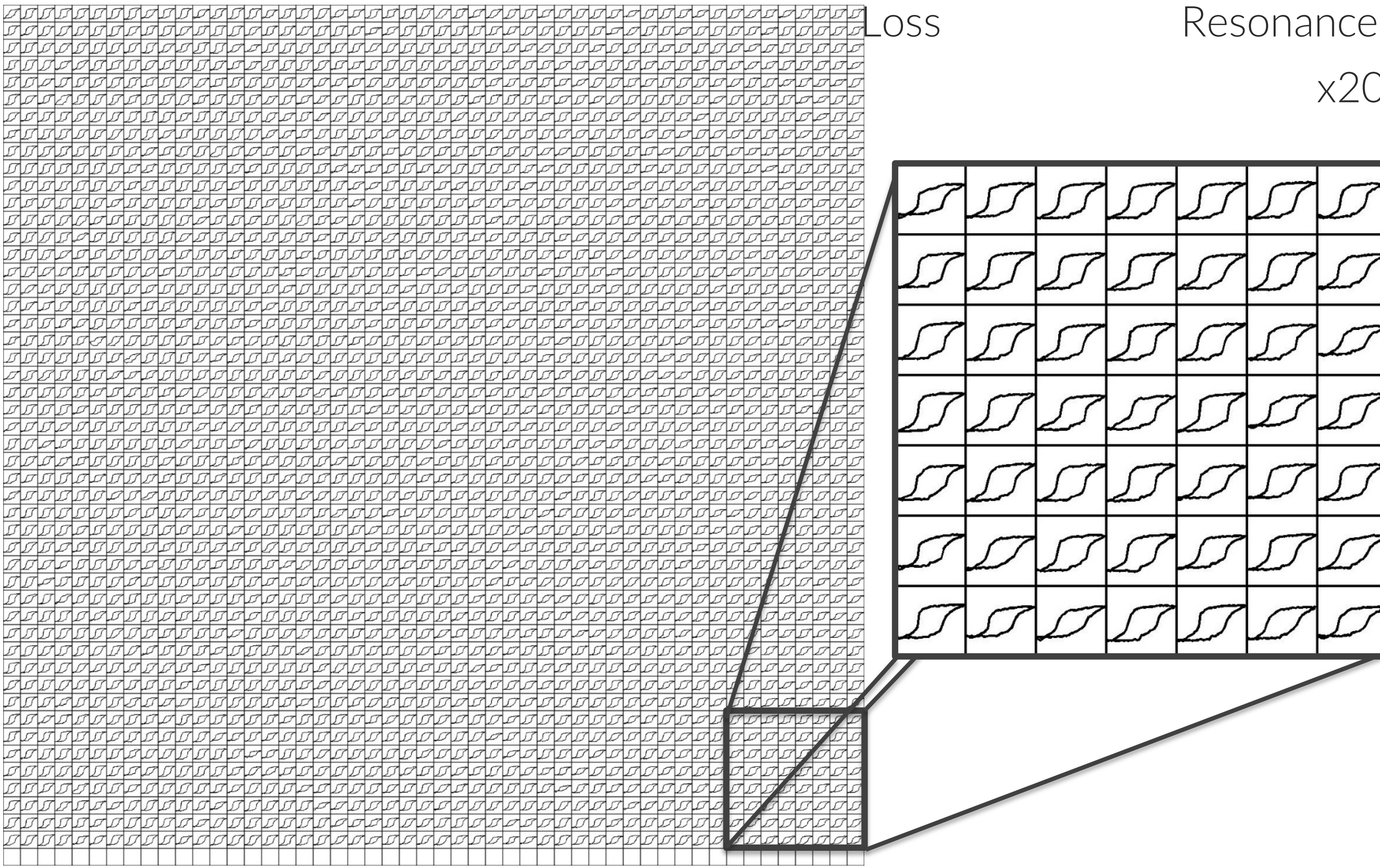
Loss

Resonance

x20-60



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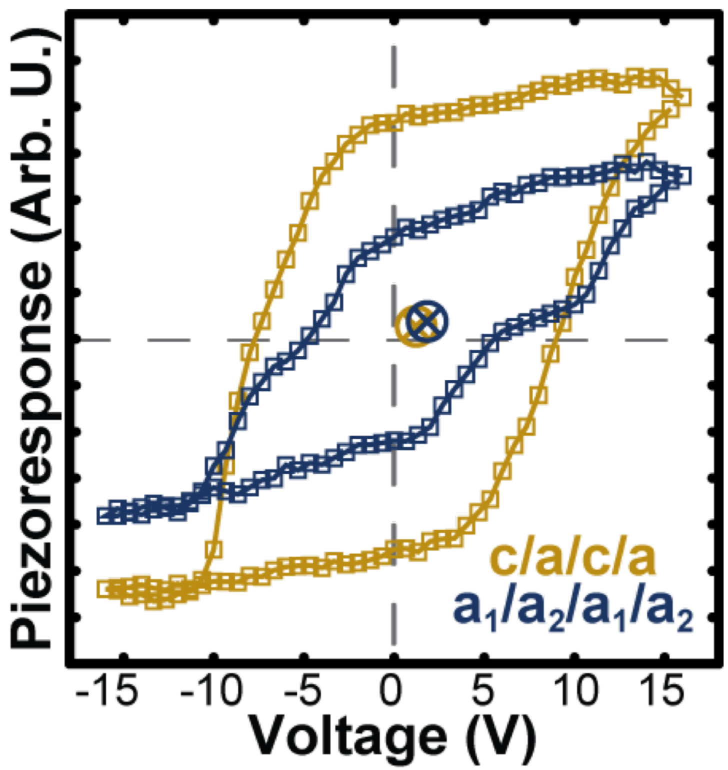
Machine Learning Ferroelastic Switching

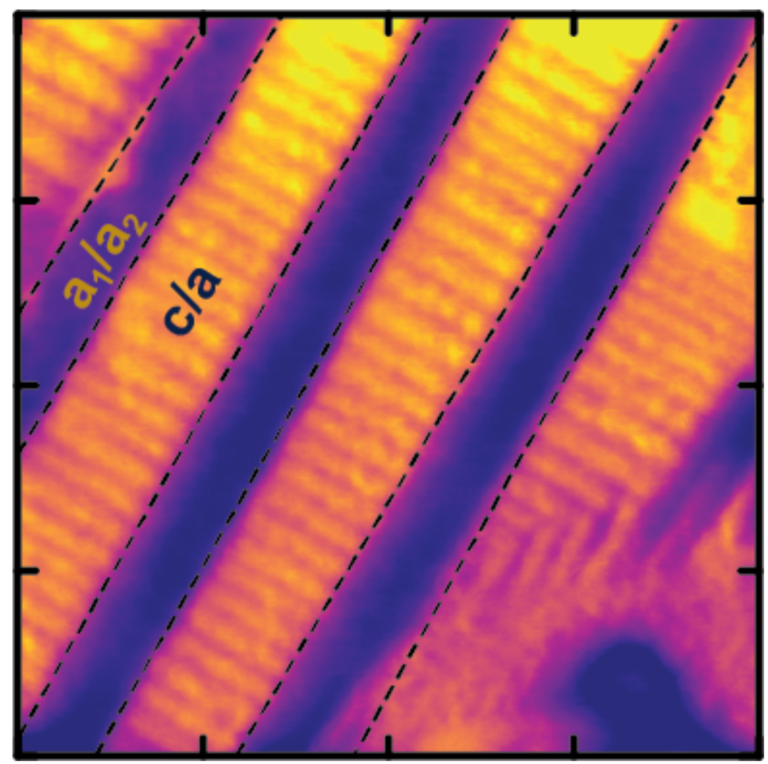
Identify characteristic features of piezoresponse hysteresis

Generalizability



- Discover unknown or difficult to quantify features

Interpretability



- Representation needs to be physically interpretable

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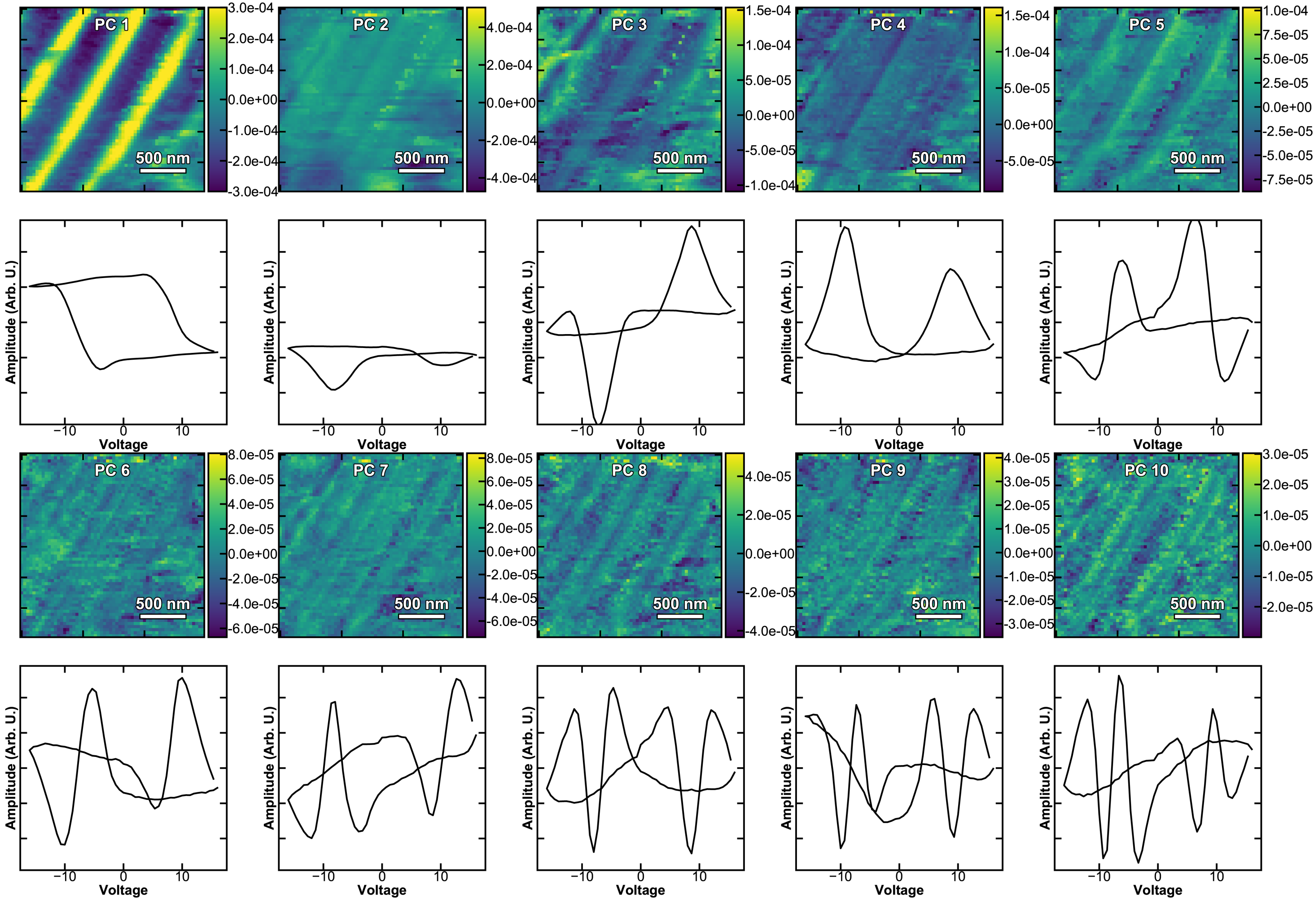
Principal Component Dimensionality Reduction

Constructs a set of orthogonal linear eigenvectors and eigenvalues ranked in terms of the variance explained



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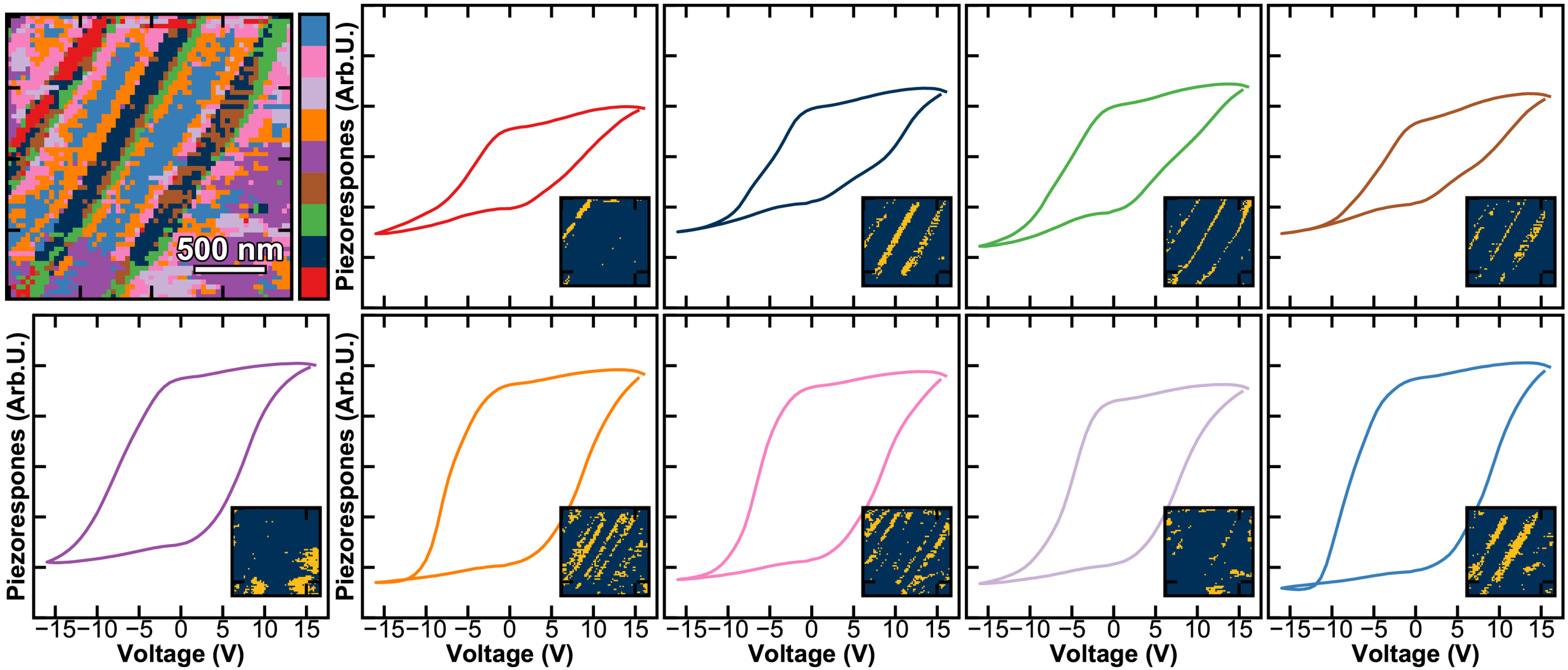
k-Means Clustering

Clusters data into k-clusters of equal variance



- Clustering identifies regions of interest which match domain structures
- Cannot account for mixed responses

Can we design a deep learning neural network for this problem?

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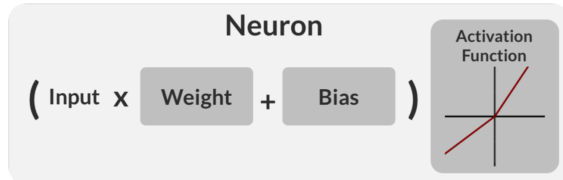
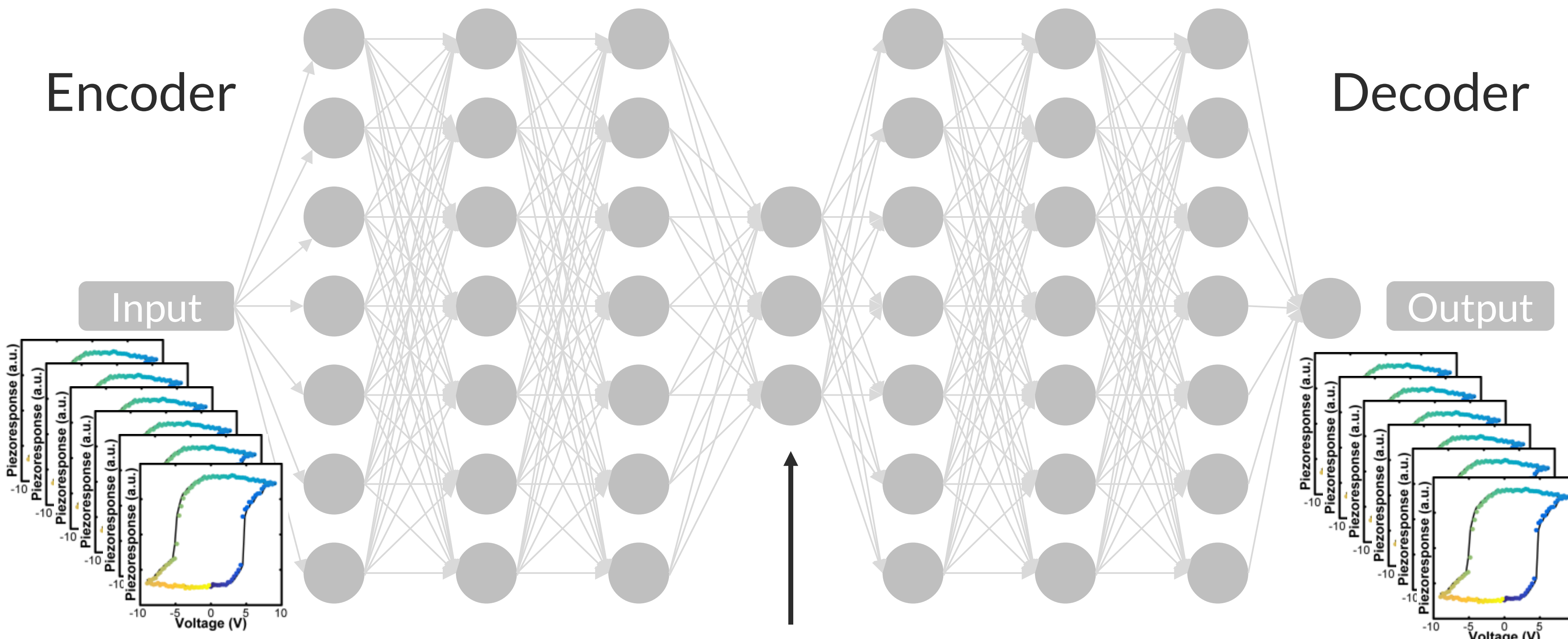
How to Make a Neural Network Learn Features?

Force a network to learn an identity function through a highly constrained layer



Autoencoder



Low-dimensional layer

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How to Train a Neural Network on Piezoelectric Hysteresis loops

Piezoelectric loops have temporal (time) dependence → need architecture which accounts for this structure

Long-Short Term Memory Recurrent Neural Networks

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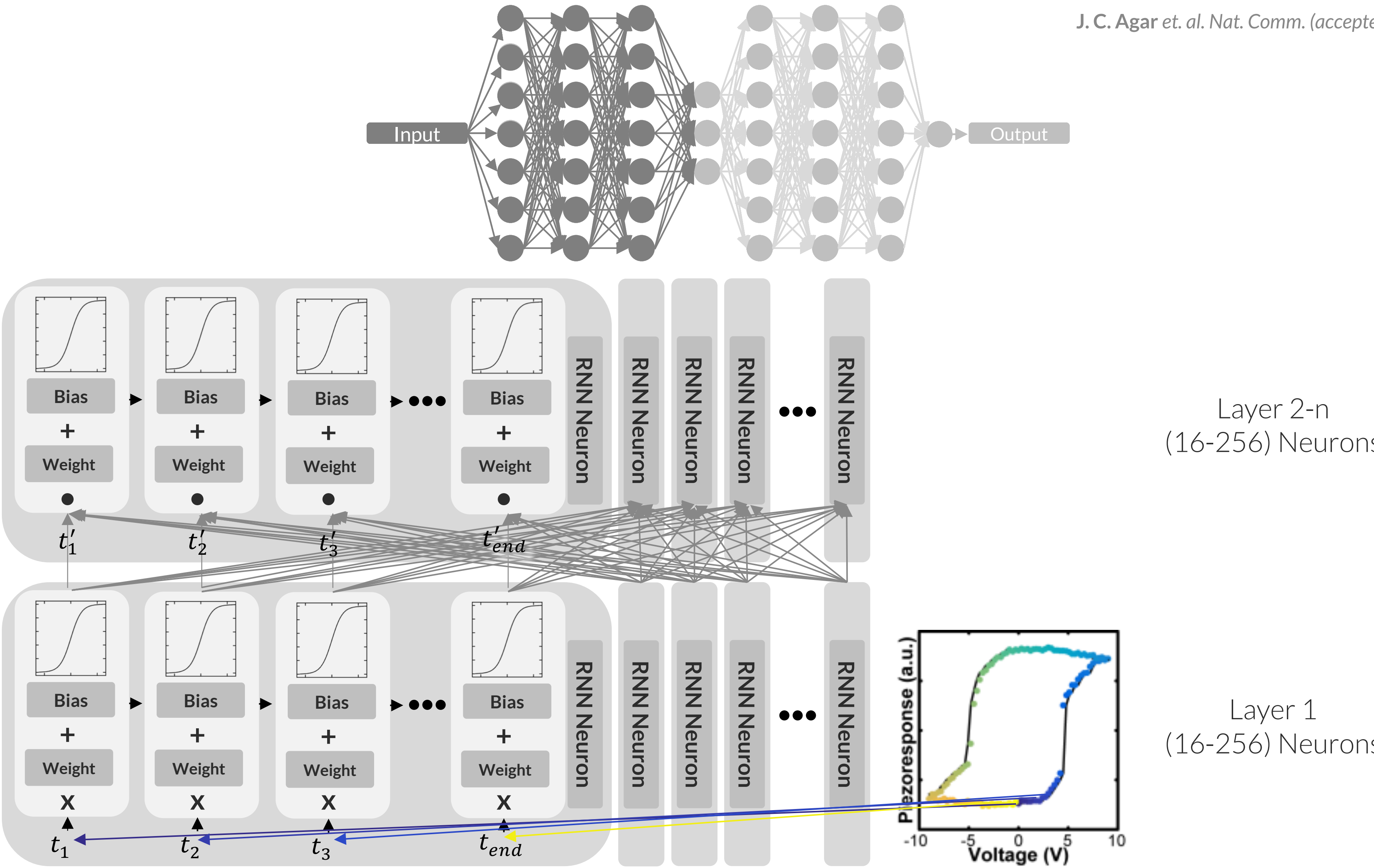

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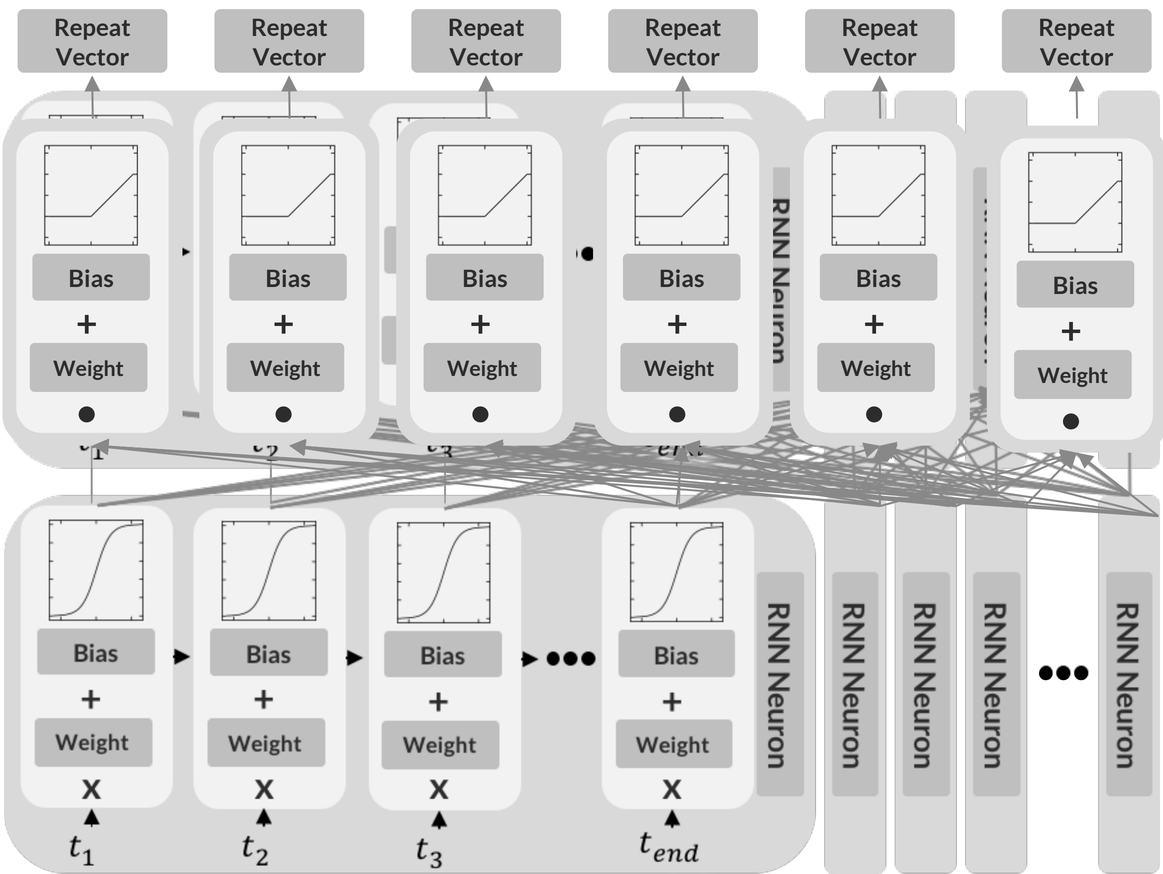

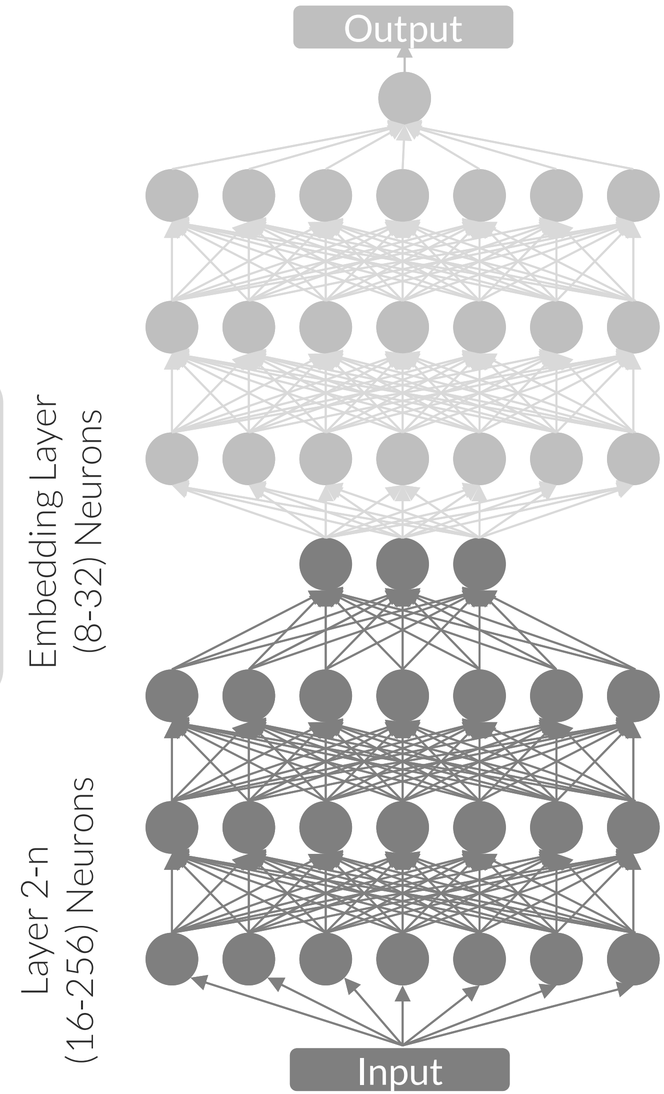

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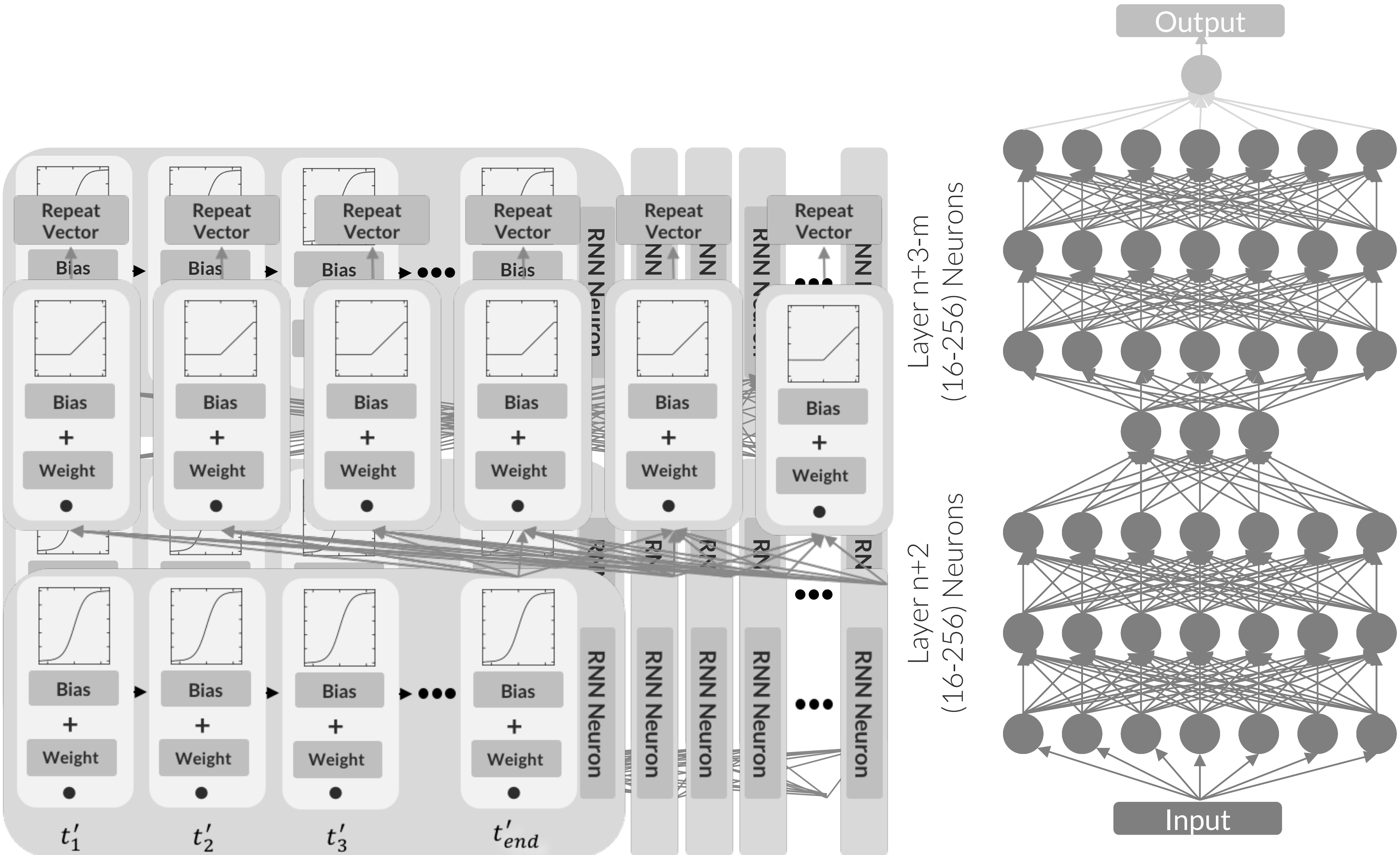

How to Train a Neural Network on Piezoelectric Hysteresis loops

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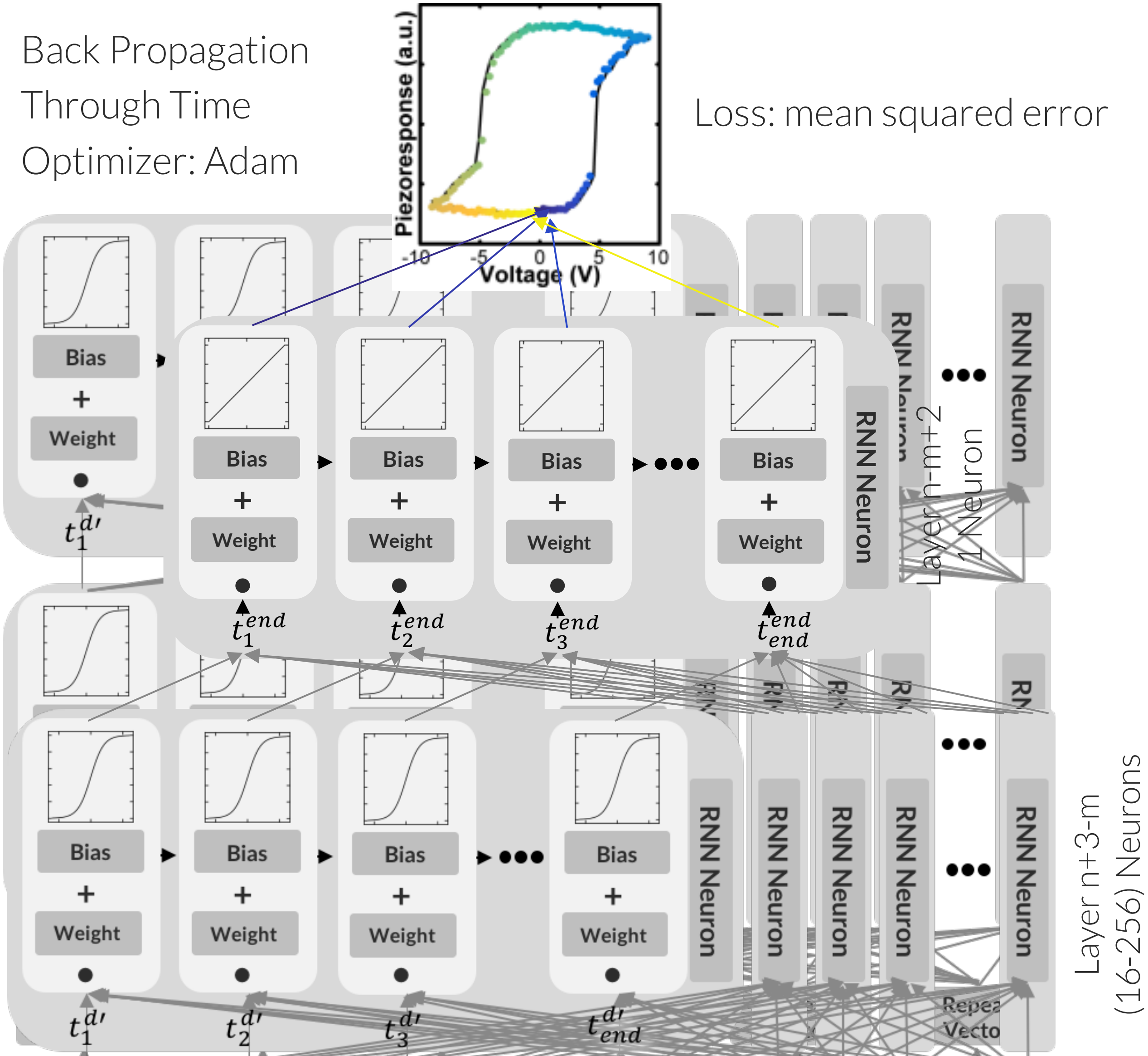
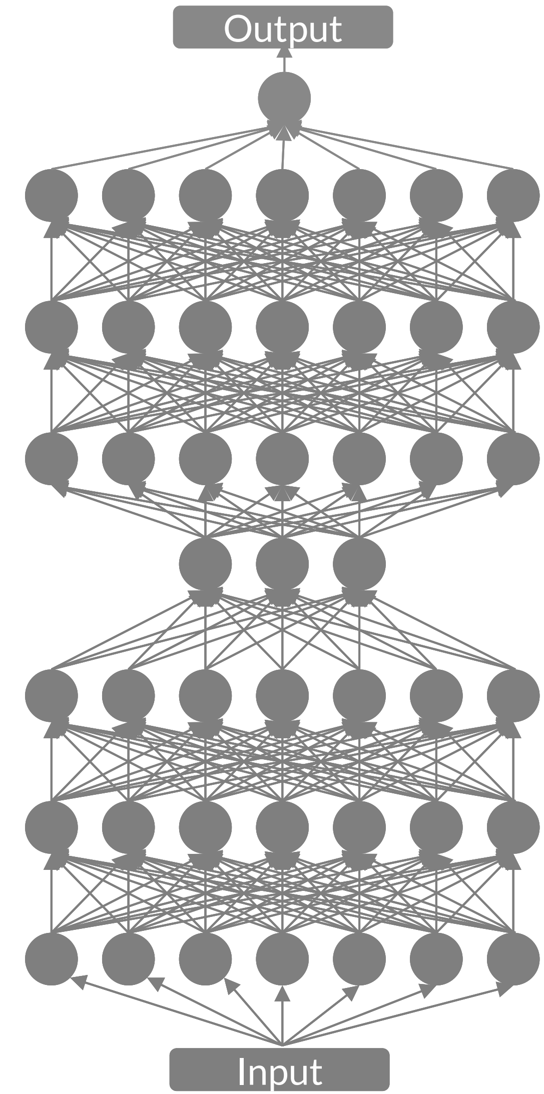
Long-Short Term Memory Recurrent Neural Networks

Back Propagation Through Time

Optimizer: Adam

Loss: mean squared error





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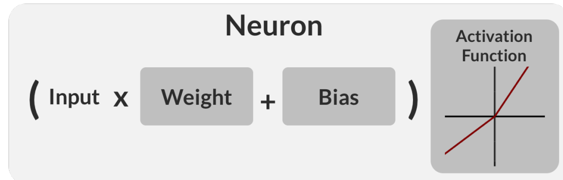

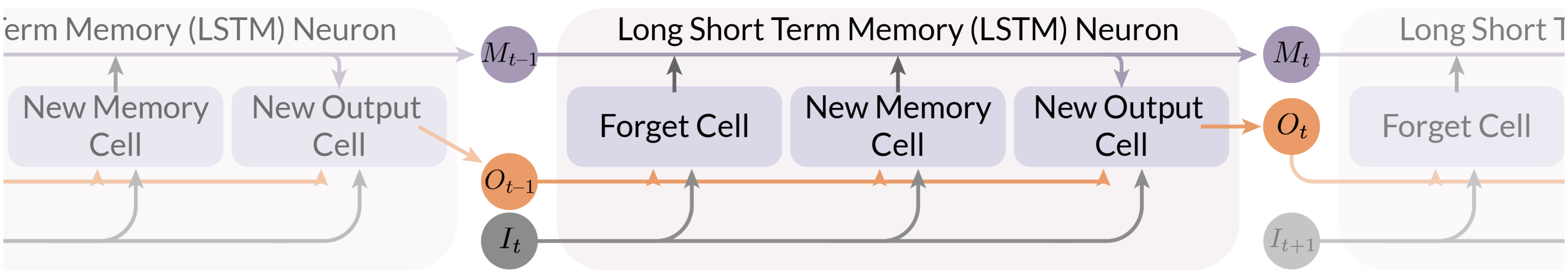

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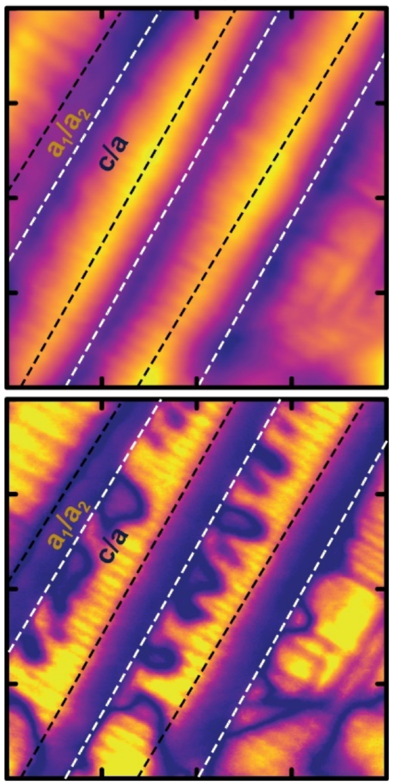
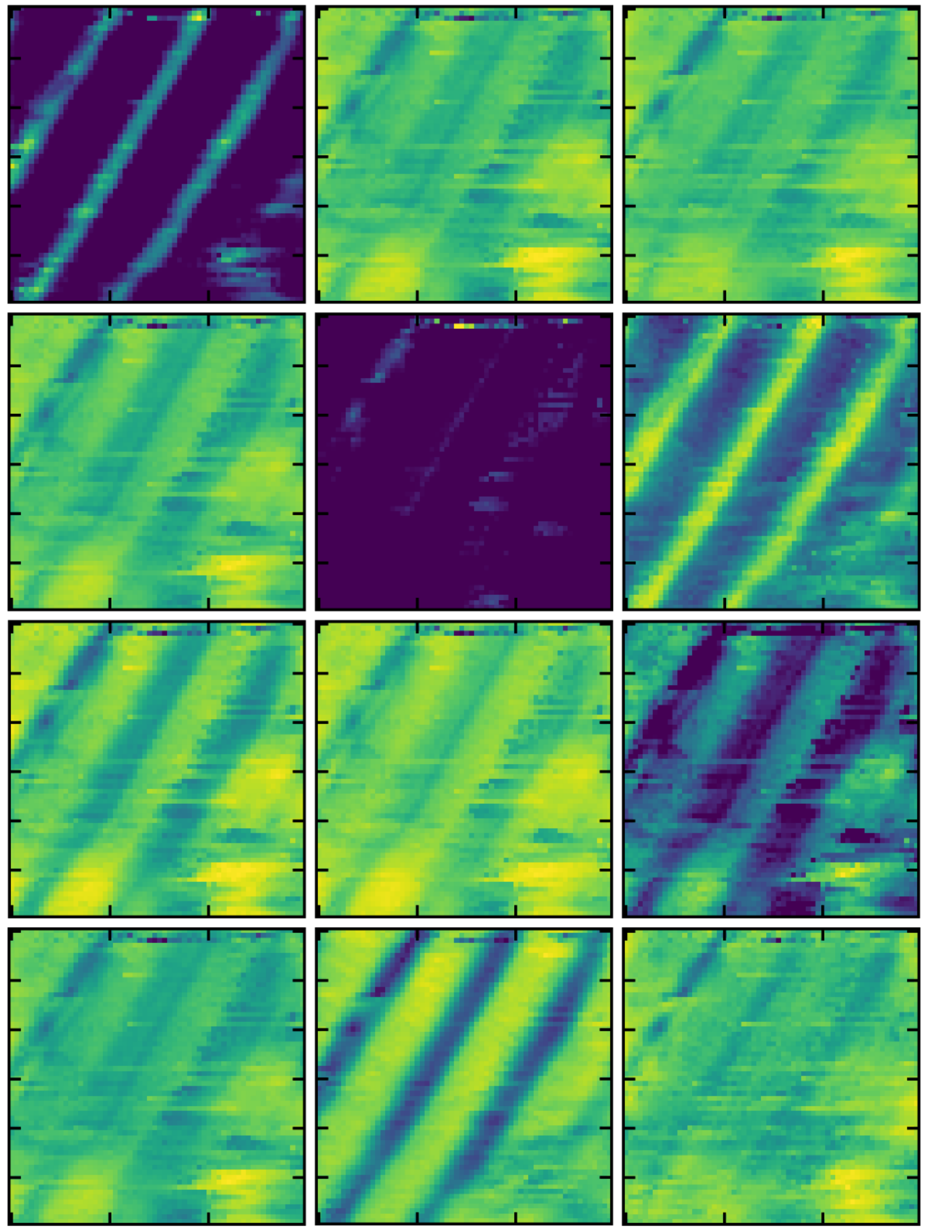
Analysis of Neural Network

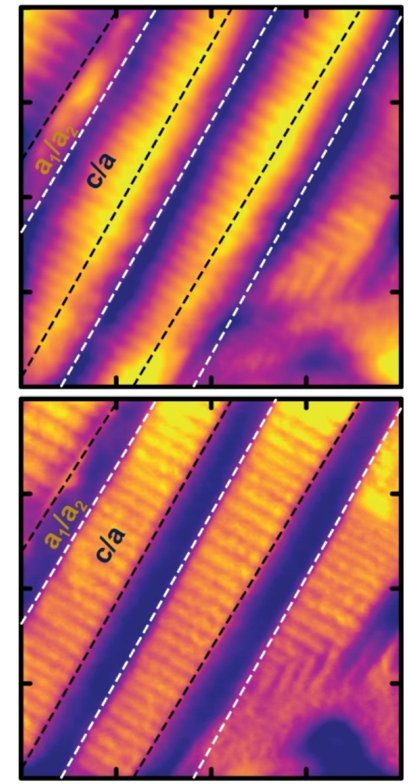
Compute activation from low-dimensional layer → reconstruct maps

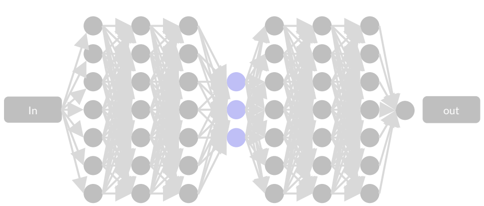
Initial





Final





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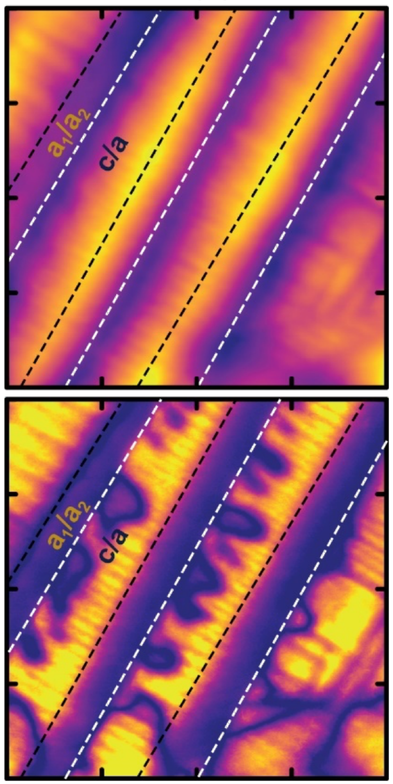


Enforcing Sparsity

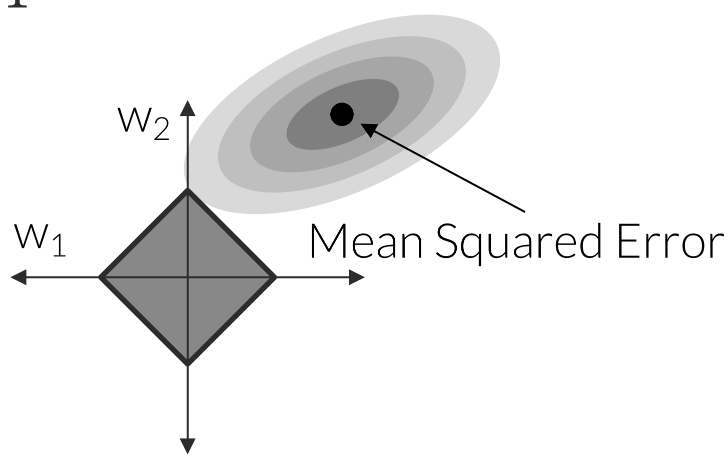
Loss Function

$$Loss = \frac{1}{n}\sum_{i=1}^{n}(Y_i - \hat{Y}_i)^2 + \lambda \sum |w_i|$$

Mean Squared Error

l_1 - normalization

l_1 = 1 isosurface



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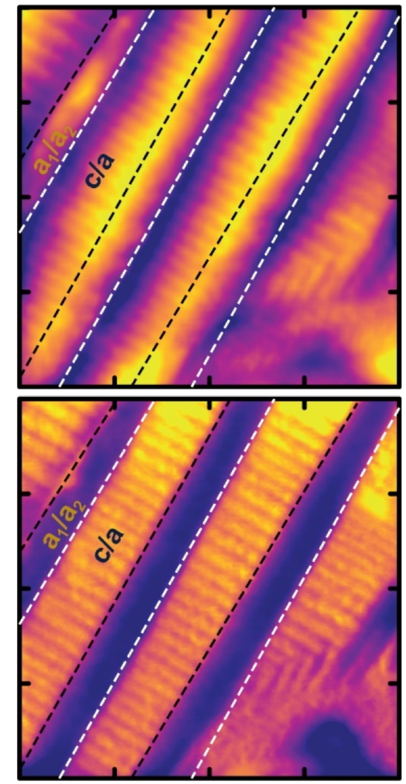

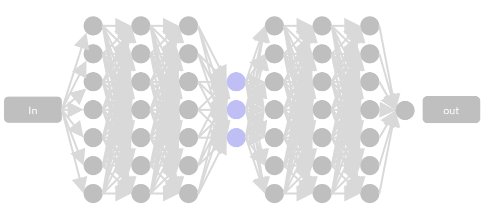

- l_x (x<1) imposes sparsity

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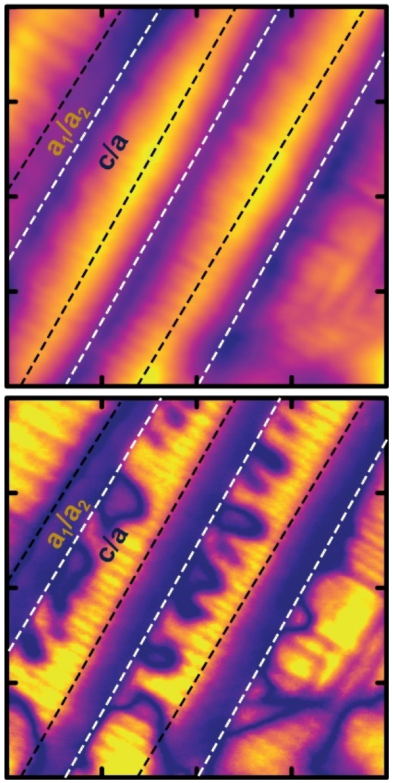

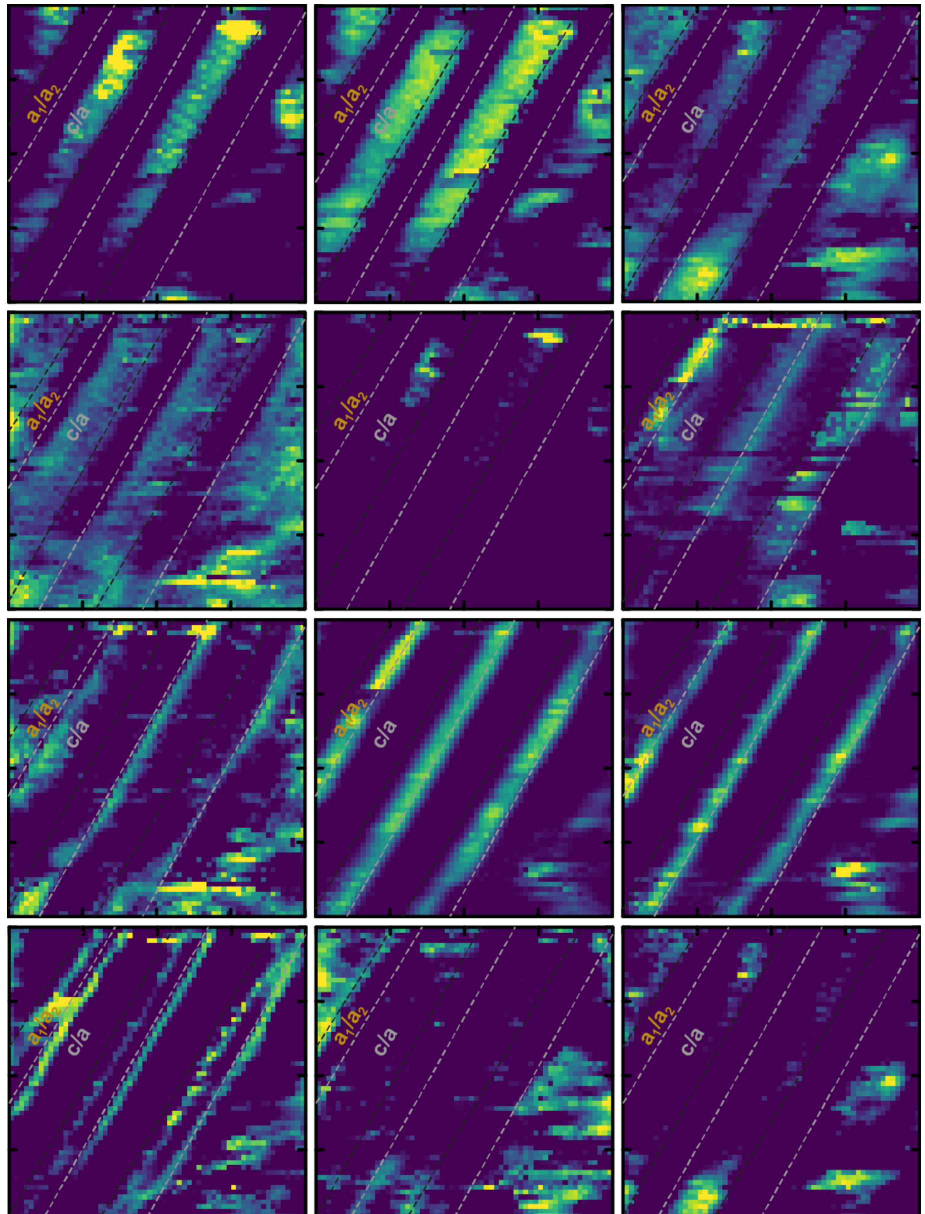

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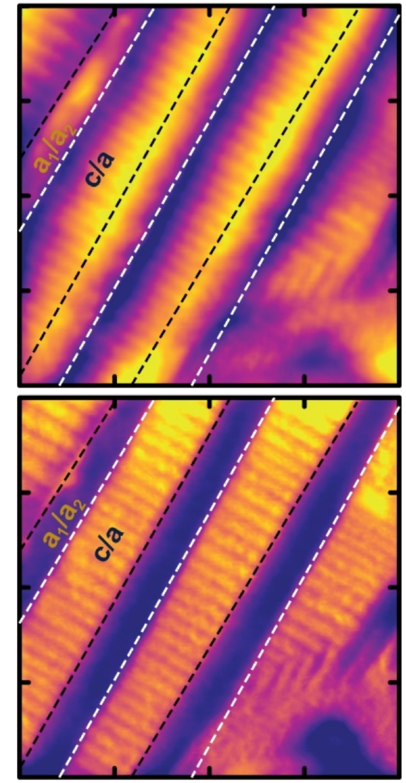

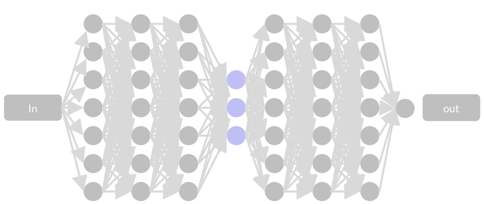

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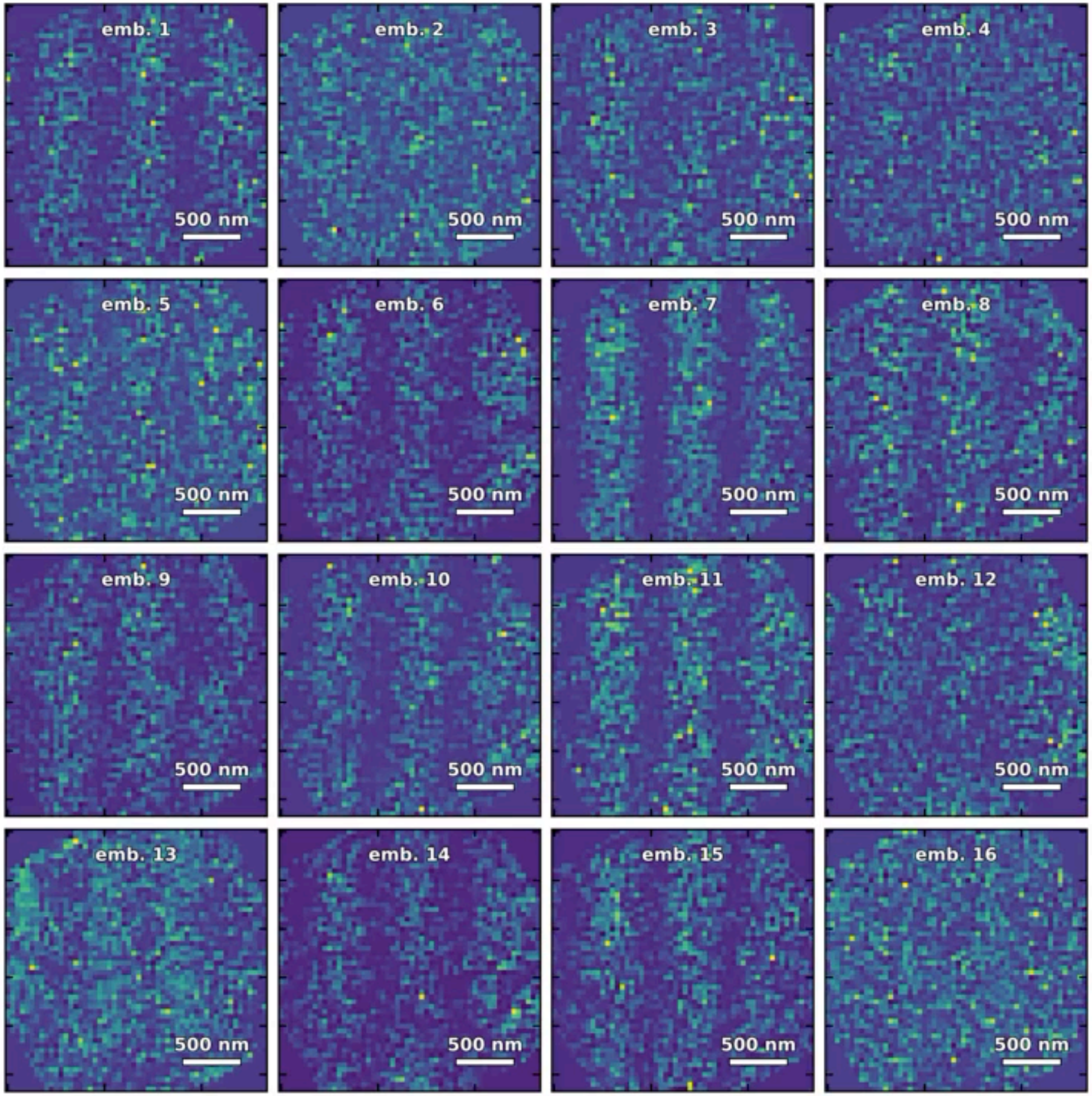
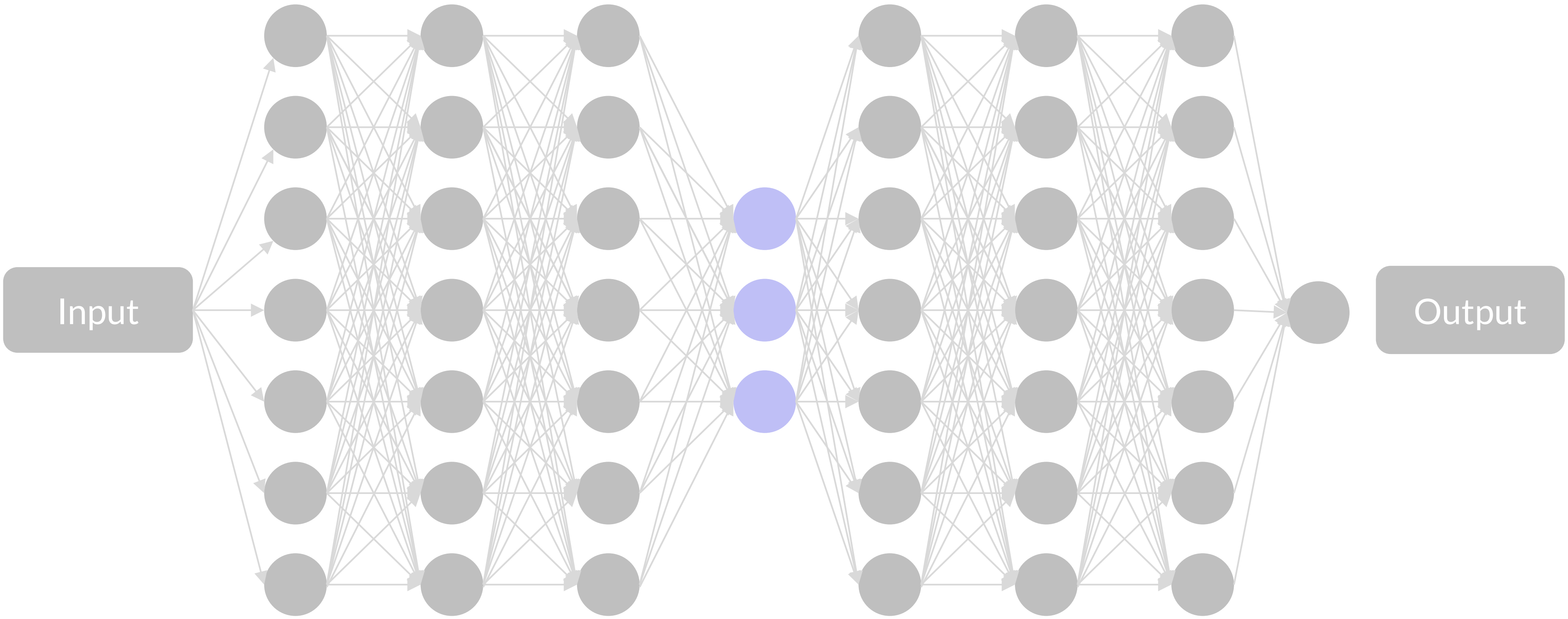
Visualization of Learning Process



Analysis of Neural Network

Compute activation from low-dimensional layer → reconstruct maps

Autoencoder as a Generator



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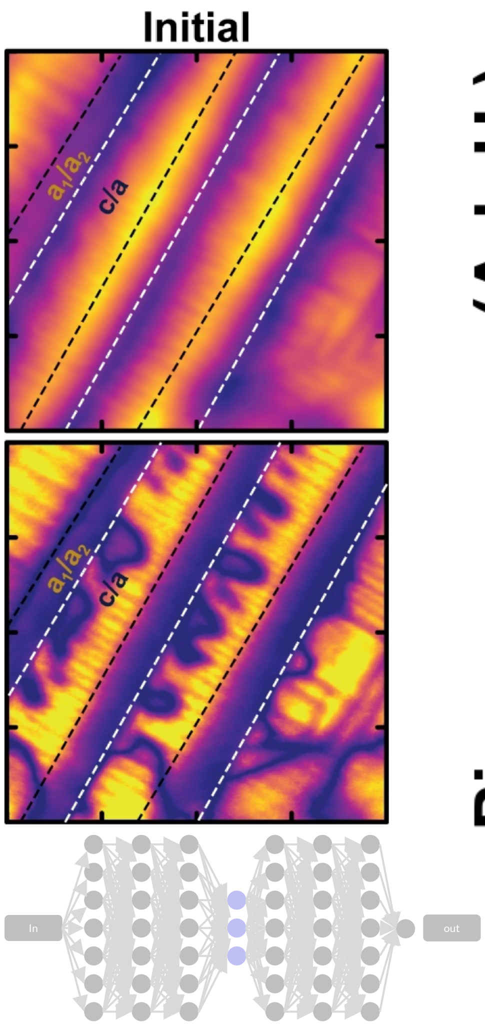

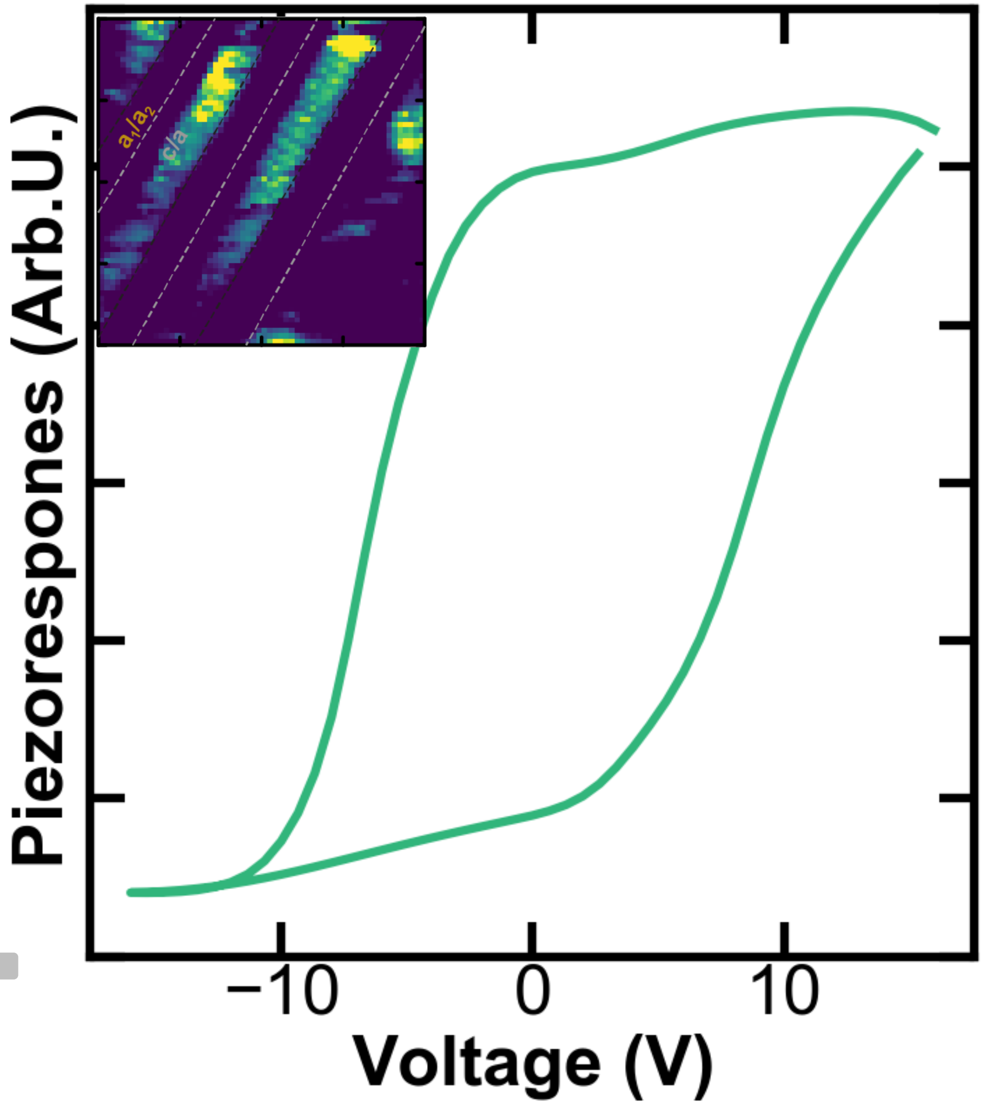

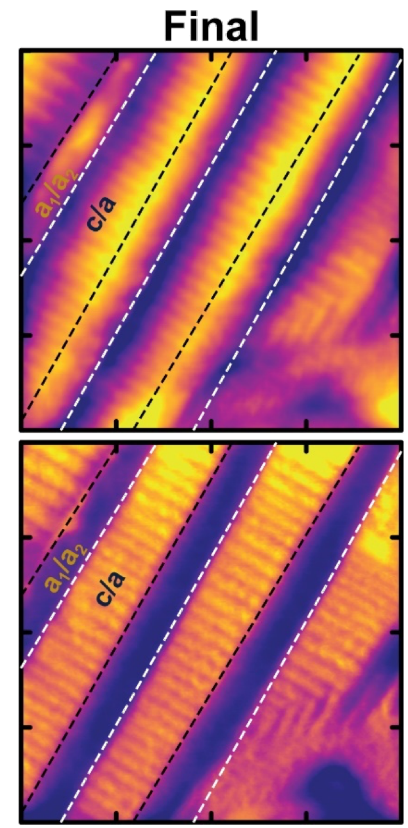

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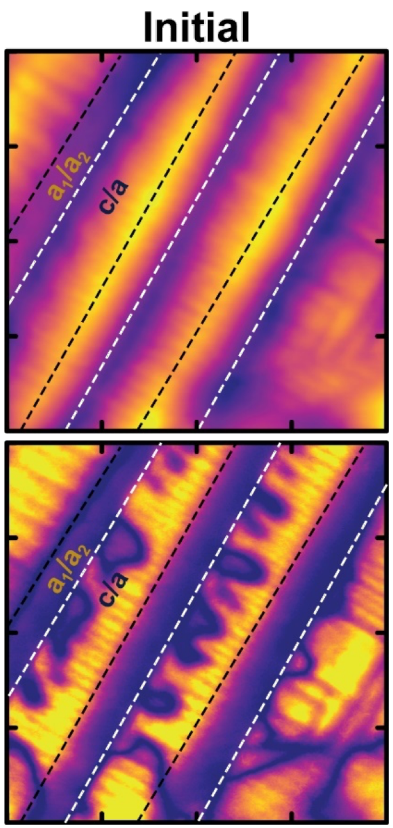

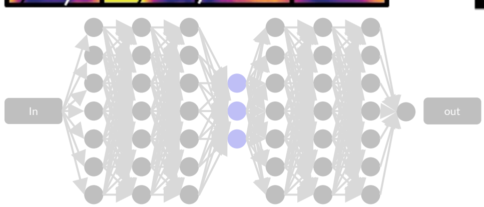

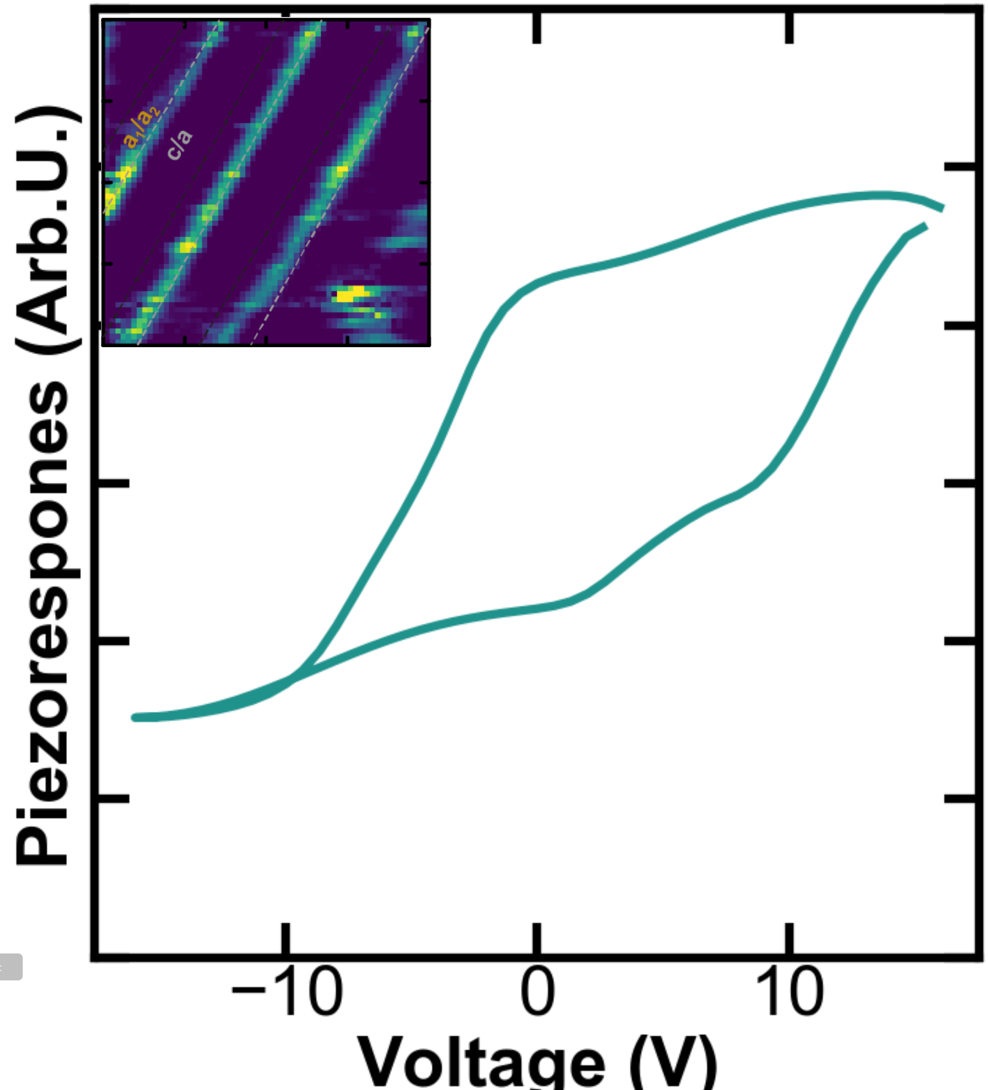

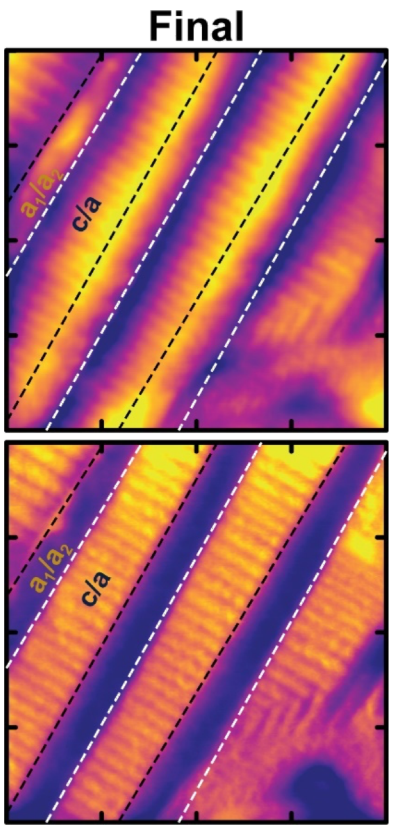

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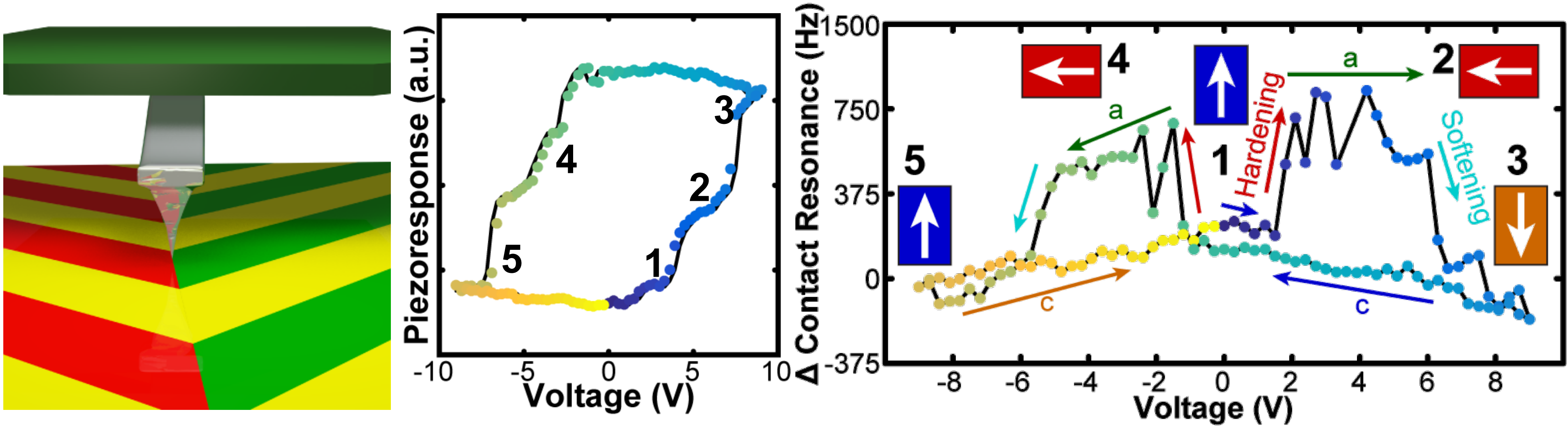
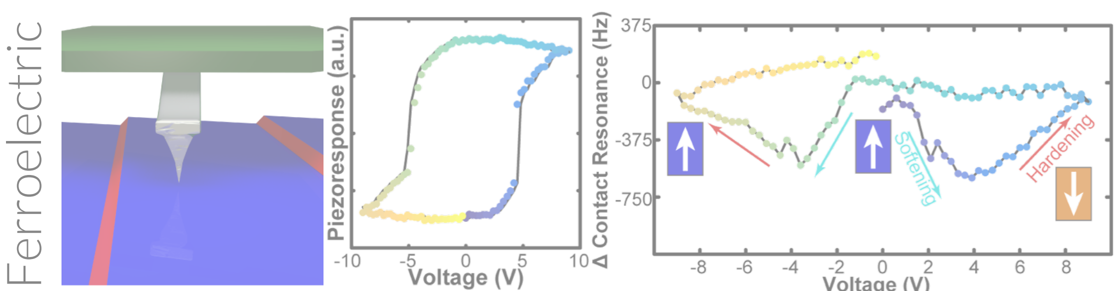
Ferroelastic Switching

Understanding Piezoelectric Concavities





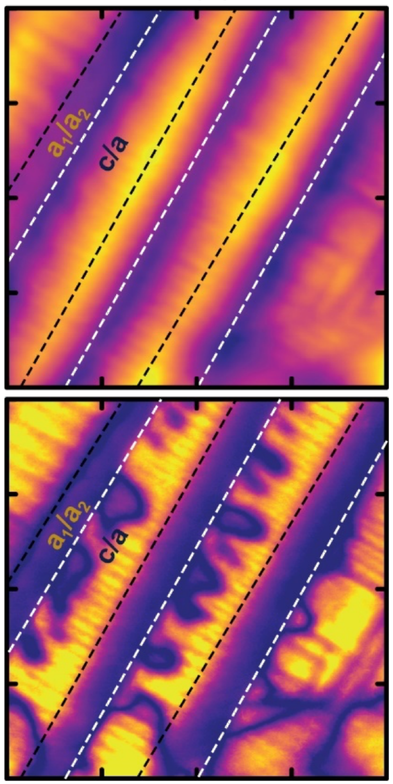
- Intermediate, stable low-piezoresponse states
- Initial elastic hardening c → a transition (low piezoresponse)
- Three-state, two-step ferroelastic switching process, >1% electromechanical response

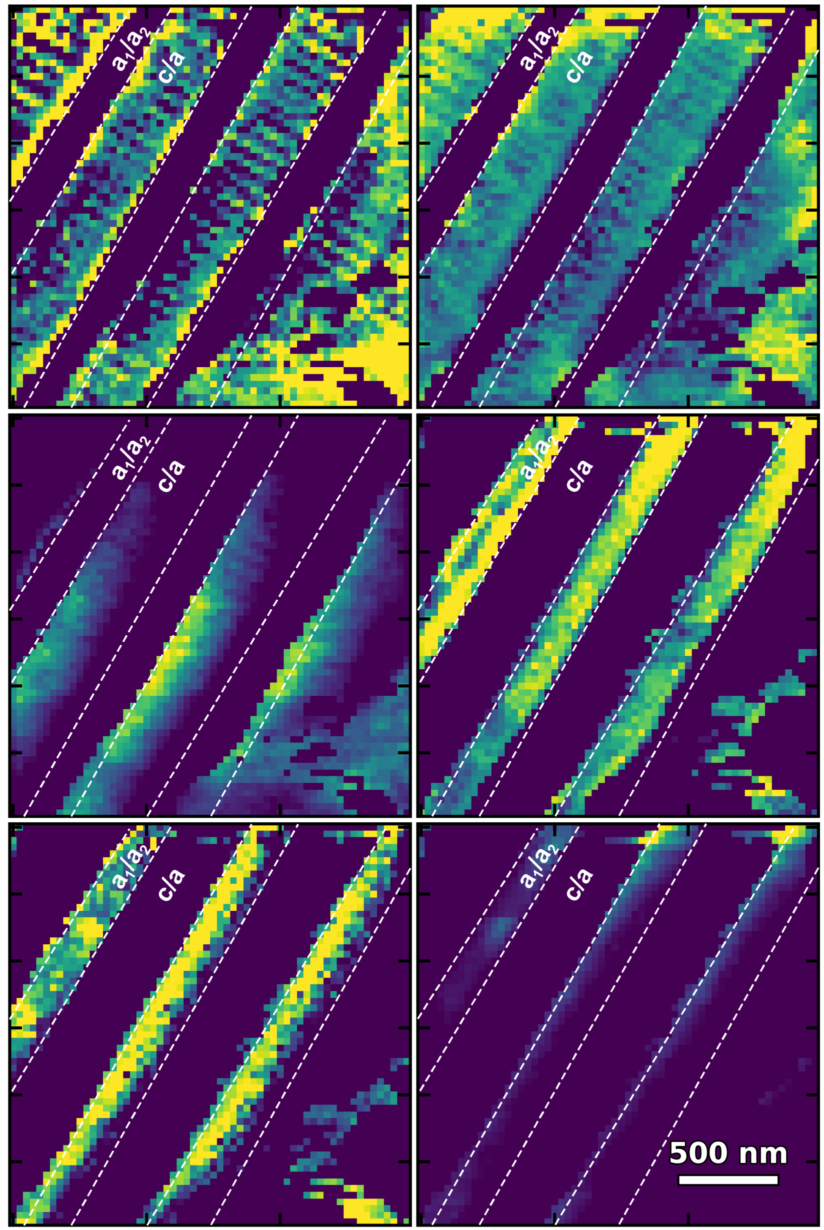
J. C. Agar *et. al. Adv. Mater.* 29, 37, 1702069 (2017)

Analysis of Neural Network

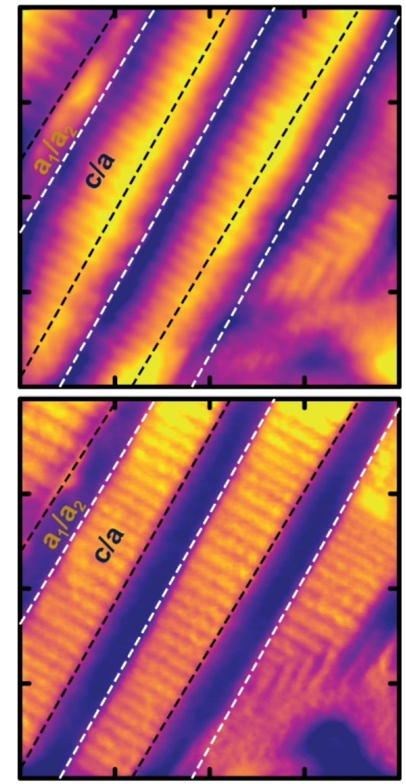
Compute activation from low-dimensional layer → reconstruct maps

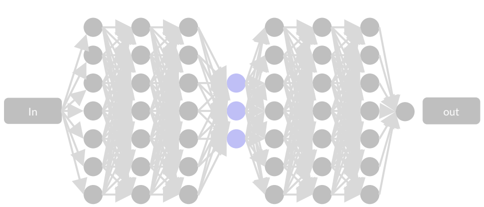
Initial





Final





J. C. Agar *et. al. Nat. Comm. (accepted)*

Analysis of Neural Network

Compute activation from low-dimensional layer → reconstruct maps

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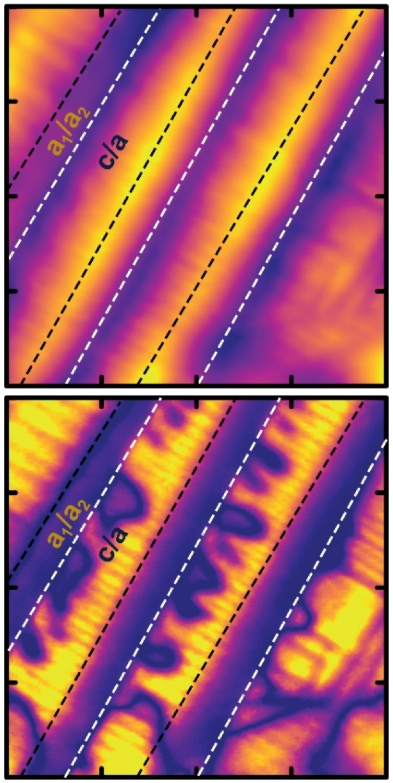
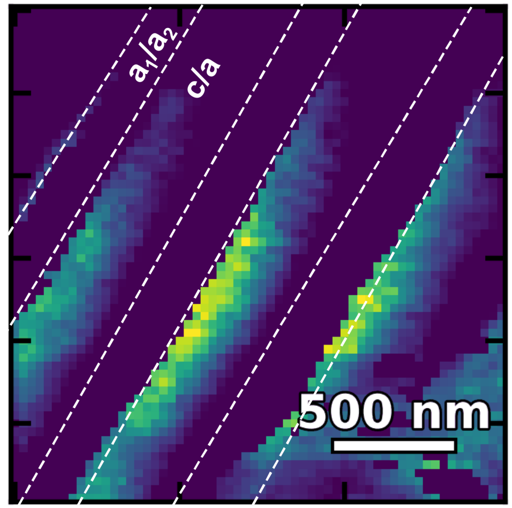
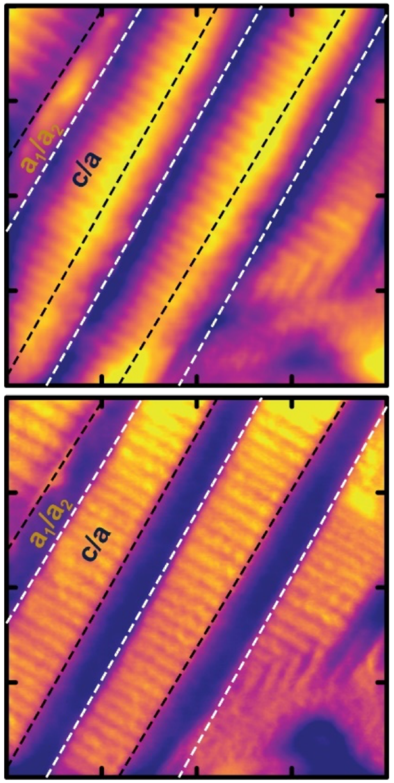
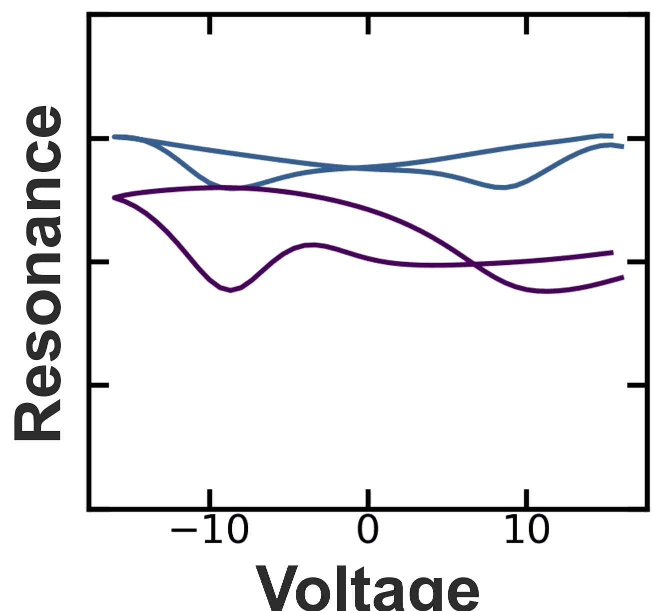
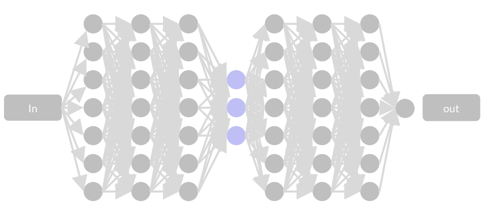




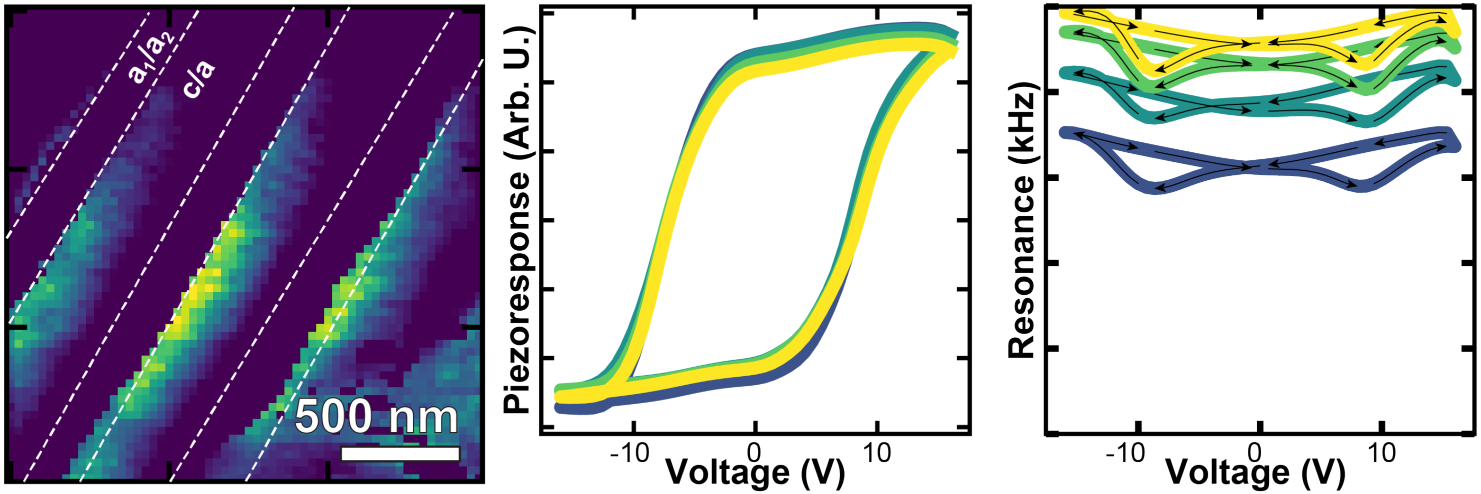
Final

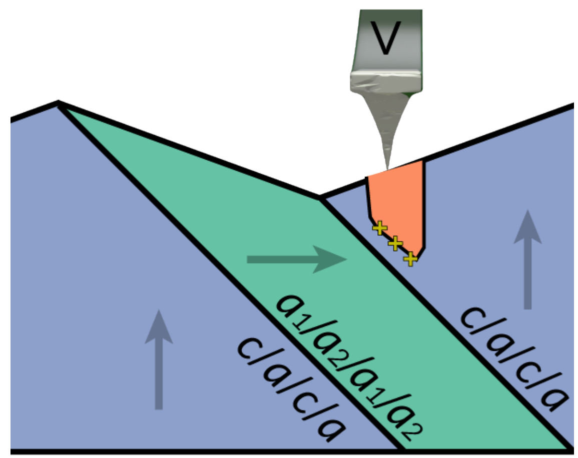






Analysis of Neural Network





- Electromechanical stiffening caused by electrostatic repulsion → charged domain front grows

Collaborators: Y. Cao (UT-Arlington)

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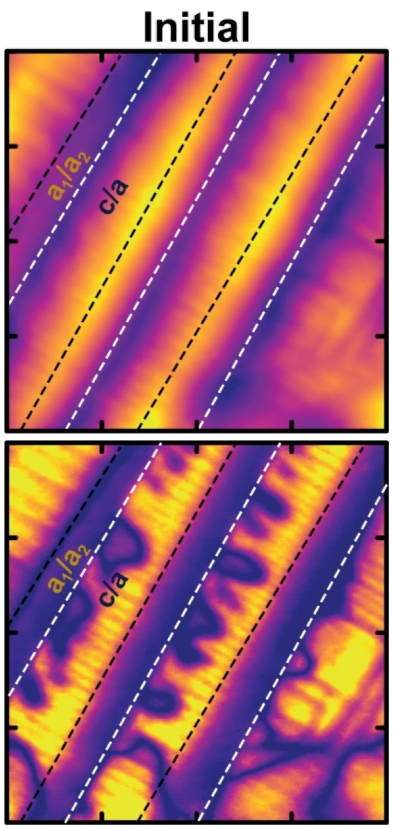
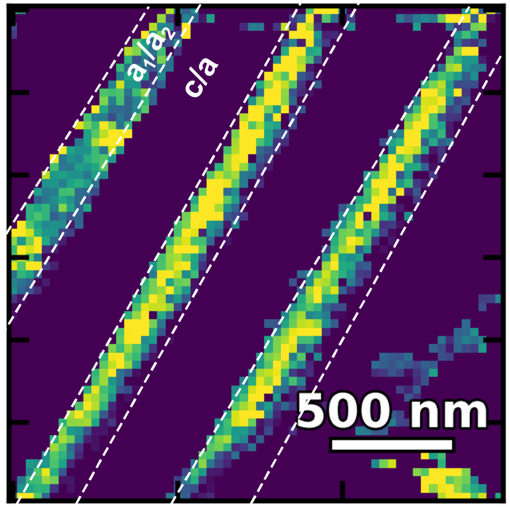
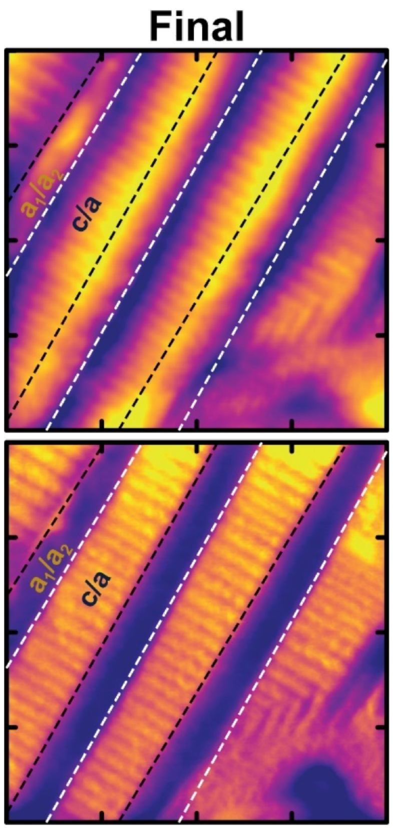
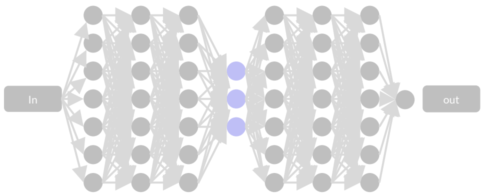
Analysis of Neural Network

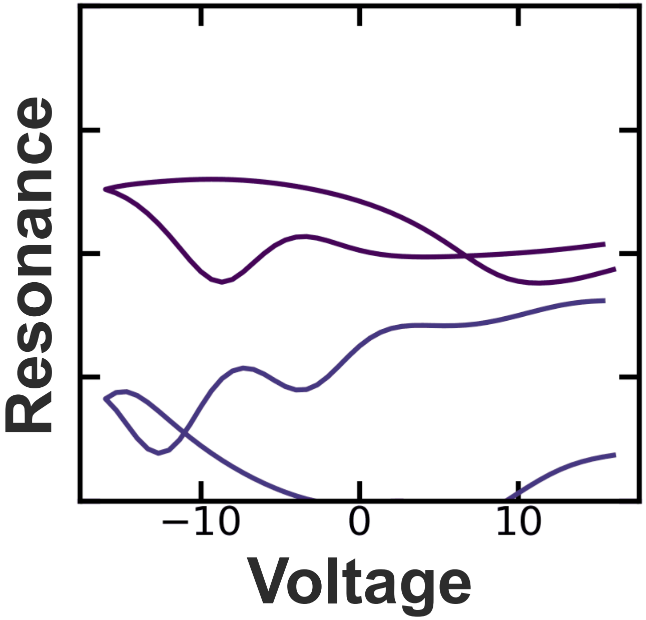
Compute activation from low-dimensional layer → reconstruct maps





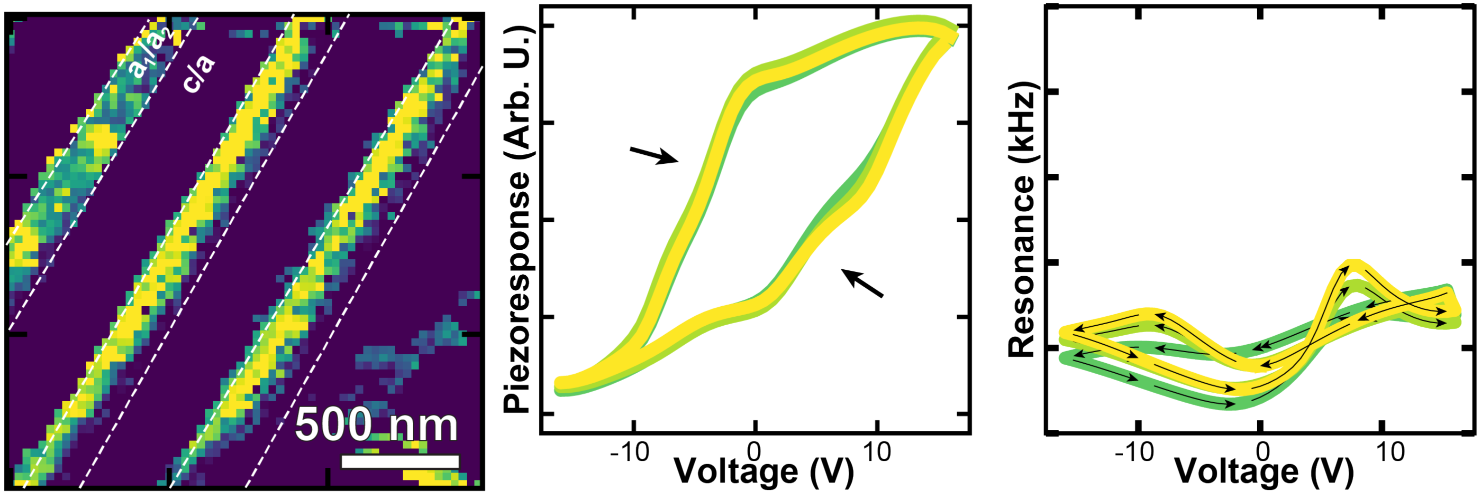


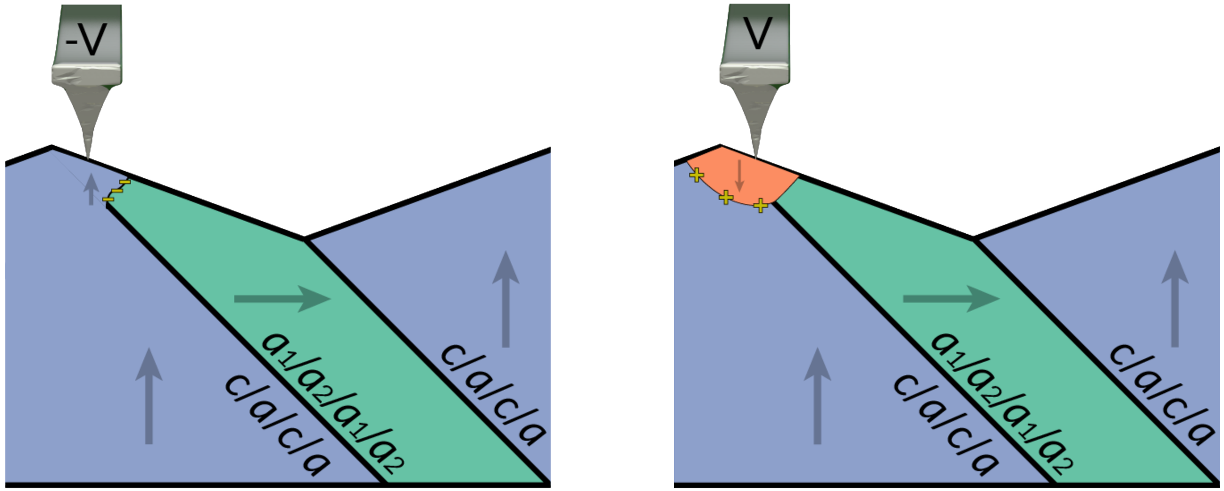




J. C. Agar et. al. Nat. Comm. (accepted)

Analysis of Neural Network





- a → c switching with growing charged domain wall

Collaborators: Y. Cao (UT-Arlington)

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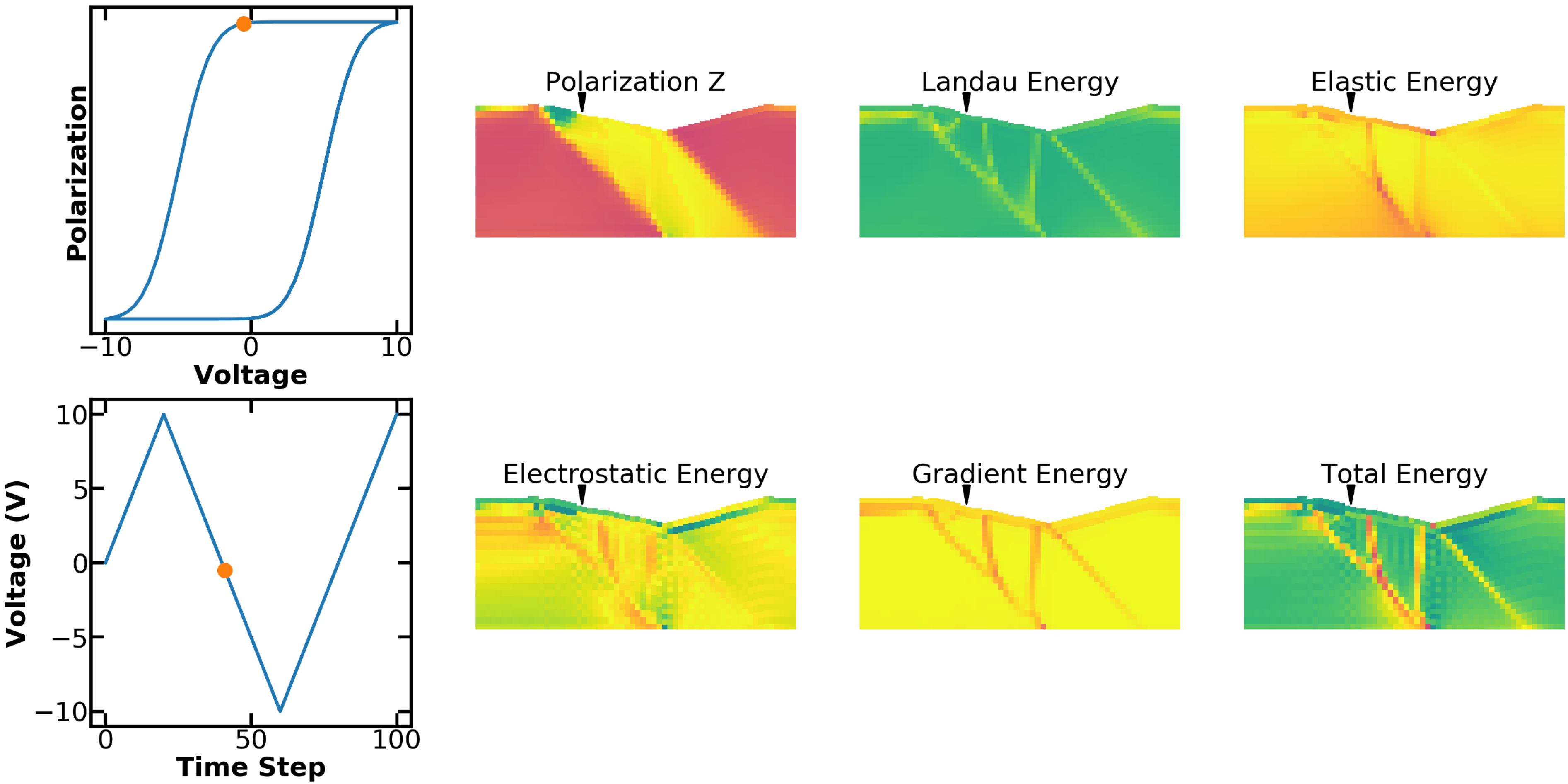
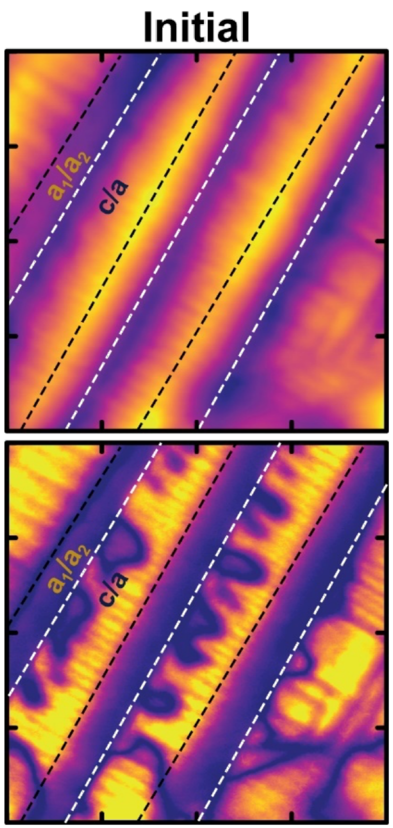
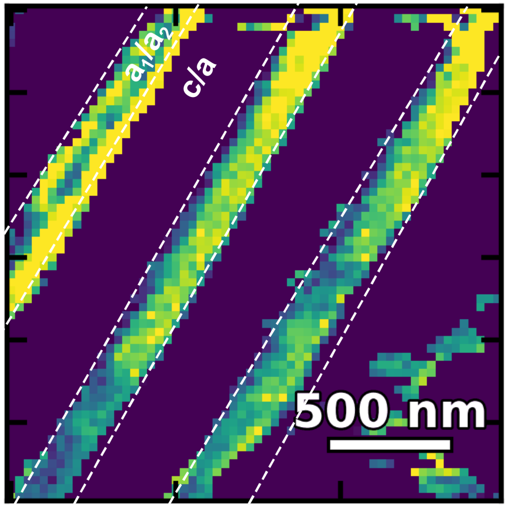
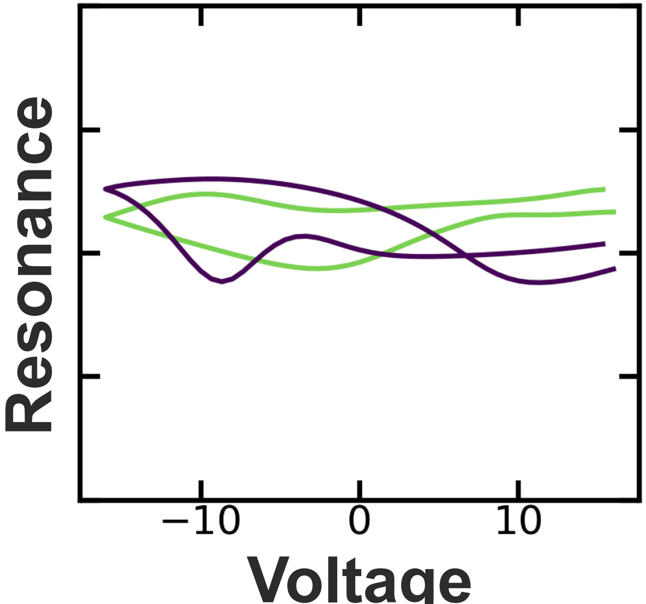

Polarization
−10
0
10
Voltage
Voltage (V)
10
5
0
−5
−10
0
50
100
Time Step
Polarization Z
Landau Energy
Elastic Energy
Electrostatic Energy
Gradient Energy
Total Energy

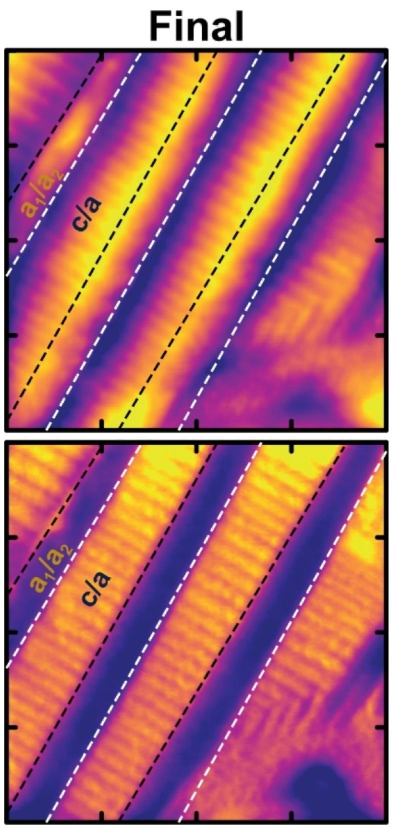
Analysis of Neural Network

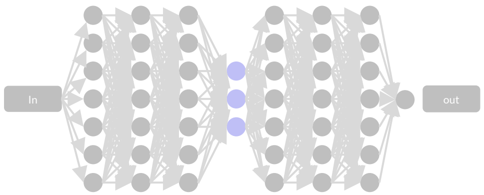
Compute activation from low-dimensional layer → reconstruct maps





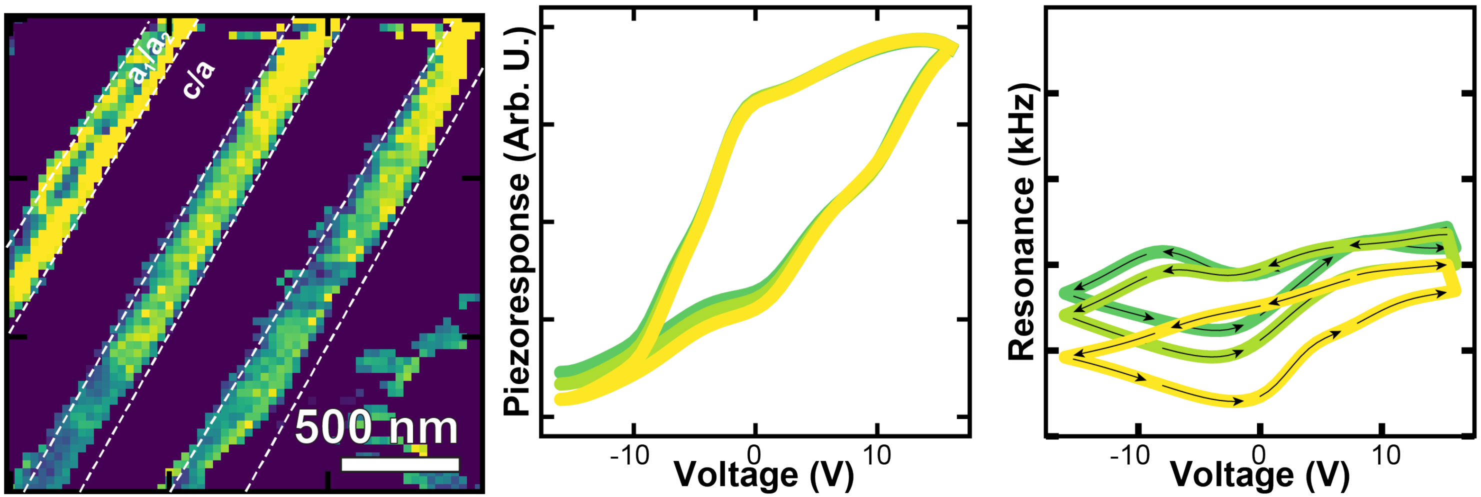


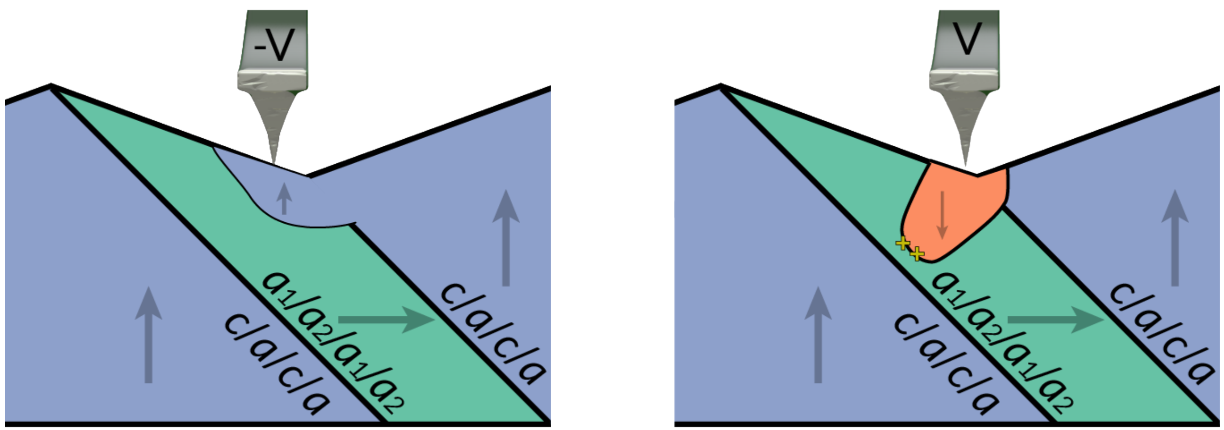




J. C. Agar *et. al.* (in preparation)

Analysis of Neural Network

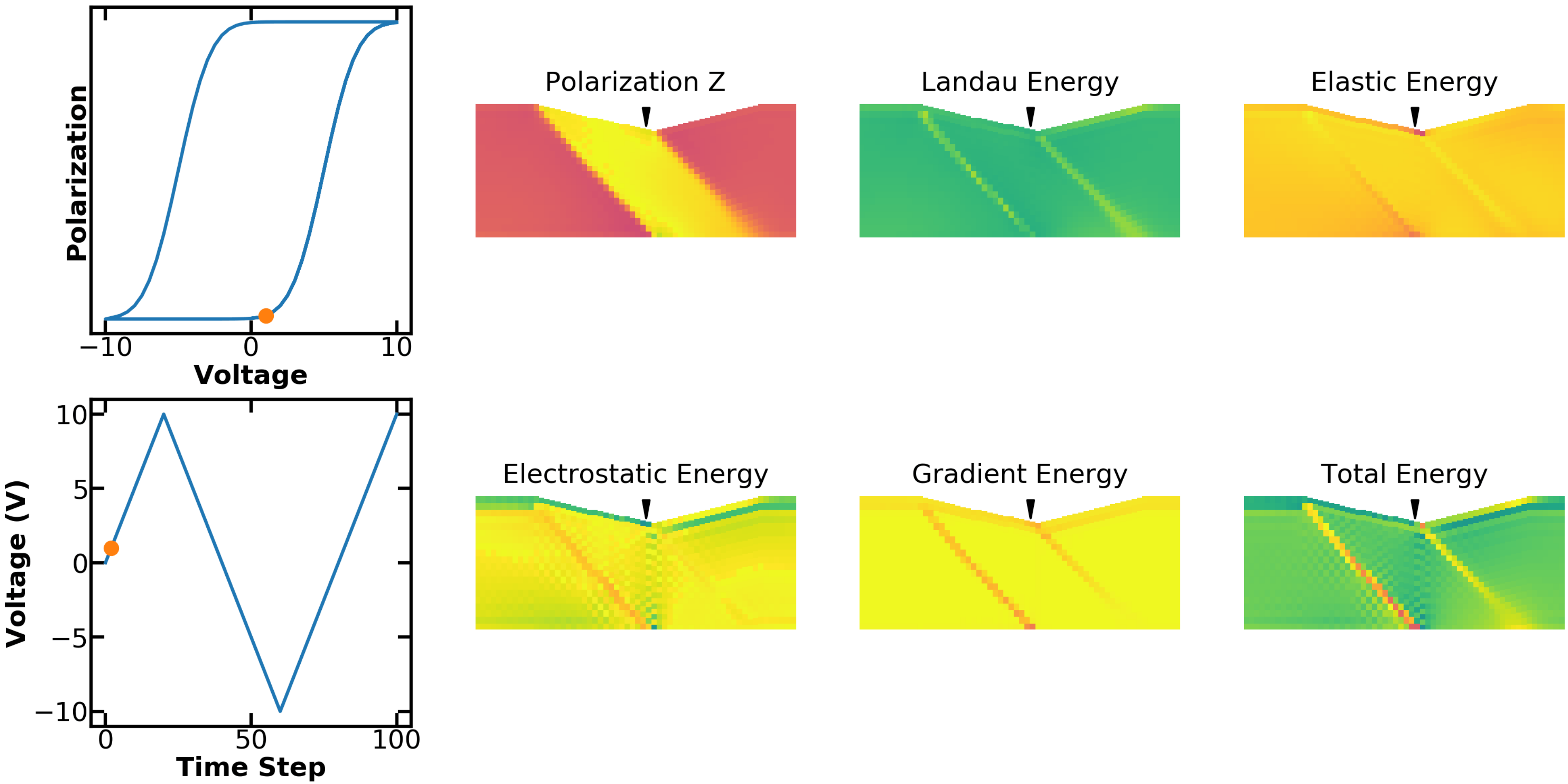




- + bias c→a switching w/ charged domain wall, - bias w/o charged domain wall

Collaborators: Y. Cao (UT-Arlington)

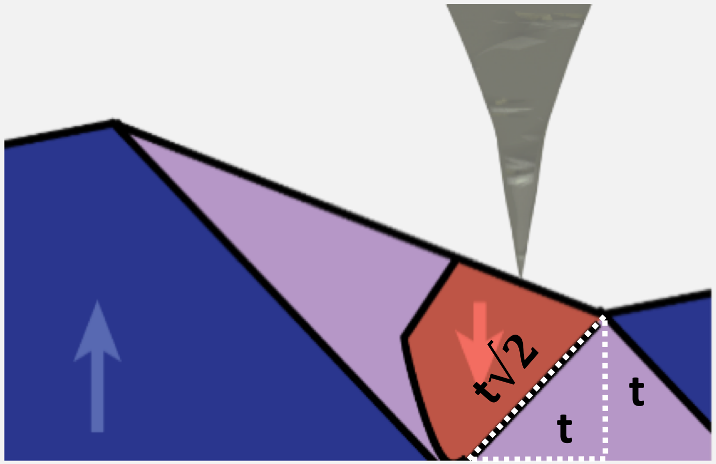
J. C. Agar *et. al. Nat. Comm. (accepted)*



Collaborators: Y. Cao (UT-Arlington)

J. C. Agar *et. al. Nat. Comm. (accepted)*

Extracting Further insight



Machine learning approaches…

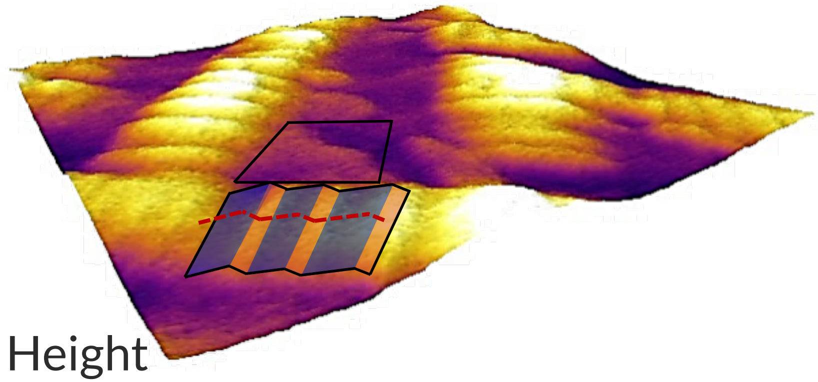
- Enable real-time classification of switching processes
- Identify intermediate stages of switching
- Find optimal geometry to favor ferroelastic switching and electromechancial response

- Concavities → well behaved in voltage space
- Area of concavities represent significance of transition → fit with mixture of Gaussians
- Enhanced ferroelectric or ferroelastic character at different c/a/c/a-$a_1/a_2/a_1/a_2$ boundaries
- **Quenched cantilever resonance (dampening) along valley boundary →** increased electromechanical energy absorption

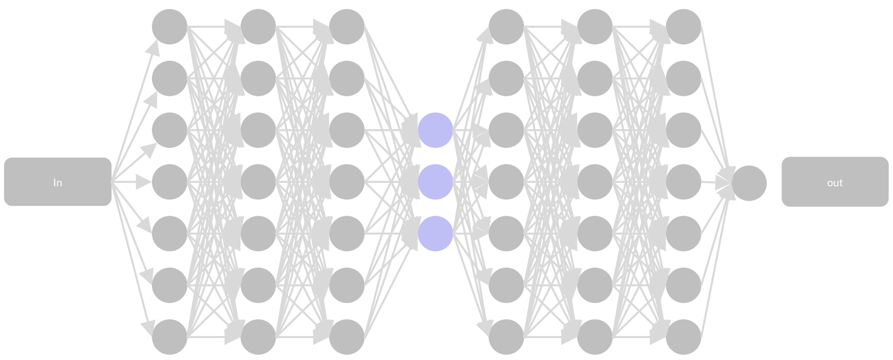
J. C. Agar *et. al. Adv. Mater. 30*, 28, 1800701 (2018)

Conclusions

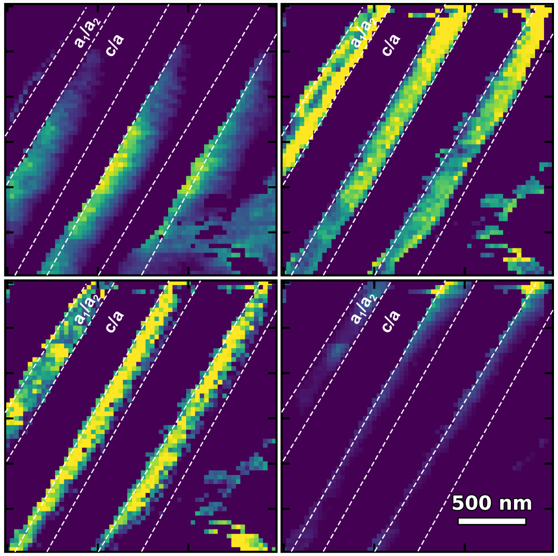
Complex-Domain Structures



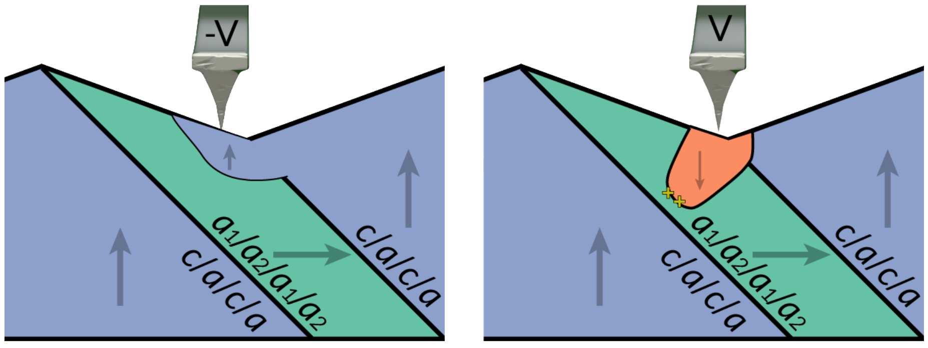
LSTM-Neural Network



Identify Features of Switching

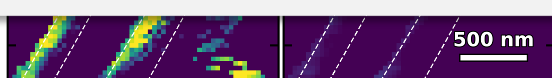


Understand Switching Mechanisms

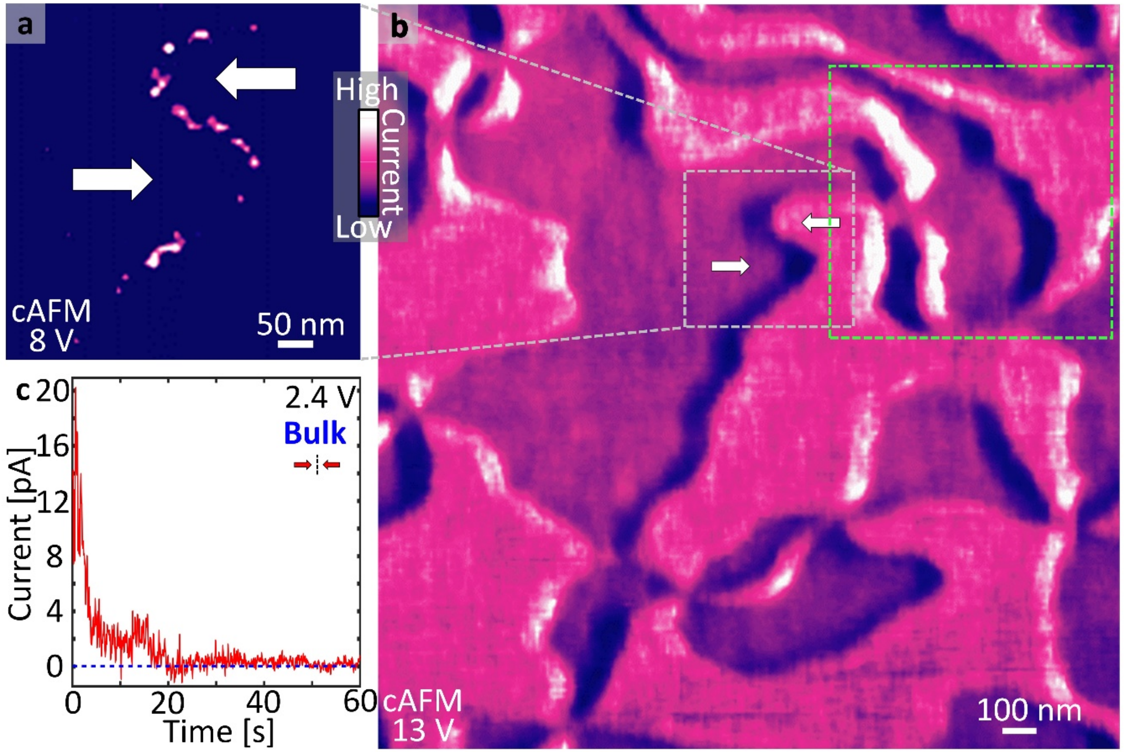


Conclusions

Can we apply this technique to other experimental techniques and materials systems?



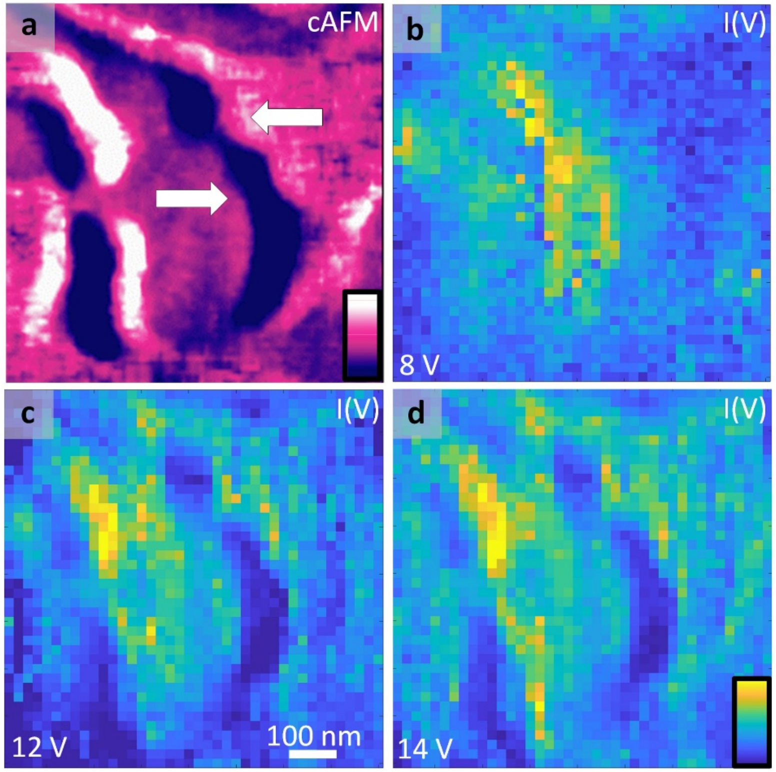
Conductive Domain Walls in $ErZrMnO_3$



- Conductivity and conduction mechanisms of domain wall are dependent on the polar topology

Trygve Ræder Tor Grande Dennis Meier

Conductive Domain Walls in $ErZrMnO_3$



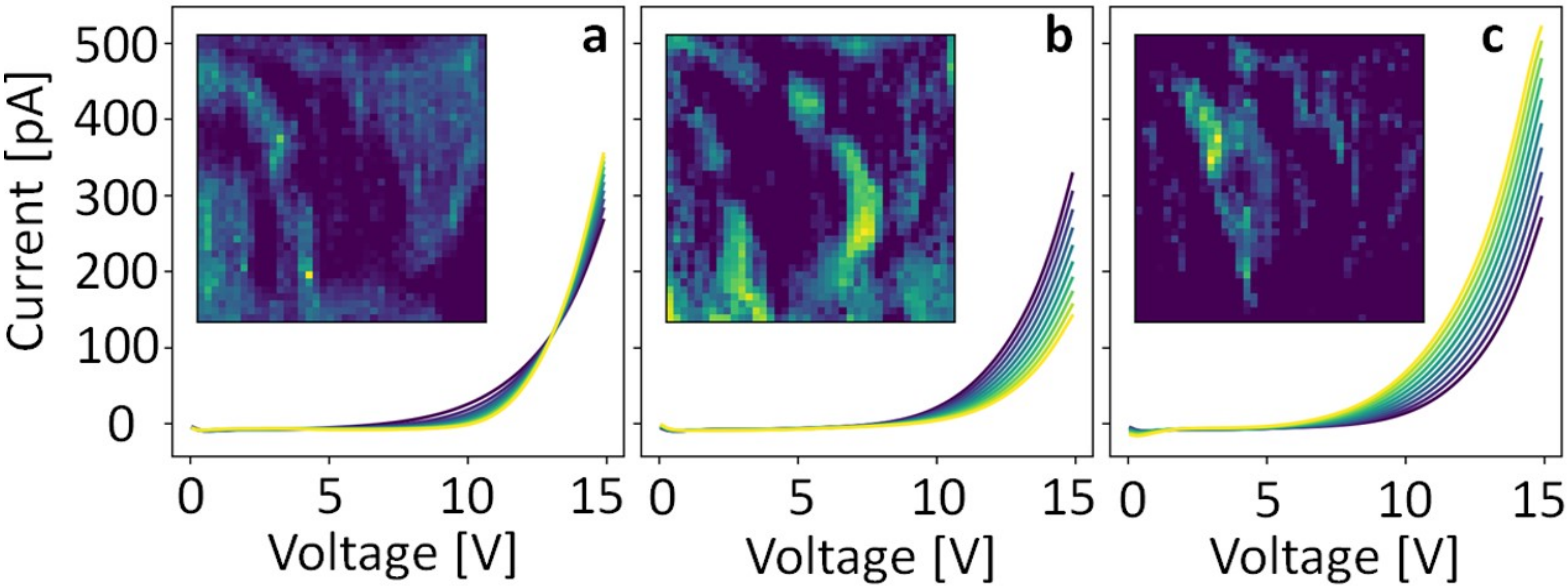
- Recurrent autoencoder extracts features of IV response relating to different conduction mechanisms

Trygve Ræder

Tor Grande

Dennis Meier

Conductive Domain Walls in $ErZrMnO_3$

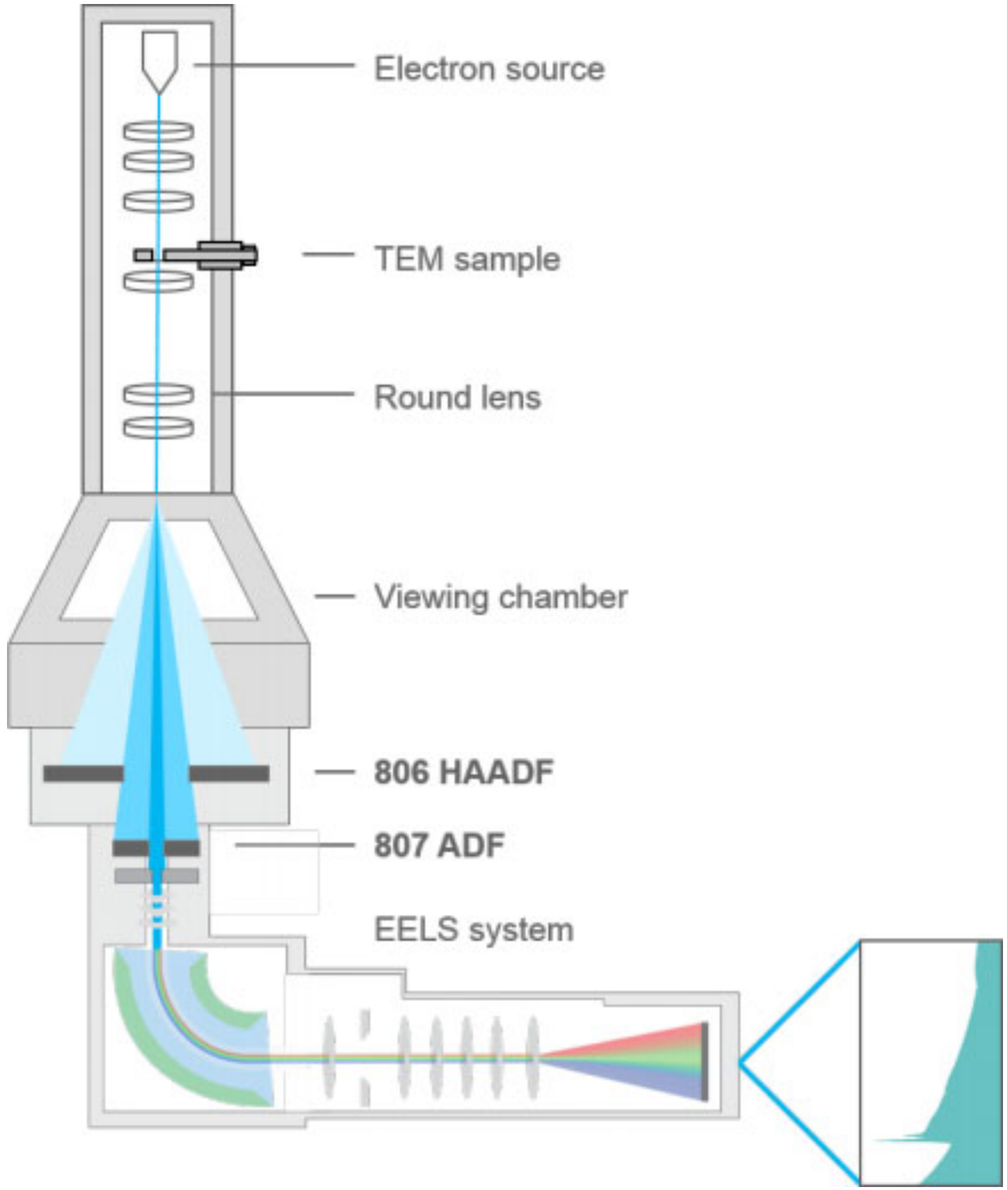


- Recurrent autoencoder extracts features of IV response relating to different conduction mechanisms

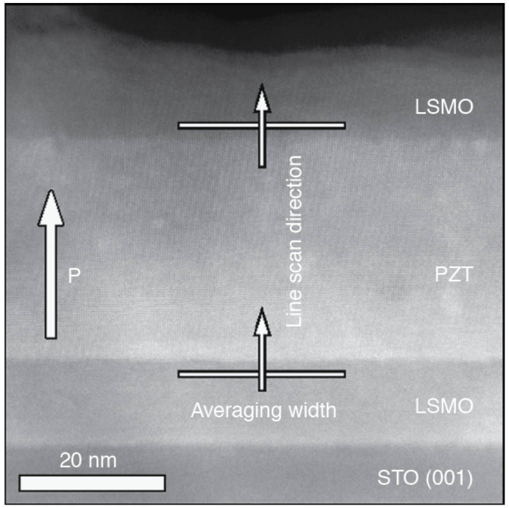
Trygve Ræder Tor Grande Dennis Meier

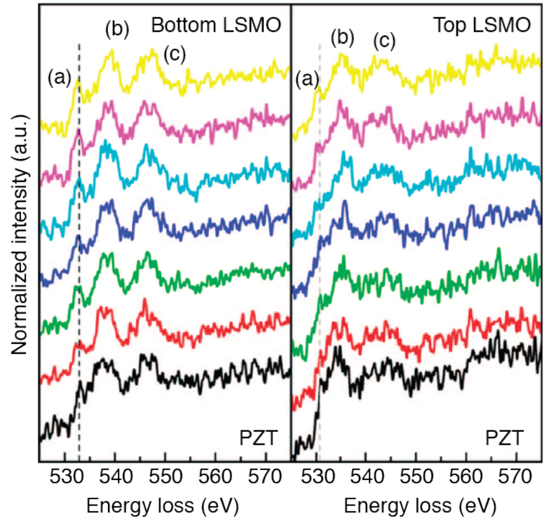


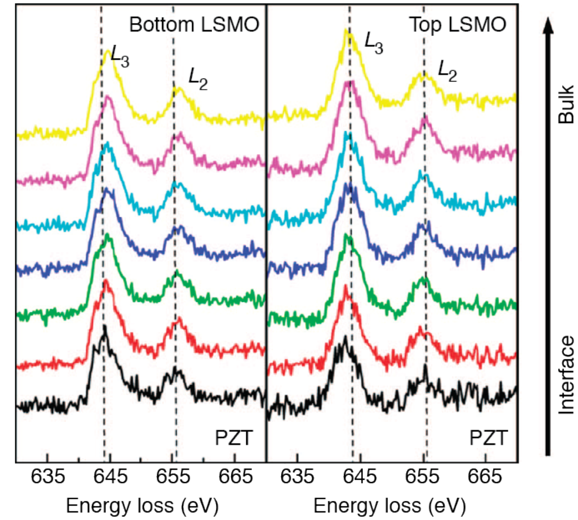
Electron Energy Loss Spectroscopy



Applying this Concept to EELS







- Provides spatially resolvable insight about coordination chemistry
- Information about charge asymmetry at interface that drives emergent properties

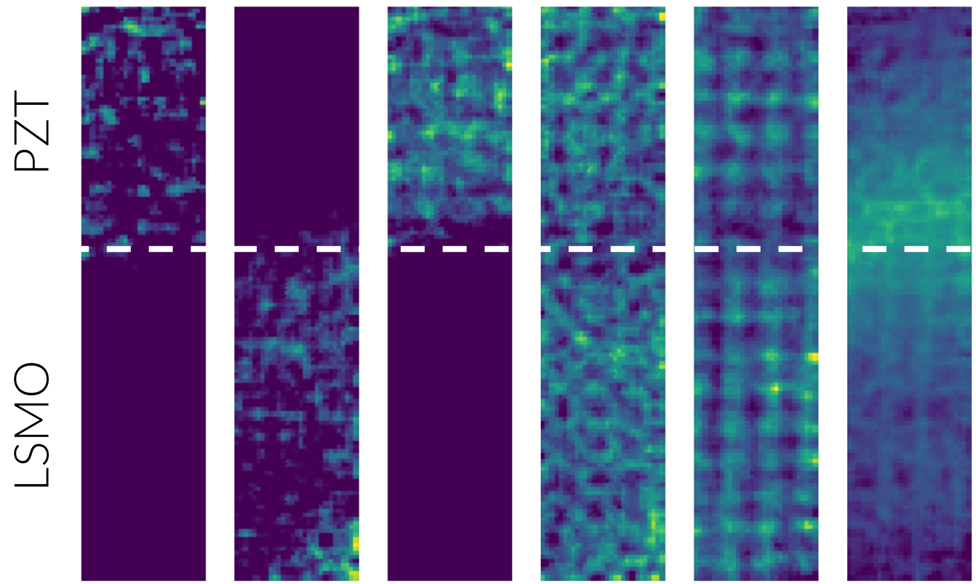
Spurgeon et al. Nat. Comm. 6, 6735 2015

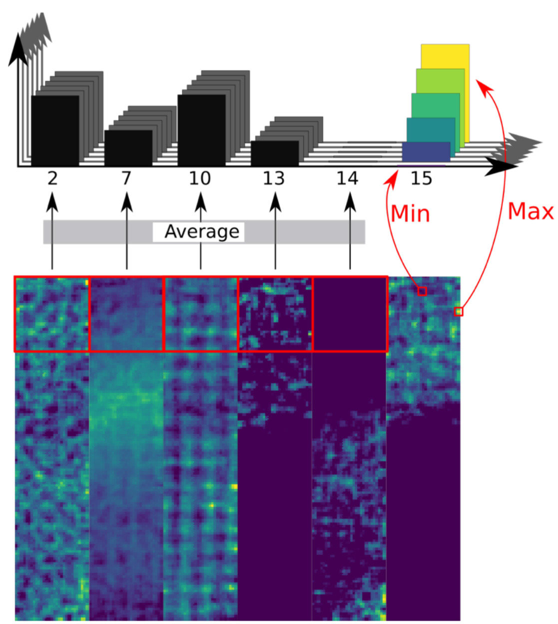
Jamie Hart

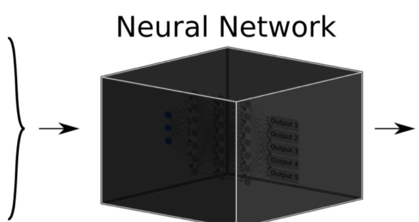
Steve Spurgeon

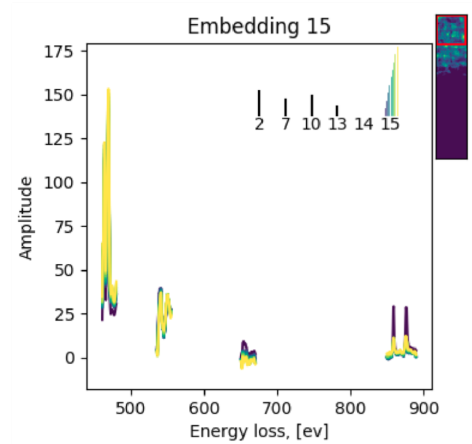
Mitra Tahari

Applying this Concept to EELS

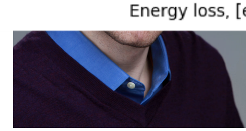




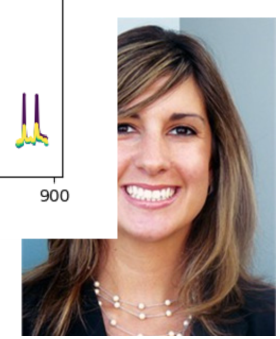




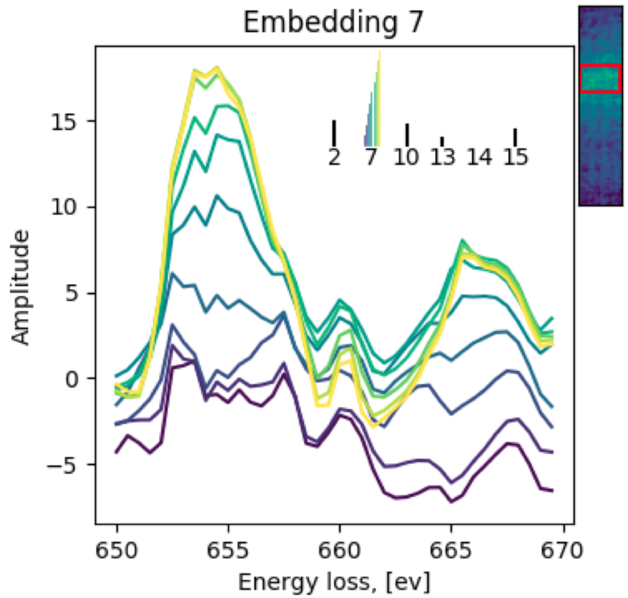
Jamie Hart

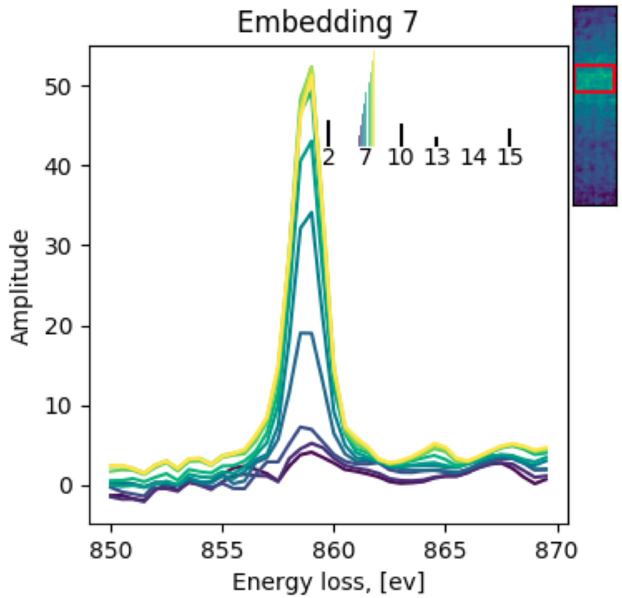


a Tahari

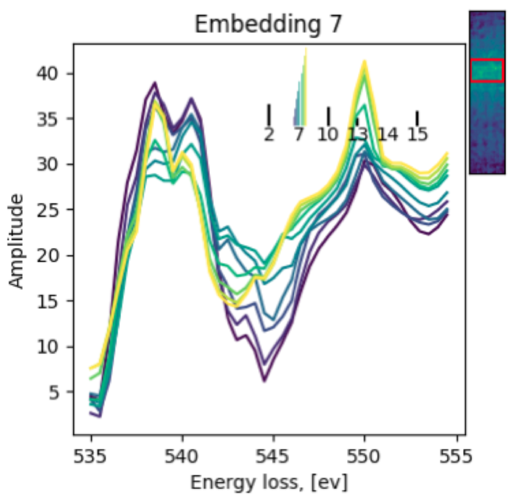


Autoencoder on EELS



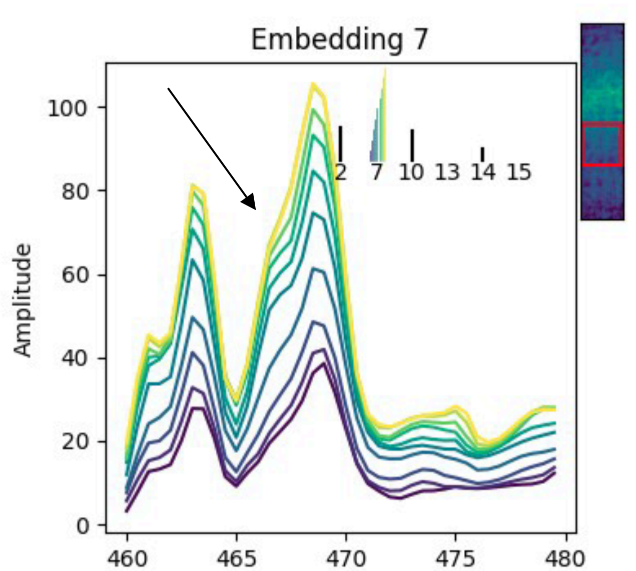


- La and Mn mixing at the interface → shows "sharpness" of interface

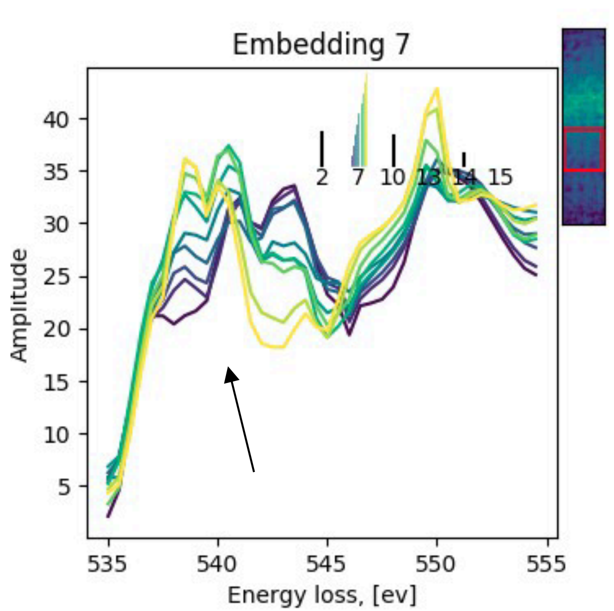


- Oxygen changing its screening at the interface

Autoencoder on EELS



- Ti intermixing and changing valence state

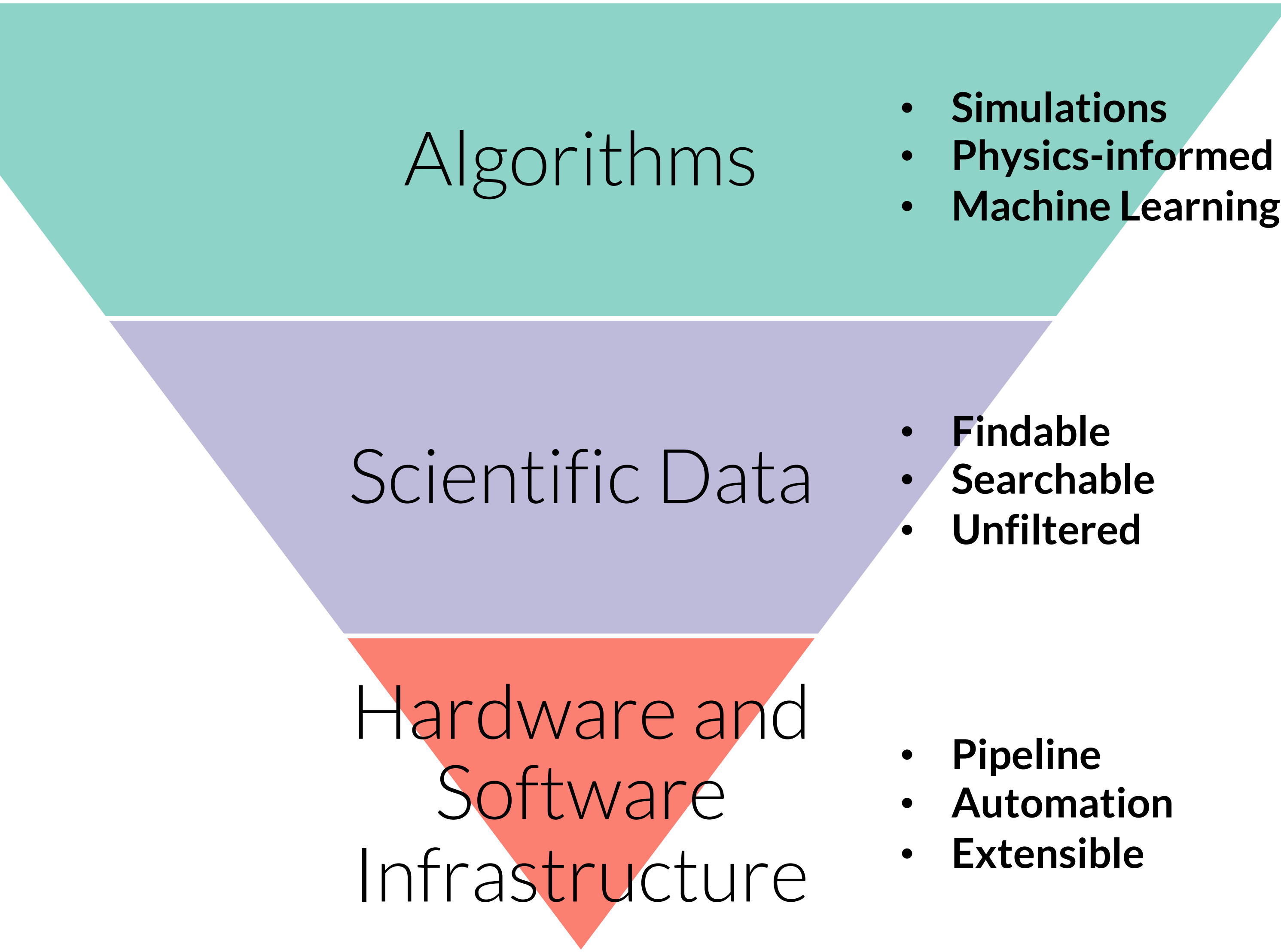


- Oxygen changing coordination

What is next?

How can we leverage data-driven approaches in experimental science?

Experimental Scientific Data Infrastructure

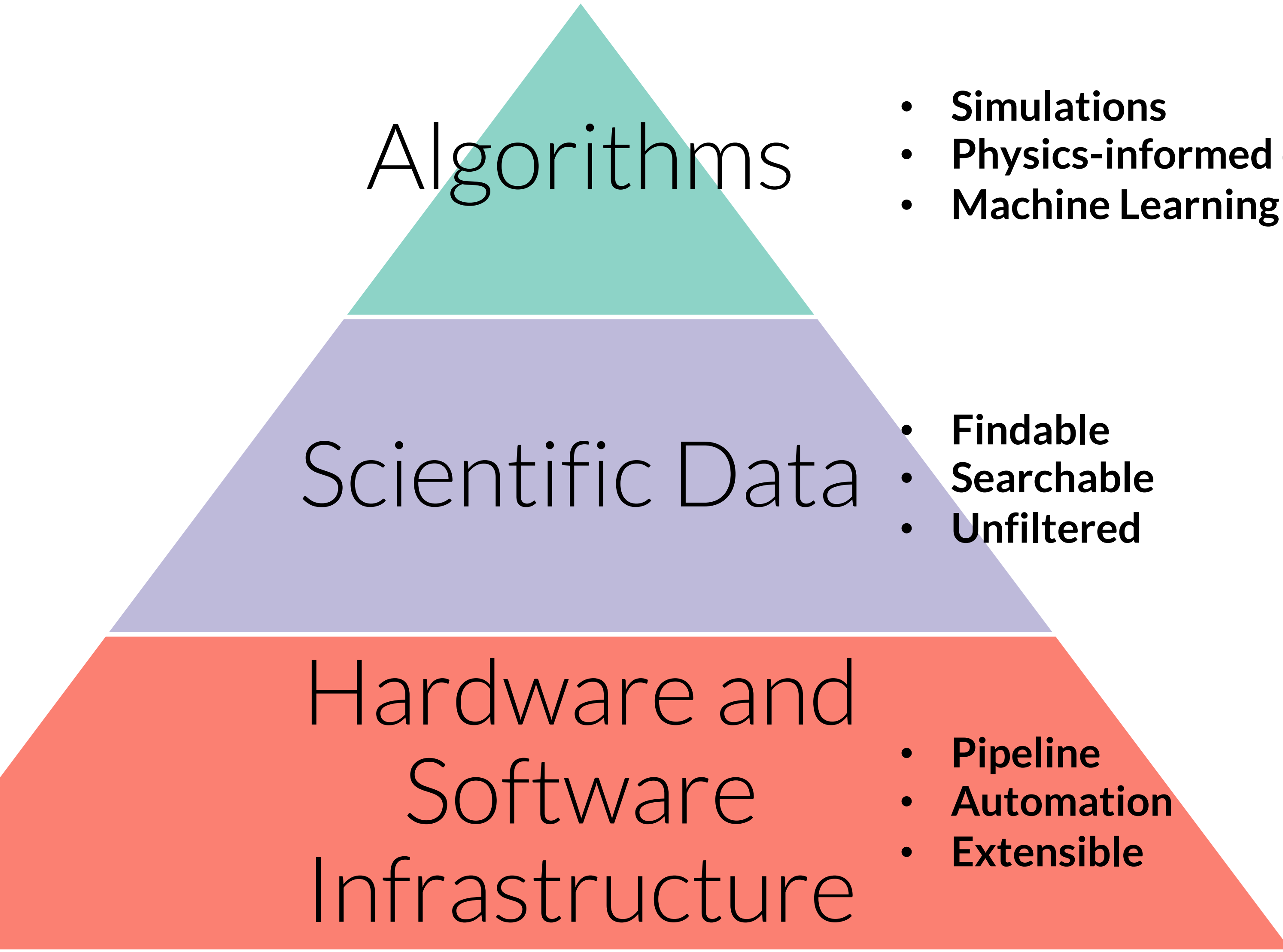


- **Simulations**
- **Physics-informed empirical models**
- **Machine Learning**

- **Findable**
- **Searchable**
- **Unfiltered**

- **Pipeline**
- **Automation**
- **Extensible**

Experimental Scientific Data Infrastructure



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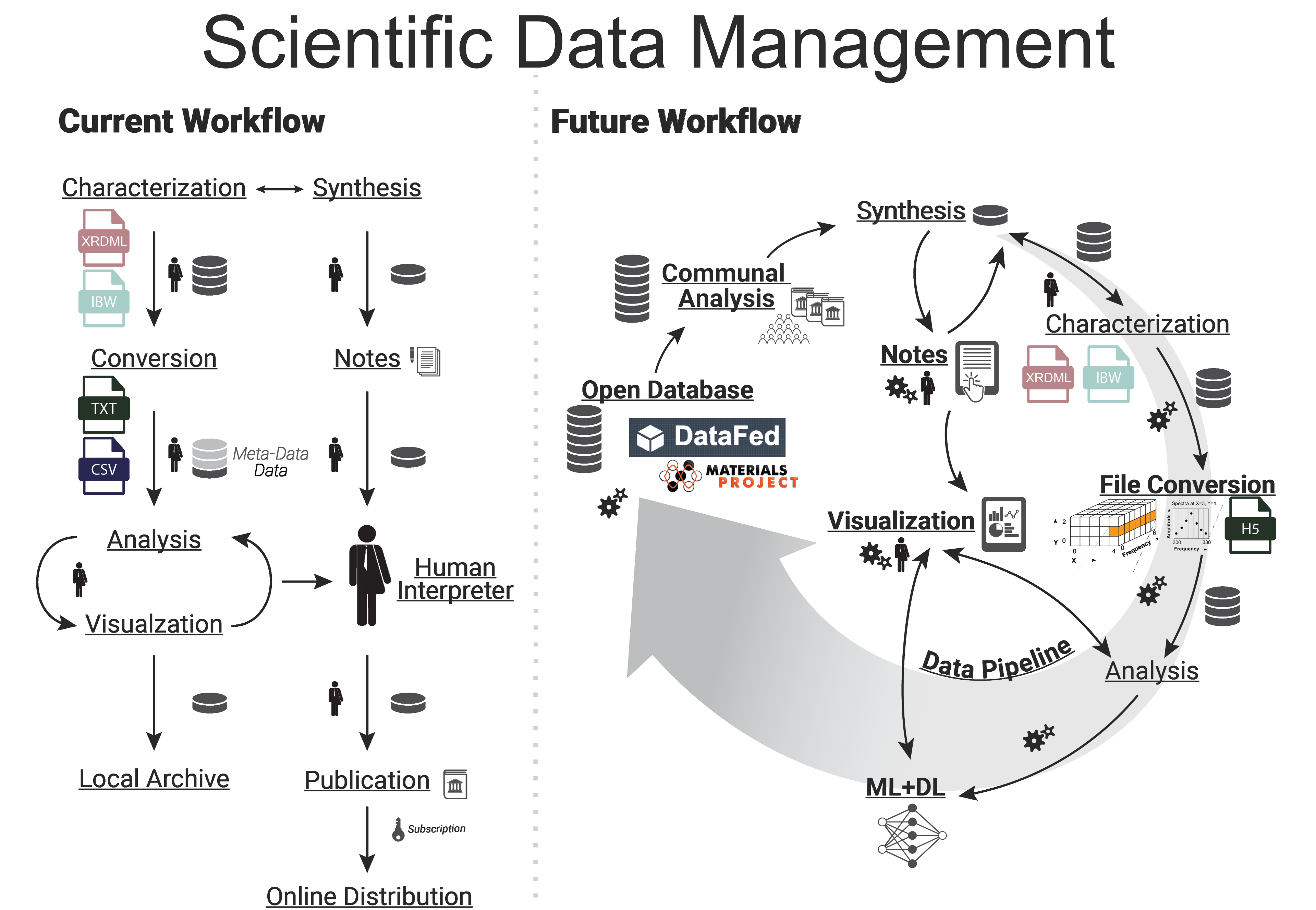
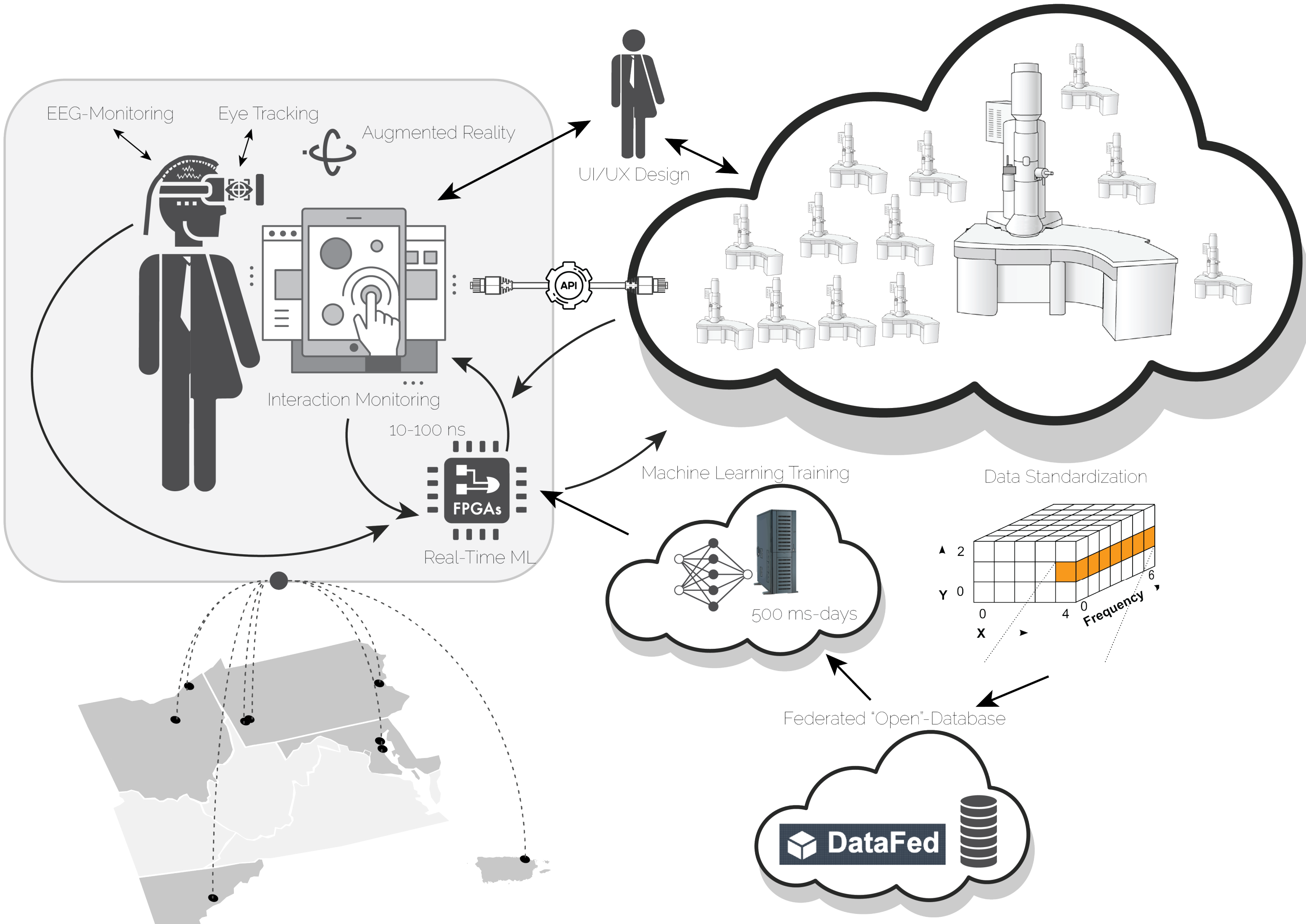
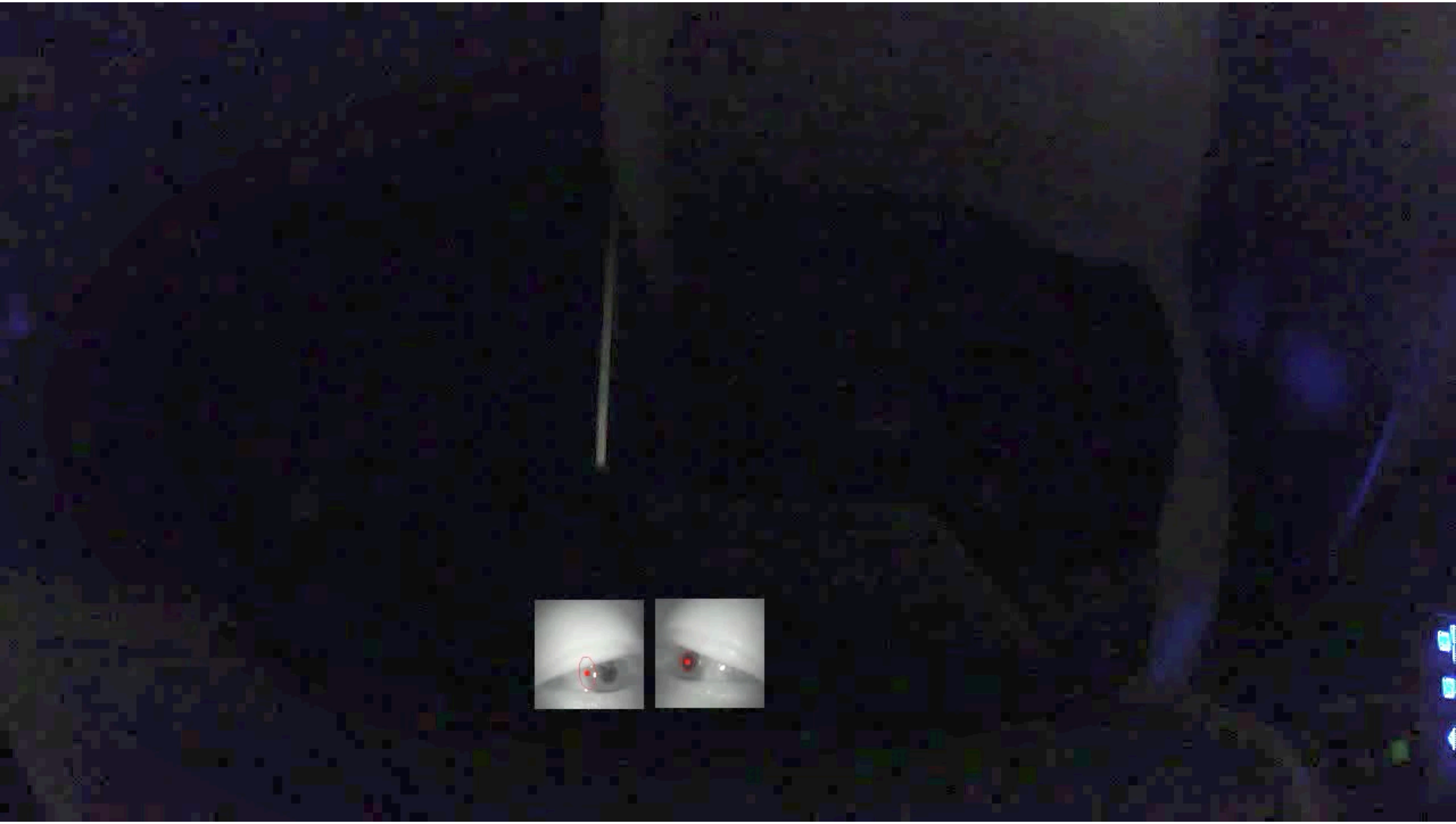


Scientific Data Management
Current Workflow
Future Workflow
Characterization
Synthesis
XRDML
IBW
Conversion
Notes
TXT
CSV
Meta-Data
Data
Analysis
Visualzation
Human
Interpreter
Local Archive
Publication
Subscription
Online Distribution
Communal
Analysis
Open Database
DataFed
MATERIALS
PROJECT
Notes
Characterization
XRDML
IBW
File Conversion
H5
Visualization
Data Pipeline
Analysis
ML+DL

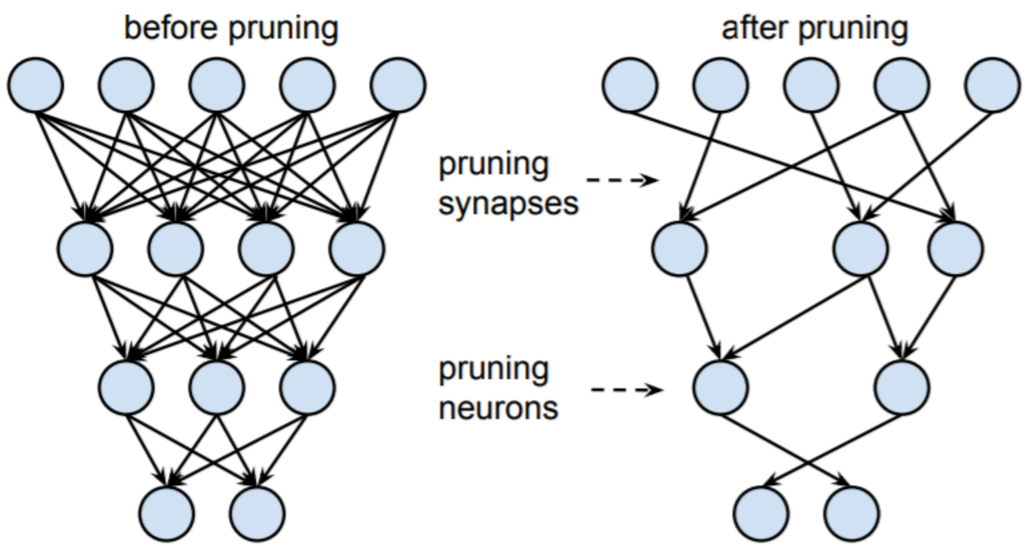
Lehigh University Nano-Human Interfaces Initiative

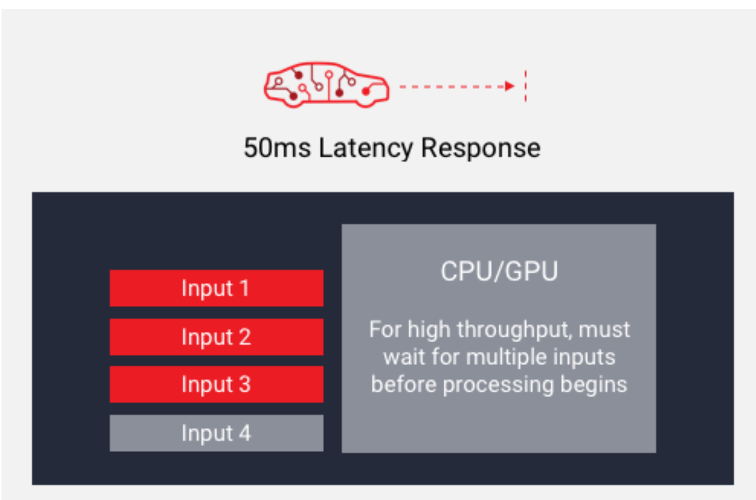


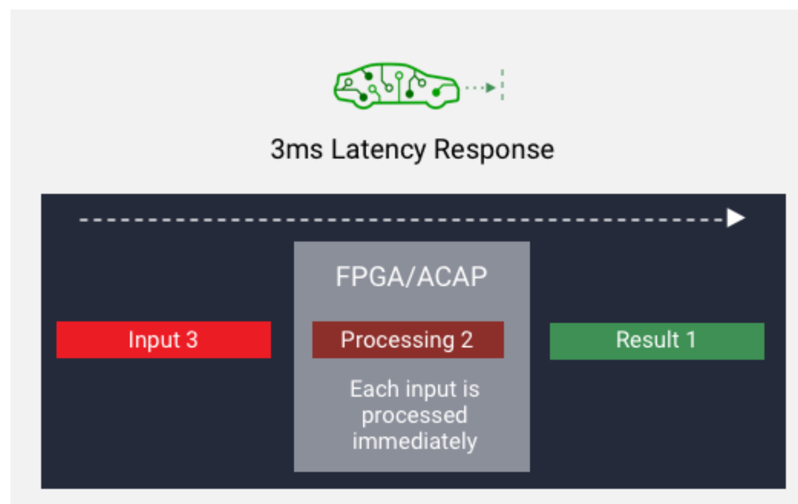
Optimizing Human-Machine Interactions



High Speed Machine Learning





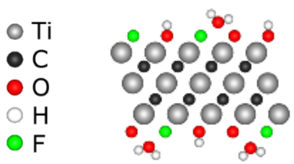


Controlling Properties of 2D Materials via Termination and Intercalation:
Tracking $Ti_3C_2T_x$ -F Surface Terminations In Situ

Initial state:

Intercalants: H_2O

T_x: $OH_{0.4}F_{0.4}O_{0.5}$

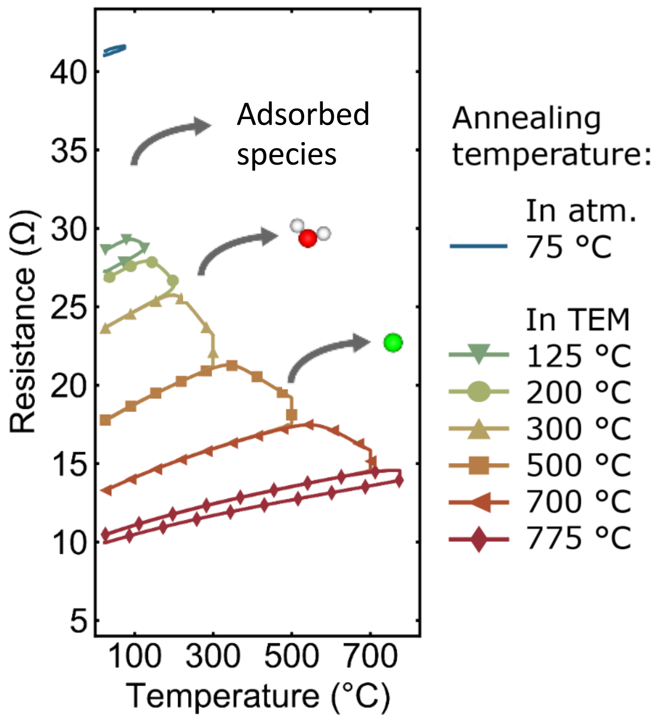


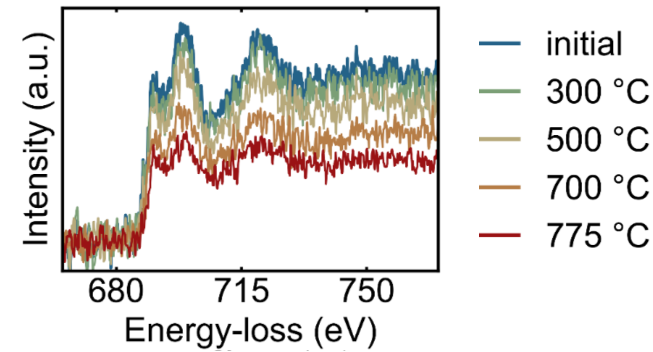
Final state:

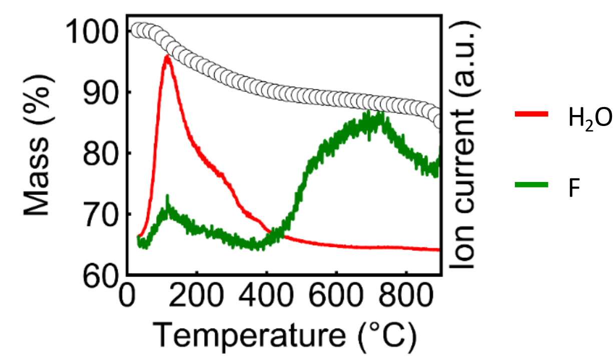
Intercalants: -

T_x: $F_{0.2}O_{0.5}$

$\Delta\sigma = 4\times$







Hart, J.L., Hantanasirisakul, K., Lang, A.C., Anasori, B., Pinto, D., Pivak, Y., van Omme, J.T., May, S.J., Gogotsi, Y. and Taheri, M.L., 2019. Nature communications, 10(1), p.522.

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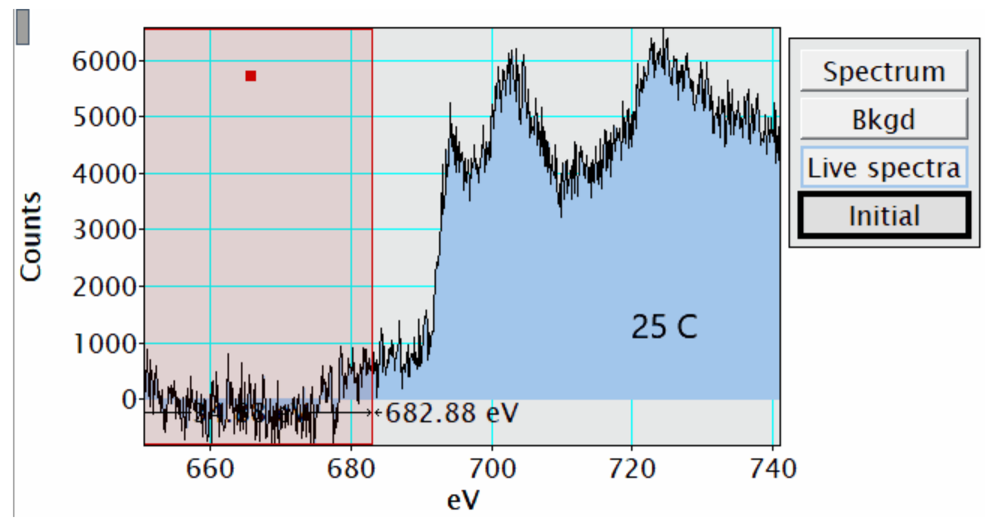
Tracking $Ti_3C_2T_x$ -F Surface Terminations In Situ, Continued: That's a lot of data!

- **DATA RATES:**
- Imaging data rate is 26 Gb/s (>1.5TB/min);
- spectroscopy data rate is 6 Mb/s
- Storage rate (SS drives) is at least 26 Gb/s.

- **DETAILS:**
- F K edge of Ti_3C_2
- Data recorded while heating from 25C → 650 C.
- F edge starts decreasing around 400 C (defunctionalizing)
- Spectra were initially acquired every 2 seconds, then summed for sufficient SNR
- Each frame in the video is sum of 10 spectra



In situ acquisition (up to 400 fps)

Hart, J.L., Hantanasirisakul, K., Lang, A.C., Anasori, B., Pinto, D., Pivak, Y., van Omme, J.T., May, S.J., Gogotsi, Y. and Taheri, M.L., 2019. Nature communications, 10(1), p.522.

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Building a Community for Integrated Microscopy

- Second NexTEM workshop (organized by Taheri, Spurgeon, and Kepastaglou from SuperSTEM (UK)) held at the Microscopy and Microanalysis meeting in Portland, Oregon (pre-meeting congress).

- **Third NexTEM being planned for Johns Hopkins University in late 2020/early 2021. Join us! We welcome experts in:**
  - Data science/AI/Analytics
  - High performance computing
  - Microscopy
  - Physics, Materials Science, Chemistry, Biology, Manufacturing….we all need intelligent microscopy!

NexTEM
Next-Generation Transmission Electron Microscopy

Next-Generation Transmission Electron Microscopy Workshop

Beyond Current Limits of Resolution, Environments, and Data Analysis

Topics

Advanced Detector and Spectroscopy Developments

- Design and use of novel detectors to investigate material structure and functionality, including 4D STEM and ptychography.
- Vibrational and phonon spectroscopies at unprecedented spatial and energy resolution.
- Methods to conduct high-resolution imaging and spectroscopy of beam-sensitive samples.
- Examination of materials structure and chemistry at cryogenic temperatures.

Frontiers of In Situ / Operando Microscopy

- Advances in S/TEM methods and instrumentation to capture the dynamics of complex materials systems, including alloys, thin films, nanoparticles, and liquids.
- Investigation of materials under stimulus across a range of sample environments and temperatures.
- New workflows for in situ experimentation to ensure reliability, reproducibility, and improve data quality.

Data-Driven Microscopy and Analysis

- Machine learning-based analysis of materials structure, dynamics, and defects.
- Integration of multiple large-scale imaging and spectroscopic data streams to elucidate physical descriptors of complex systems and phenomena.
- High-throughput simulation approaches to guide the interpretation of experimental datasets.

Invited Speakers

David Muller
Cornell University

Naoya Shibata
University of Tokyo

Stig Helveg
Haldor Topsoe

Quentin Ramasse
SuperSTEM

Luiz Tizei
Université Paris Sud

Paul Voyles
University of Wisconsin–Madison

Hamish Brown
Lawrence Berkeley National Laboratory

Rama Vasudevan
Oak Ridge National Laboratory

Chongmin Wang
Pacific Northwest National Laboratory

Pacific Northwest NATIONAL LABORATORY

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