OAK RIDGE National Laboratory

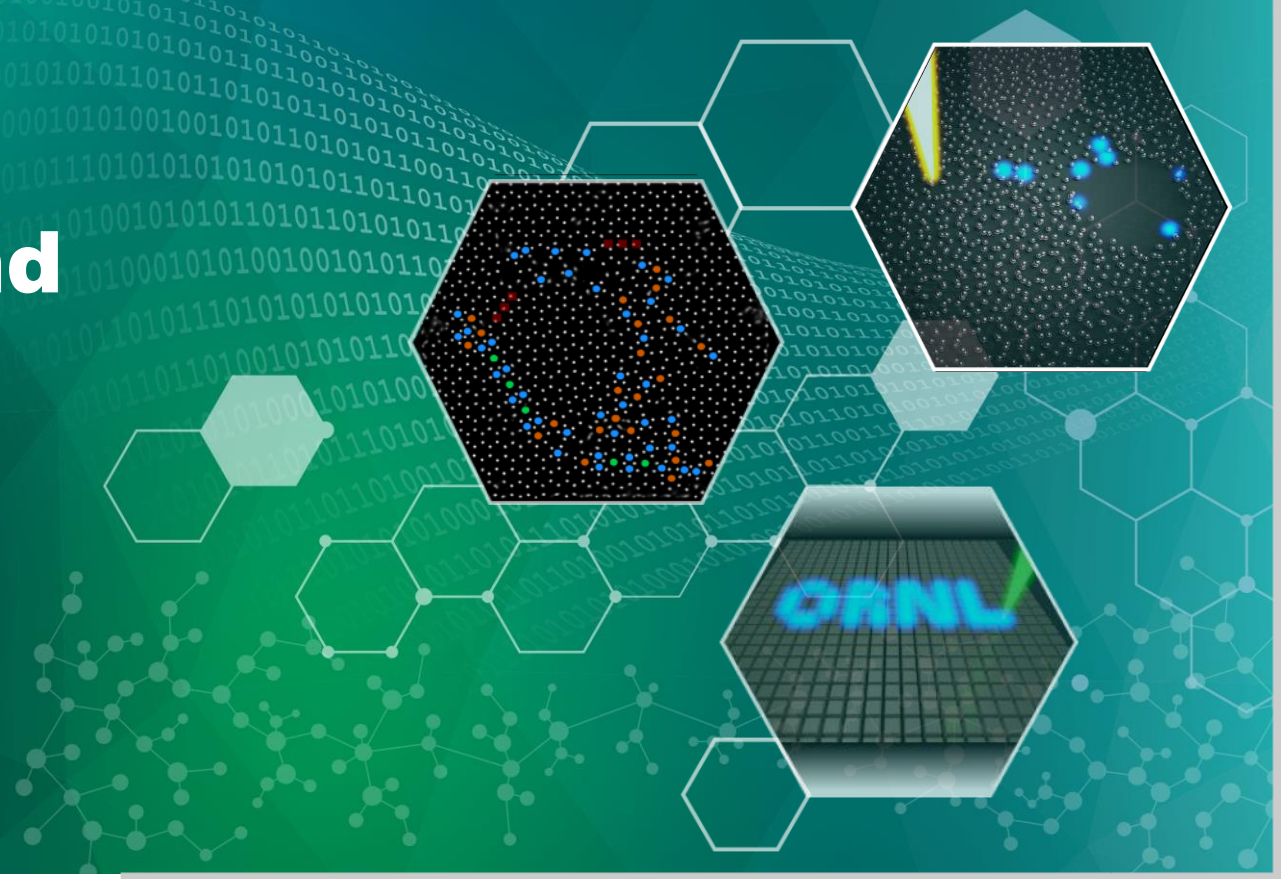
Lecture XVIII

Encoders-decoders and structure-property relationships

Kevin M. Roccapriore, Sergei Kalinin

August 04, 2023

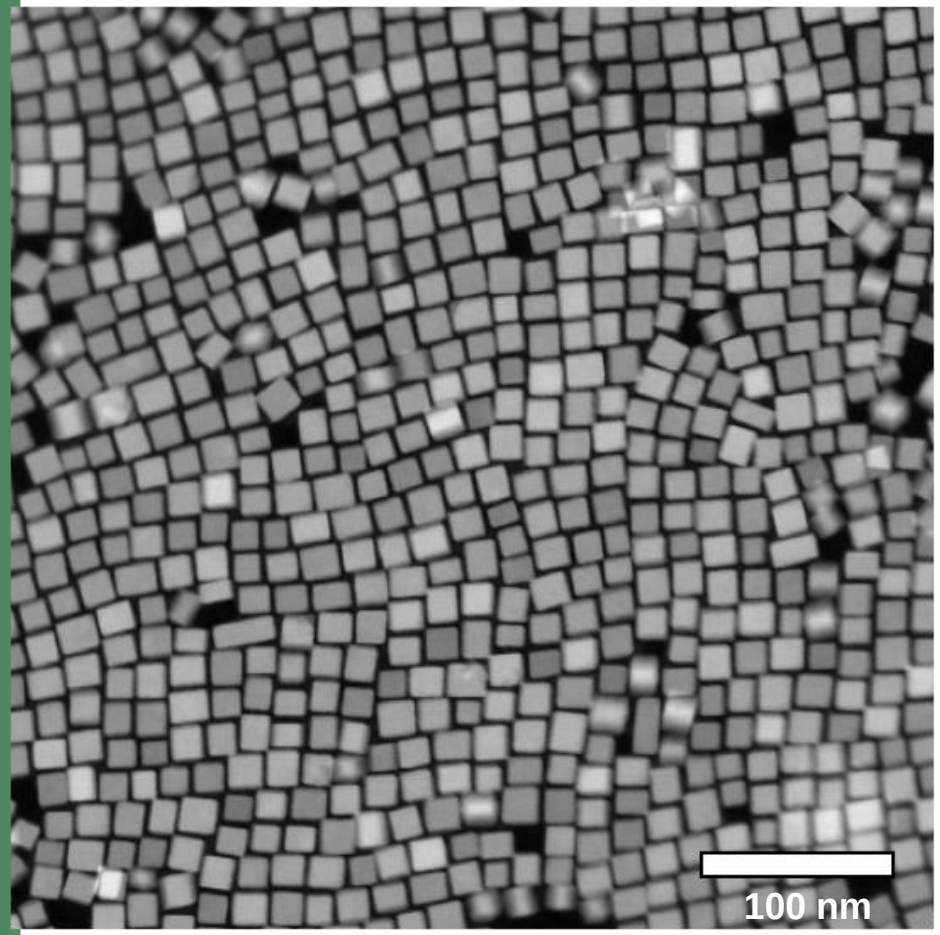
Center for Nanophase Materials Sciences
Oak Ridge National Laboratory



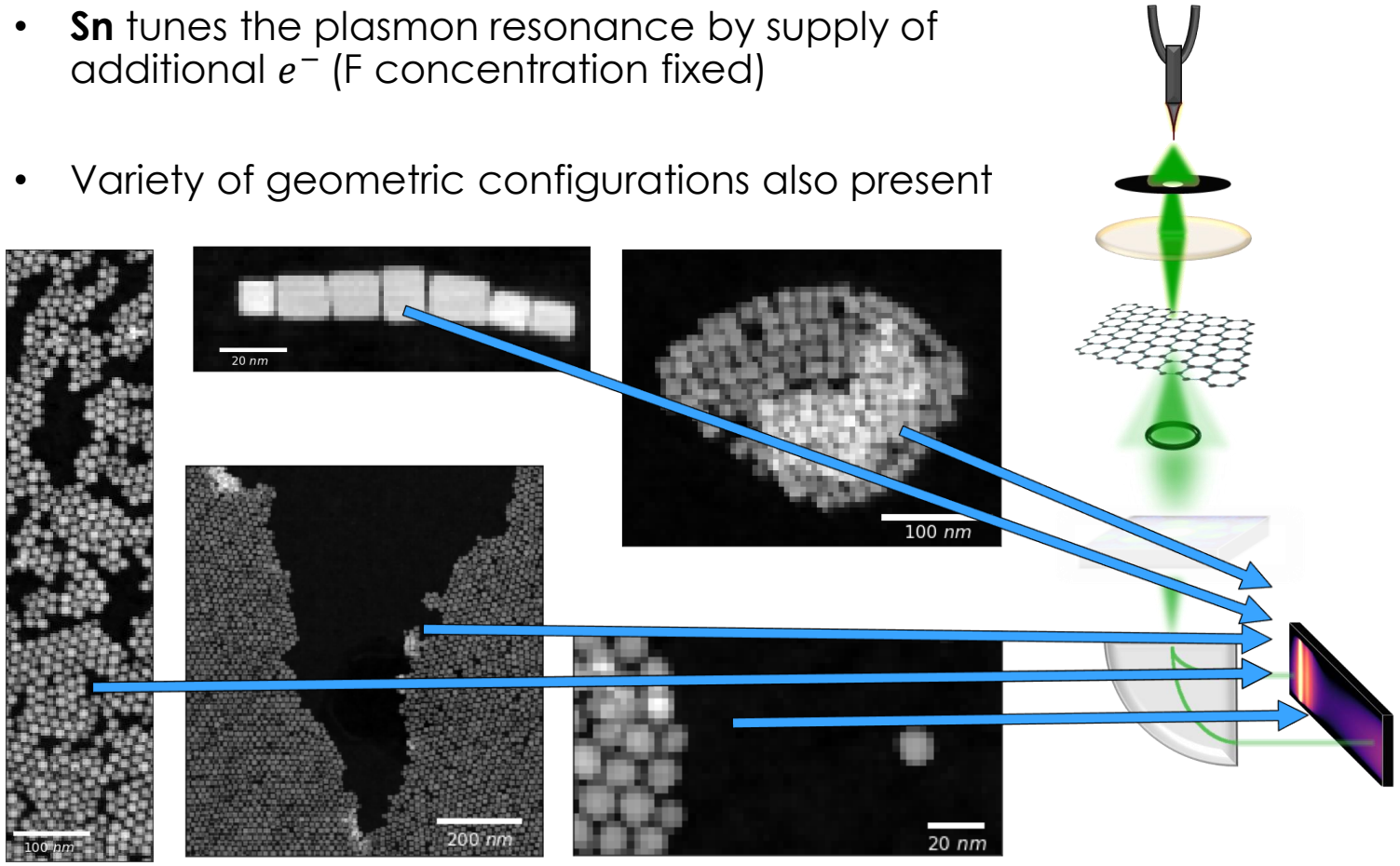
ORNL is managed by UT-Battelle LLC for the US Department of Energy

U.S. DEPARTMENT OF ENERGY

A system to realize structure-property relationships
Complex nanoparticle assemblies

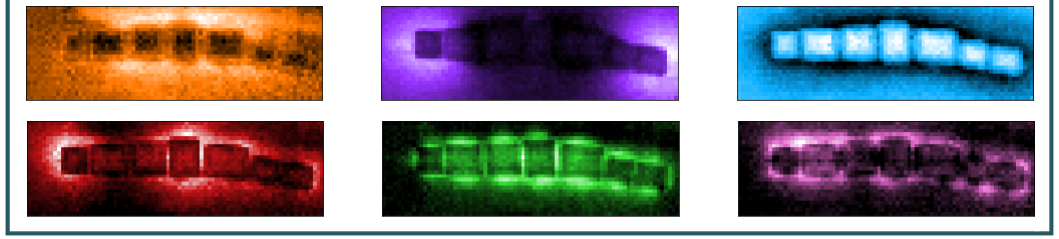


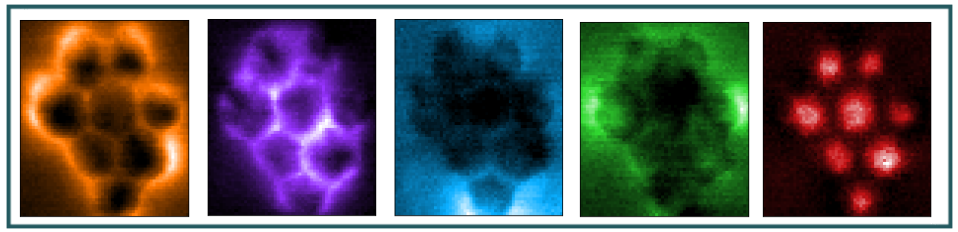
- Self-assembled monolayer of **metal oxide nanoparticles** (**F**,**Sn** co-doped indium oxide)
- **Sn** tunes the plasmon resonance by supply of additional e^- (F concentration fixed)
- Variety of geometric configurations also present

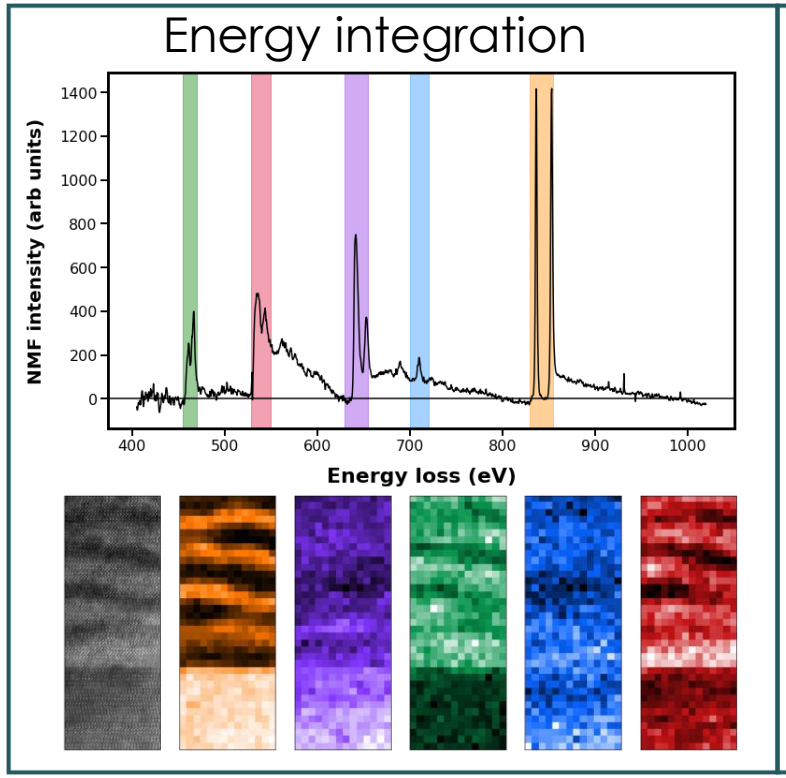


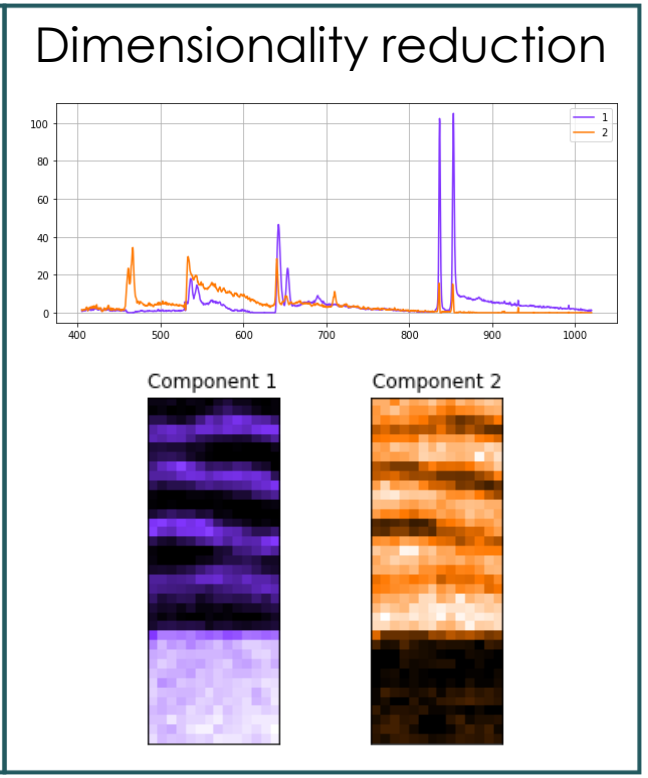
Electron energy loss spectroscopy (EELS)

- Can consider to be another signal in the form of a **1D spectrum**
- Collect EEL spectrum in an (x,y) grid: EELS imaging
- To better visualize / understand these 3D signals, can integrate specific spectral bands, or dimensionally reduce (PCA, NMF) them
- Applications in plasmonics & nanophotonics

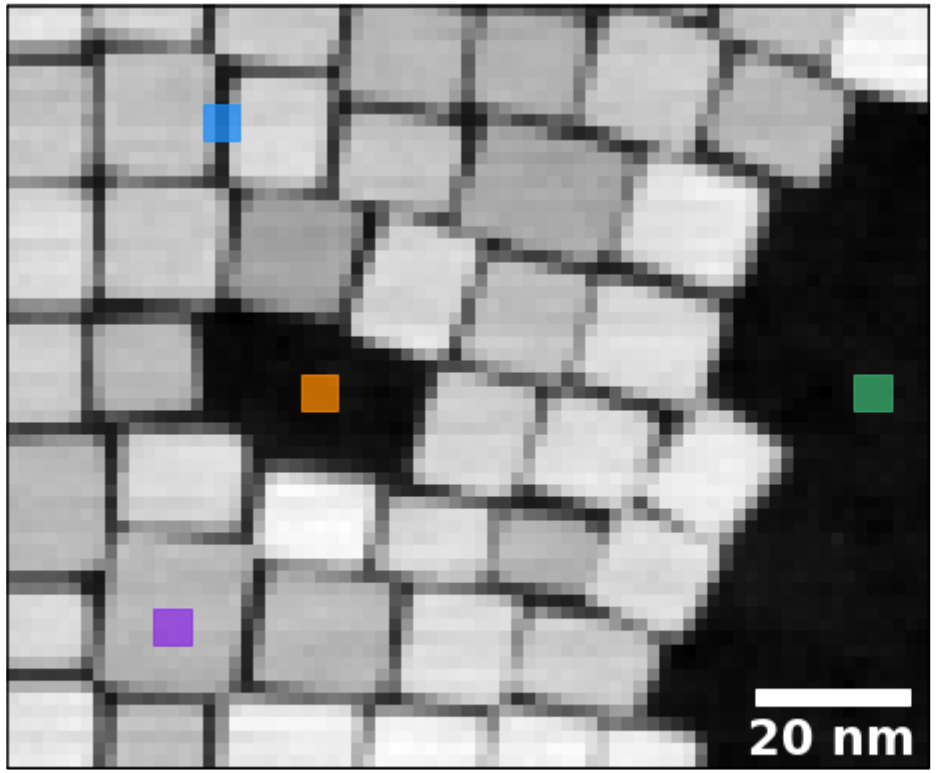


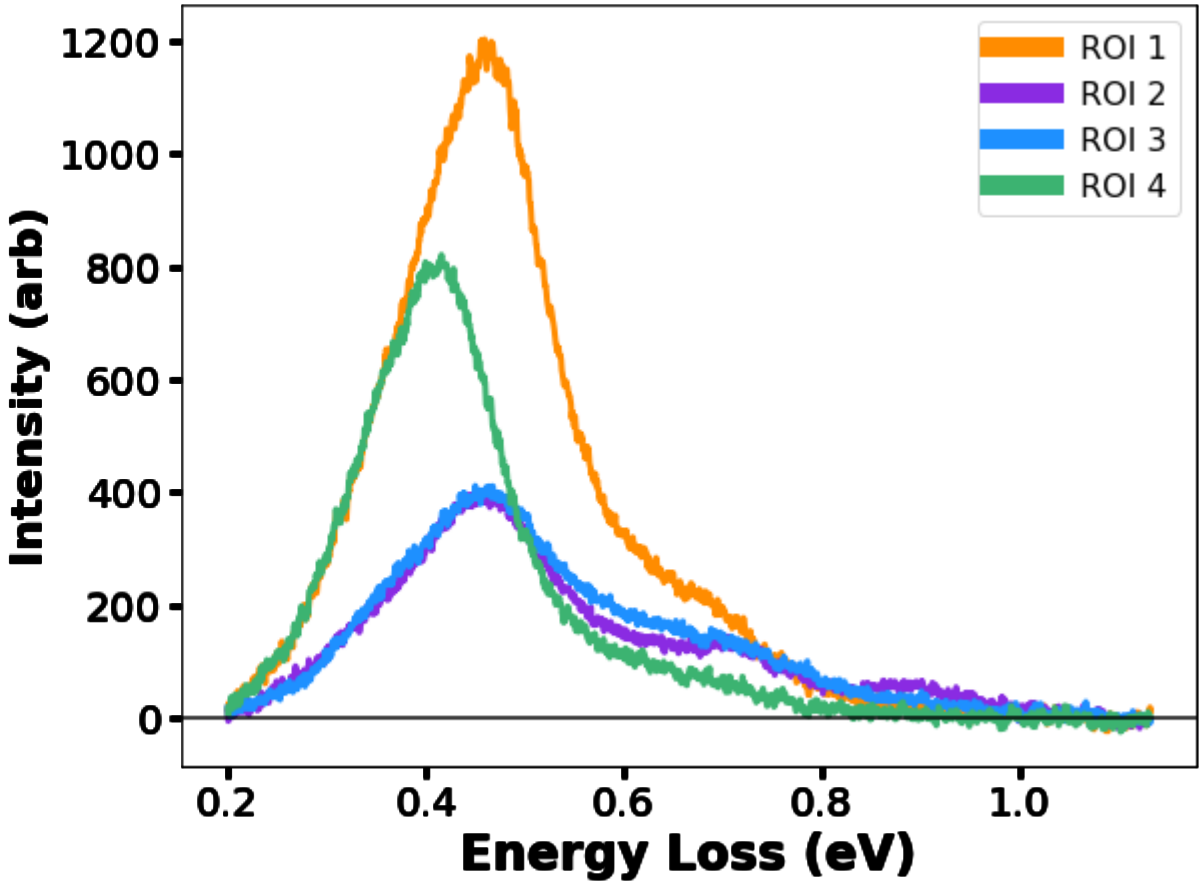




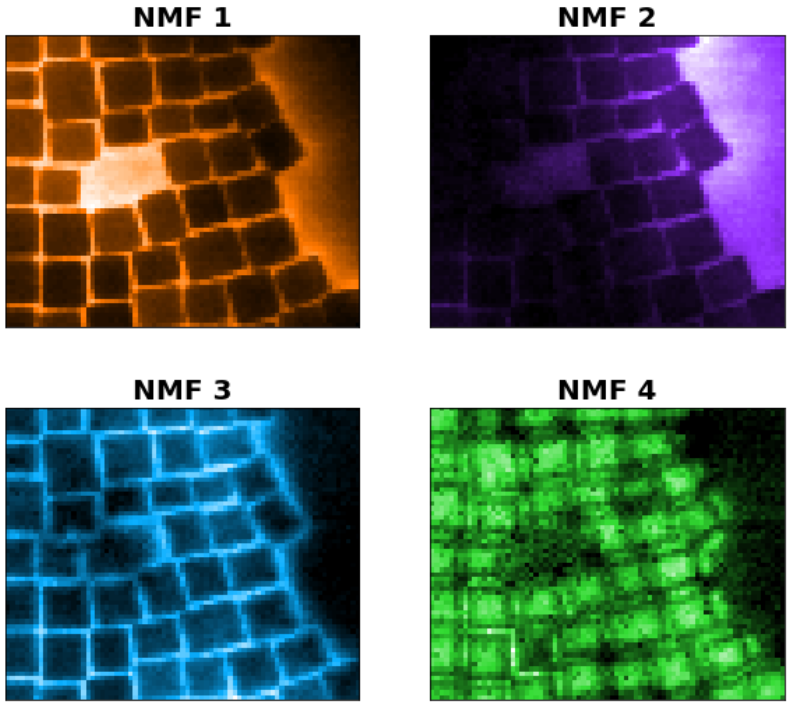


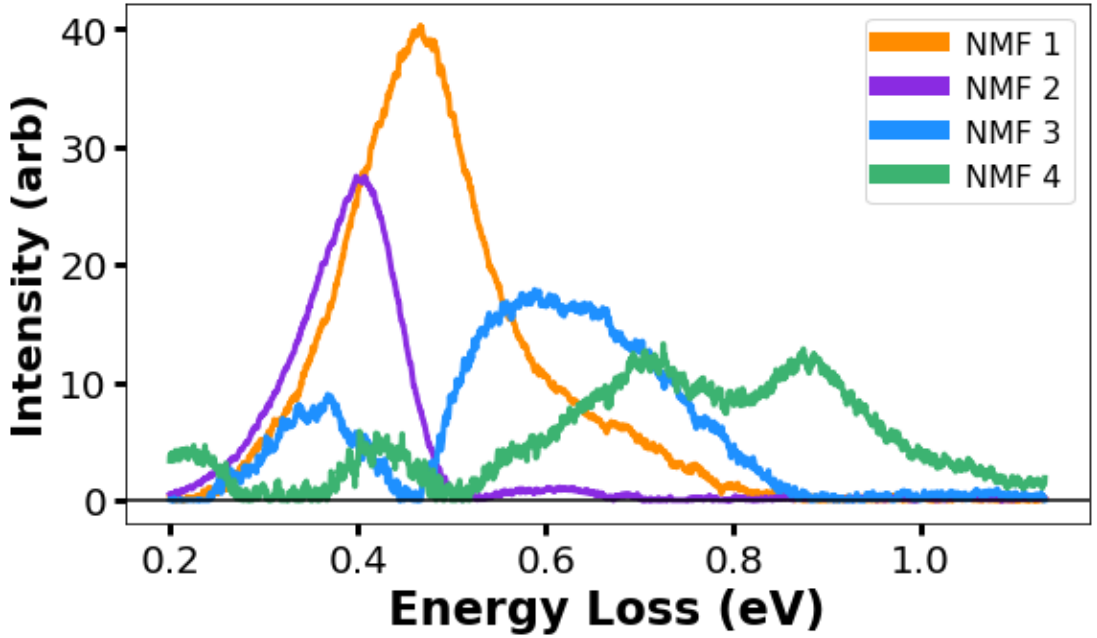
Plasmonic response in complex geometries





Plasmonic response in complex geometries
NMF



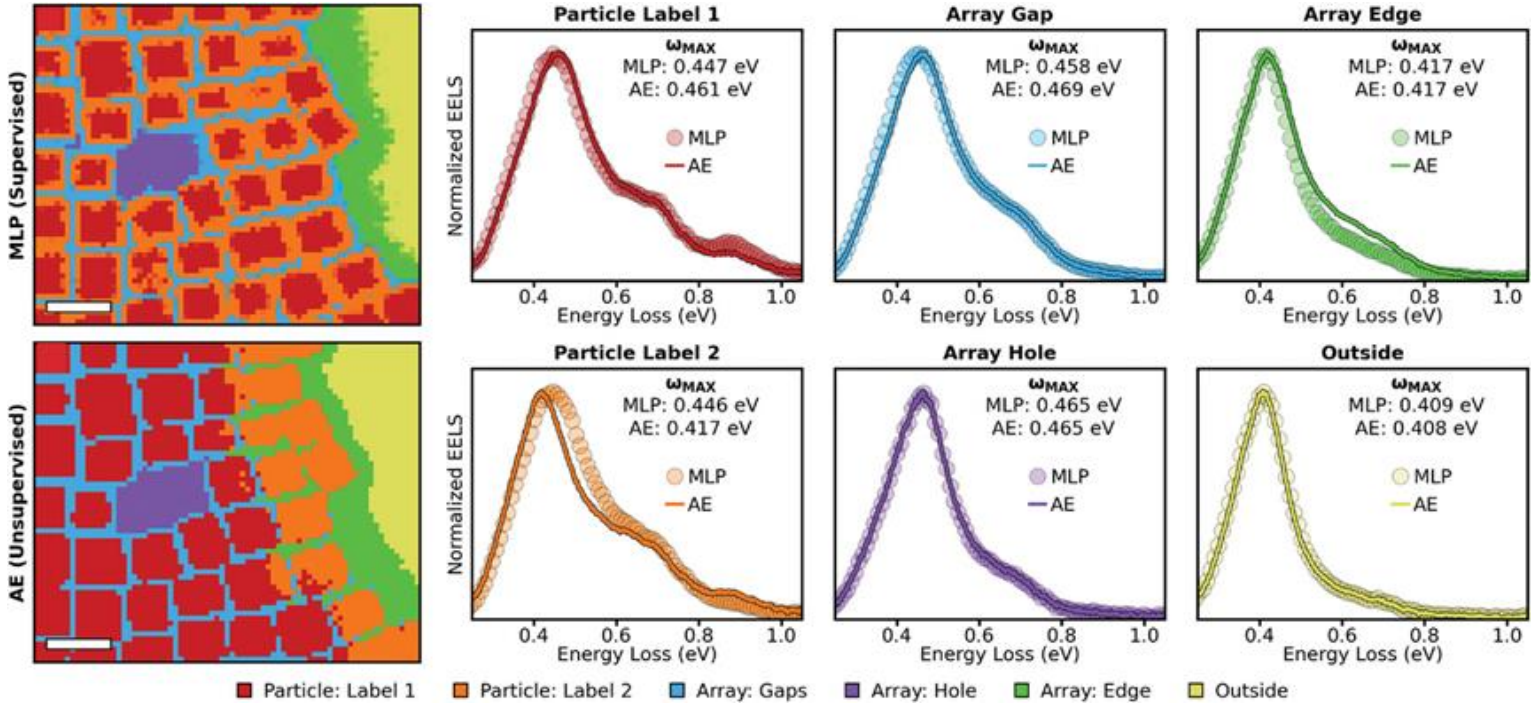


- Great way to explore system
- Visualization of hyperspectral data

- Multiple modes per pixel
- Non-physical extraction
- No relationship between geometry established

Separating effects: pixel labeling

Nonlinear ML methods: Multilayer Perceptron, Autoencoder network

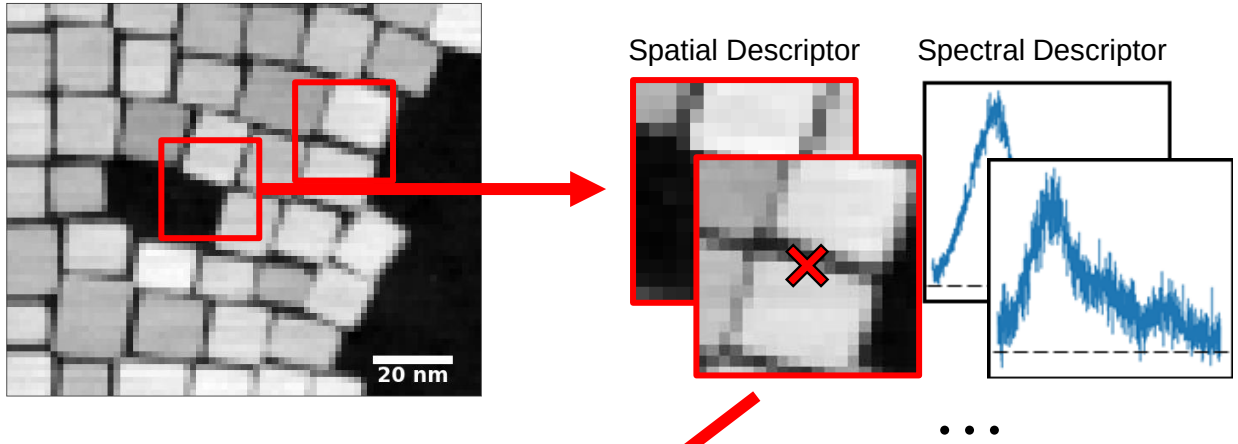


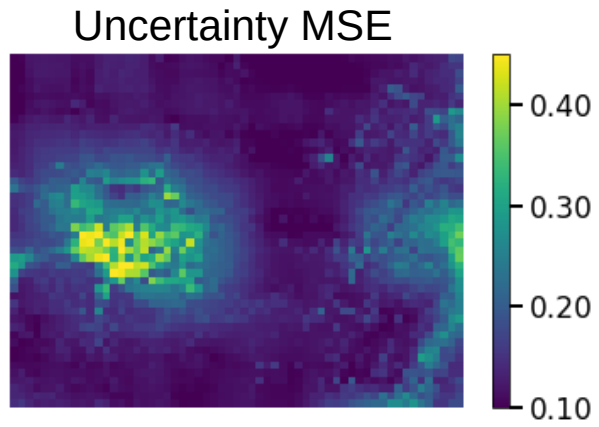
- Can separate effects
- Knows no physics
- **No relationship between geometry established**

OAK RIDGE National Laboratory

S.V. Kalinin, **K.M. Roccapriore**, S.-H. Cho, D.J. Miliron, R. Vasudevan, M. Ziatdinov, J.A. Hachtel, *Adv. Opt. Mater.* **2021**, 2001808

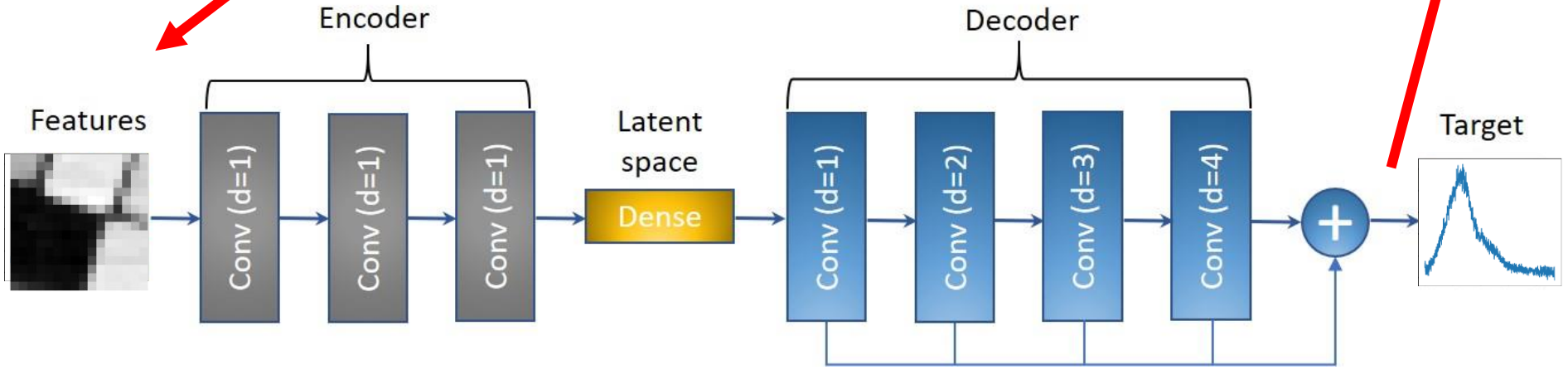
Structure-property correlations:
Autoencoder neural network





Training

Evaluation



im2spec: Latent space visualization and error mapping

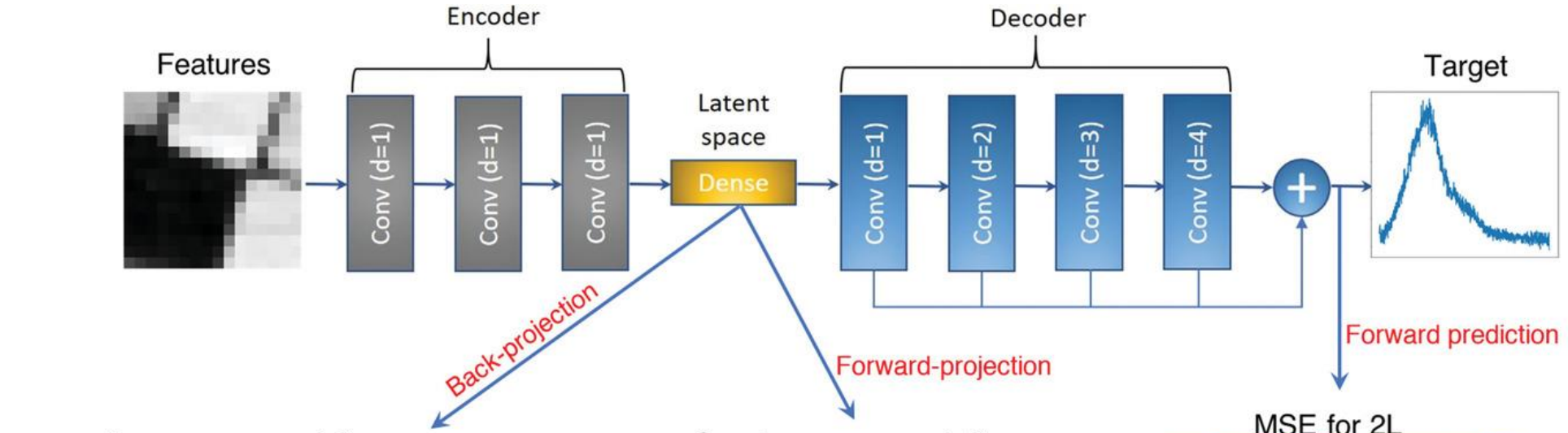
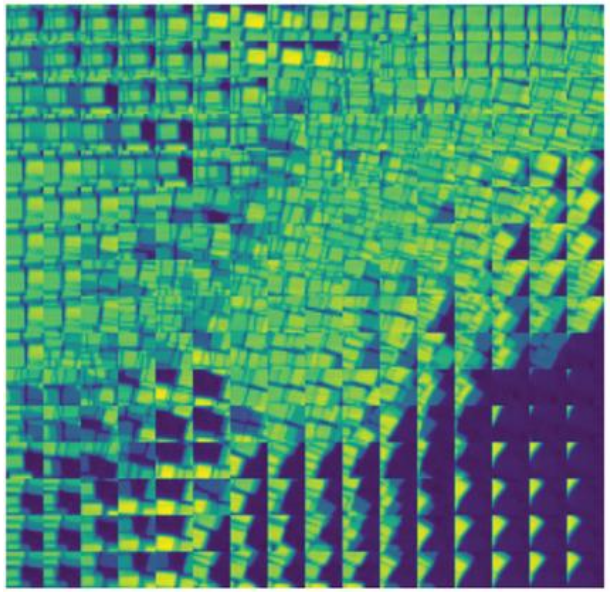
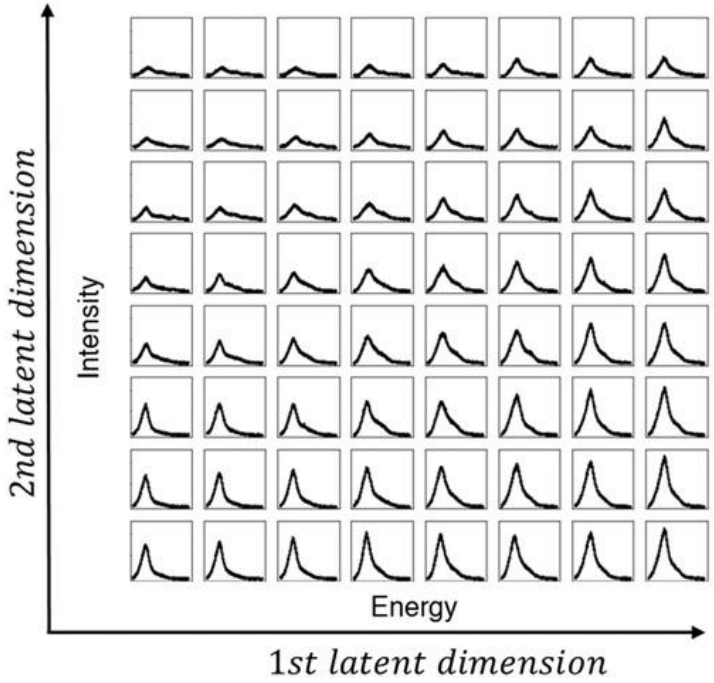


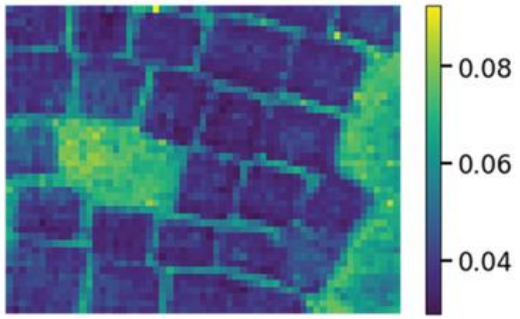
Image representation



Spectrum representation



MSE for 2L

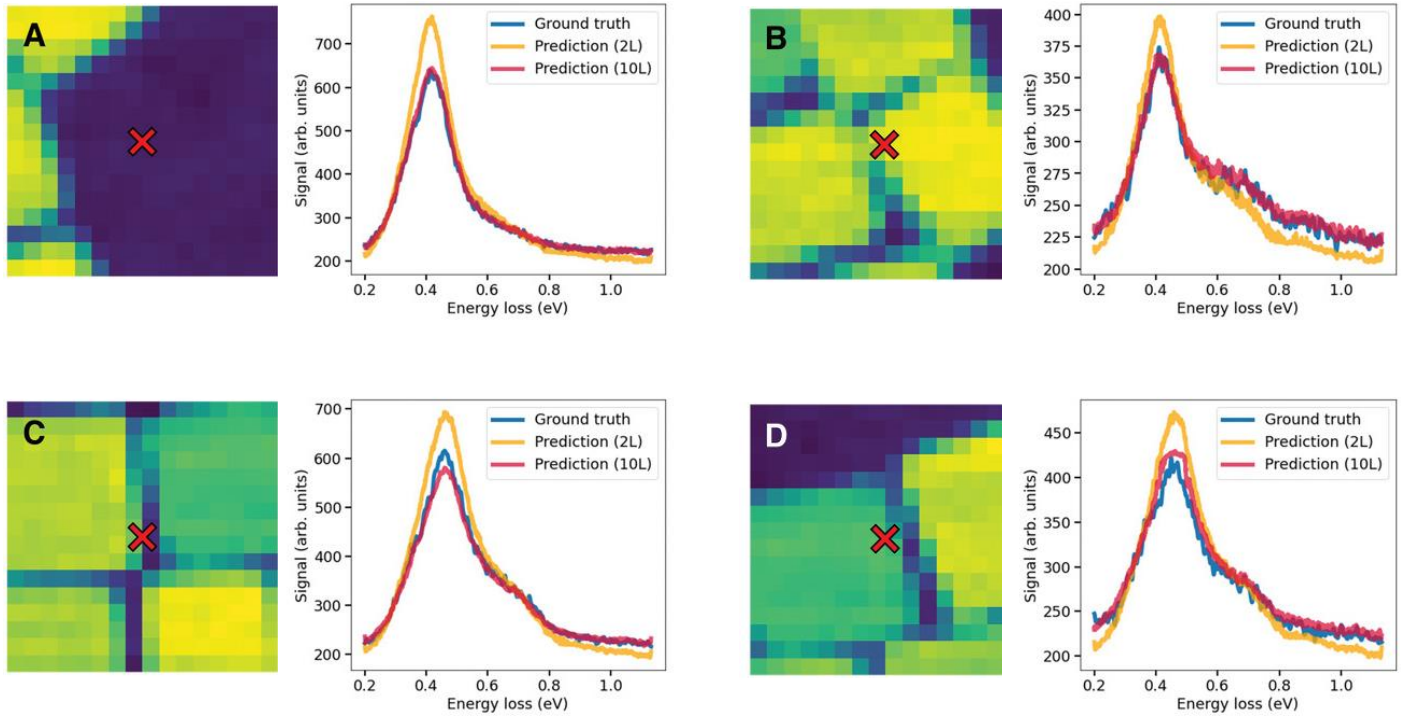


MSE for 10L

OAK RI
National Laboratory

Encoder-decoder neural networks: Predictions

"*im2spec*"



- After training, predict the spectral response of a geometric arrangement that the network has **never encountered**
- **2L** and **10L** refers to number of latent dimensions chosen
- # spectral channels
  - can we describe spectra by simpler means?
  - (for future thoughts…)

- **Library** of geometric-plasmonic relationships
- Towards solution of inverse design in nanophotonics and other fields

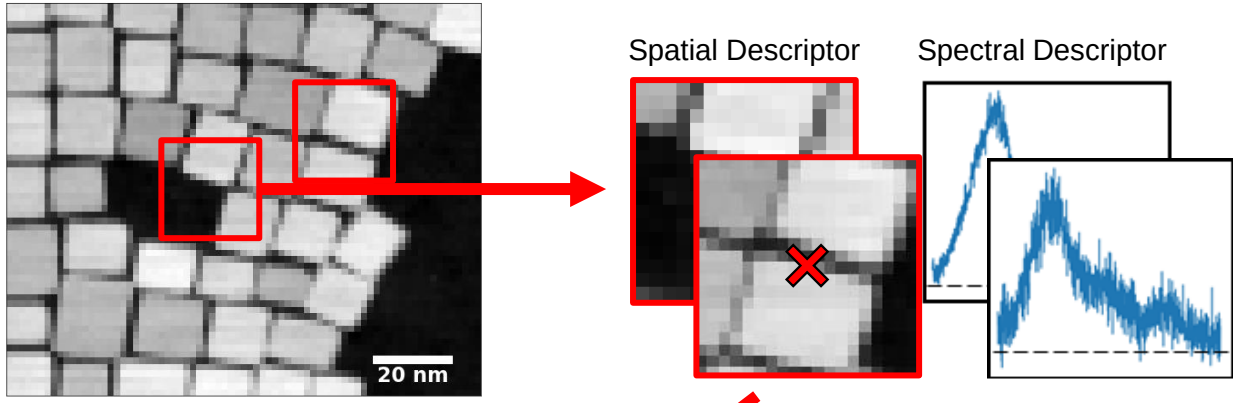
Limitations

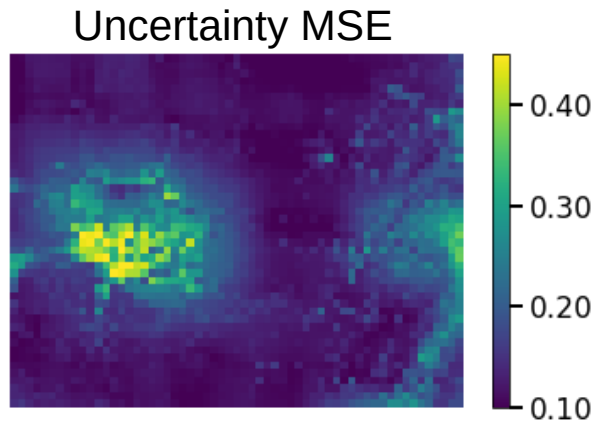
- System-dependent (currently fails to generalize)
- Requires pre-acquired training data
- Working with *as-fabricated* systems

OAK RIDGE National Laboratory

Roccapriore, K. M., Ziatdinov, M., Cho, S. H., Hachtel, J. A., Kalinin, S. V., Predictability of Localized Plasmonic Responses in Nanoparticle Assemblies. *Small* 2021, 2100181.

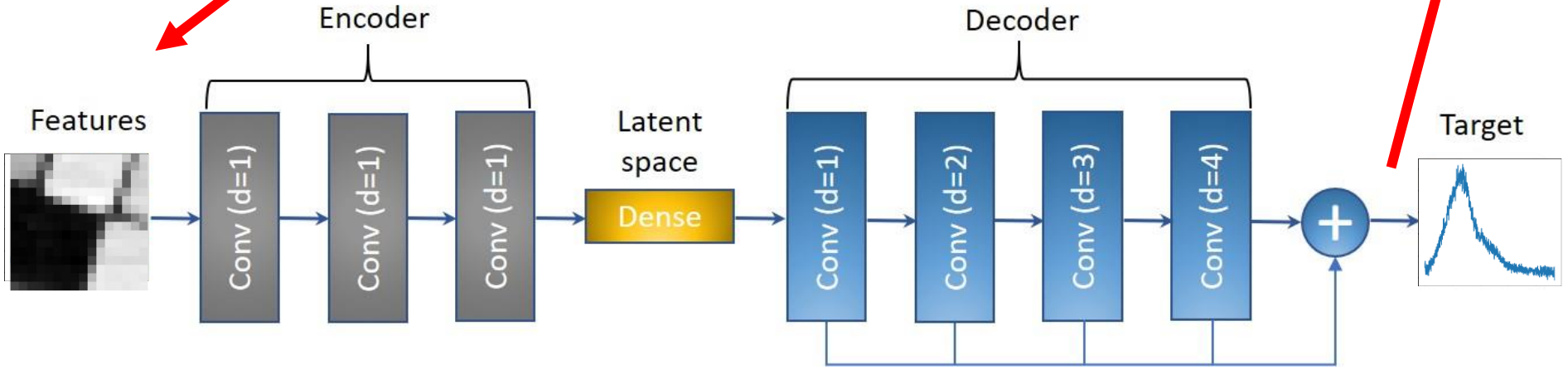
Structure-property correlations:
Autoencoder neural network



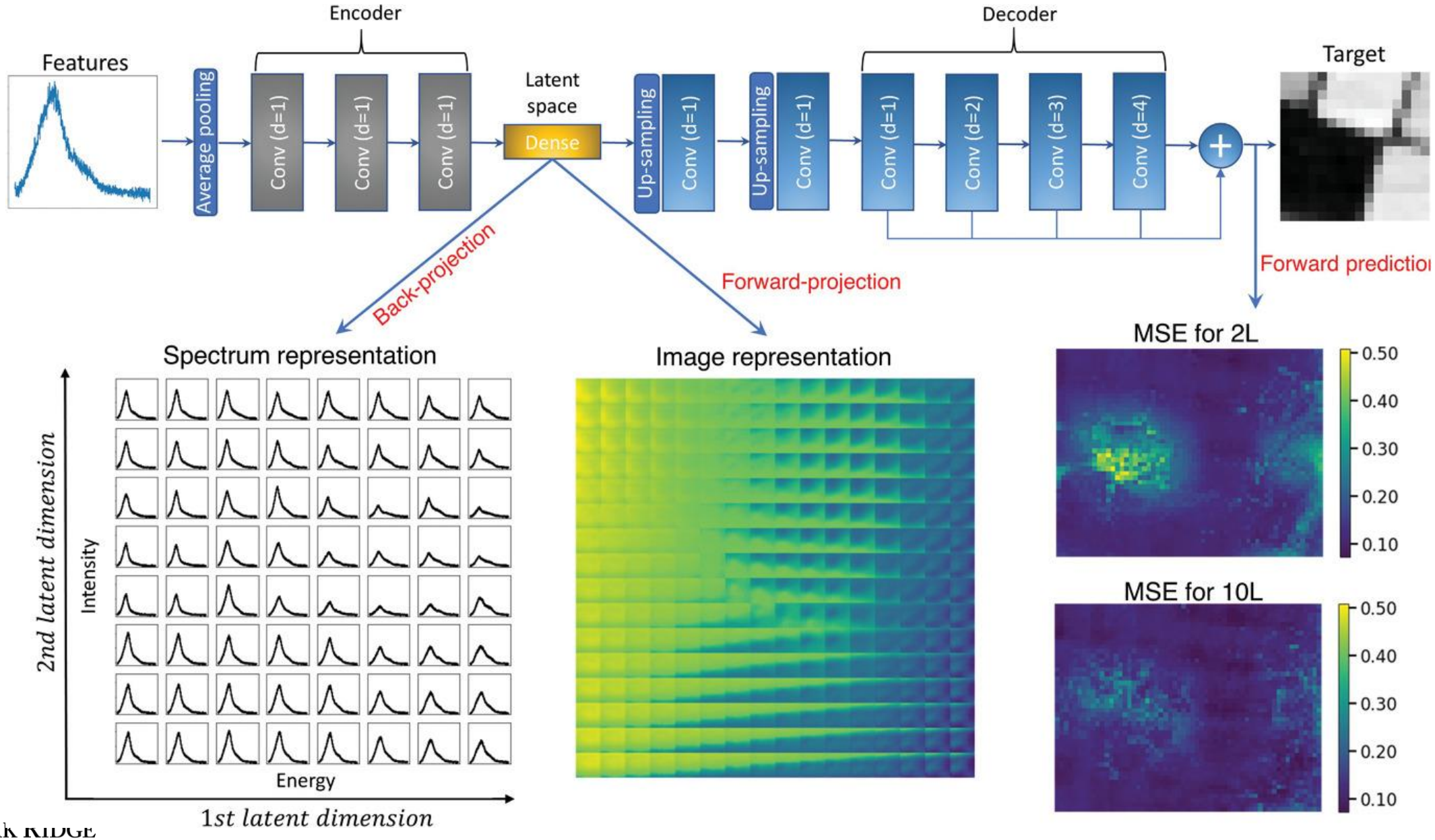


Training

Evaluation

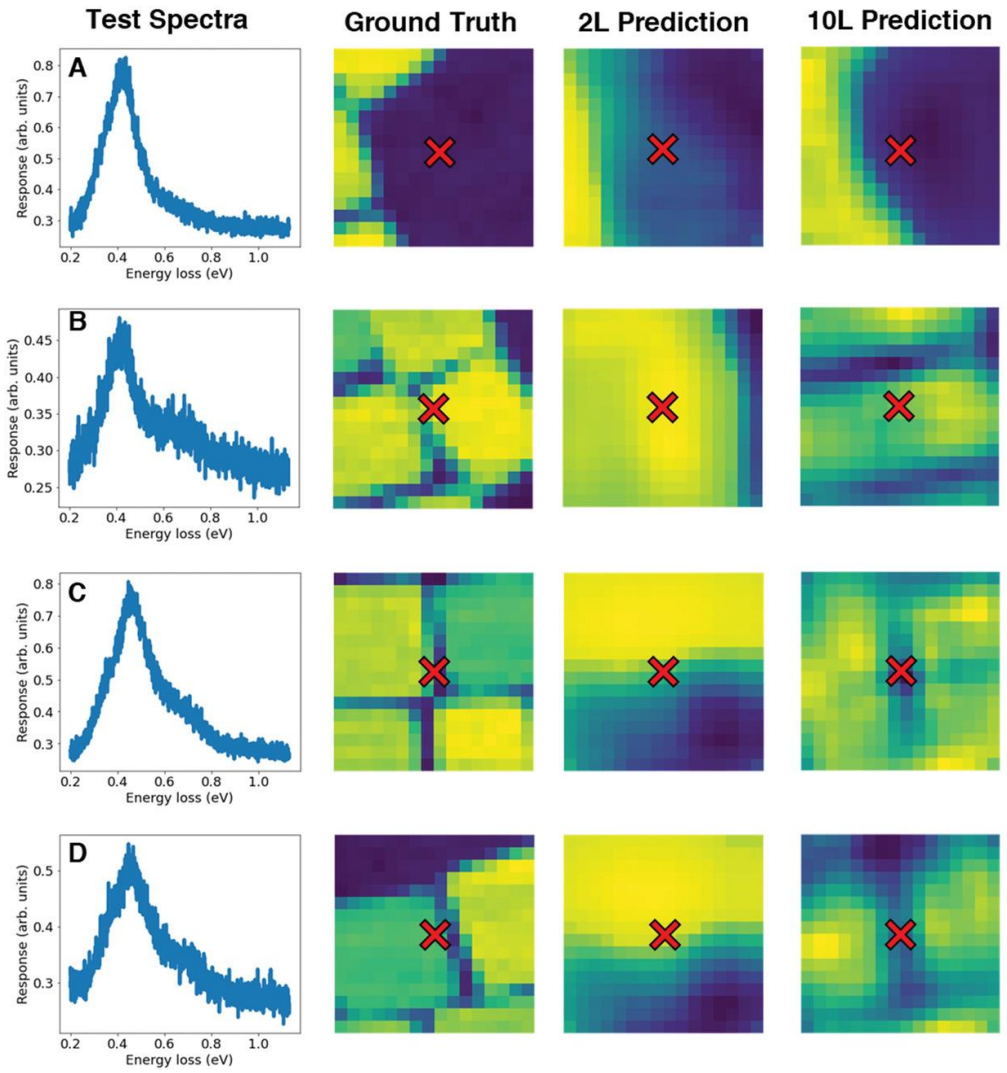


spec2im: Latent space visualization and error mapping



Encoder-decoder neural networks: Predictions

"*spec2im*"



- After training, predict the spectral response of a geometric arrangement that the network has **never encountered**
- **2L** and **10L** refers to number of latent dimensions chosen
- # spectral channels
  - can we describe spectra by simpler means?
  - (for future thoughts…)

Limitations

- System-dependent (currently fails to generalize)
- Requires pre-acquired training data
- Working with *as-fabricated* systems

OAK RIDGE National Laboratory

Roccapriore, K. M., Ziatdinov, M., Cho, S. H., Hachtel, J. A., Kalinin, S. V., Predictability of Localized Plasmonic Responses in Nanoparticle Assemblies. *Small* 2021, 2100181.

In summary:
Correlative structure property relationships

- We can **learn the relationship** between local geometry and plasmonic response

Aim

Desired spectral response

↓

Prediction of particle geometry

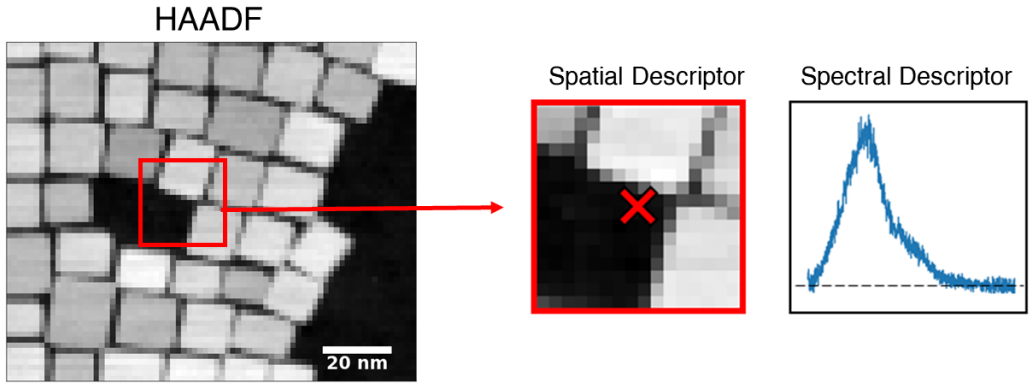
↓

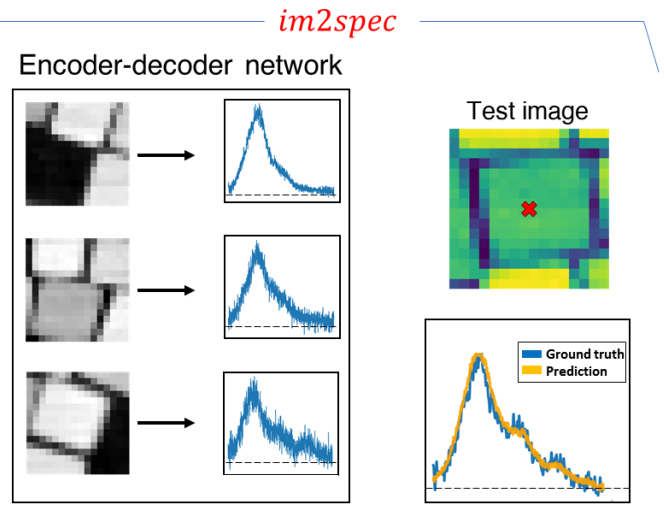
Fabricate geometry

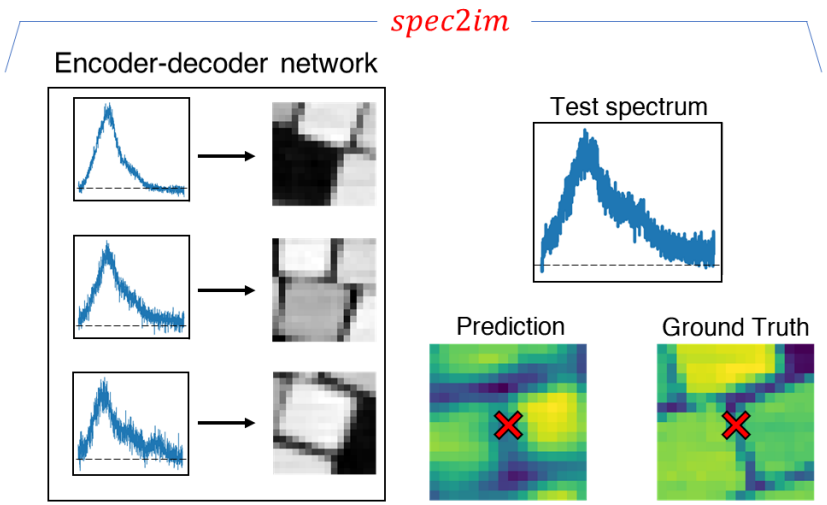
↓

Evaluate response

- We operate with **complete (preacquired)** 3D data
- Random particle geometries



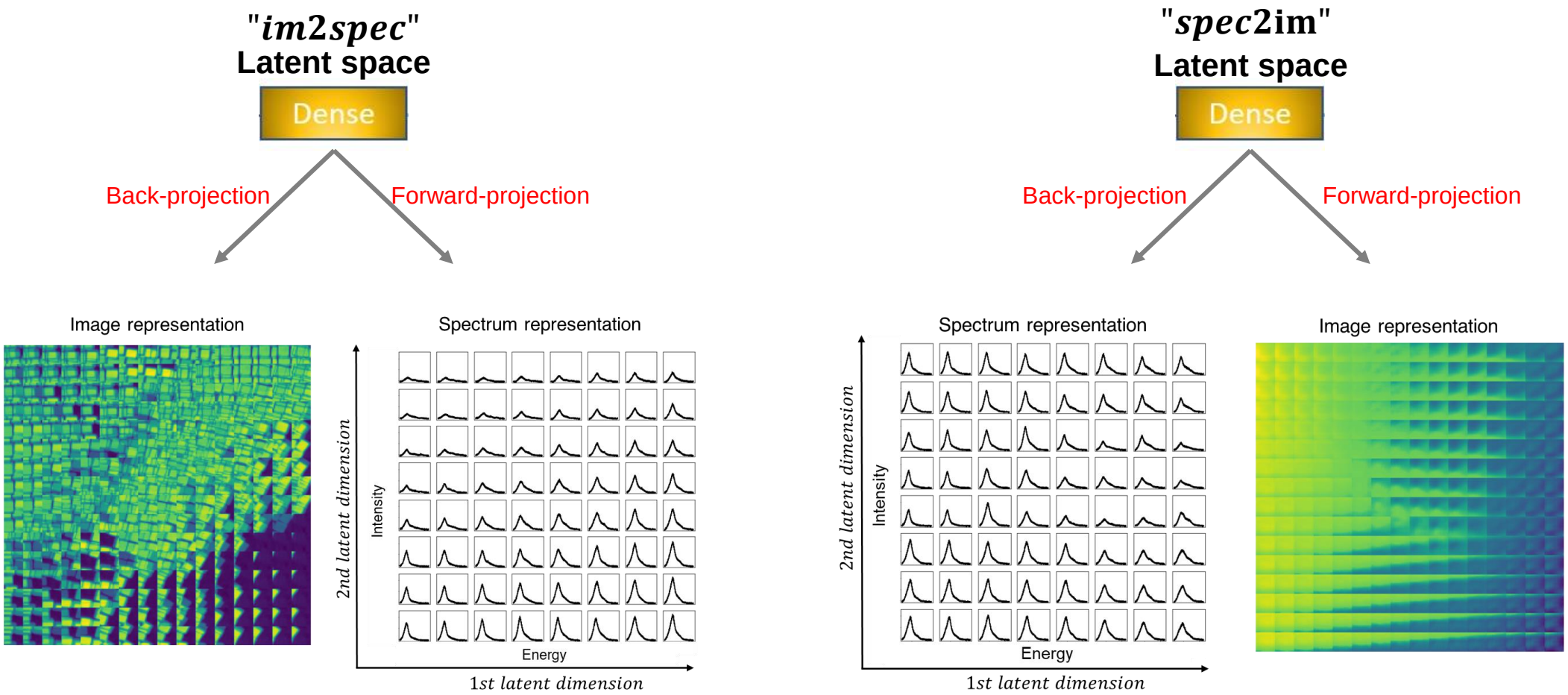




https://doi.org/10.1002/smll.202100181

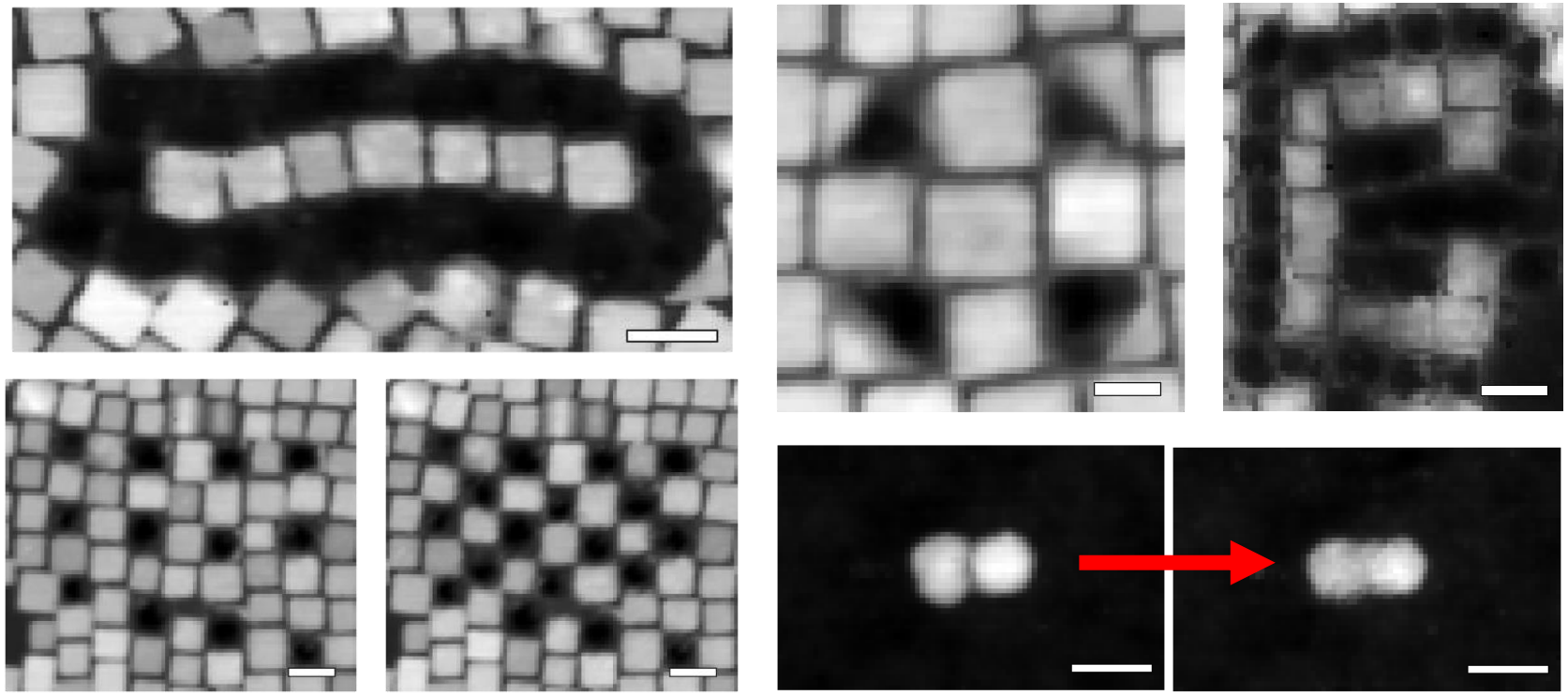
In summary:

Correlative structure property relationships (Visualizing)



Can we engineer the ones that exhibit spectral properties we desire? **under certain conditions, yes!**

- High current density (~10^6 A/cm^2)
- 60 – 200 kV beam energies
- Monitor plasmonic responses dynamically (EELS)



- Build a library of structure-property relationships to guide inverse design?

Going forward

- Increased geometric variation, fields of view…
- Moving beyond the *as-fabricated* particle restrictions: Designing and testing other geometries
- "inverse problem"
- Automated experiments **(on the fly)**!
  - *deep kernel learning*
- **Colab!**

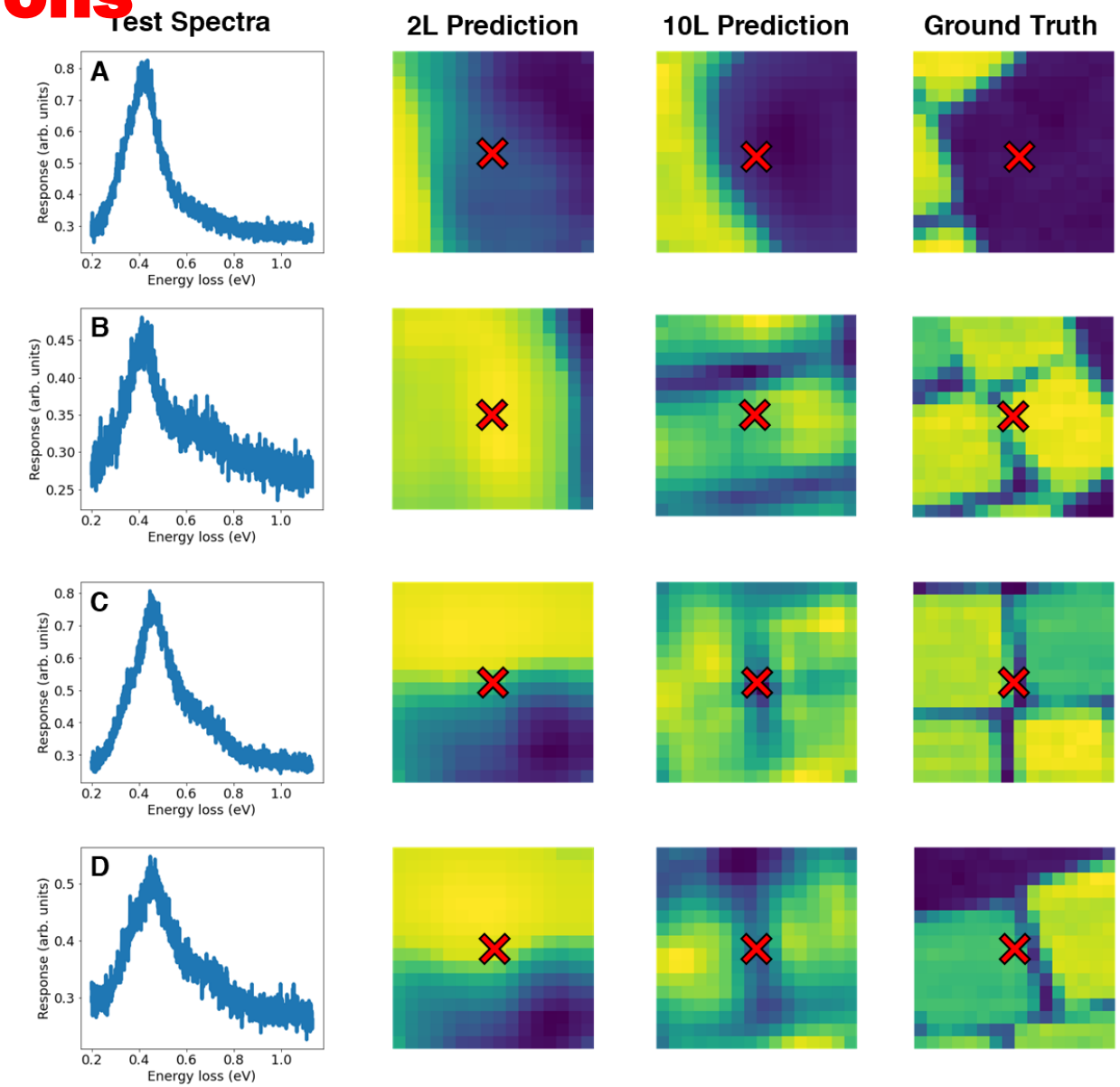
Open slide master to edit

im2spec paper link for STEM-EELS

https://onlinelibrary.wiley.com/doi/full/10.1002/smll.202100181

Encoder-decoder neural networks: Geometric Predictions

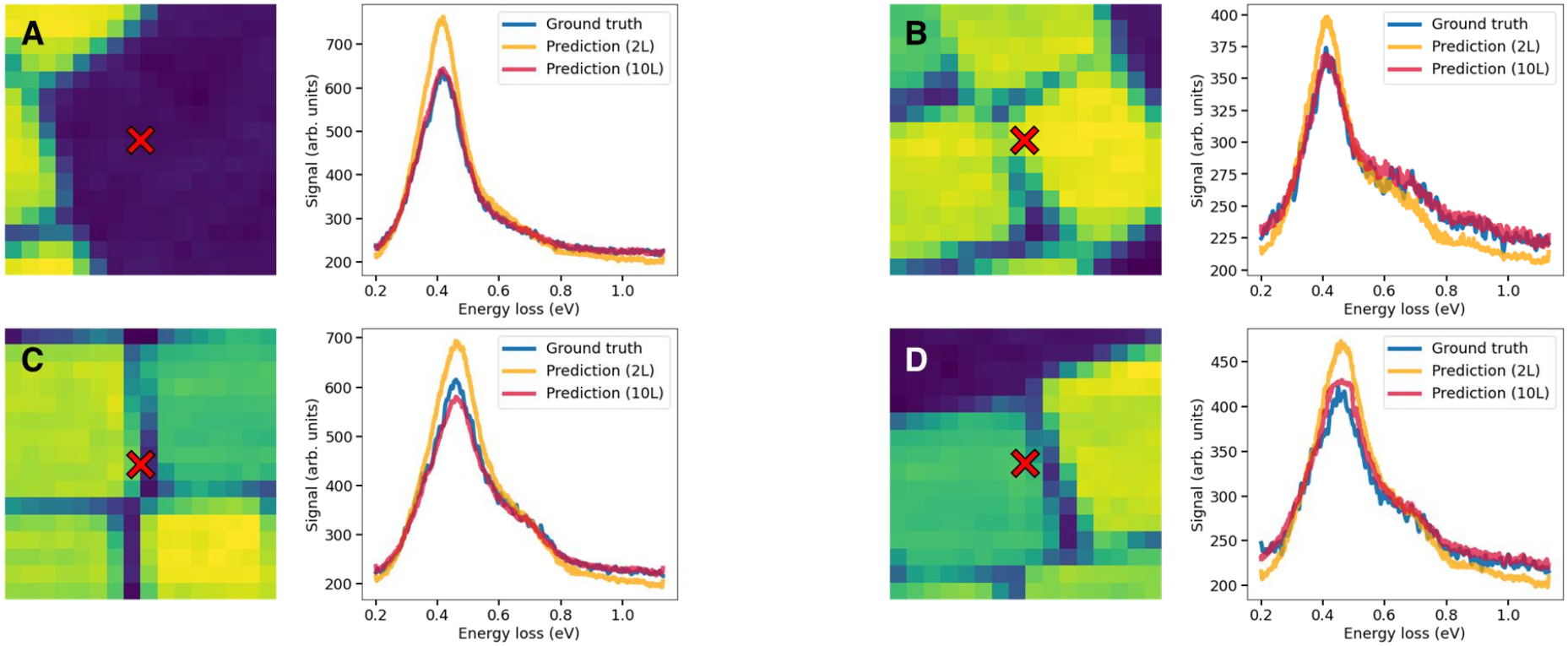
- Predict the geometric arrangement given a spectrum that the network has **never encountered**
- **2L** and **10L** refers to number of latent dimensions chosen



Roccapriore, K. M., Ziatdinov, M., Cho, S. H., Hachtel, J. A., Kalinin, S. V., Predictability of Localized Plasmonic Responses in Nanoparticle Assemblies. *Small* 2021, 2100181.

Encoder-decoder neural networks: Spectral Predictions

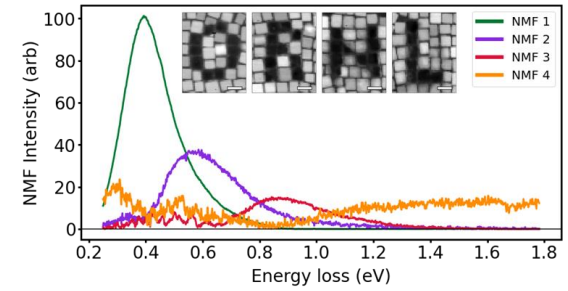
- After training, predict the spectral response of a geometric arrangement that the network has **never encountered**
- **2L** and **10L** refers to number of latent dimensions chosen

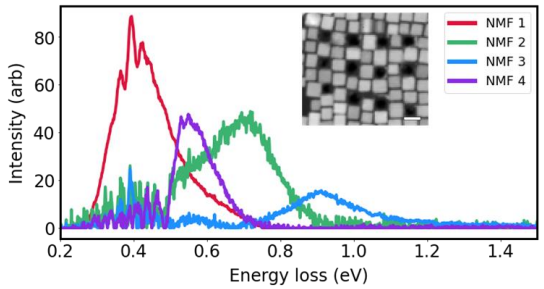


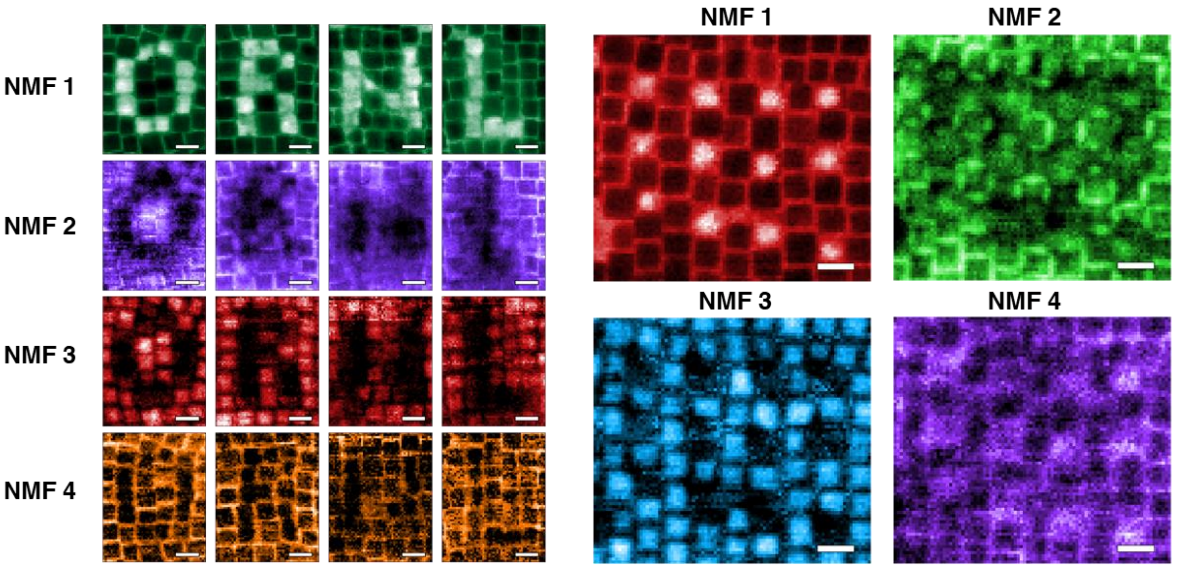
Roccapriore, K. M., Ziatdinov, M., Cho, S. H., Hachtel, J. A., Kalinin, S. V., Predictability of Localized Plasmonic Responses in Nanoparticle Assemblies. *Small* 2021, 2100181.

Electron beam modification of plasmon response

Space

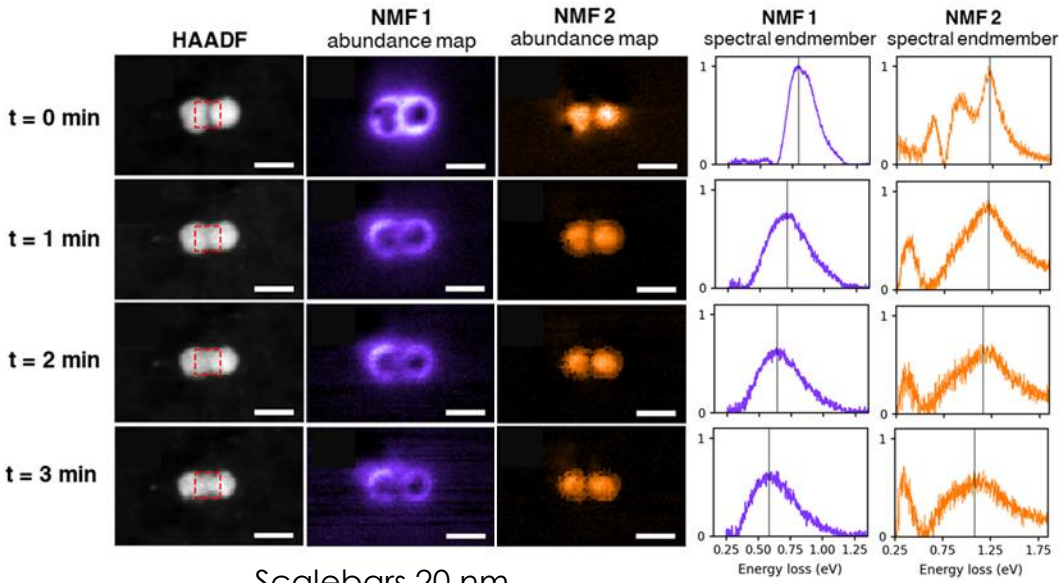






Scalebars 20 nm

Energy

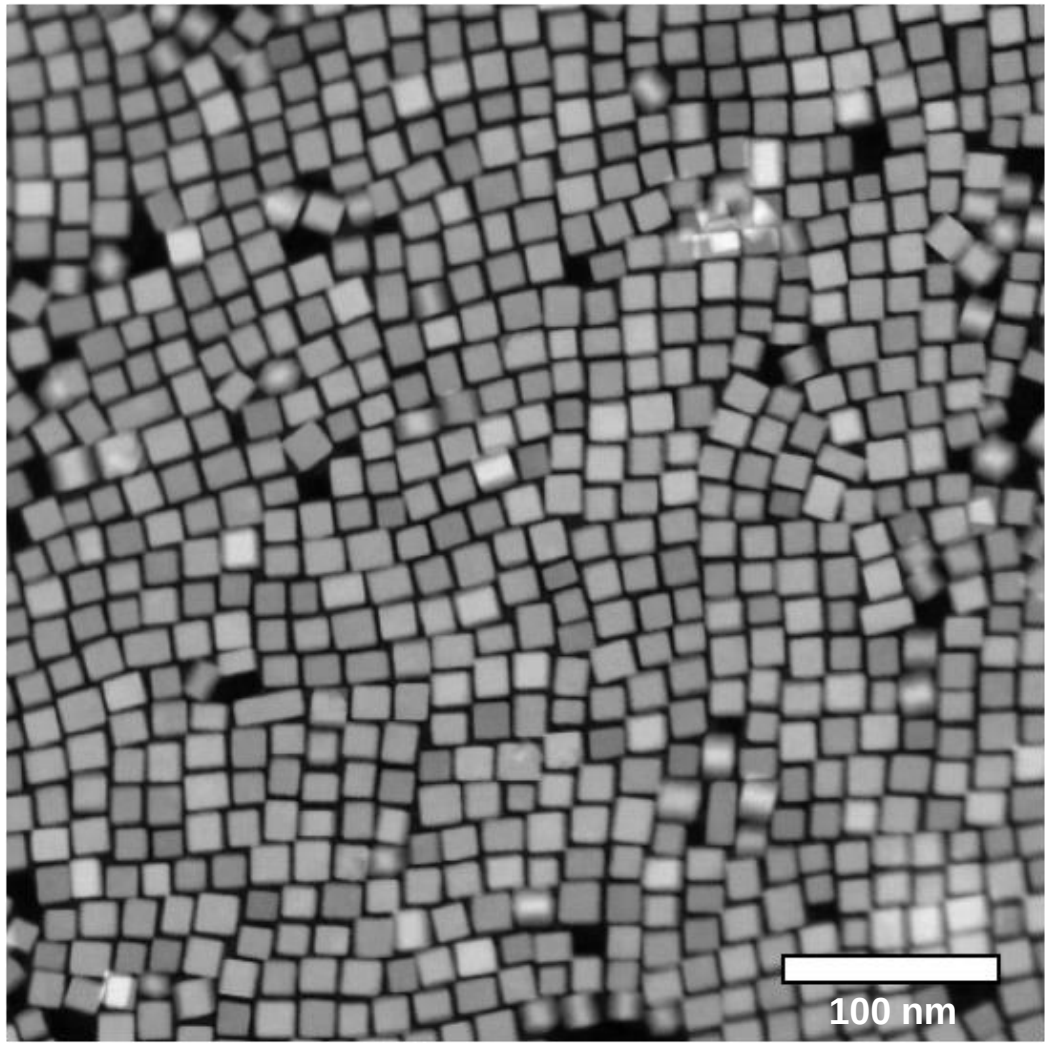


Scalebars 20 nm

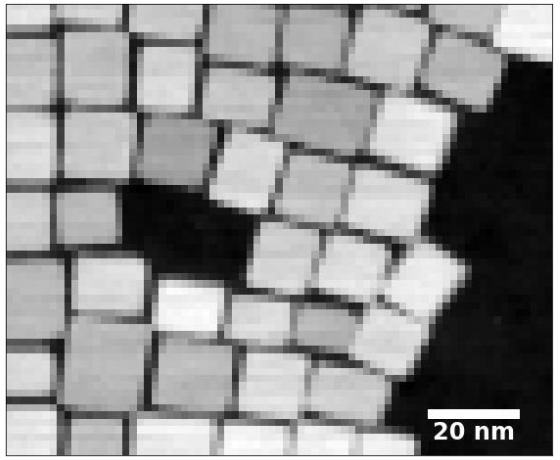
OAK RIDGE National Laboratory

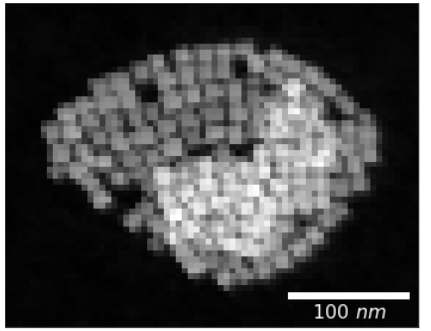
Roccapriore K.M., Cho S.-H., Lupini A.R., Milliron D.J., Kalinin S.V., arXiv preprint: 2104.02165, *under review Adv. Funct. Mater.*

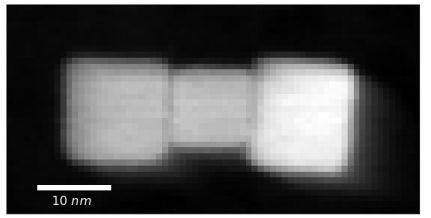
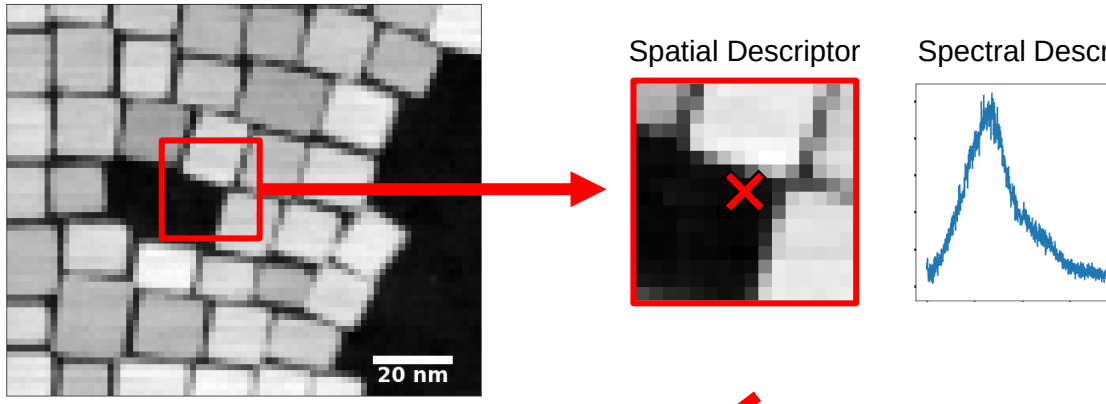
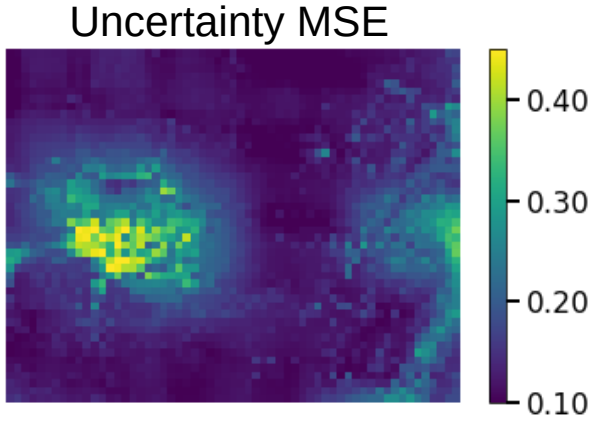
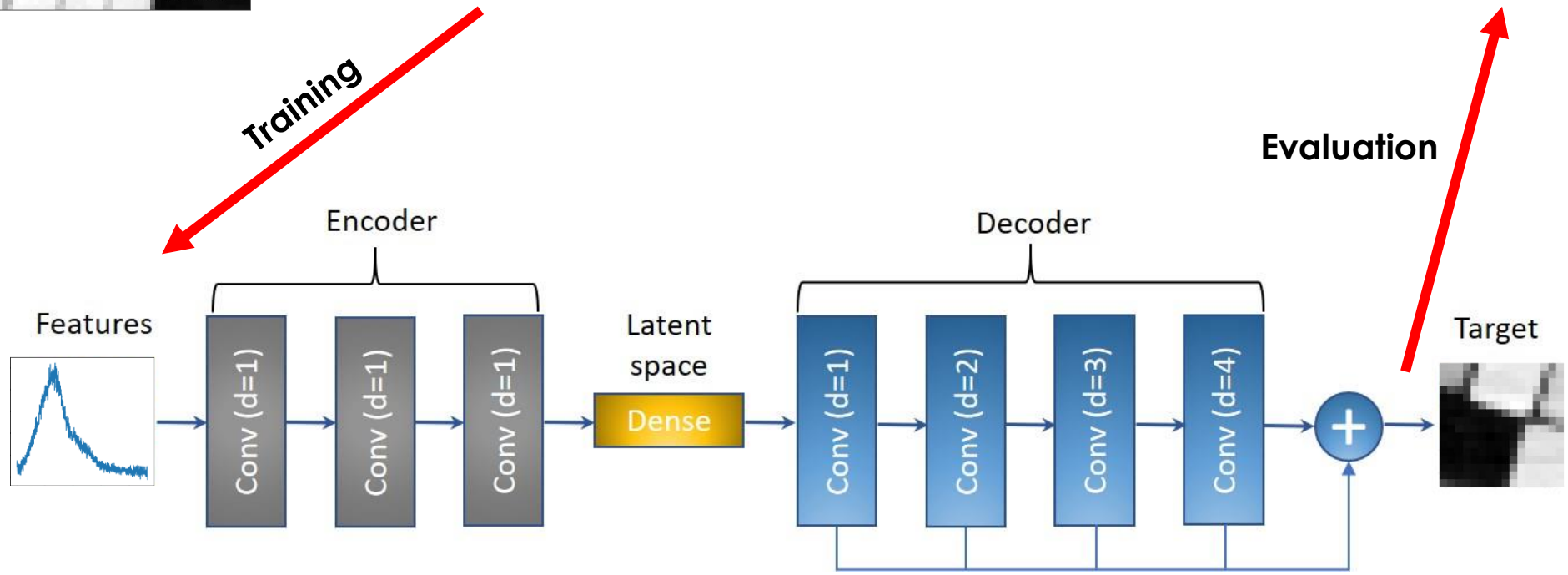
Material system: doped indium oxide



- Self assembled monolayer
- co-doped with F and Sn
- **Sn** tunes (*during synthesis*) the plasmon resonance by supply of additional e^-
- Supported on silicon nitride membrane







Structure-property correlations:
Autoencoder neural network