OAK RIDGE National Laboratory

Artificial Intelligence Methods for Microscopy Analysis and Knowledge Extraction

IMC-20 Pre-Congress Workshops
Spectral and Image Processing

Rama K. Vasudevan

Center for Nanophase Materials Sciences,
Oak Ridge National Laboratory

10th September 2023
Busan, Korea

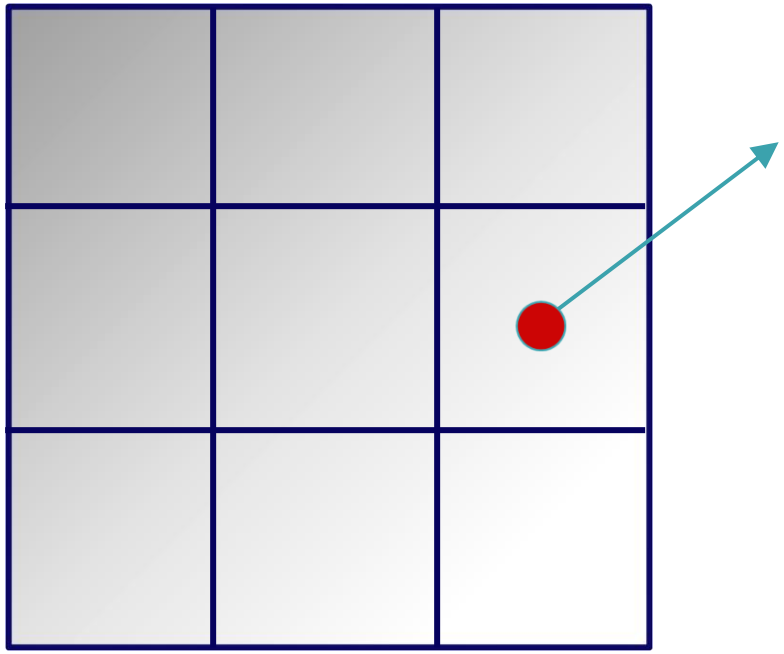
ORNL is managed by UT-Battelle, LLC for the US Department of Energy

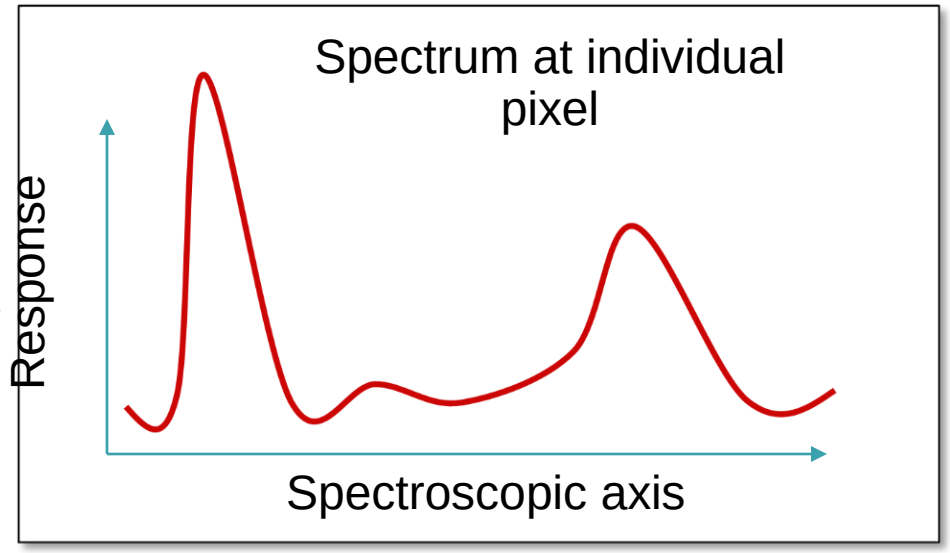
U.S. DEPARTMENT OF ENERGY

Spectral Data

- Many imaging modalities have spectroscopy components
- By capturing spectra across an image, it allows us to correlate features of the spectra with specific regions of the sample

Image (MxN pixels)



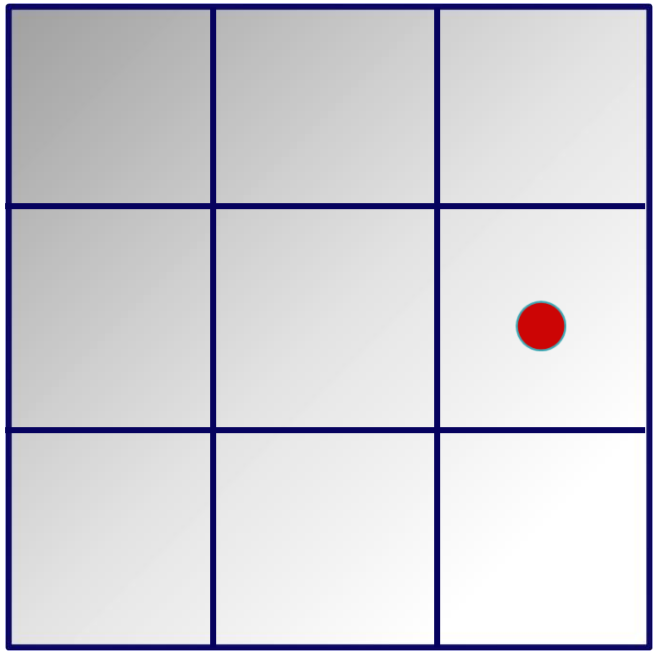


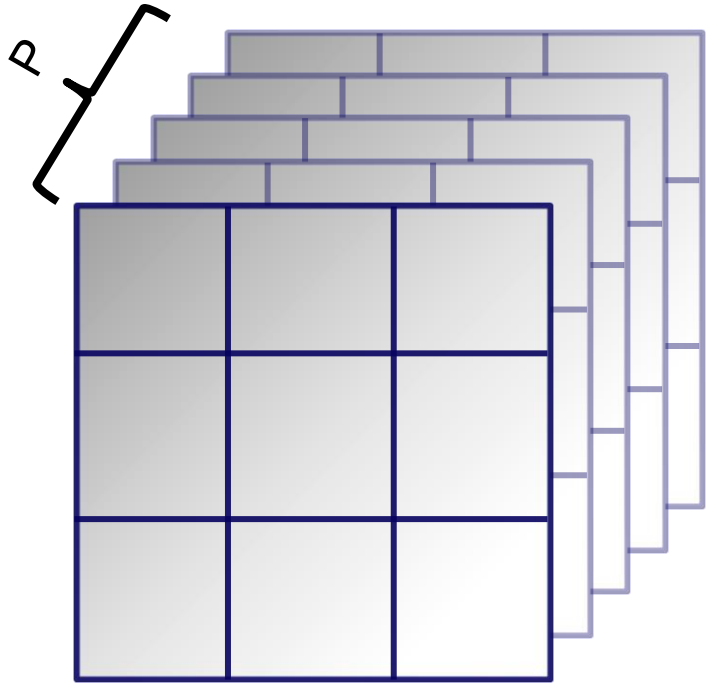
M (rows) x N (columns) x P (spectral points)

Spectral Data

- Many imaging modalities have spectroscopy components
- By capturing spectra across an image, it allows us to correlate features of the spectra with specific regions of the sample

Image (MxN pixels)





(Repeat MxN matrix P times to make 3D cube)

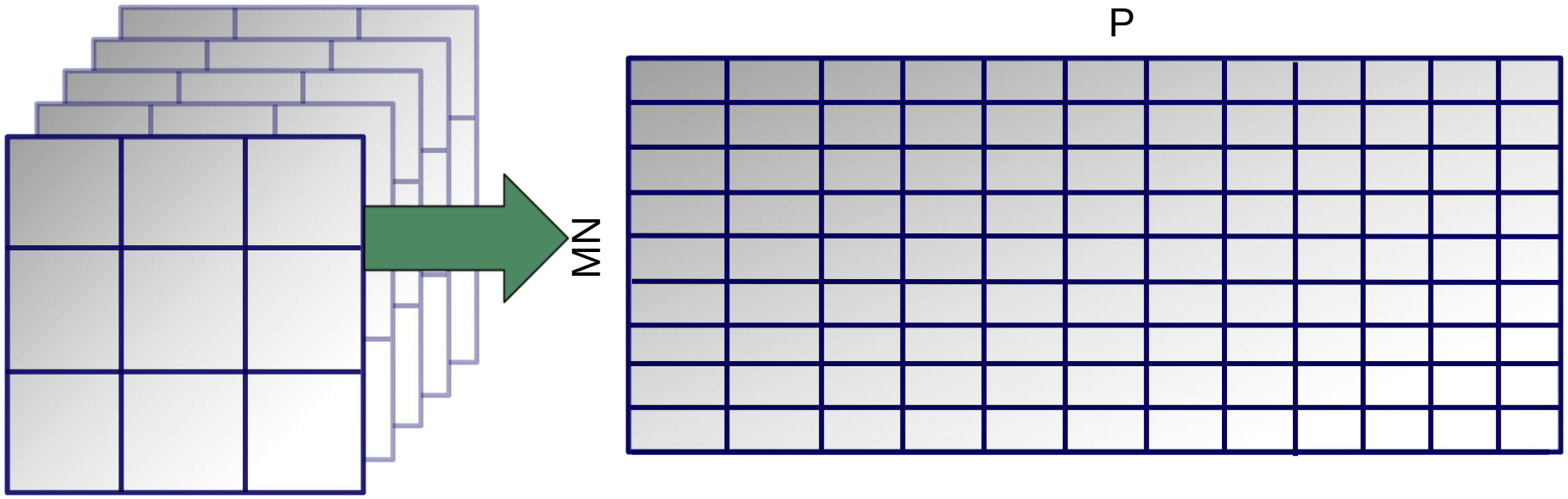
Spectral Data

- Examples include EELS, micro Raman imaging, force-distance curve maps in AFM, Scanning tunneling spectroscopy, etc.
- Usually, the 3D matrix is converted to a 2D matrix

M (rows) x N (columns) x P (spectral points)

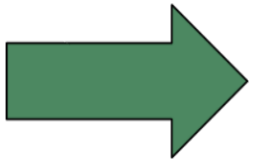
= (MN) x P

Pixels x spectral points



Matrix Factorization

- We now have a 2D matrix (call it **X**) of size (MN,P)
- If **X** is ‘small’ (e.g., (10,10)) we can comprehend it easily. But if the size of X is (2500, 32), then this becomes more difficult
- The number of spectral points **P** is often termed the “number of dimensions”.
- To see trends in our data, we want to visualize them in some manner. But the 3D data cube is big, so what can we do?



Matrix Factorization

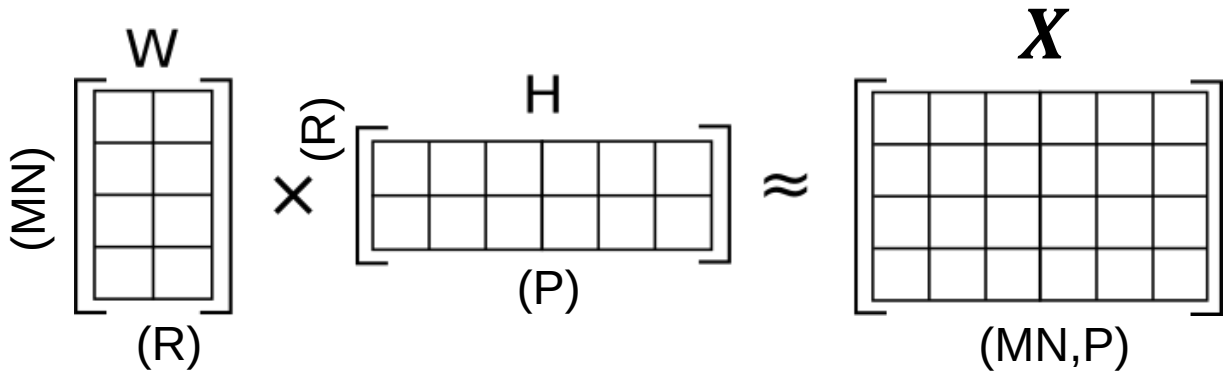
Dimensionality Reduction

- The reason it is difficult to visualize is because there are too many spectral points *P* per pixel. What we need is to reduce the dimensionality, while preserving most of the data structure.
- What if we could write our matrix **X** (size MN,P) as

$$\boldsymbol{X} \approx \boldsymbol{W}\boldsymbol{H}$$

Where W is size (MN,R)
H is size (R,P)

R <P



Dimensionality Reduction

- The reason it is difficult to visualize is because there are too many spectral points *P* per pixel. What we need is to reduce the dimensionality, while preserving most of the data structure.
- Alternative viewpoint: Represent the spectrum at pixel z by a linear expansion

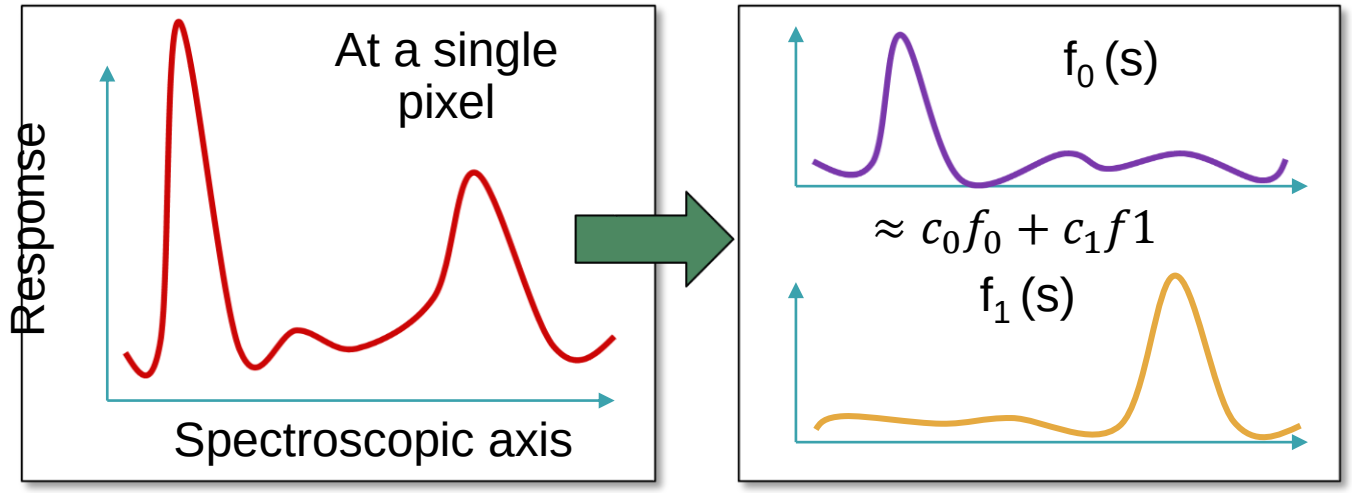
$$f(z_i, s) = c_0(z_i)f_0(s) + c_1(z_i)f_1(s) + \ \ldots + c_n(z)f_n(s)$$

$$f(z, s) = \sum_{i=1}^{MN} c_i(z_i)f_i(s)$$

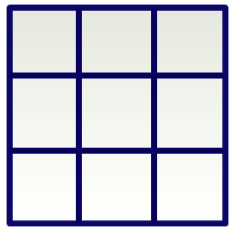
The task boils down to determining the functions $f_n(s)$, and determining the coefficients c_n

Dimensionality Reduction

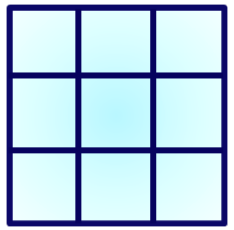
- The reason it is difficult to visualize is because there are too many spectral points P per pixel. What we need is to reduce the dimensionality, while preserving most of the data structure.
- Alternative viewpoint: Represent the spectrum at pixel z by a linear expansion



$c_0(x, y)$



$c_1(x, y)$



(1) Singular Value Decomposition

$$\boldsymbol{X = USV^T}$$

Note: In the full decomposition, r = n. Practically, r<<n

X is of size (m,n)
U is of size (m,r)
S is of size (r,r)
V is of size (n,r)

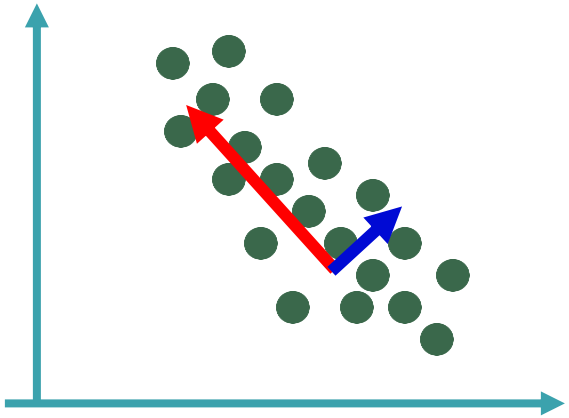
The columns of U,V form the basis
S scales them, in descending order

By the SVD theorem, it is always possible to decompose real matrix **X** into USV^T where

U,S,V: unique
U,V: column orthonormal
- $U^TU = I$, $V^TV = I$
- Columns are orthogonal unit vectors

S: diagonal
- Positive, sorted in descending order

Finding the main 'directions' in the data



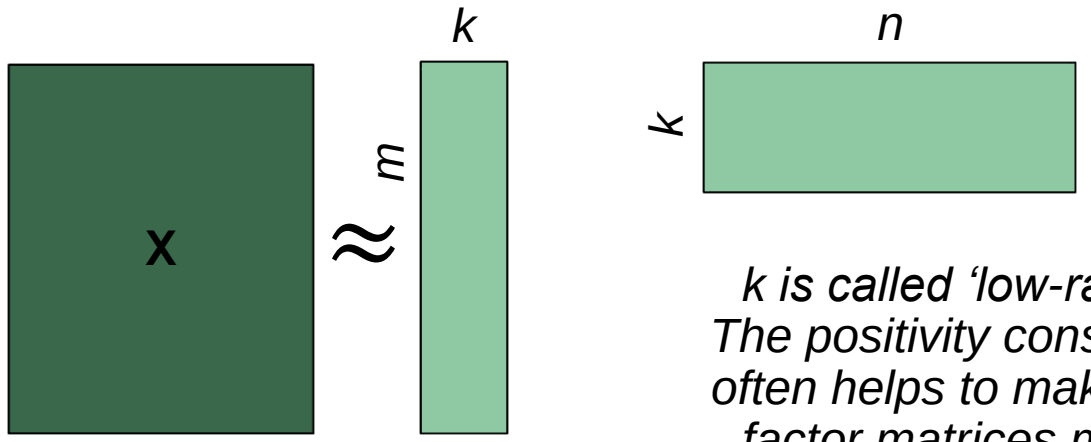
Non-negative Matrix Factorization

- SVD is an extremely powerful tool to visualize data and observe trends, but it has weaknesses. Mainly, it often produces highly unphysical behavior, for instance negative intensity values in spectra where only positive values are physically realizable
- Therefore, we can turn to other methods, such as non-negative matrix factorization approaches

Matrix Form

$$\boldsymbol{X} \approx \boldsymbol{WH}$$

Here, we enforce that X, W and H are all non-negative matrices



k is called 'low-rank'. The positivity constraint often helps to make the factor matrices more interpretable.

Spectral Unmixing: N-FINDR

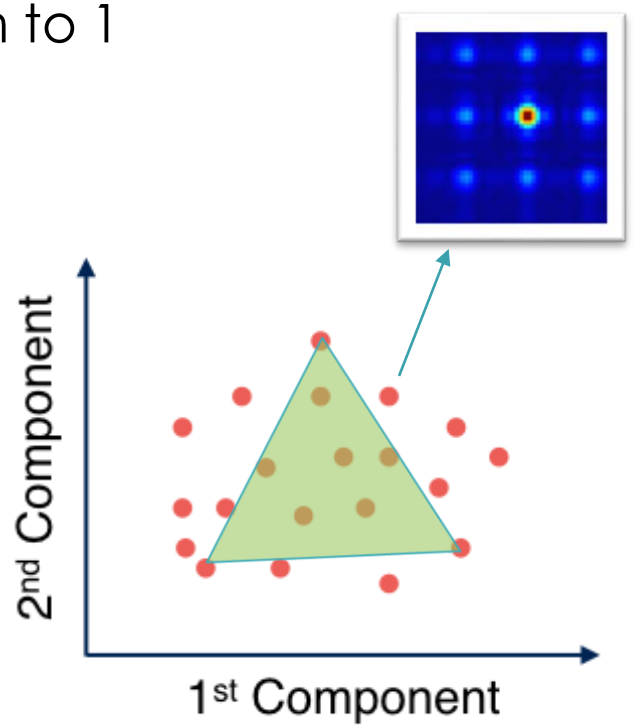
- Spectra for a given pixel is assumed to be a linear combination of the end-member spectra (+ Gaussian noise). The mixing proportions sum to 1

Physics constraint

$$p_{ij} = \sum_k e_{ik} c_{kj} + \varepsilon \qquad \sum_k c_{kj} = 1$$

- Let E be the matrix of end-members (here, 3).

$$\boldsymbol{E} = \left[\overrightarrow{e_1} \quad \overrightarrow{e_2} \quad \overrightarrow{e_3}\right] \qquad V\left(\frac{\mathbf{1}}{(\boldsymbol{l}-1)!}\right)|\det(E)|$$



- Iteratively select endmembers, accepting the new selection if the volume increases

Segmenting based on phases

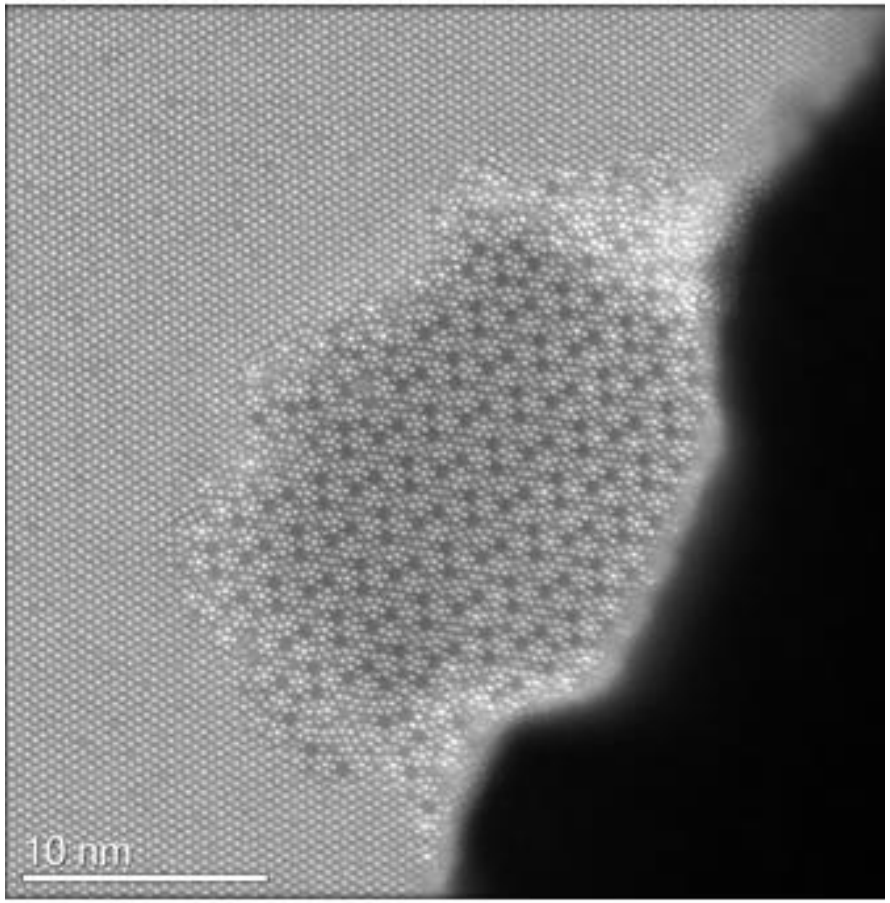
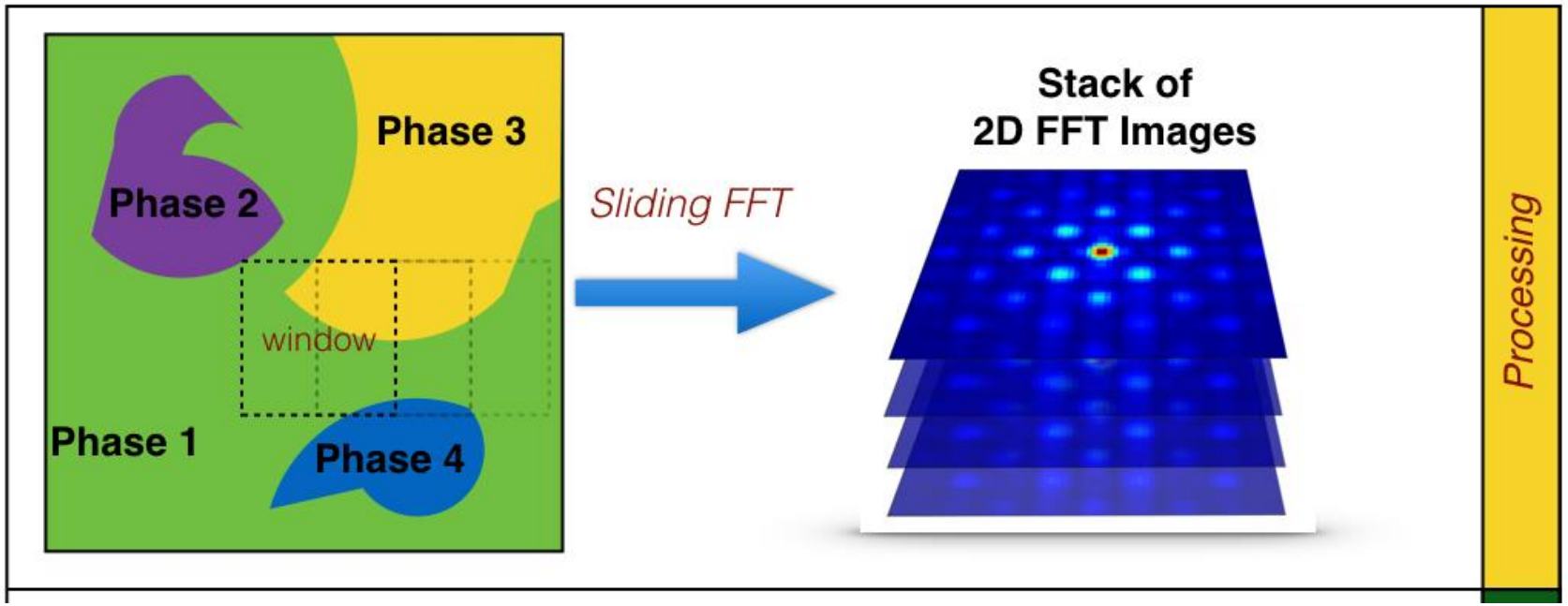


Image courtesy of
A. Borisevich (ORNL)
and
Q. He
(ORNL/Manchester)

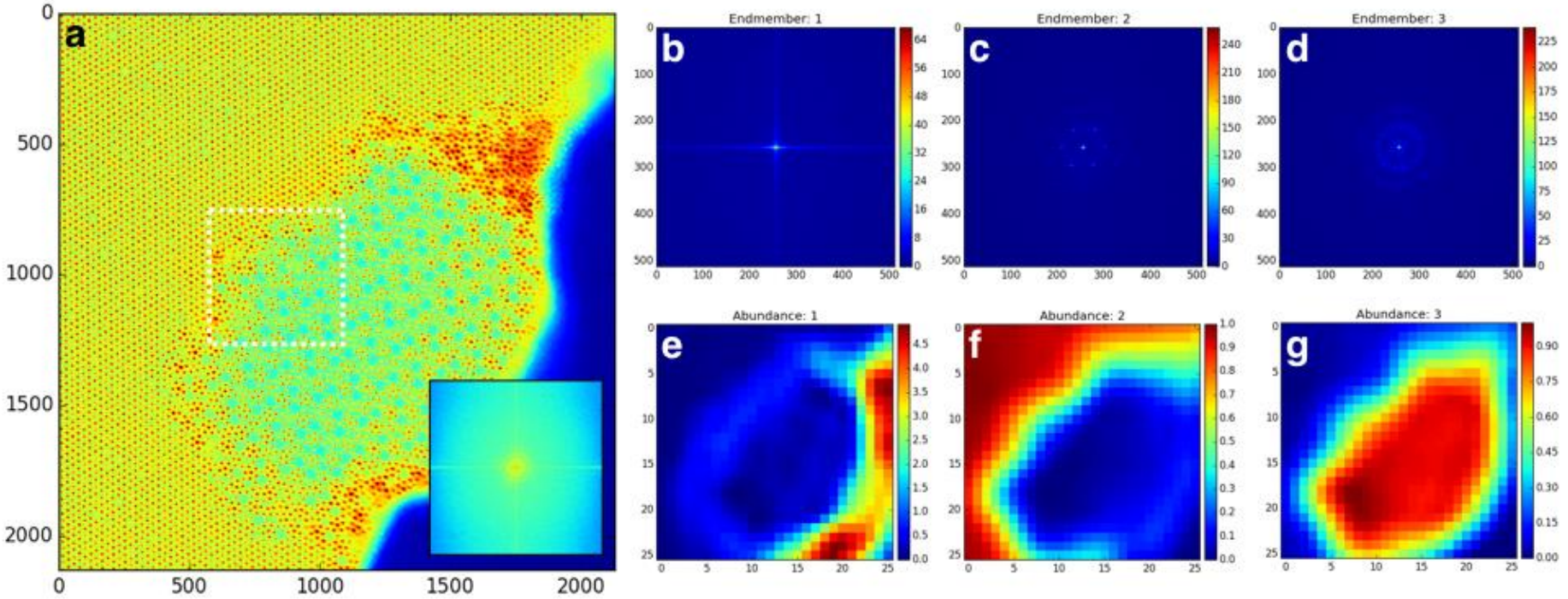
How can we segment this image based on the phases present?

Sliding Window Method



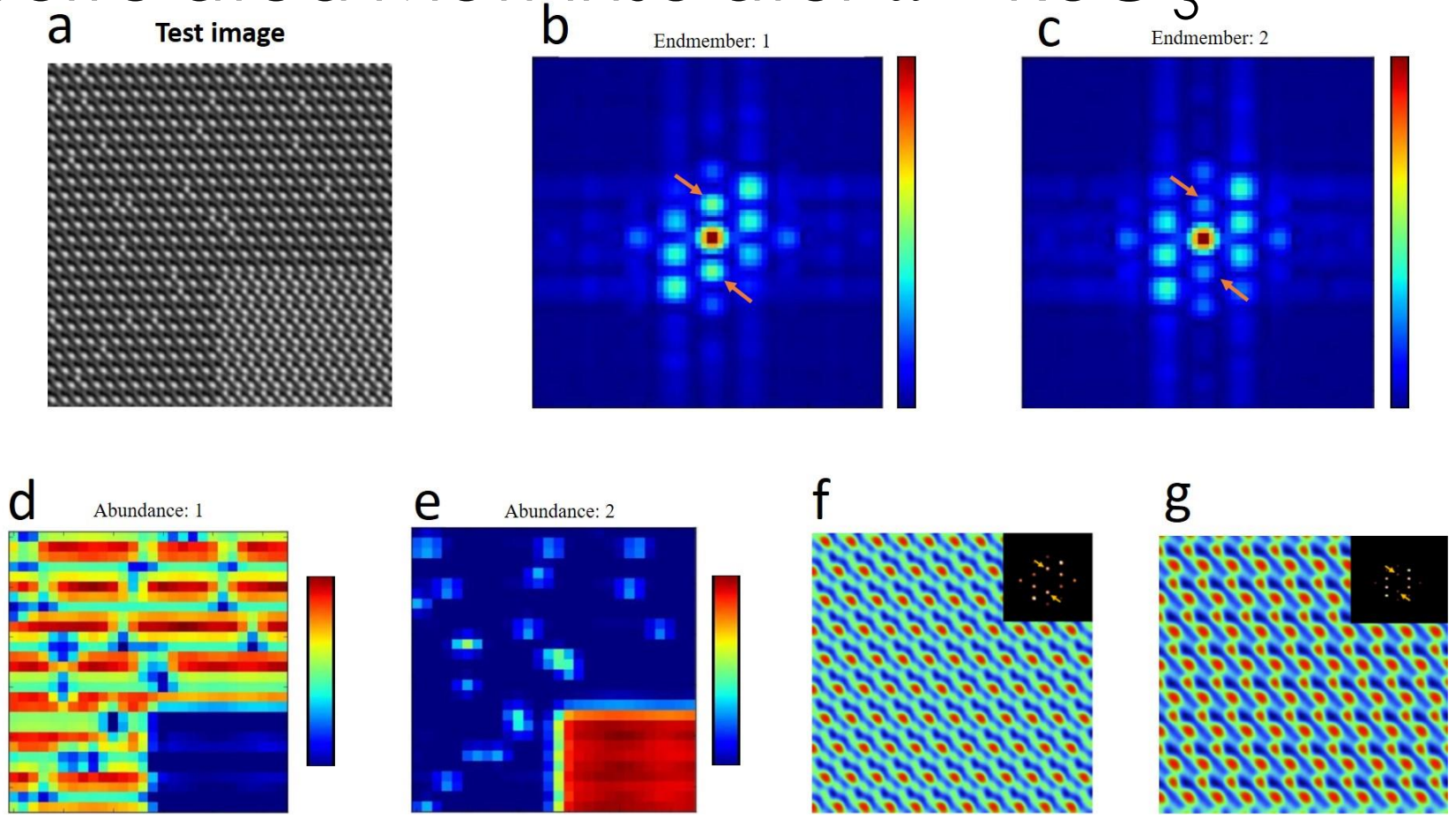
Then, we can try to do spectral unmixing on the 2D stack of FFT images

Sliding FFT on an oxide catalyst



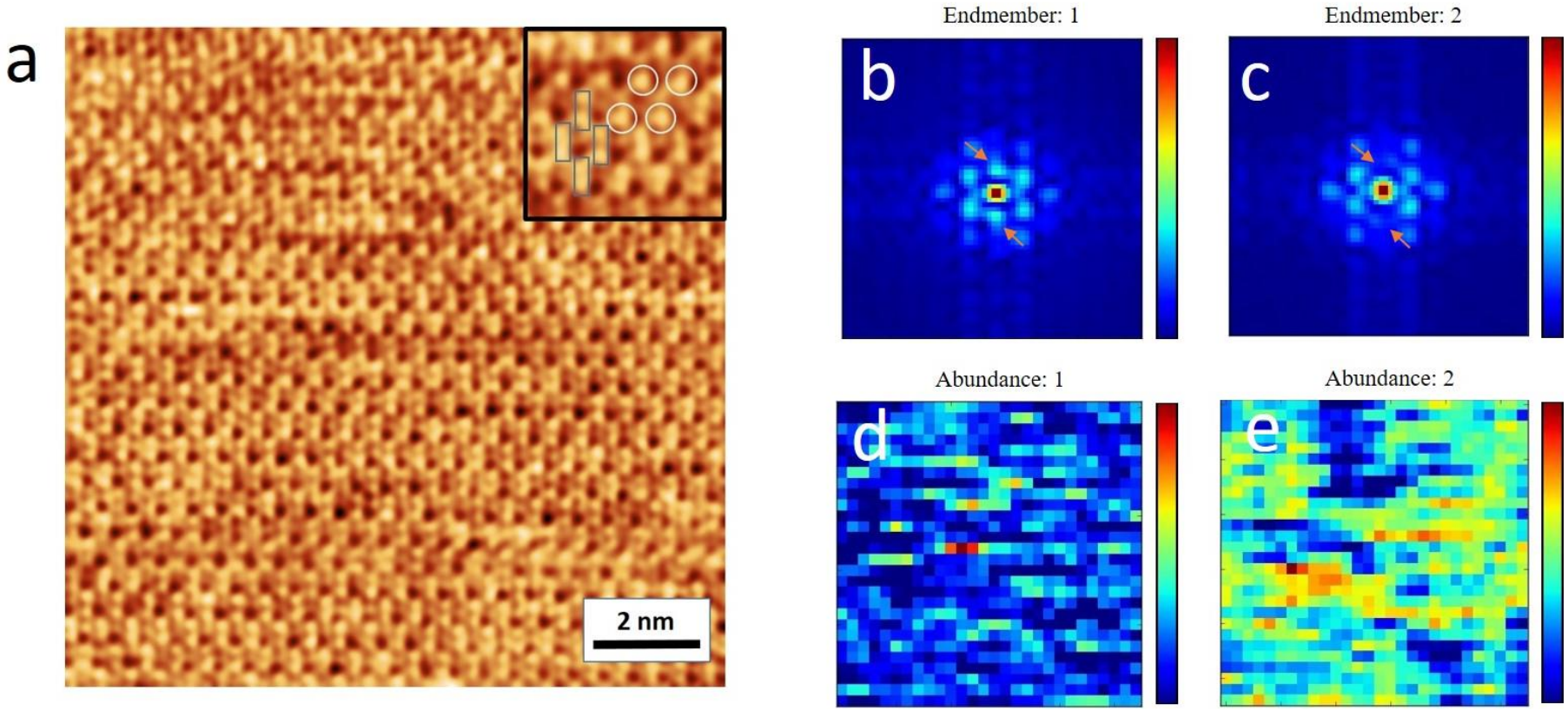
- Combination of N-FINDR and Sliding FFT can quickly and automatically determine the two phases present and their spatial abundance.

Fine structures in Electronic Superlattice in a correlated Mott insulator α –$RuCl_3$



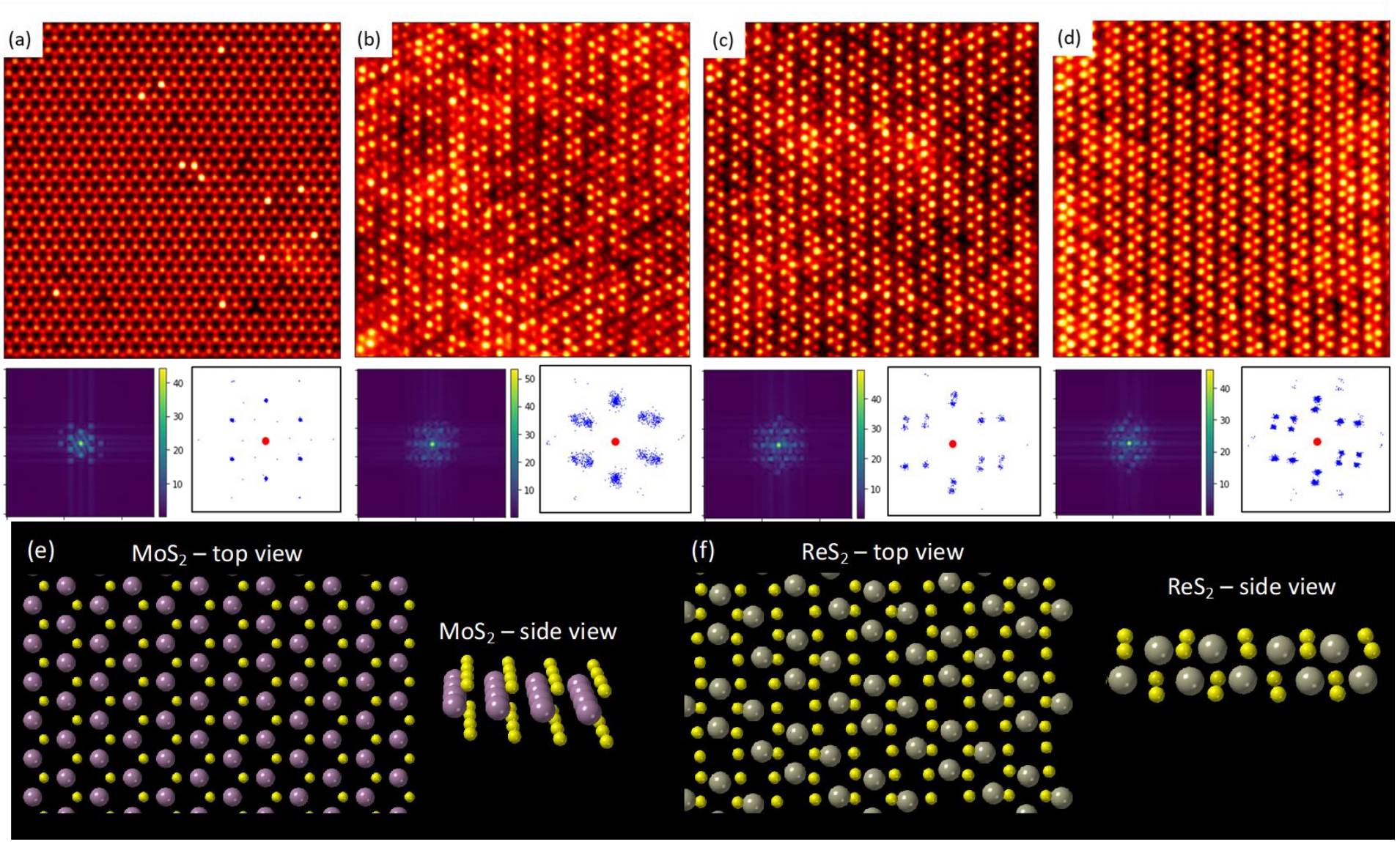
- What would dimers overlayed on hexagonal lattice look like?
- Strong suppression of vertical spots in FFT for dimers
- Scale bar: 0 to 1

Fine structures in Electronic Superlattice in a correlated Mott insulator



- Clear evidence of dimerization in $RuCl_3$ through Sliding FFT/N-FINDR

Another example: MoS_2-ReS_2

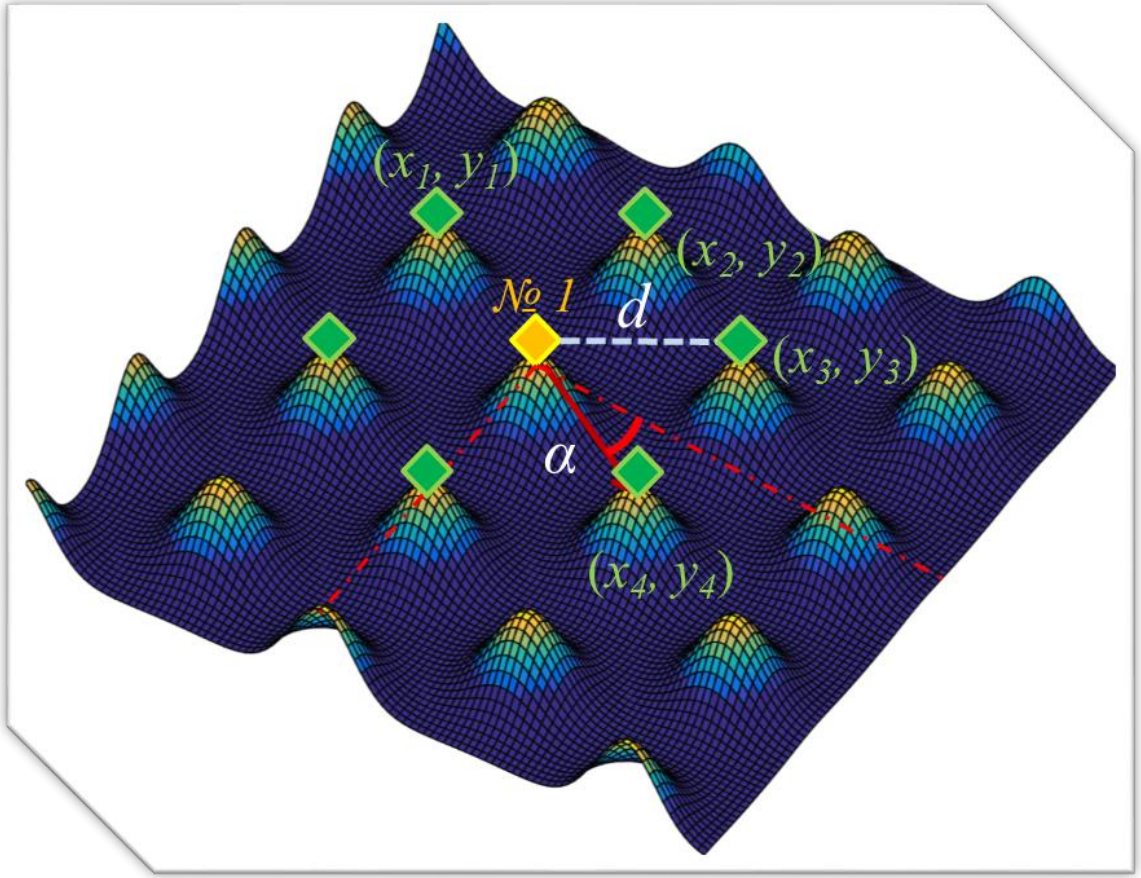


Phase transition between MoS_2 ("H") and ReS_2 ("T").

Note that there must exist broken bonds/defects

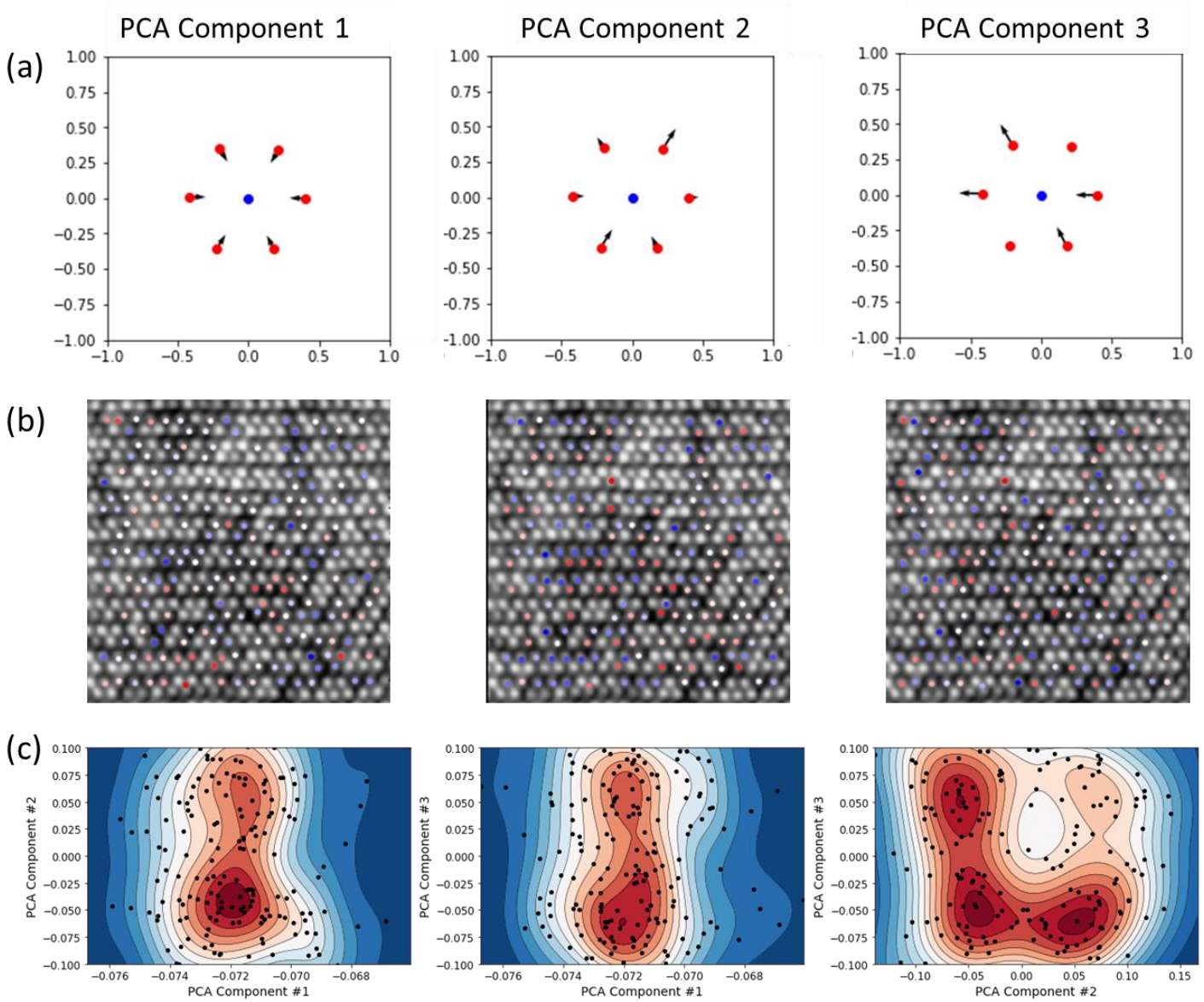
Question: Does transformation occur from global composition, or is it locally driven?

Atomic positions: Obtaining statistics



- Find atoms using some algorithms
- Determine the distance from each individual atom to the *J* nearest neighbors, and compute the angles
- Iterate over all *N* atoms in image to build the matrix of size *N* x *J* x 2
- Compute PCA over *N* x *J* (angle or distance metric)

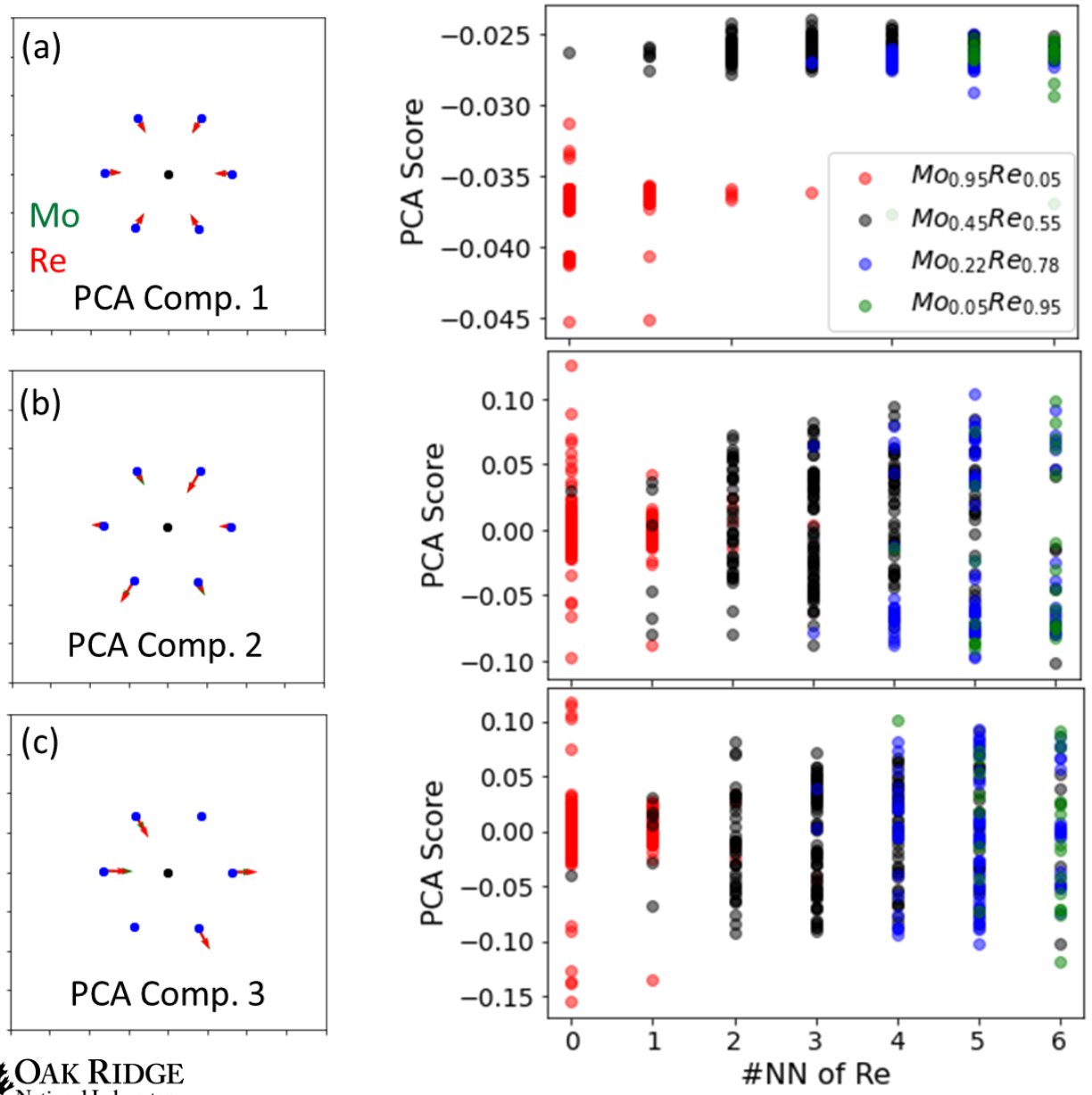
Principal Component Analysis



- PCA of local atomic neighborhoods shows progression of distortions across the phase transition

Vasudevan et al. Appl. Phys. Rev. **8**, 011409 (2021).

Principal Component Analysis



- The second component appears to show a splitting as the number of Re neighbors increases, irrespective of composition

Vasudevan et al. Appl. Phys. Rev. **8**, 011409 (2021).

Colab Notebook

- In the notebook we will be exploring spectral unmixing methods
- If we have time, then we can also take a look at the unsupervised learning of atomic scale distortions in the Jupyter paper by Maxim Ziatdinov

https://colab.research.google.com/github/jupyter-papers/mock-paper/blob/master/FerroicBlocks_mockup_paper_v3a.ipynb