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# Linear Unmixing Methods for Image Analysis

Sergei V. Kalinin

Inputs from Mani Valleti and Maxim Ziatdinov are gratefully appreciated

# Linear Unmixing methods

1. **Why analyzing images and image stacks?**

2. **Workflows and pipelines**

3. **Building the descriptors**
   - Uniform sampling grid
   - Feature-centered sampling
   - Time-delayed sampling

4. **Principal component analysis (PCA) pipelines**
   - Ferroelectrics on the atomic scale
   - Analyzing mesoscopic images

5. **K-means clustering pipelines**
   - Analyzing ferroelectric domains
   - Analyzing mesoscopic images

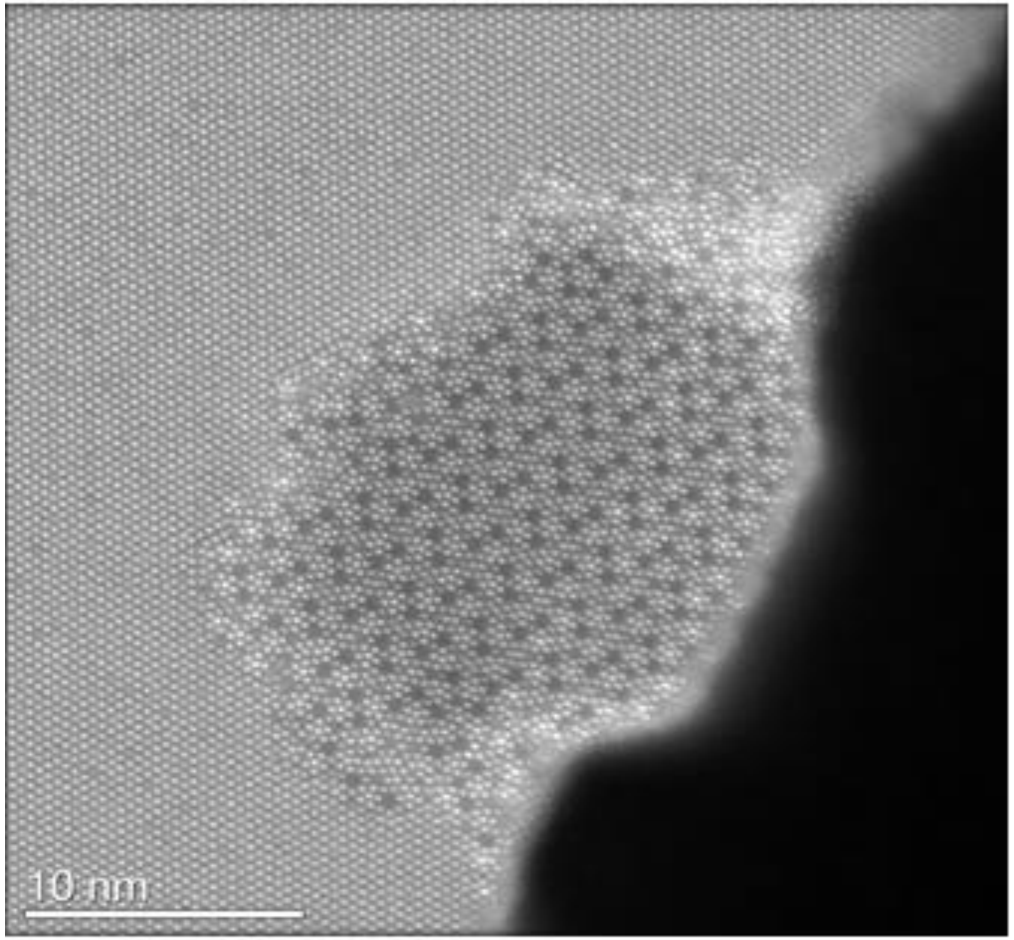
6. **BLU and ICA**

7. **Analyzing video data**

8. **4D STEM**

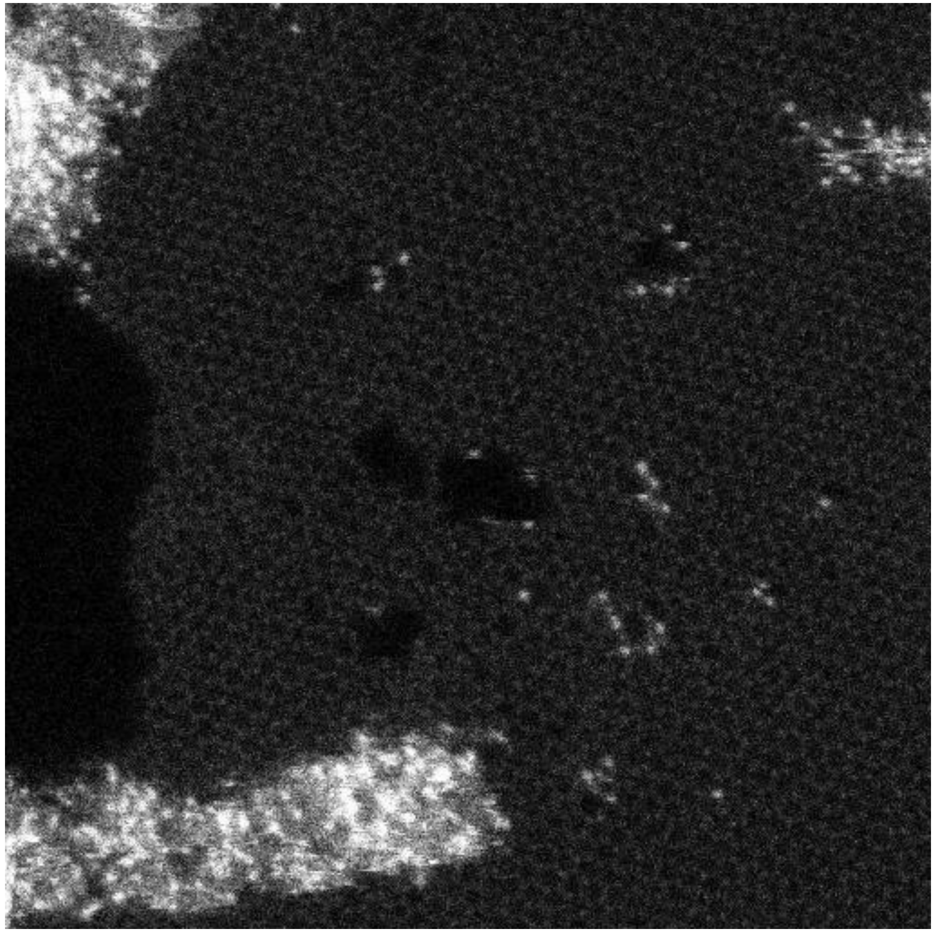
# Chemically disordered systems

**Mo-V-Ta complex oxide**




Q. He et al, ACS Nano 9, 3470-3478

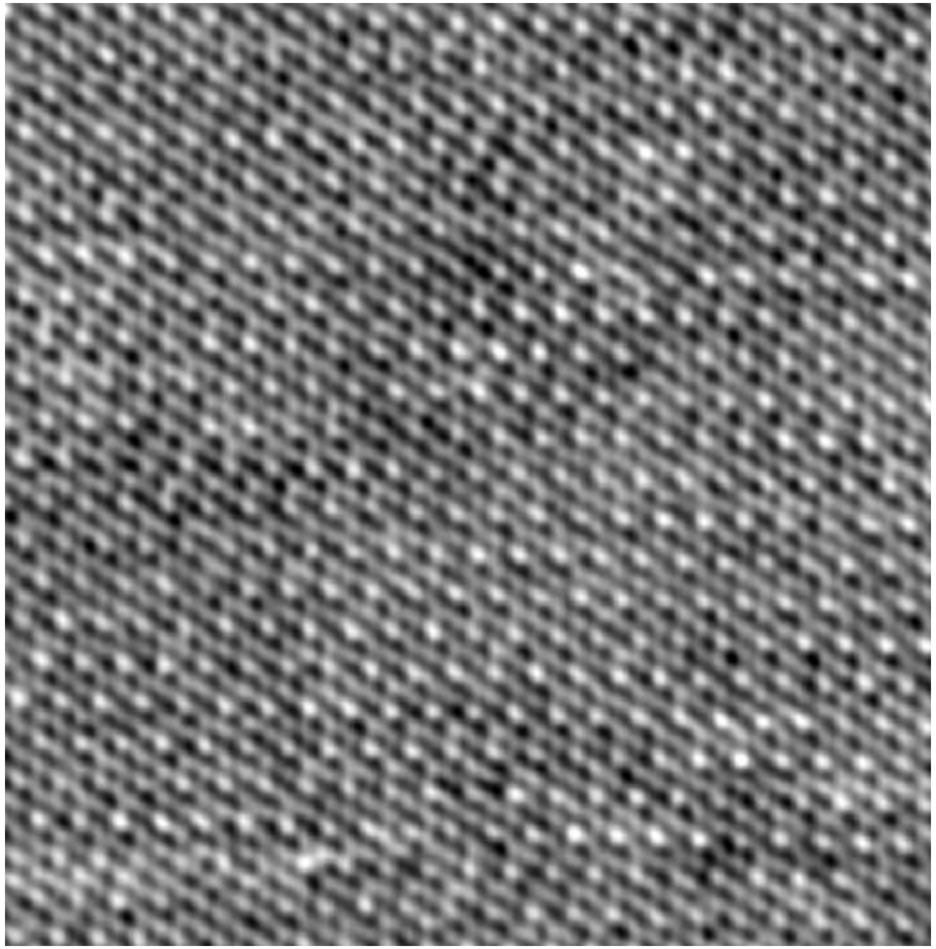
**Si in graphene**



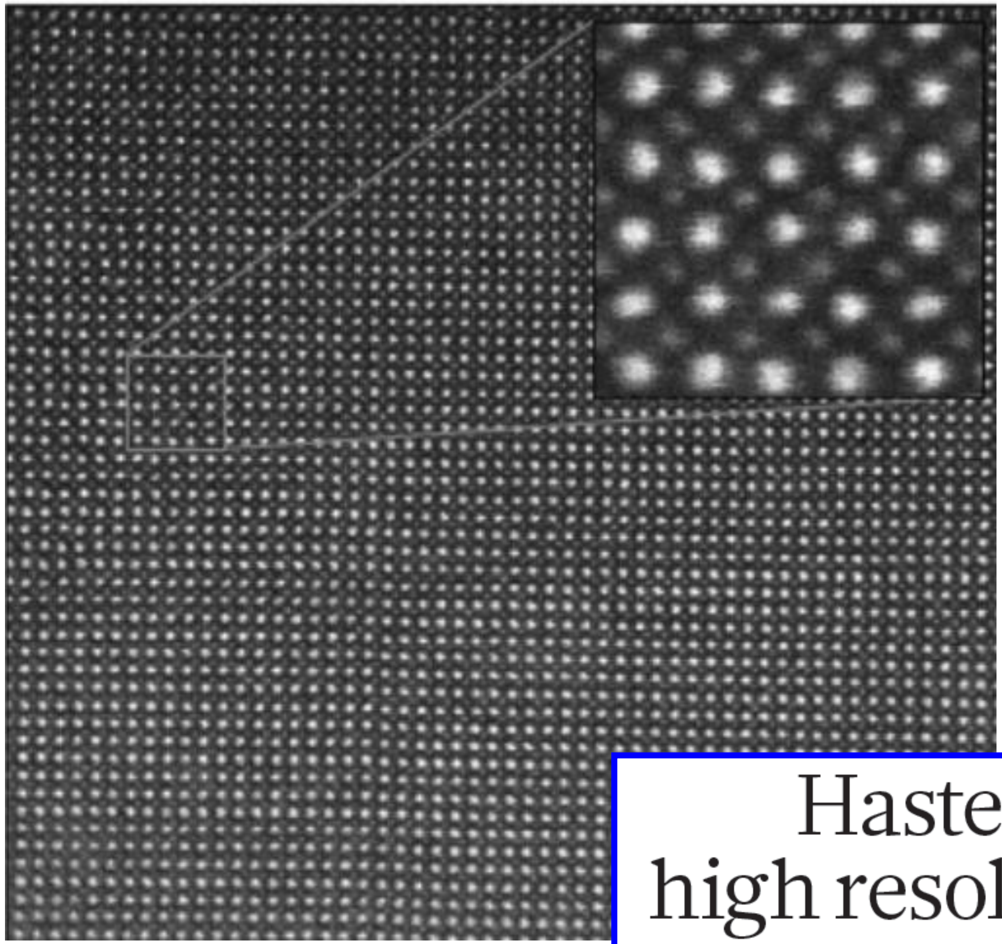
Data collected by O. Dyck (ORNL)

- What is the nature of the building blocks and relevant atomic configurations?
- Can we define single-phase regions and phase boundaries?

**Electronic structure in $RuCl_3$**



**$BiFeO_3$ on $SrRuO_3$**



Hasten high resolution

Build precision microscopes to map atoms, say **Stephen J. Pennycook** and **Sergei V. Kalinin**.

Nature 515, 487 (2014)

- Can we identify ferroelectric and ferroic variants and associated topological defects?
- What is the nature of the phases?

# Can global FFT help?

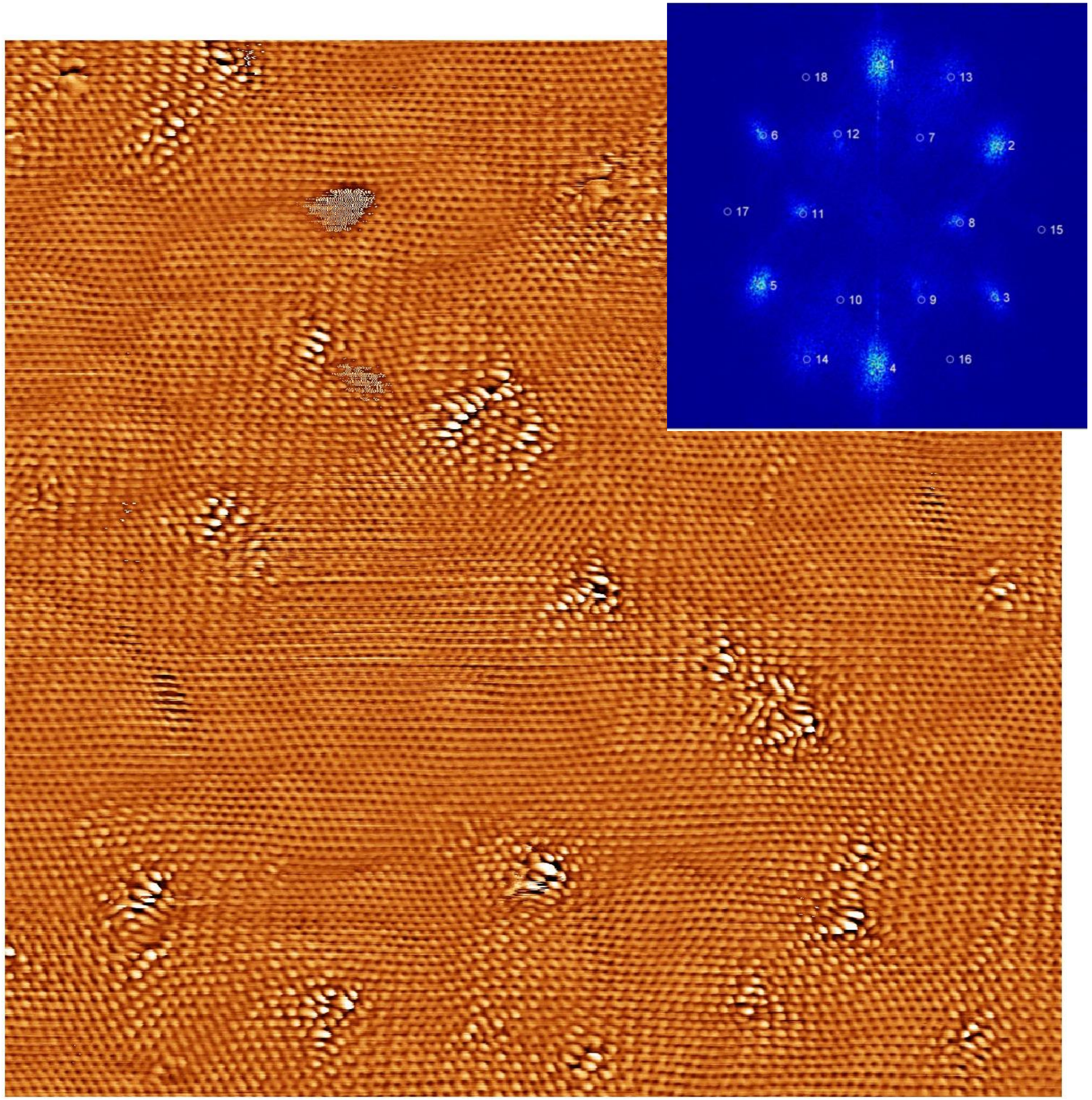


- Global FFT – everything is averaged:
  - Drift
  - Extended defects
  - Multiple grains

We are averaging out all interesting phenomena except for small spatially uniform structural distortions

- Solution – sliding window approaches:
  - Fit FFT peaks: amplitudes, positions
  - Multivariate analysis

Note – window can be also tied to a specific feature, such a selected atom. Then we explore atomic neighborhood



# General linear unmixing

$$S(\mathbf{x}, \mathbf{R}) = \sum_i a_i(\mathbf{x})\, w_i(\mathbf{R}) + \mathrm{N}$$

**We start with:**

- **x** is the spatial variable, x = (*x*,*y*)
- R is the (vector) parameter variable

**We aim to get:**

- $a_i(\mathbf{x})$ are loading maps
- $w_i(\mathbf{R})$ are endmembers/eigenvectors
- N is noise

The M pixel 2D image is transformed to M/N pixel image of more complex structure.

Our loading map is 2D image, and endmembers/eigenvectors are 2D images

**Image analytics**

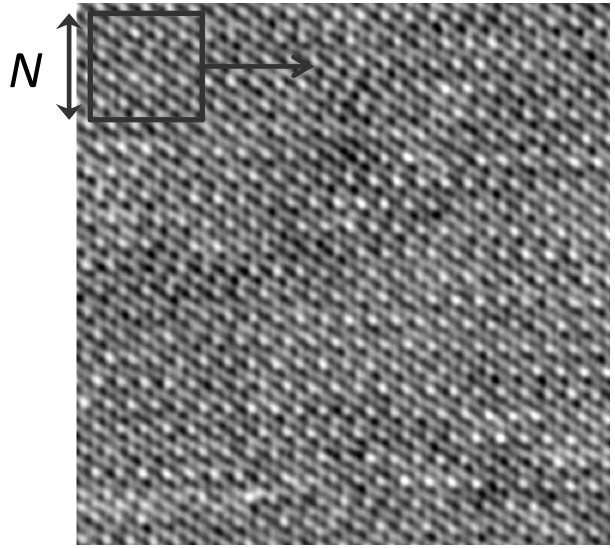
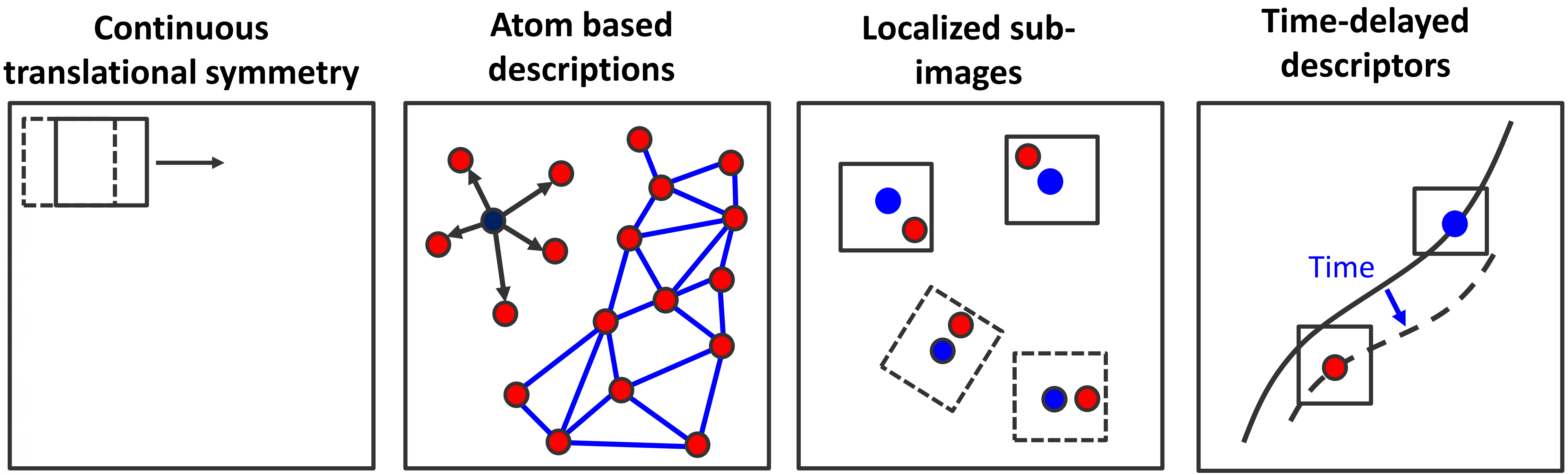



Figure by M. Ziatdinov

**Sliding image transforms:**

- Fast Fourier Transforms
- Correlation functions
- Intensity histograms
- Structural descriptors

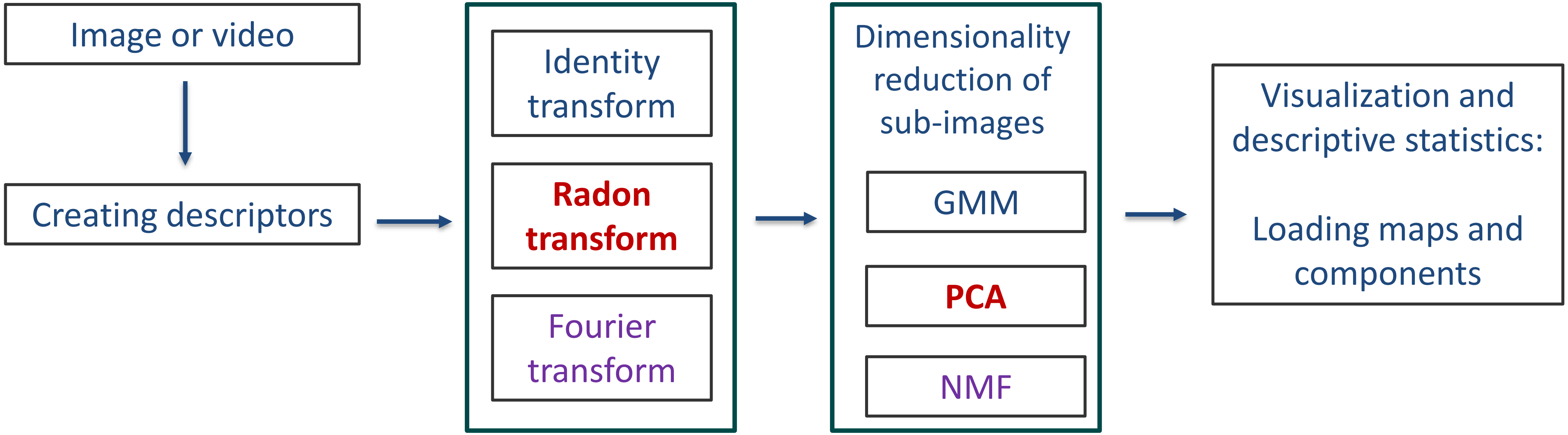
# Constructing the descriptors




The choice of the descriptor:

- Defines physical inferential biases and allows to introduce prior knowledge
- Determines the physical meaning of the analysis
- Establishes the analysis pipeline

# Example of analysis pipeline




**Pipelines are defined to**

- Make analysis traceable, repeatable, explainable, and transferable
- Allow for hyperparameter tuning and optimization
- Efficiently use the memory

# Principal Component Analysis

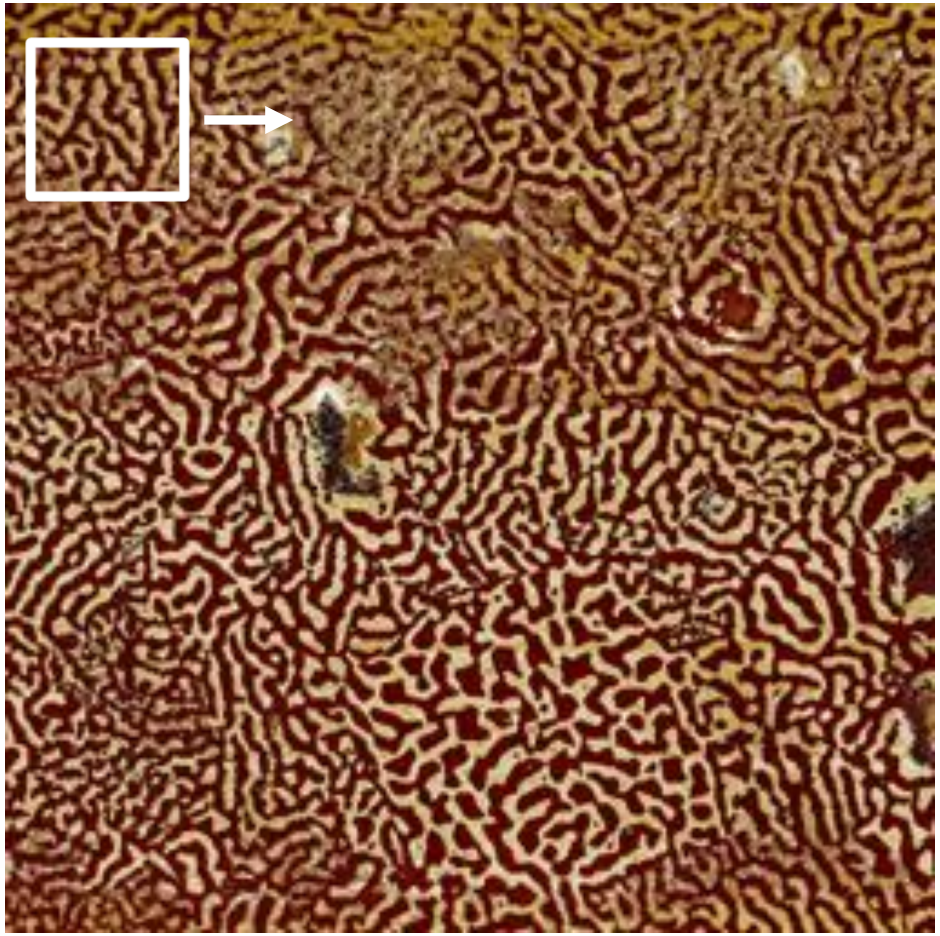
$$S(\mathbf{x}, \mathbf{R}) = \sum_i a_i(\mathbf{x})\, w_i(\mathbf{R})$$

In PCA, the eigenvectors $w_i(\mathbf{R})$ are orthonormal and are arranged such that corresponding eigenvalues are placed in descending order by variance

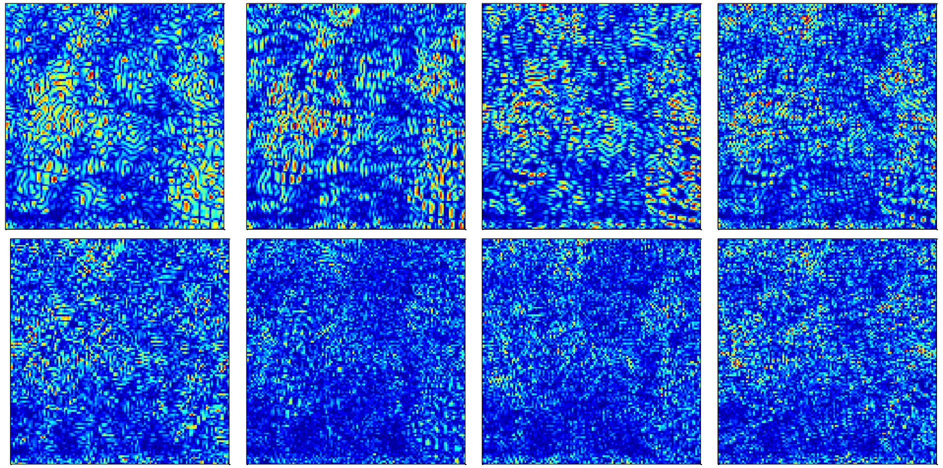
- Reveals internal structure of the data that best explains variance in the data set
- Since data often moves in clusters, PCA reveals those variables that drive the variance
- PCA transforms the data such that the greatest variance by any projection lies on the first coordinate

# Sliding PCA-FFT

Can we use PCA of FFT transform in sliding windows to find periodicity?



First 8 maps



First 16 eigenvectors

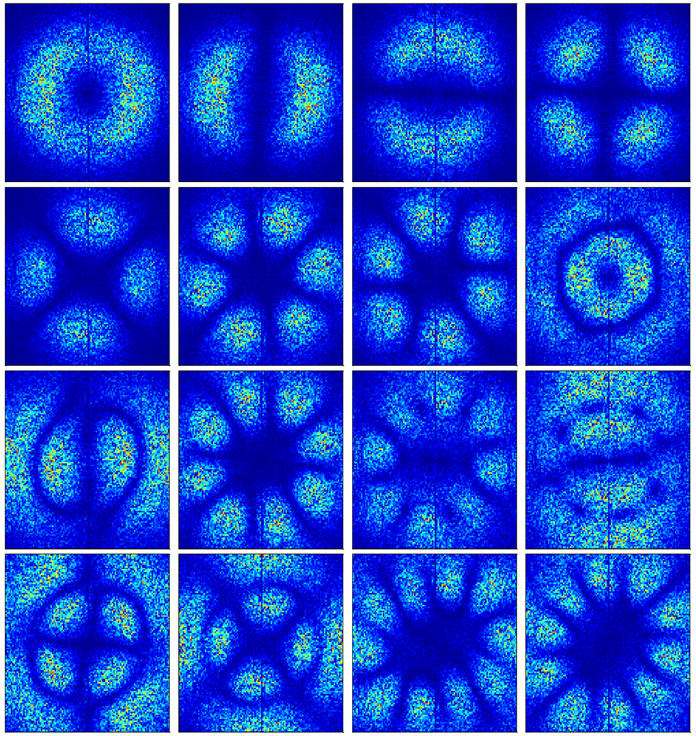


Figure by S. Jesse, data D. Gobelic

# 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).

$$\boldsymbol{E} = \begin{bmatrix} 1 & 1 & 1 \\ \vec{e}_1 & \vec{e}_2 & \vec{e}_3 \end{bmatrix} \qquad V\left(\frac{\mathbf{1}}{(\boldsymbol{l}-1)!}\right)|\det(E)|$$

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

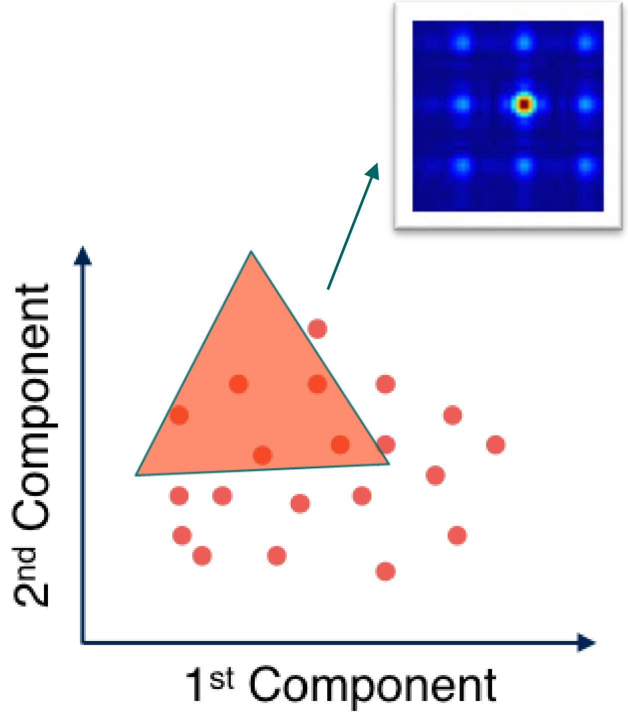



Figure by R. Vasudevan


R. Vasudevan, M. Ziatdinov, S. Jesse, and S.V. Kalinin, *Phases and interfaces from real space atomically resolved data: physics based deep data image analysis*, Nano Lett. **16**, 5574 (2016).

R.K. Vasudevan, A. Belianinov, A.G. Gianfrancesco, A.P. Baddorf, A. Tselev, S.V. Kalinin, and S. Jesse, *Big data in reciprocal space: Sliding fast Fourier transforms for determining periodicity*, Appl. Phys. Lett. **106**, 091601 (2015).

# Ideal test case

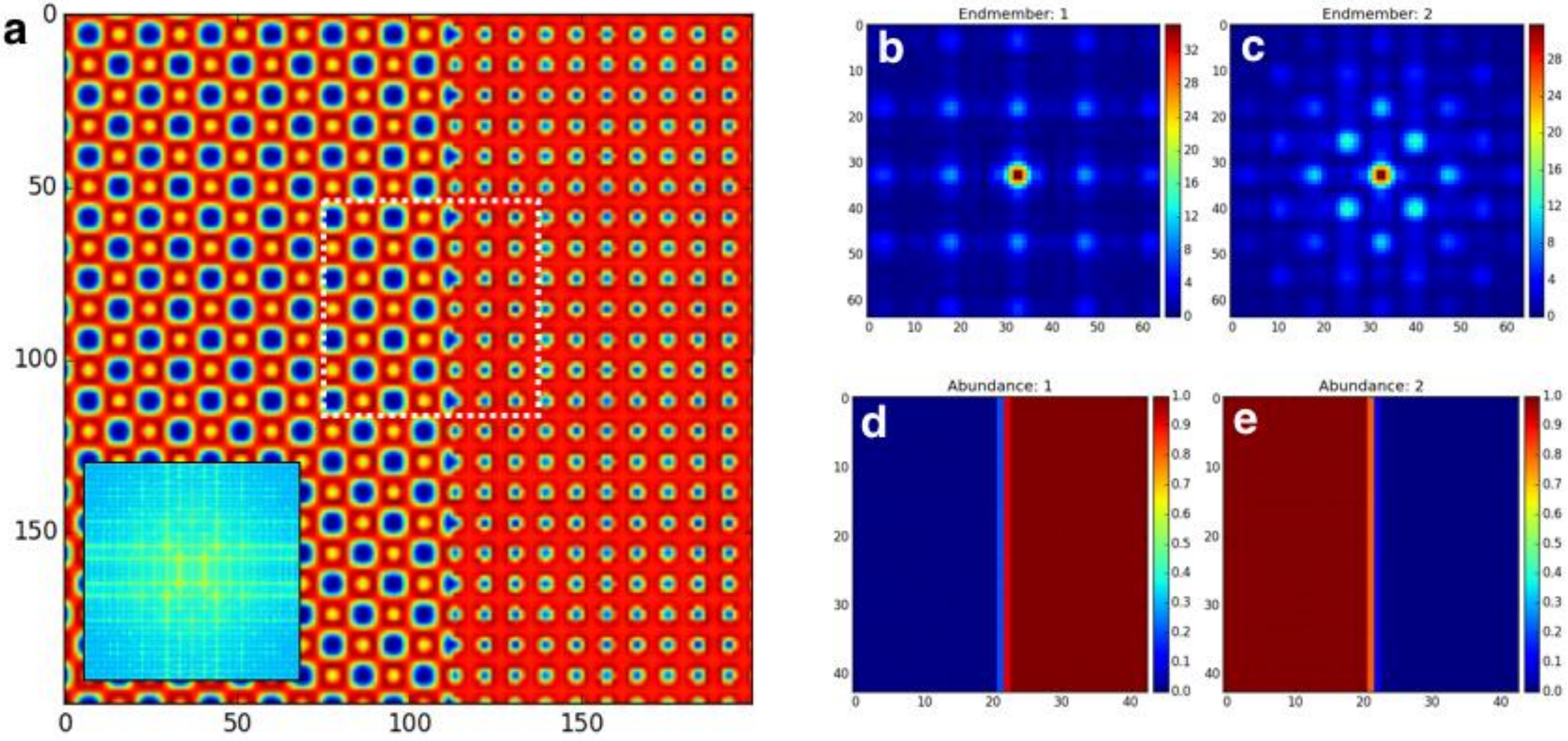
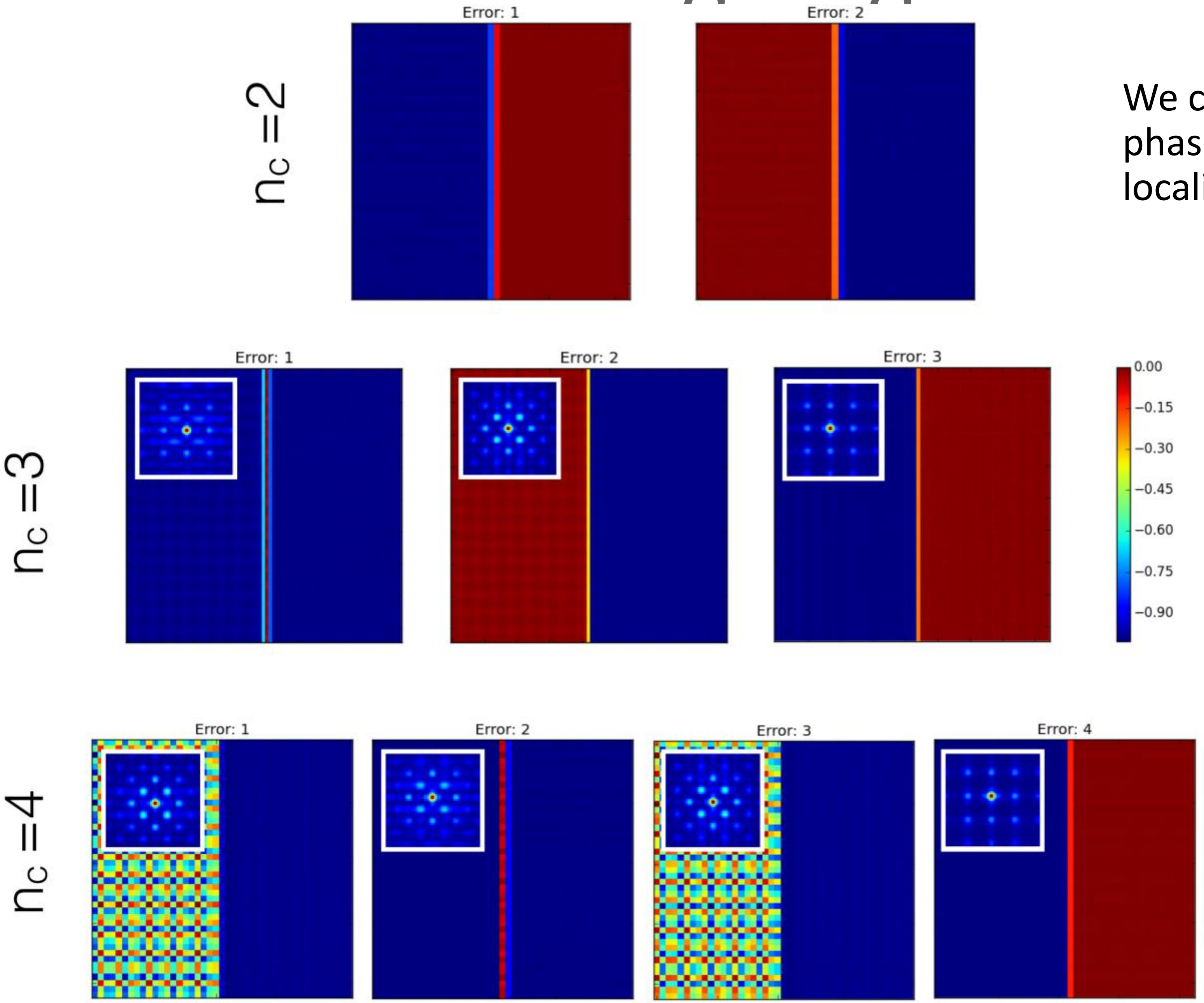



Figure by R. Vasudevan

**Main idea:**

- FFT amplitudes are non-negative;
- FFT removes translation

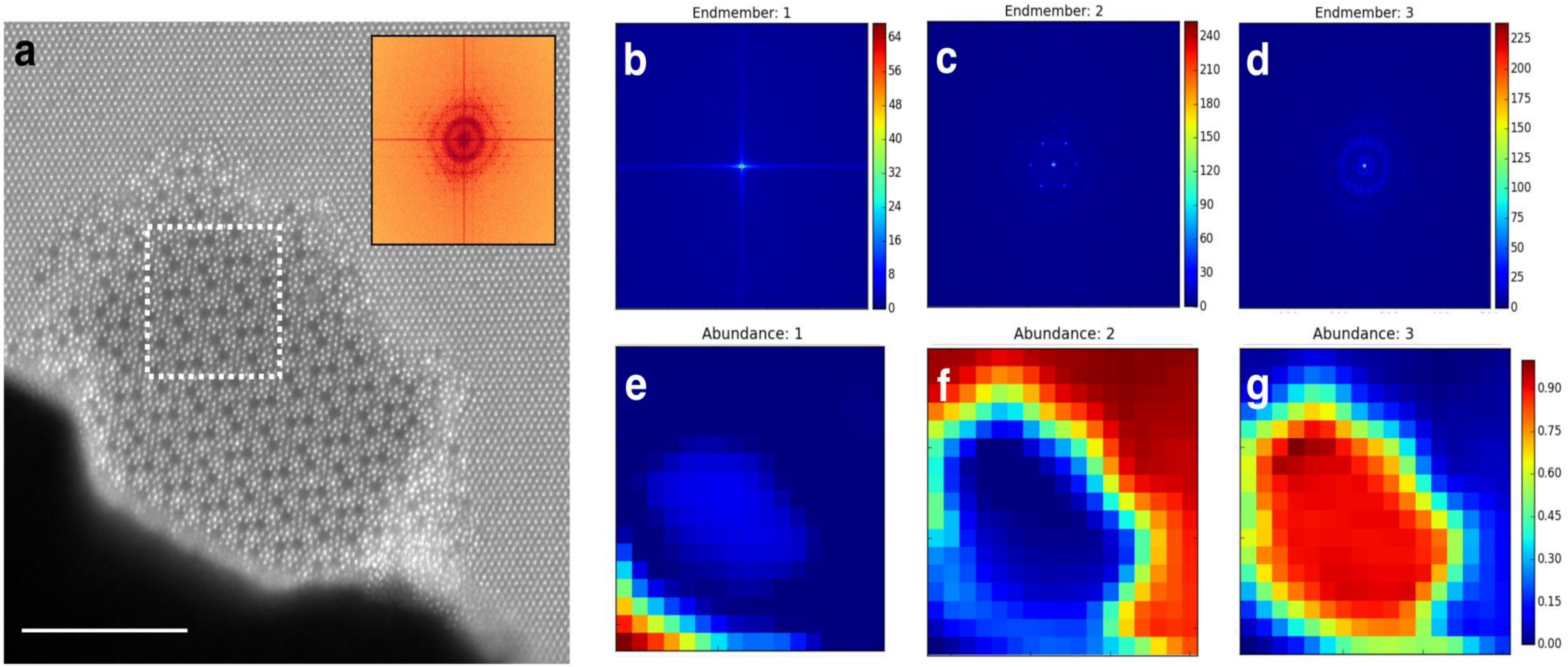
# N-FINDR for image segmentation




We can determine number of phases based on spatial localization and geometry

Figure by R. Vasudevan

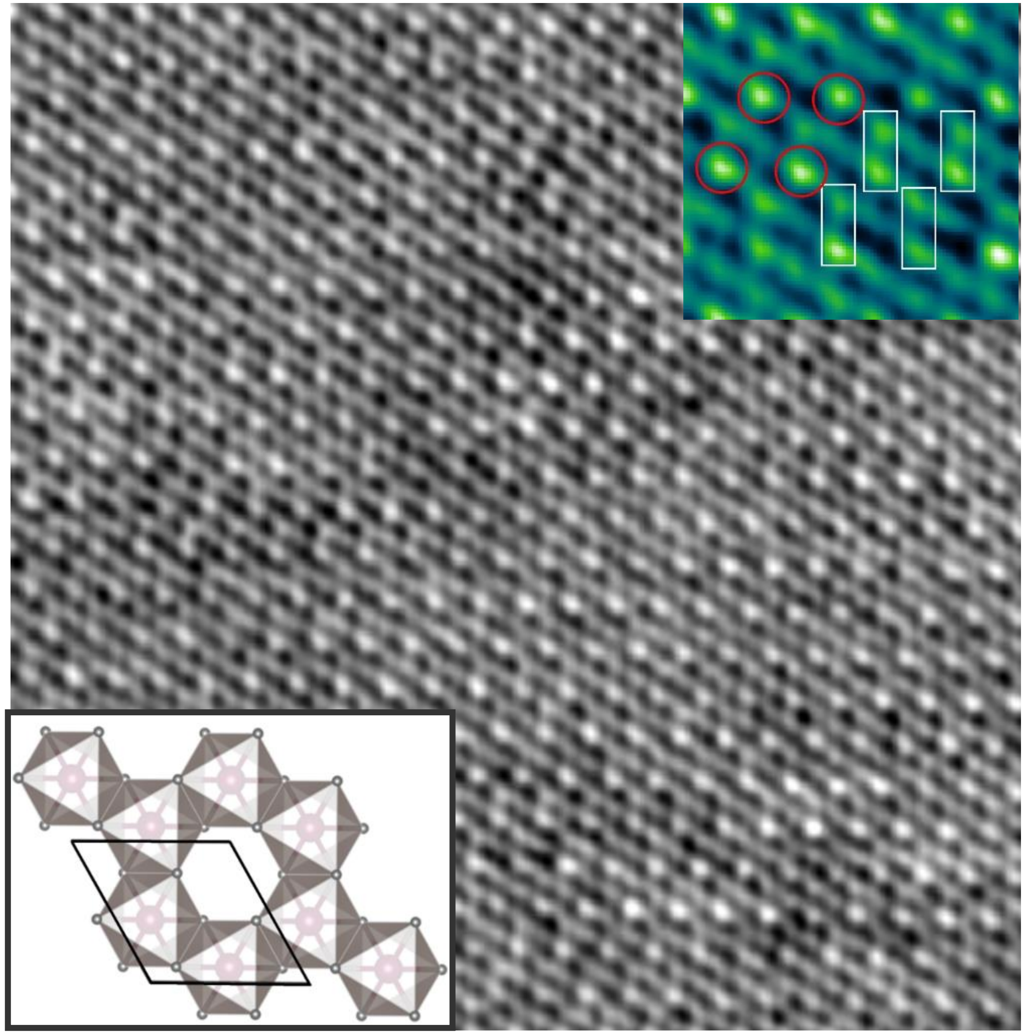
# N-FINDR for chemically separated images



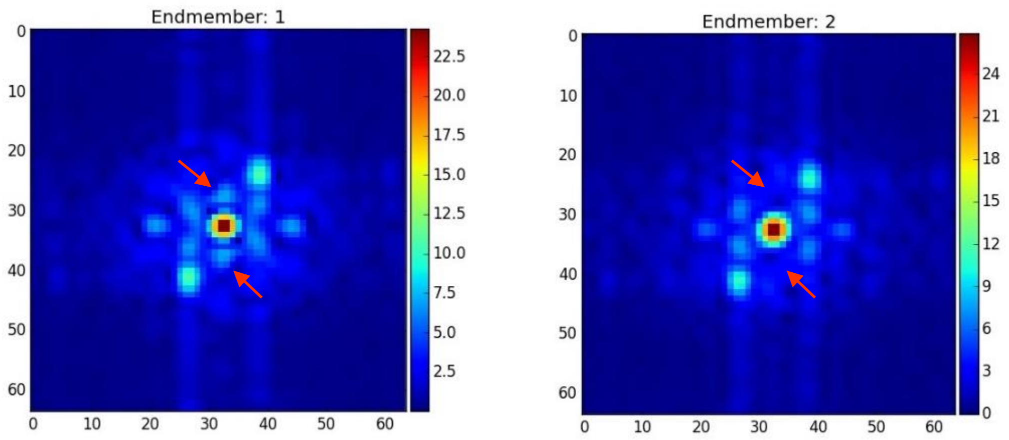

Q. He, J. Woo, A. Belianinov, V.V. Guliants, A.Y. Borisevich, Better catalysts through microscopy: mesoscale M1/M2 intergrowth in Molybdenum–Vanadium based complex oxide catalysts for propane ammoxidation
ACS Nano 9, 3470-3478

# NFINDR for coexisting order parameters

**Input experimental image**



**FFT endmembers**




**Real-space abundance maps**

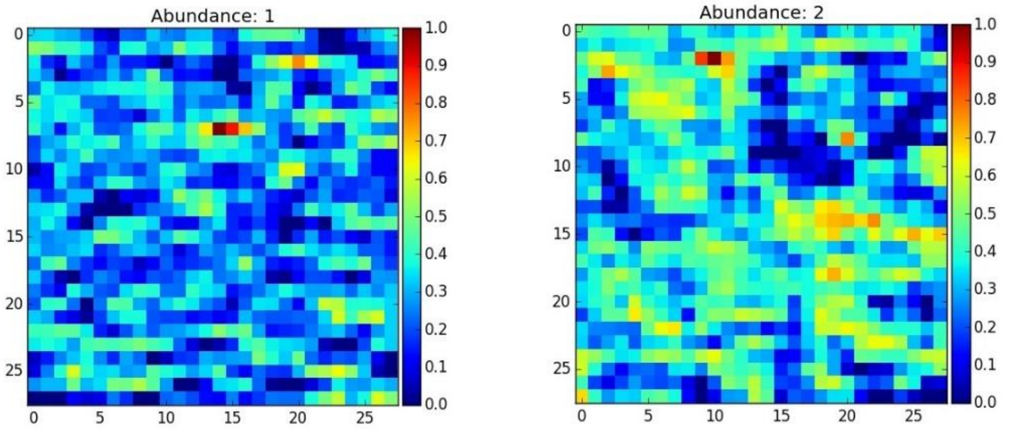
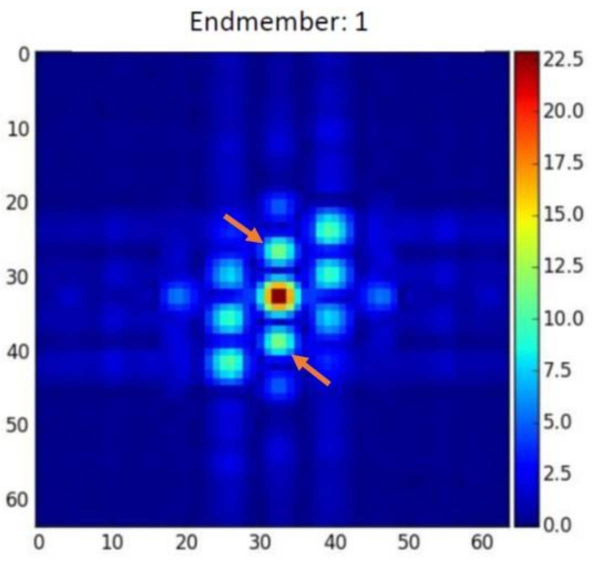


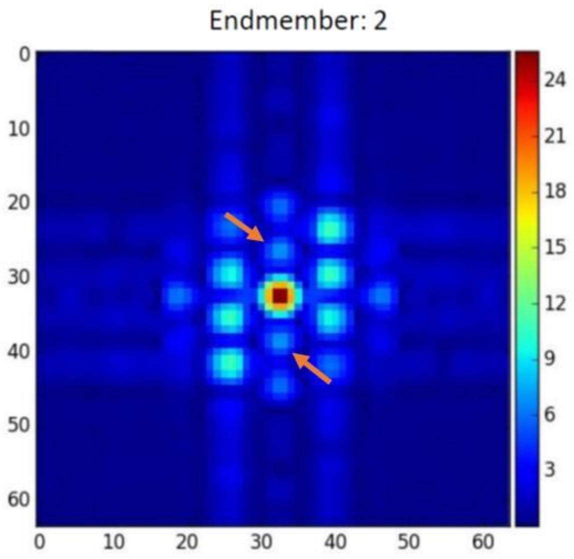

Figure by M. Zlatdinov

In a good agreement with test case, 2 spots in the “inner hexagon” are strongly suppressed in the 2nd component reflecting a fine structure of charge ordered pattern

# NFIND-R for coexisting order parameters

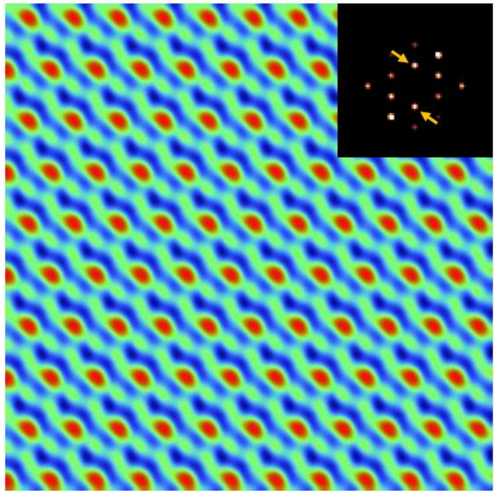
**FFT endmembers**



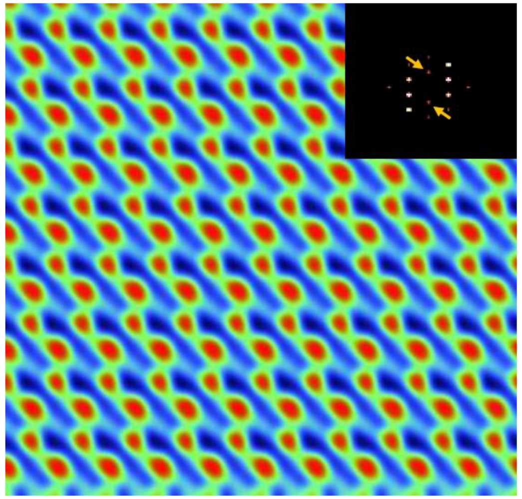




In the 2nd component, 2 spots in the "inner hexagon" are strongly suppressed reflecting a fine structure of charge ordered pattern

**Real-space images of corresponding phases**



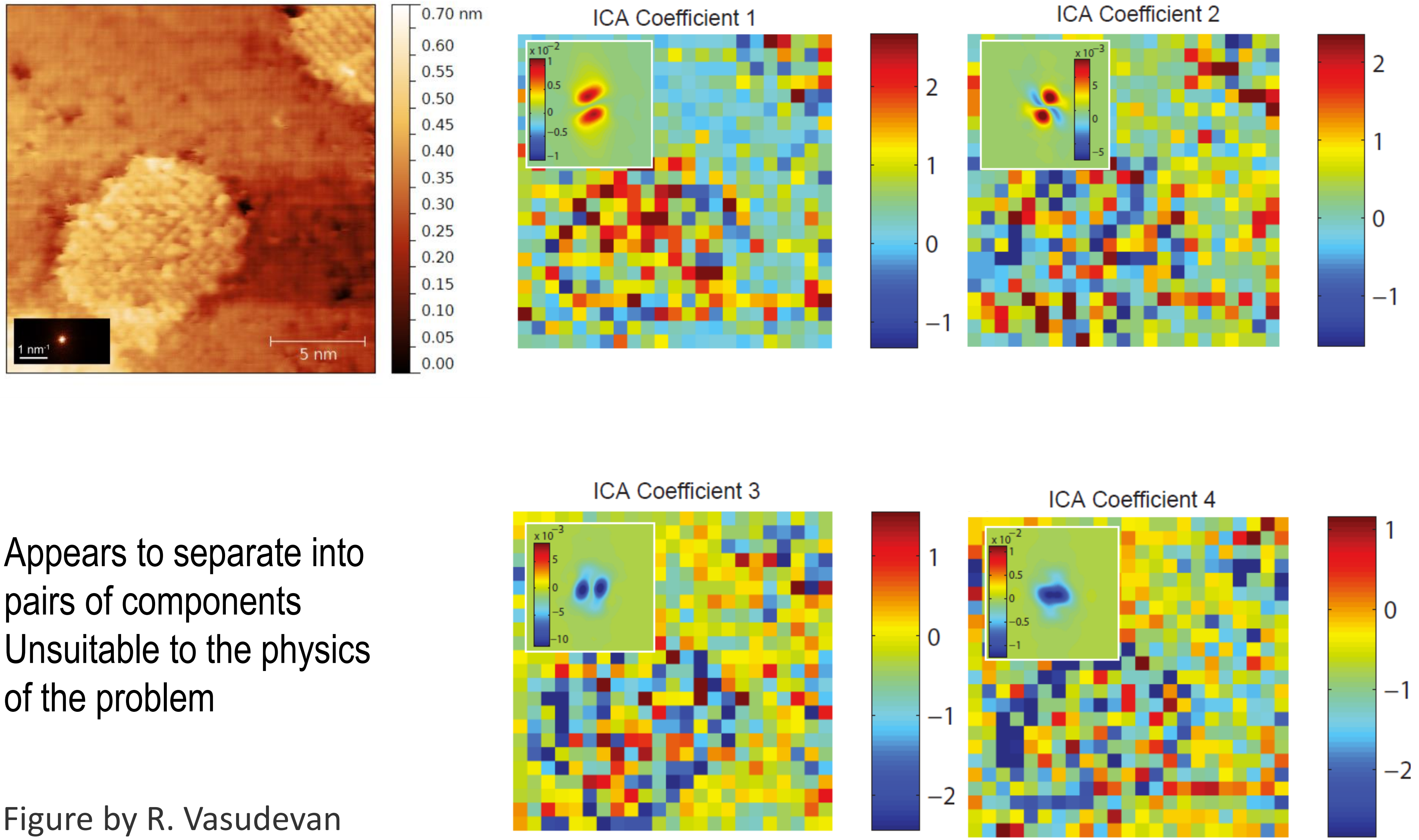
*Hexagonal superlattice*



*Dimer superlattice*

M. Ziatdinov, A. Banerjee, A. Maksov, T. Berlijn, W. Zhou, H.B. Cao, J.Q. Yan, C.A. Bridges, D.G. Mandrus, S.E. Nagler, A.P. Baddorf, and S.V. Kalinin, *Atomic-scale observation of structural and electronic orders in the layered compound* α-$RuCl_3$, Nature Comm. **7**, 13774 (2016).

# ICA on Sliding FFT




- Appears to separate into pairs of components
- Unsuitable to the physics of the problem

Figure by R. Vasudevan

# NFIND-R vs. ICA

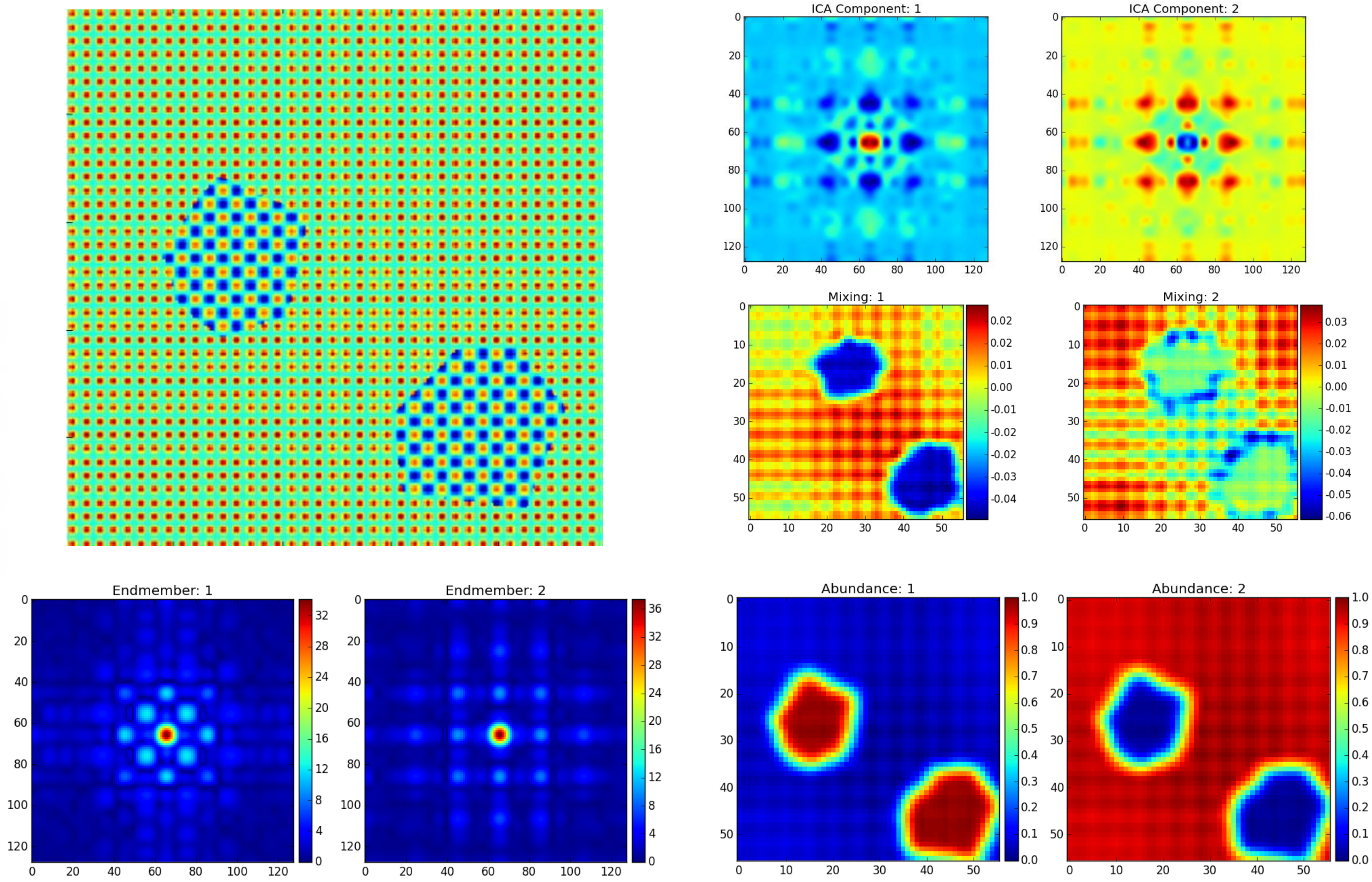



Figure by
R. Vasudevan

# Sliding FFT:



- We always have a problem of window size:
  - too large – loose spatial resolution,
  - too small – FFT behaves poorly due to edge effects
- Interpretation of FFT data is complicated (too much data if fit each peak, unclear meaning of the unmixing components)
- Natural descriptor for atomically resolved images – atomic coordinates!

# Local crystallography

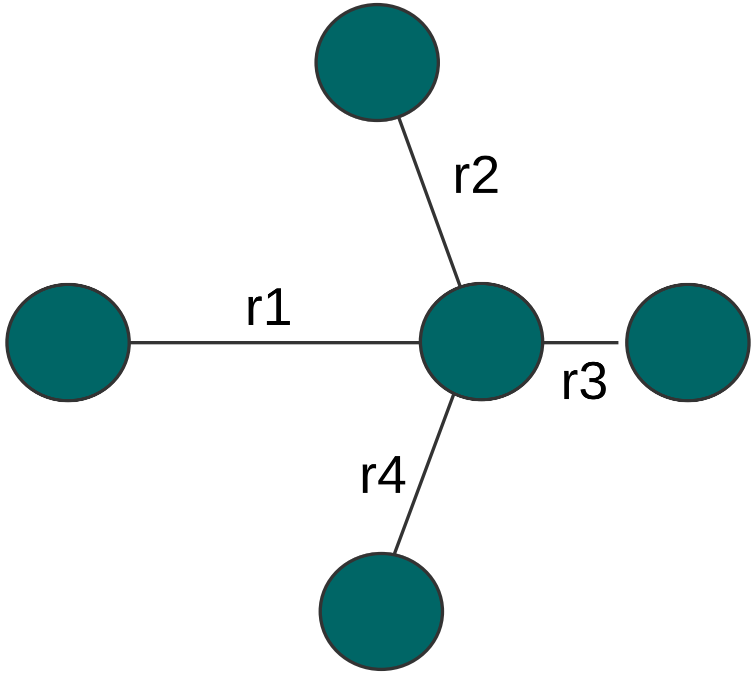


For each atom, define nearest neighbors and generate array of the corresponding radius-vectors of the form

$NA_{ij} = (rx_1,ry_1, rx_2,ry_2,rx_3,ry_3,rx_4,ry_4)_{ij}$

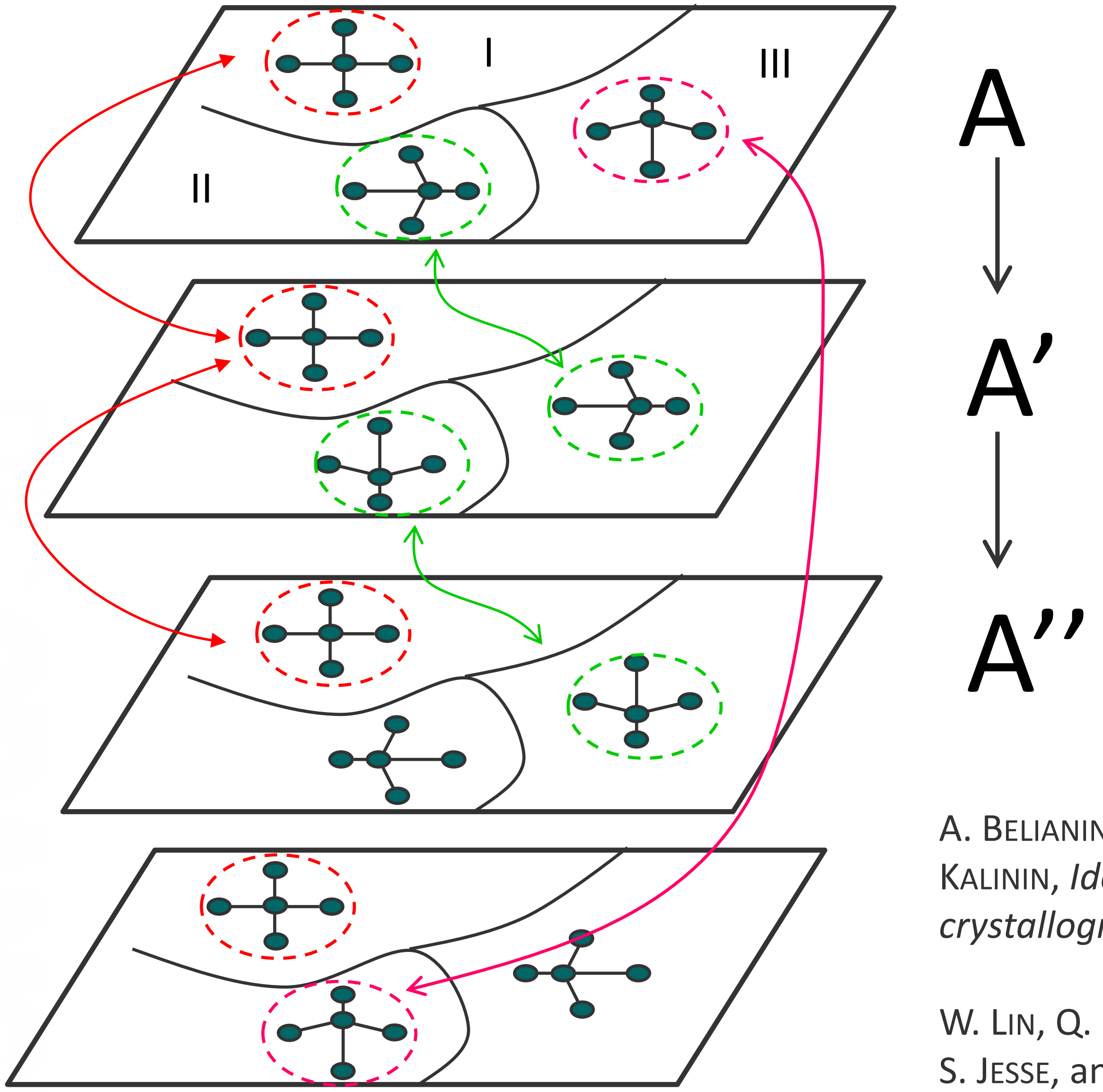
Indexes 1,2,3,4 are chosen in the same sense for all atoms (generalization for different lattice and/or next coordination sphere obvious)

Then, phase/ferroic variant identification problem can be reduced to finding equivalent (in statistical sense) groups of nearest neighbours (for limited sense, we use point groups, for general sense, we use the spatial group and add translation symmetry operations, i.e. i -> i+1 and j -> j+1 for lattice doubling)



# Local crystallography






Same cluster in all replicas:
Non-ferroic phase

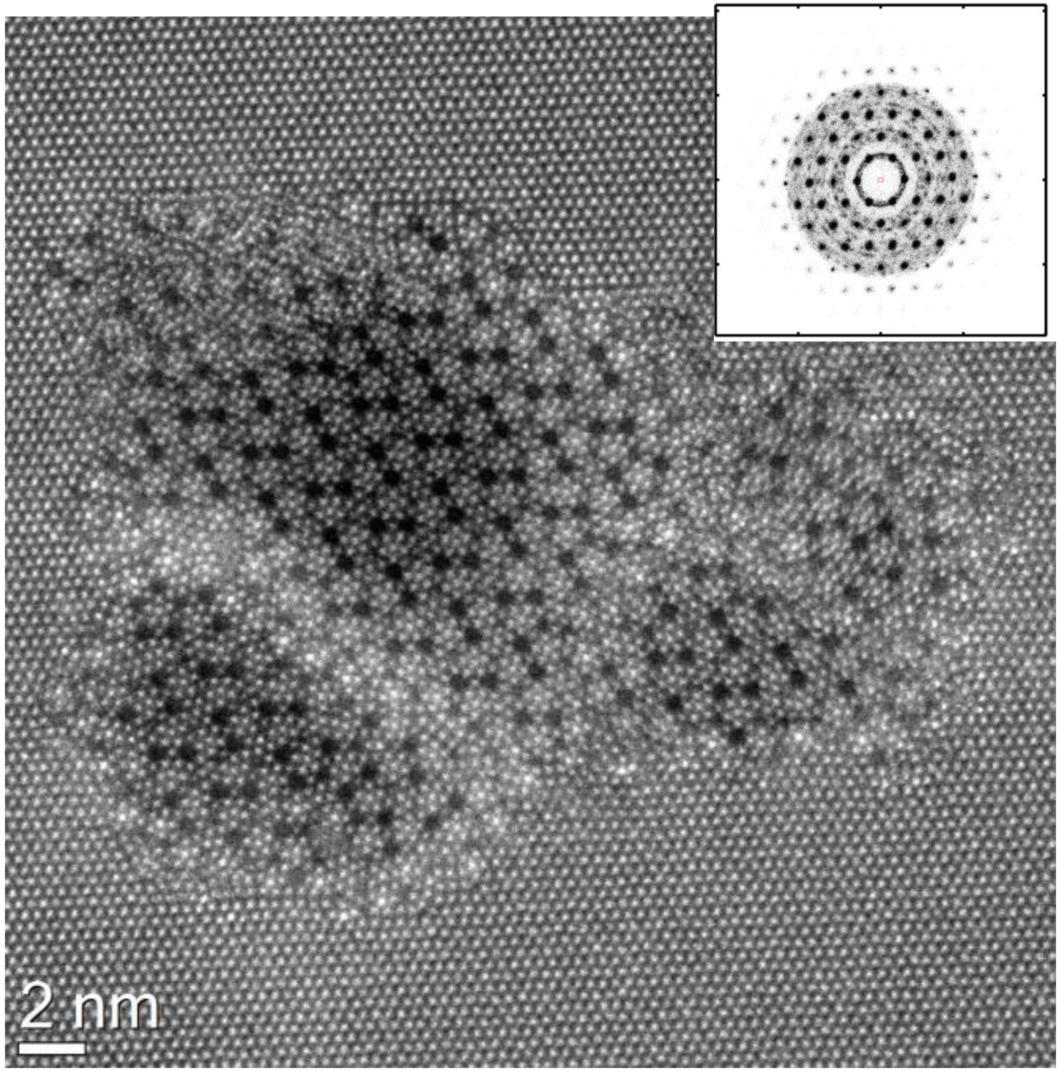
Form cluster with different regions in different replicas:
Ferroic phase

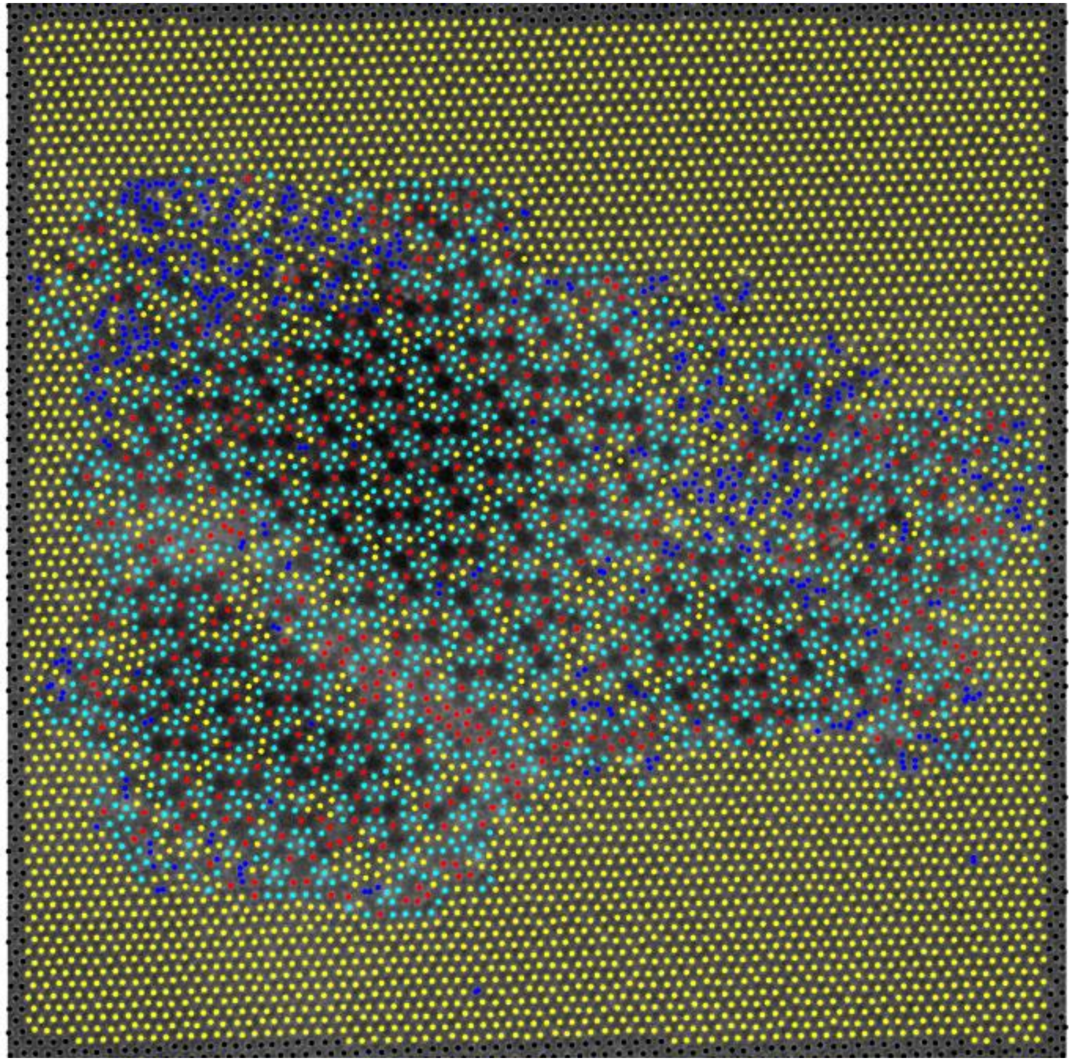
Only some of the correspondences are shown (but these are obvious)

A. Belianinov, Q. He, M. Kravchenko, S. Jesse, A. Borisevich, and S.V. Kalinin, *Identification of phases, symmetries, and defects through local crystallography*, Nat. Comm. **6**, 7801 (2015).

W. Lin, Q. Li, A. Belianinov, B.C. Sales, A. Sefat, Z. Gai, A.P. Baddorf, M. Pan, S. Jesse, and S.V. Kalinin, *Local crystallography analysis for atomically resolved scanning tunneling microscopy images*, Nanotechnology **24**, 415707 (2013).

# Local crystallography: k-means

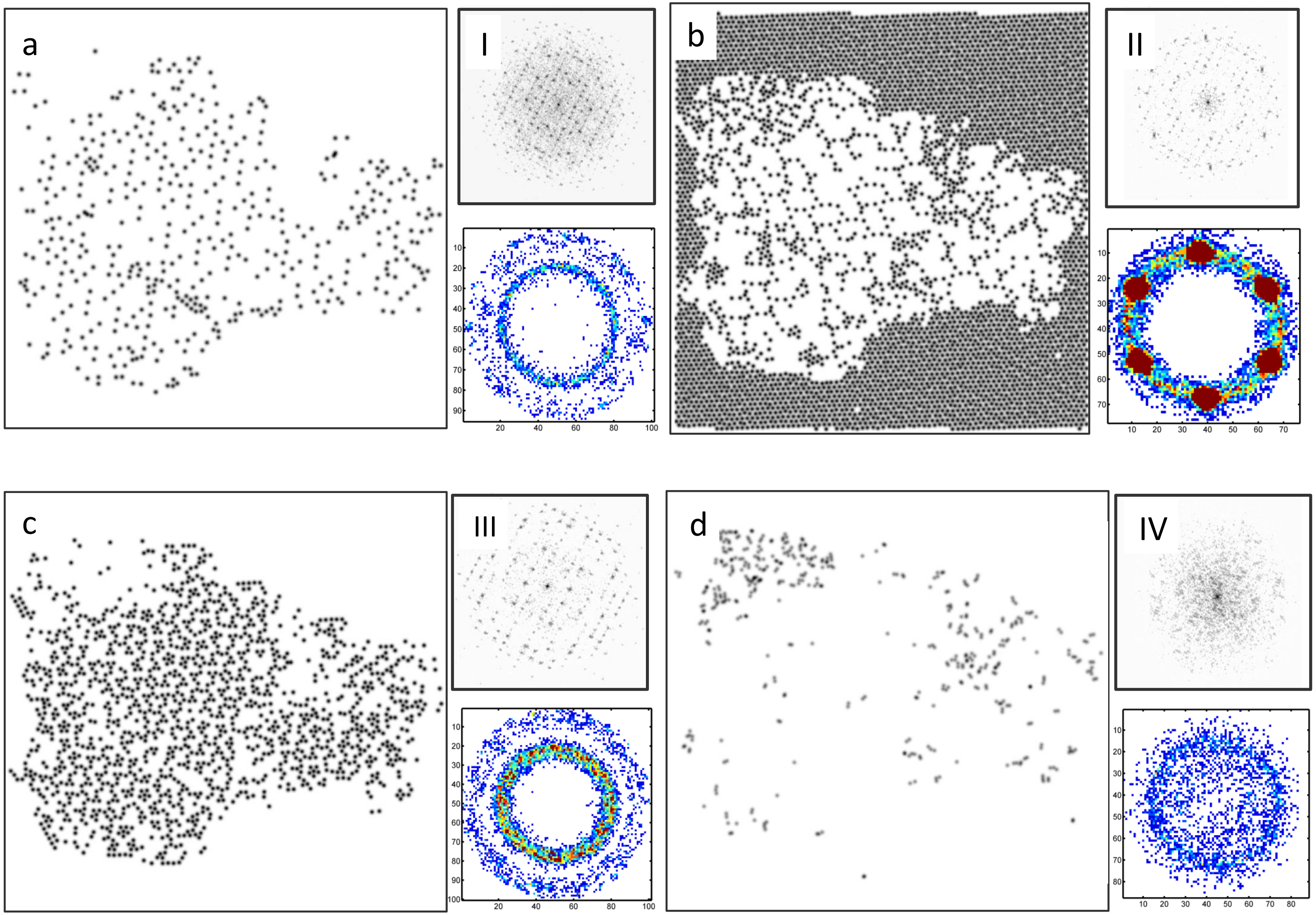


A. Belianinov, Q. He, M. Kravchenko, S. Jesse, A. Borisevich, and S.V. Kalinin, *Identification of phases, symmetries, and defects through local crystallography*, Nat. Comm. **6**, 7801 (2015).

# Local crystallography





# Local crystallography

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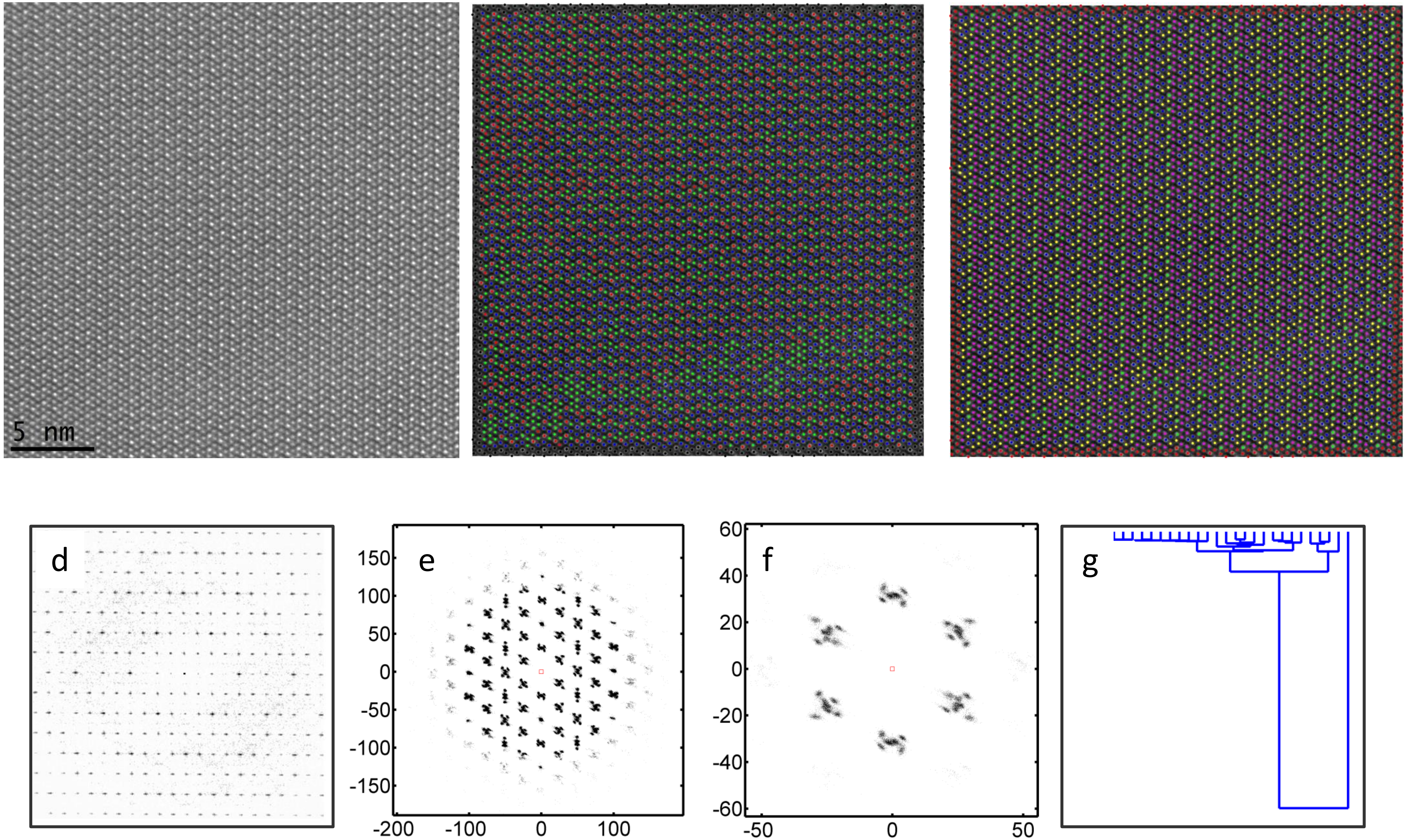
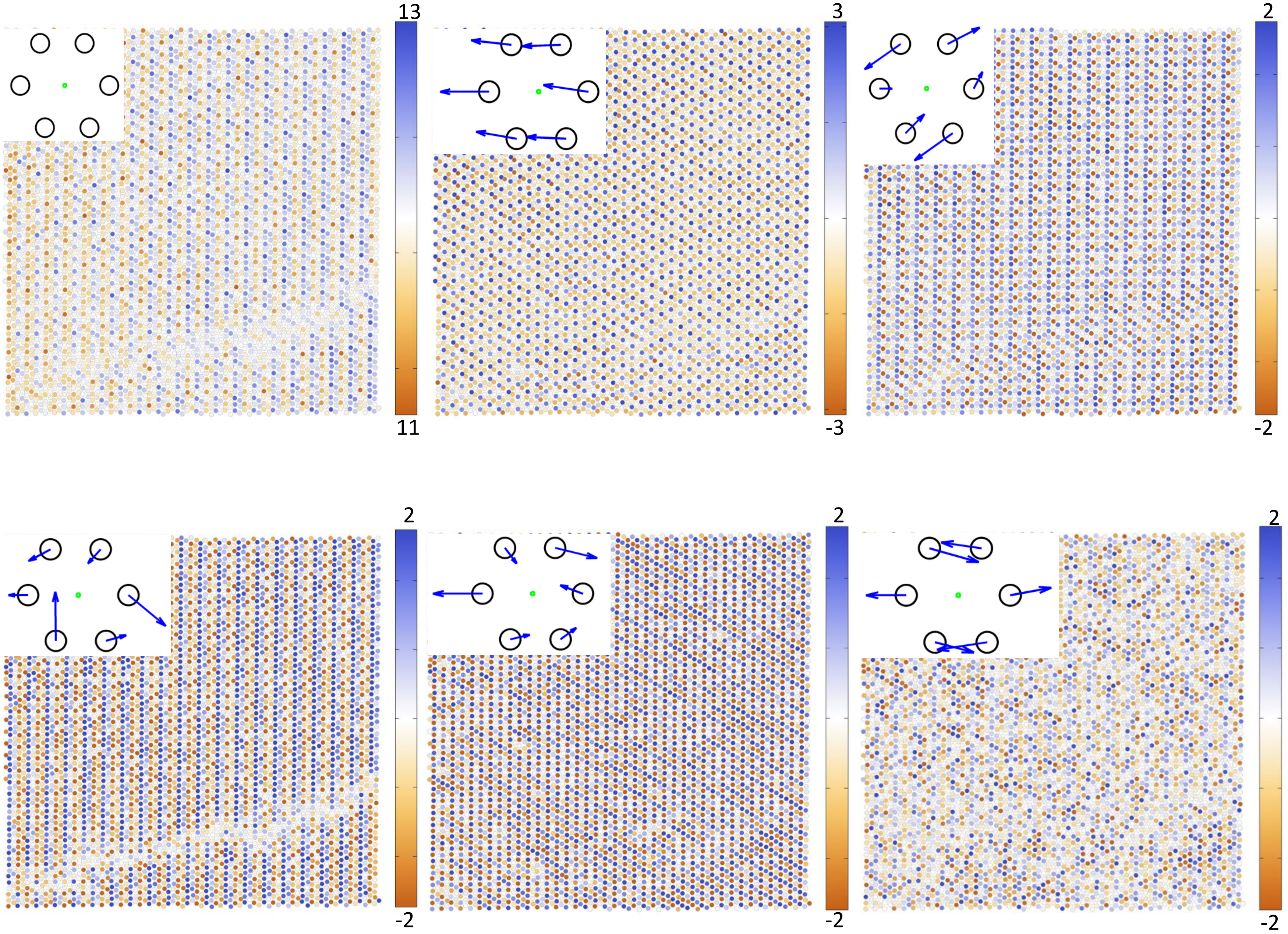


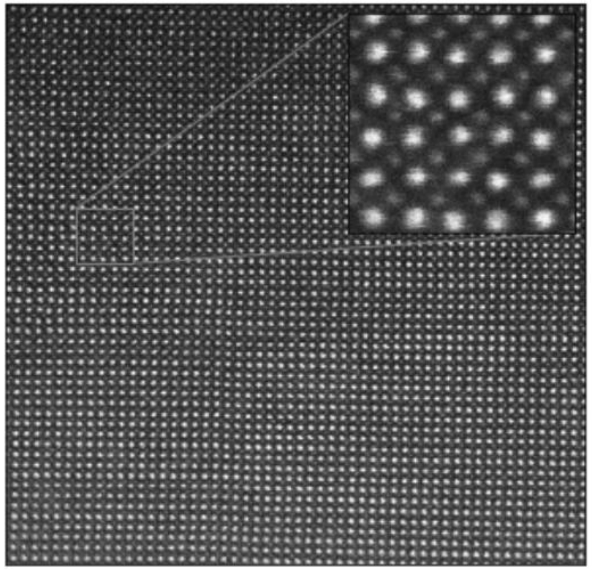
# Local crystallography



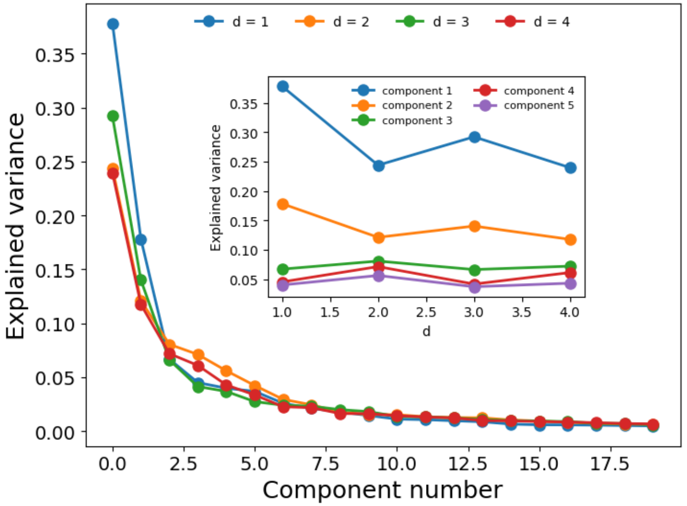


# Local crystallography: FerroNET

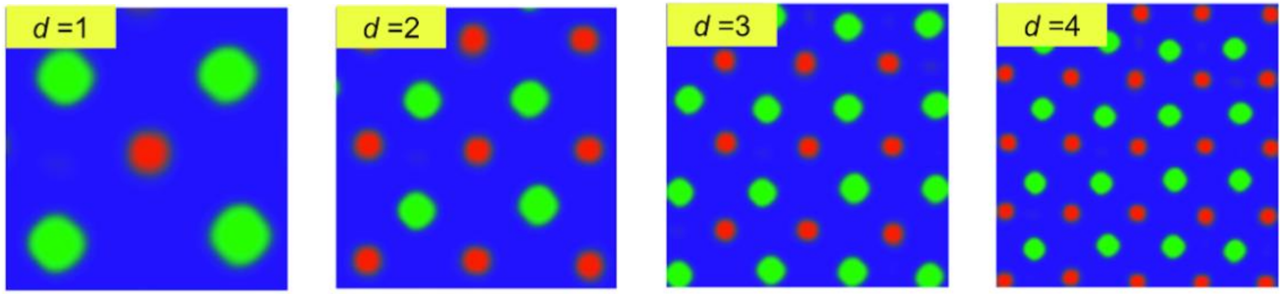
## Experimental (LBFO film)



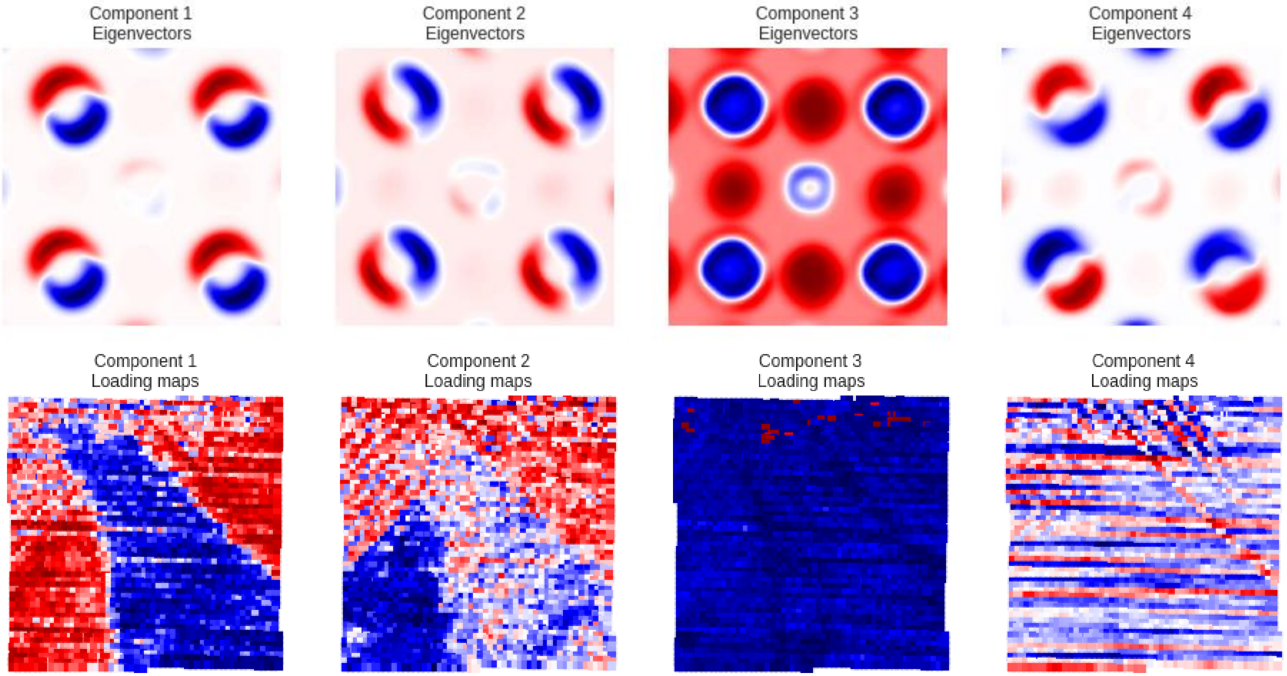
## Information Content




## Building blocks (from neural network output)




## PCA eigenvectors and loading maps



# $MoS_2$-$ReS_2$

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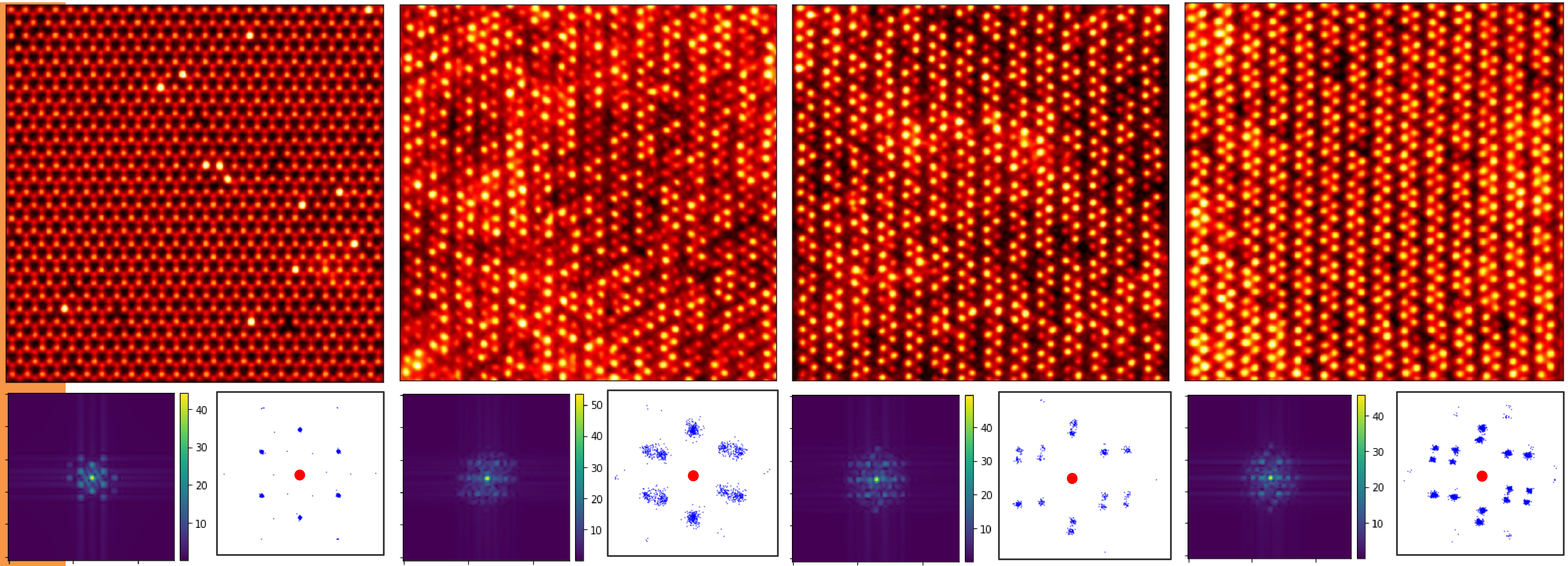
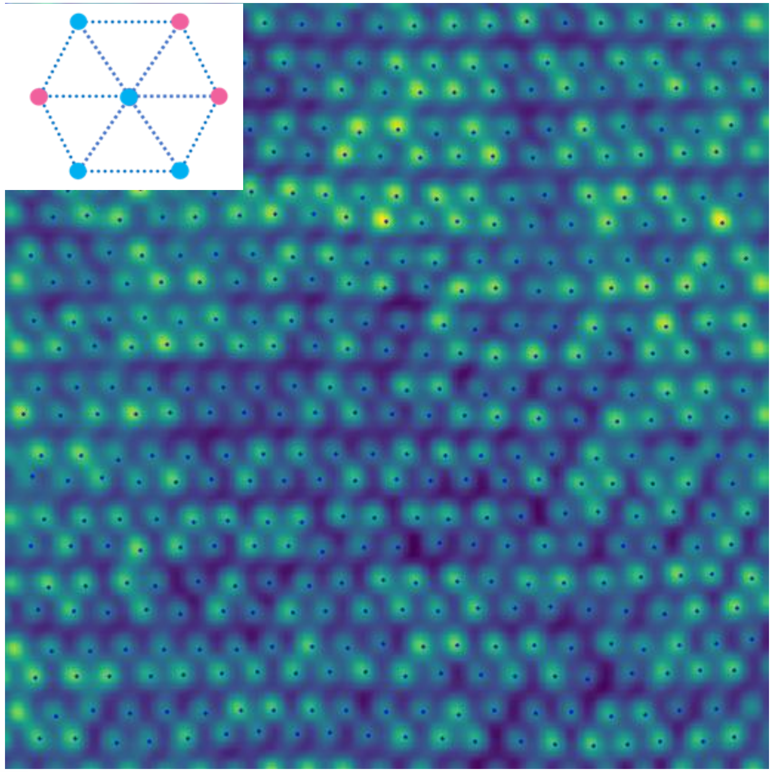
5% $ReS_2$ 55% $ReS_2$ 78% $ReS_2$ 95% $ReS_2$

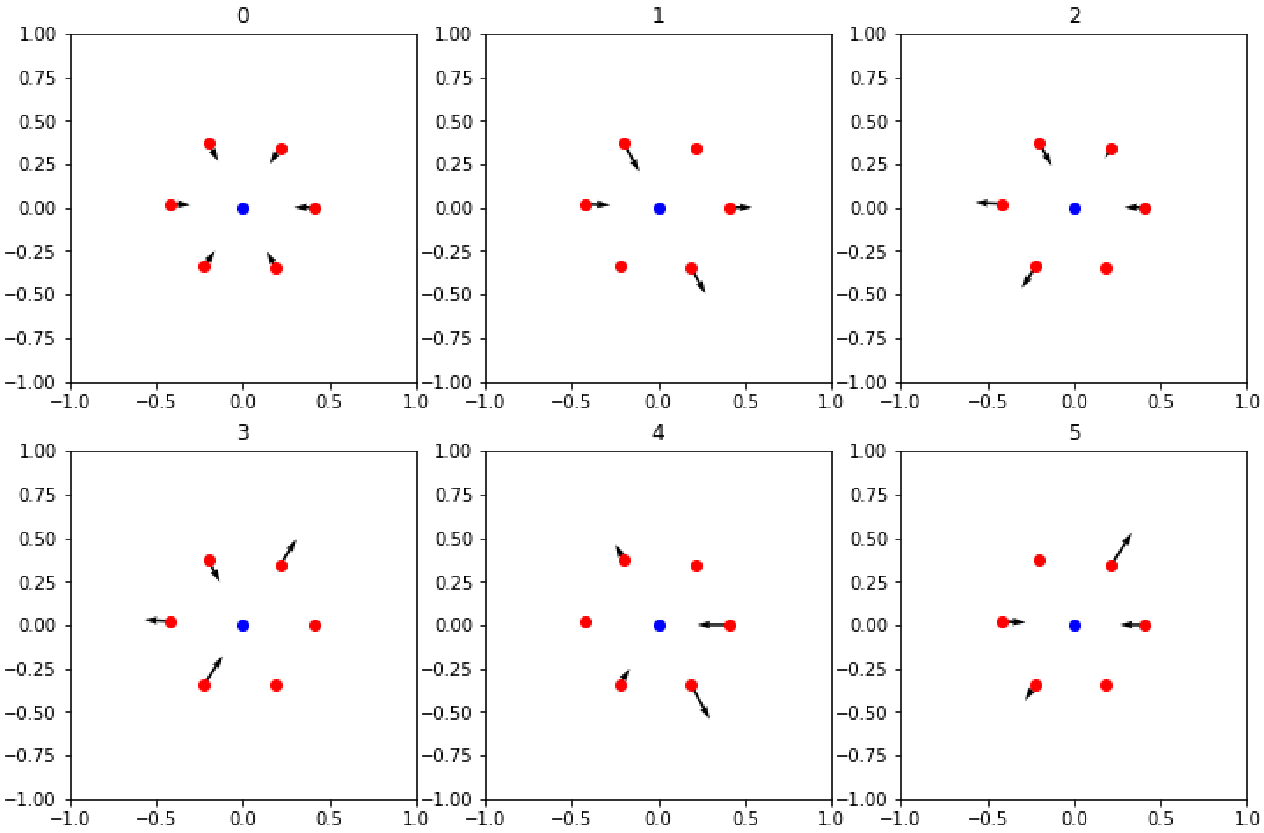


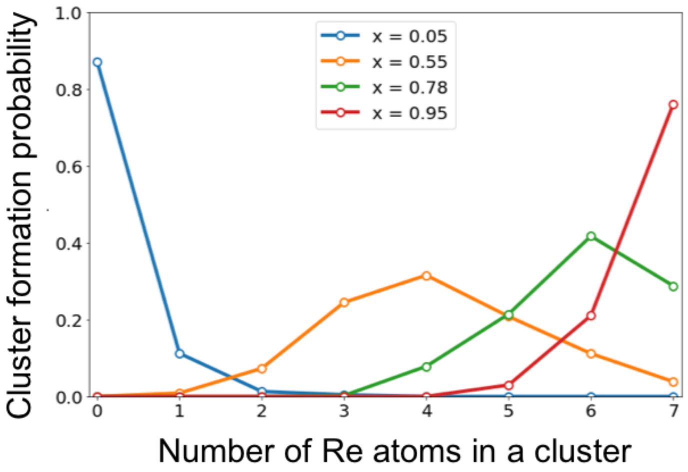

Data by Shize Yang and Matt Chisholm

# Local crystallography





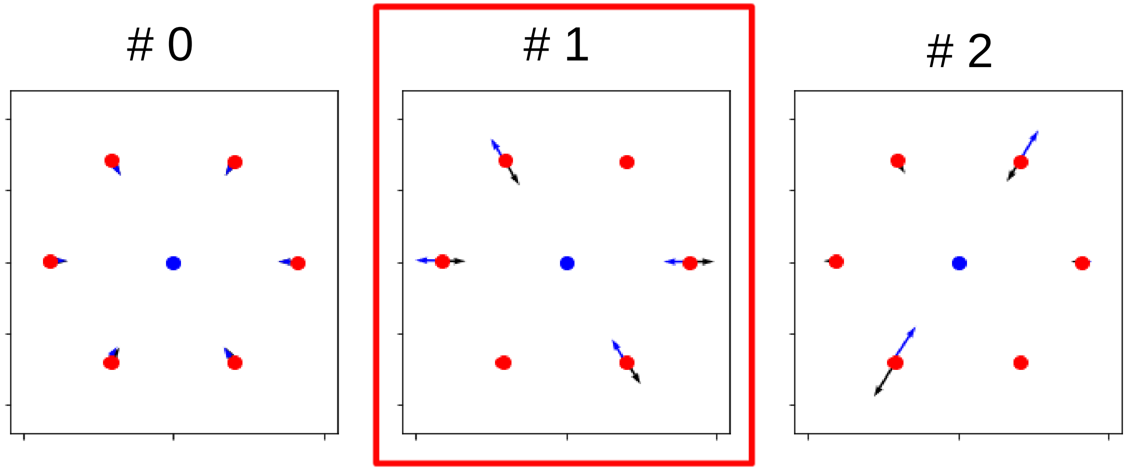


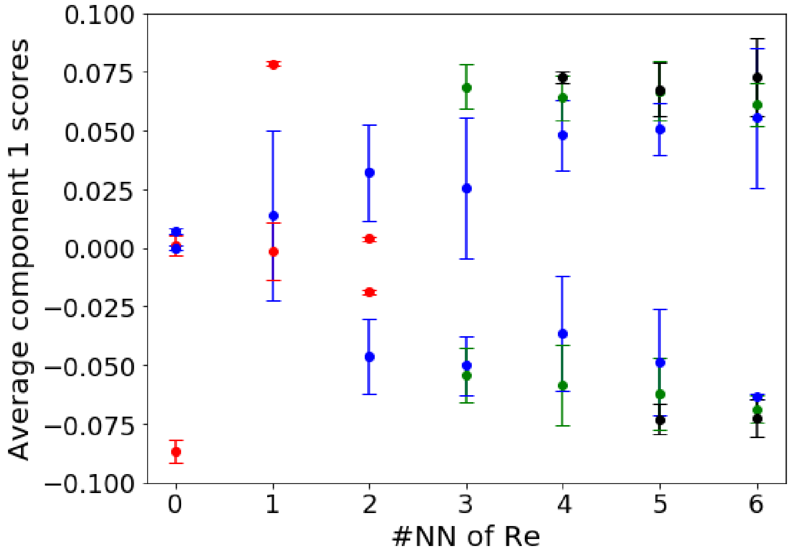

- Traditionally, the order parameter is defined based on symmetry and atomistic representation is established in the *ad hoc* manner
- But what if we define order parameter from the bottom up – based on the statistics of atomic distortions?
- And further correlate it to local chemical composition?

# Describing the phase transition

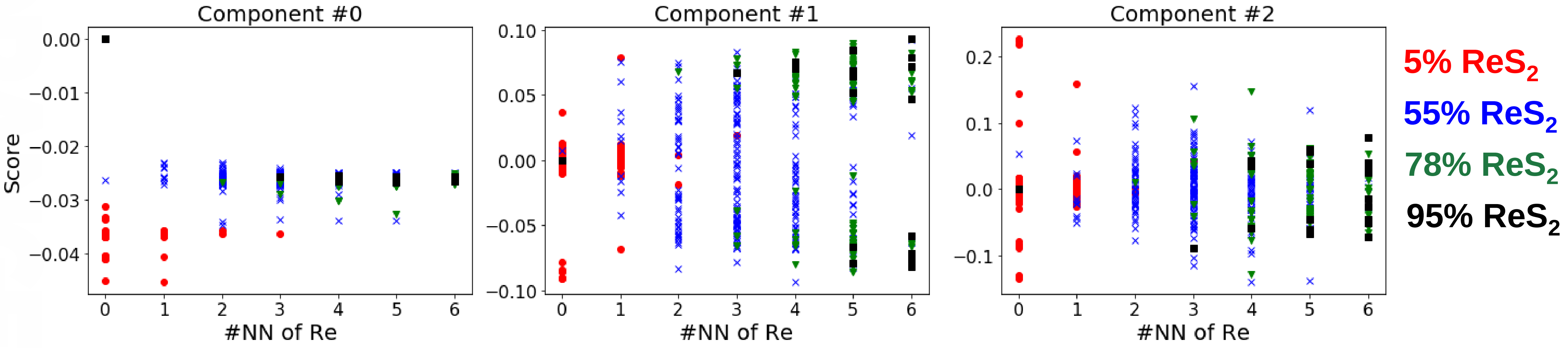
**Three dominant distortion modes**




**Local symmetry breaking!**

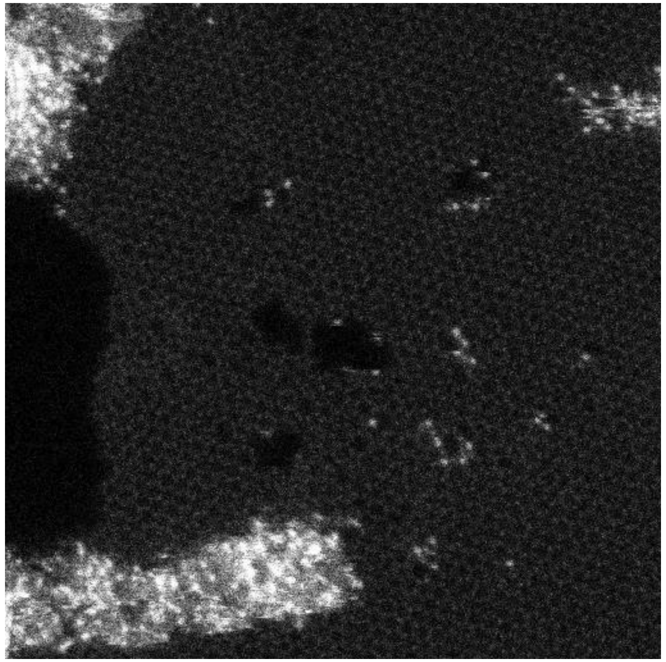



**Mode distributions vs. global and local composition**



# What about chemical dynamics?

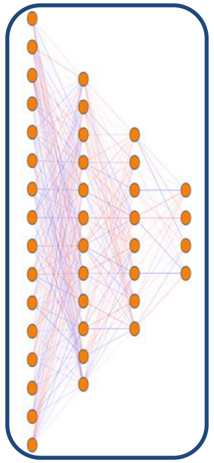
Experimental STEM movie
Graphene+Si under e-beam



Data collected by O. Dyck (ORNL)

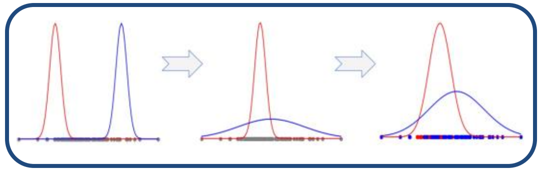
Deep & machine learning

Deep learning

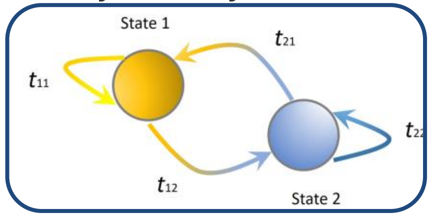


Position and type
of all atomic species

Clustering/unmixing:
Library of structural descriptors



Markov model:
System dynamics



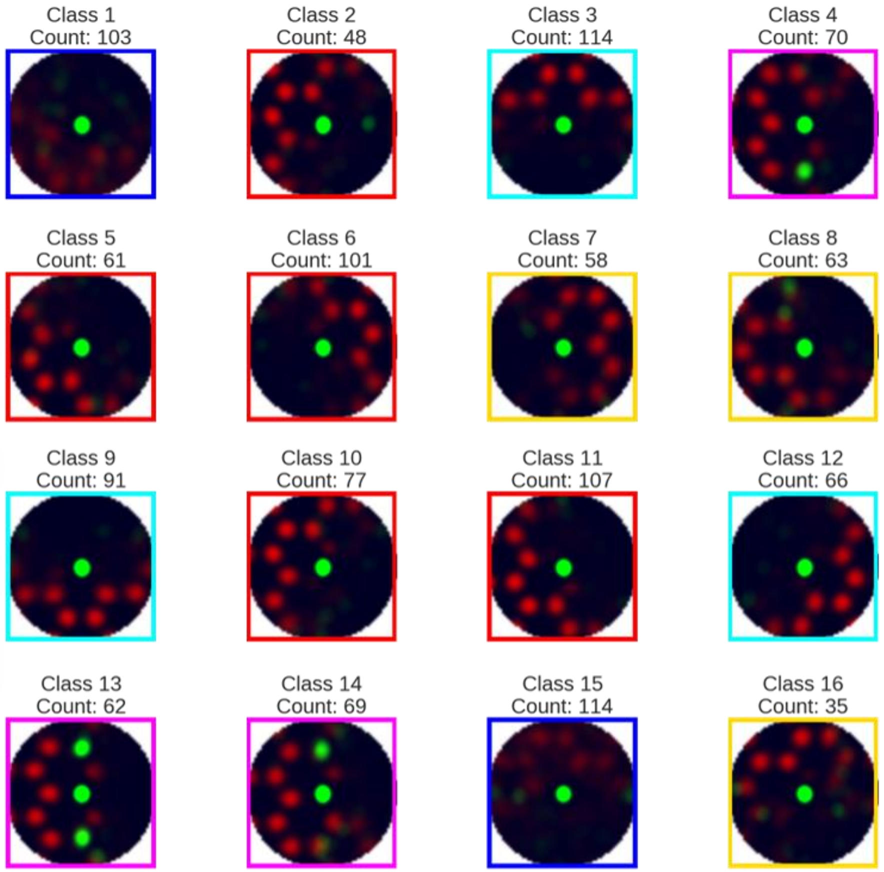

1. Convert noisy experimental data into atomic positions/trajectories → Deep convolutional neural networks
2. Create libraries of structural descriptors → Clustering/unmixing applied to the output of neural networks
3. Analysis of dynamics and transition probabilities → Markov modelling on the constructed classes

Ziatdinov et al., ACS nano 11, 12742 (2017)
Ziatdinov et al., Appl. Phys. Lett. 115, 052902 (2019)
Ziatdinov et al., npj Computational Materials 3, 31 (2017)
Maksov et al., npj Computational Materials 5, 12 (2019)

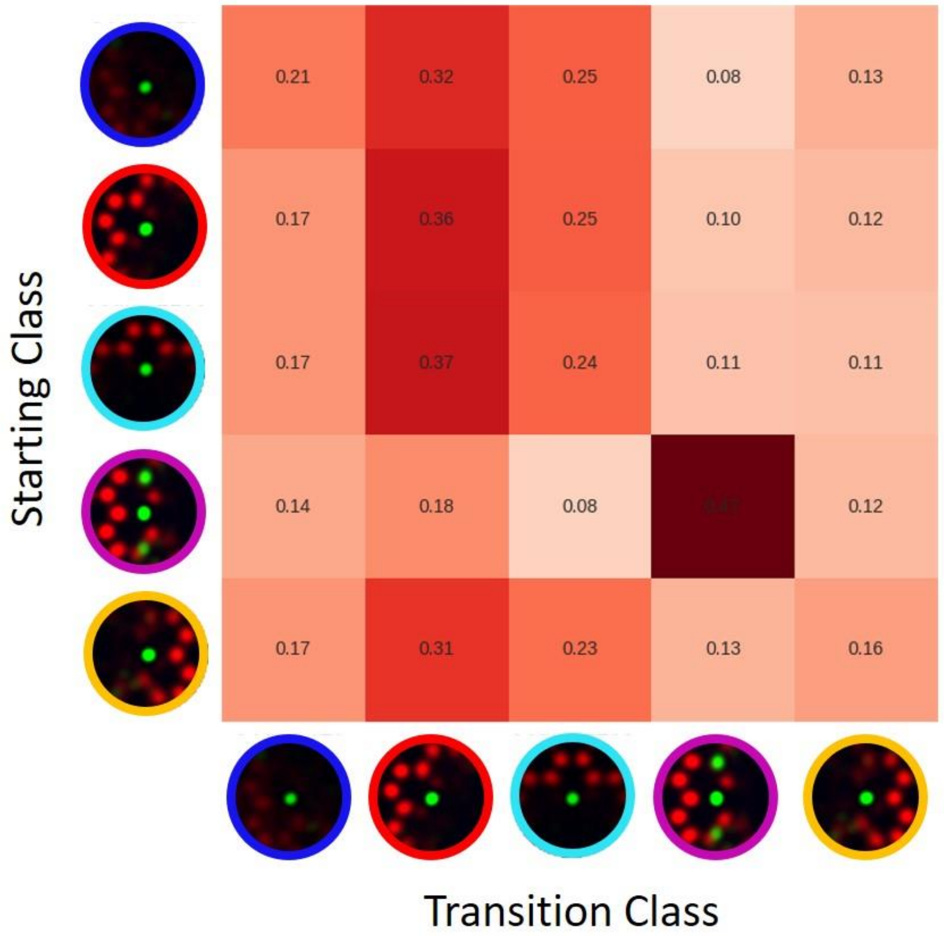
# Local crystallography



## Derived classes of Si-C edge configurations




## Transition probabilities matrix



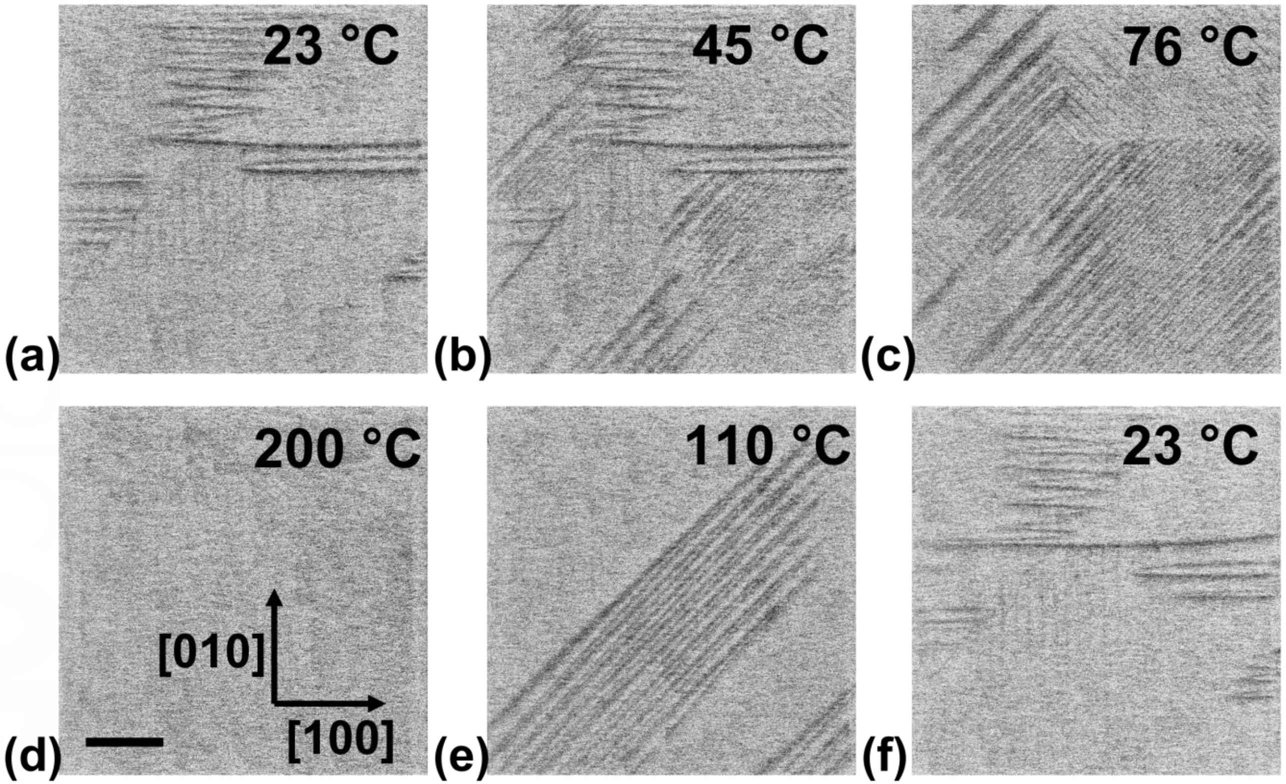

- Gaussian mixture model

- Discrete rotation symmetry + structural similarity algorithm

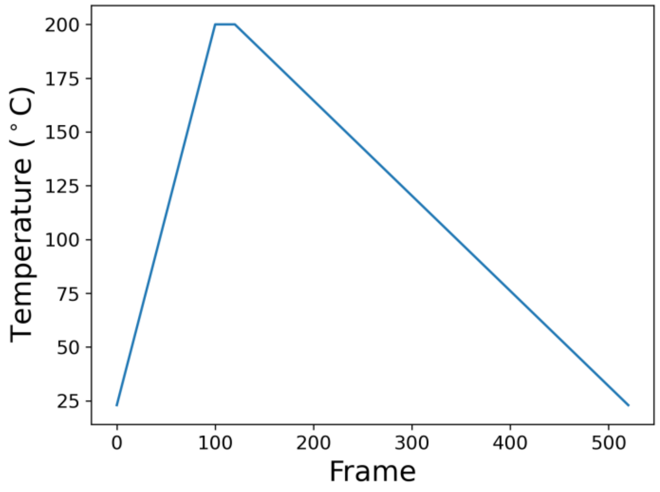
- Markov state analysis

Ziatdinov *et al.*, ArXiv:1901.09322 (2019)

# Mesoscopic STEM data




Scale: 500 nm

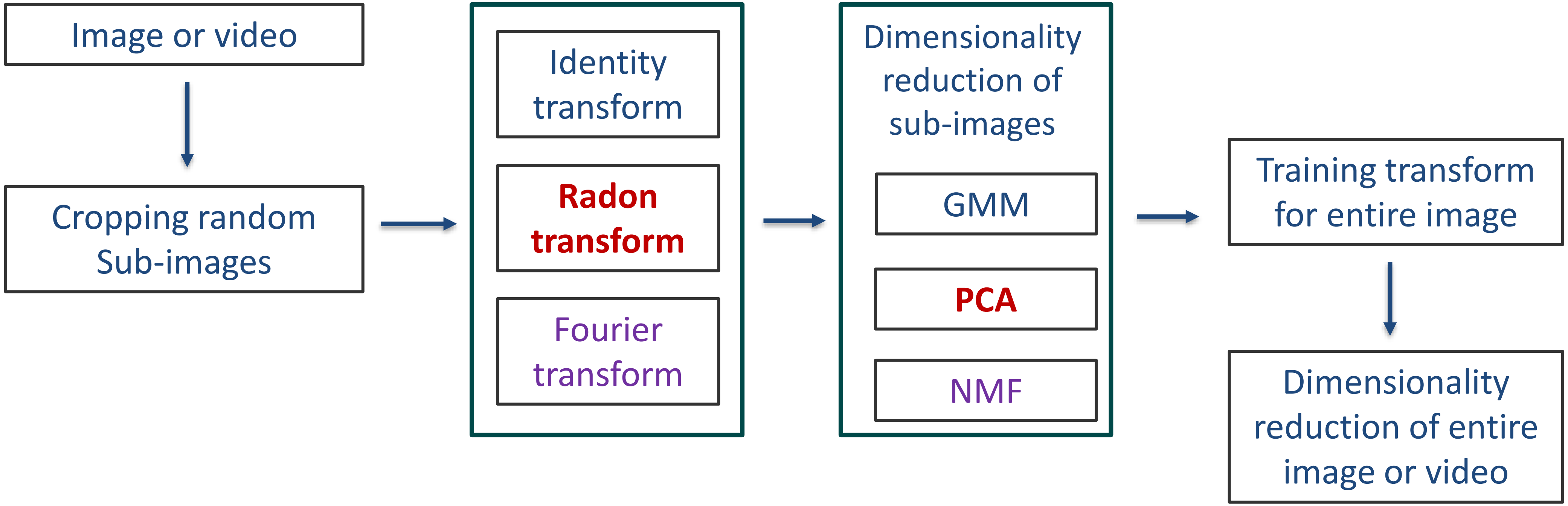


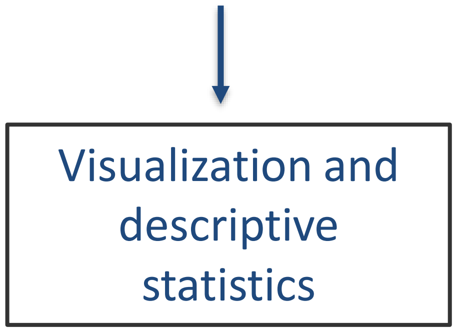

**Key Observations**

- We start with only 180° domain walls at RT (tetragonal phase)
- 90° domain walls at around 50°C (orthorhombic phase)
- Curie Temperature ($T_c$ = 137°C) (Cubic phase)
- 90° - 180° transformation at around 30°C while cooling

**Dataset:** Dr. Vasiliki Tileli, EPFL

# Extended analysis pipeline



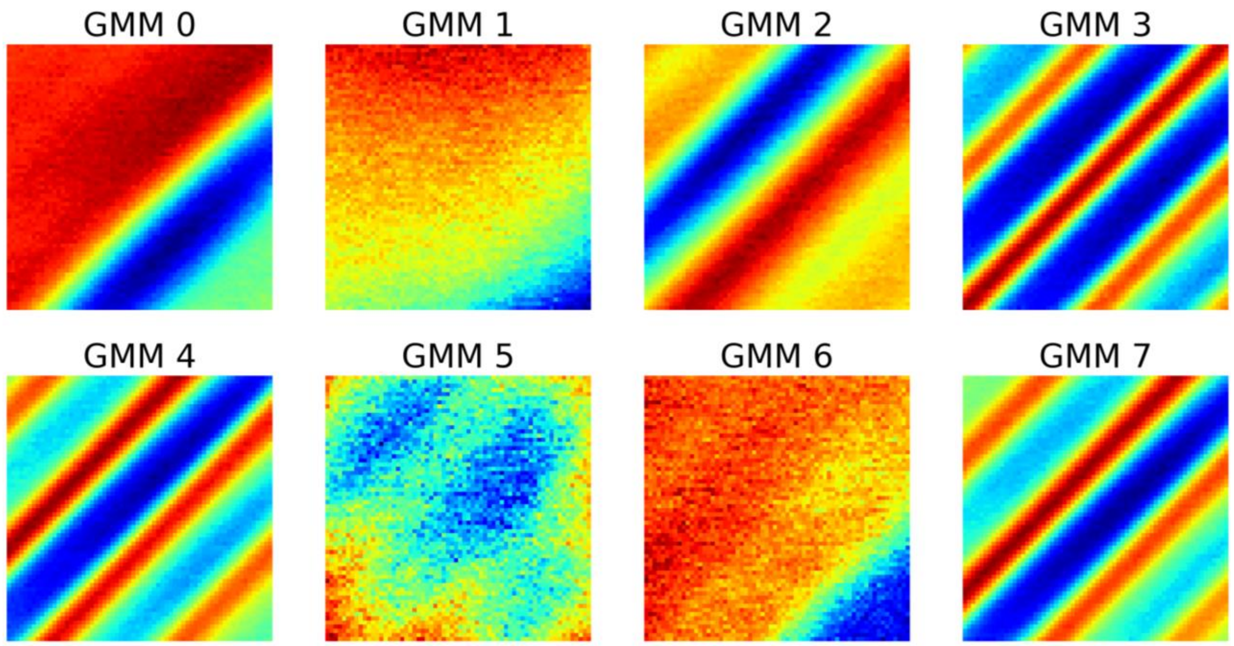

Pipelines are defined to

- Make analysis traceable, repeatable, explainable, and transferable
- Allow for hyperparameter tuning and optimization
- Efficiently use the memory

# Gaussian Mixture Model

**Workflow of the Gaussian Mixture Model (GMM) analysis**

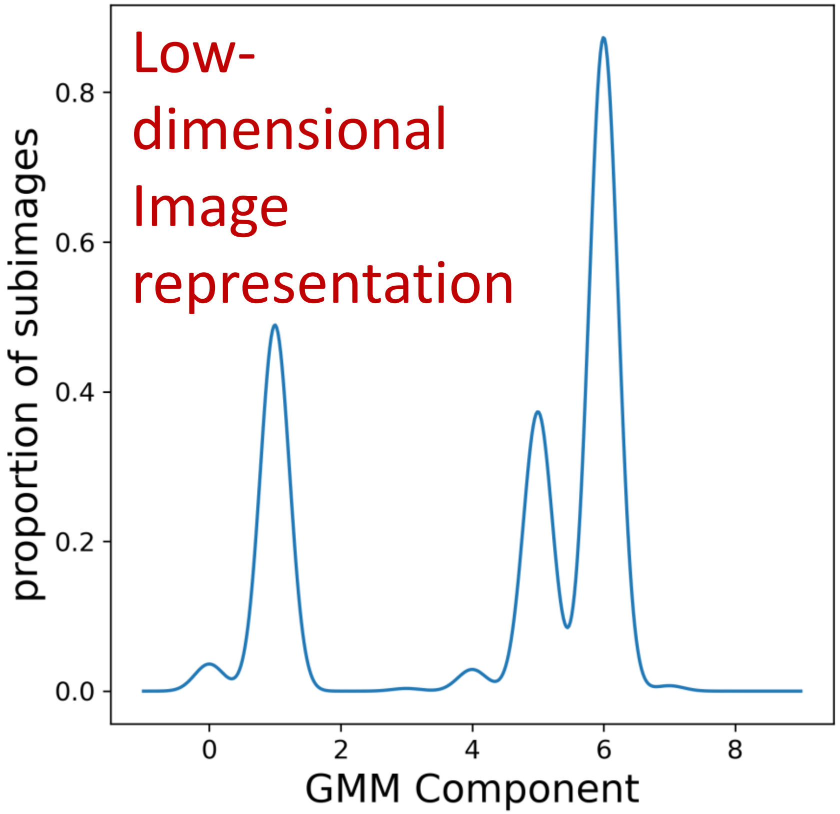
- Feature data: Create random sub-images (500 per frame) from each dataset of a given window size (64*64)
- Perform the GMM on the flattened feature data.
- And the results class centers of resulting classes:
- 90° domain are prominent, while increasing the number of classes will have classes associated with 180° domain walls
- Notice the shifts in the domain wall positions in similar looking classes



# … Continued



- **Workflow of the Gaussian Mixture Model (GMM) analysis**
  - Now each image in our dataset can be represented as a histogram of 8 bins, where the height of each bin corresponds to number of sub-images that fall into the category.
  - Or a continuous probability distribution that approximates the above-mentioned histogram.
  - We chose the latter, and an example of this representation:
  - We will call this the which is a low-dimensional representation “feature-vector” of an image.



# Et voila!

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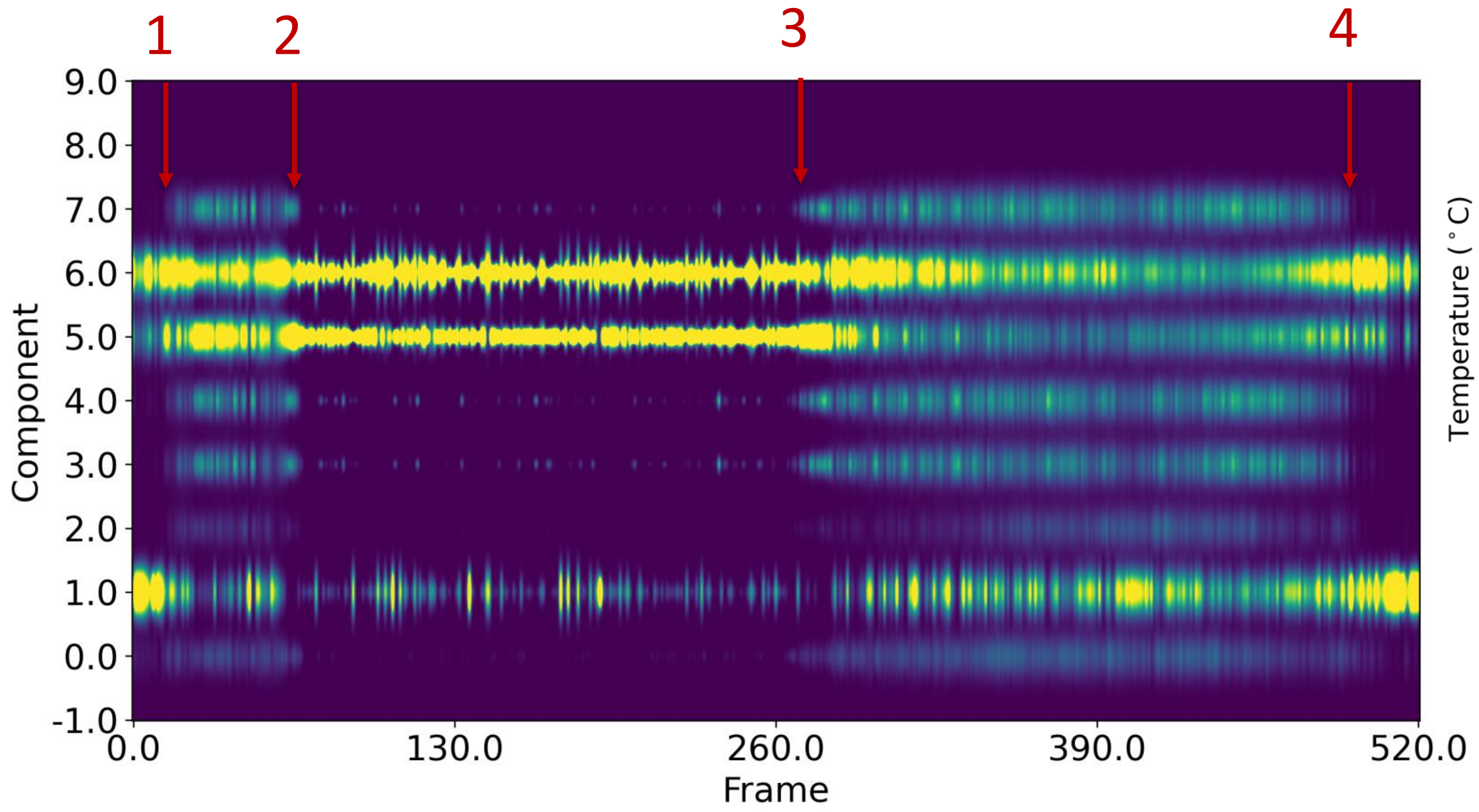
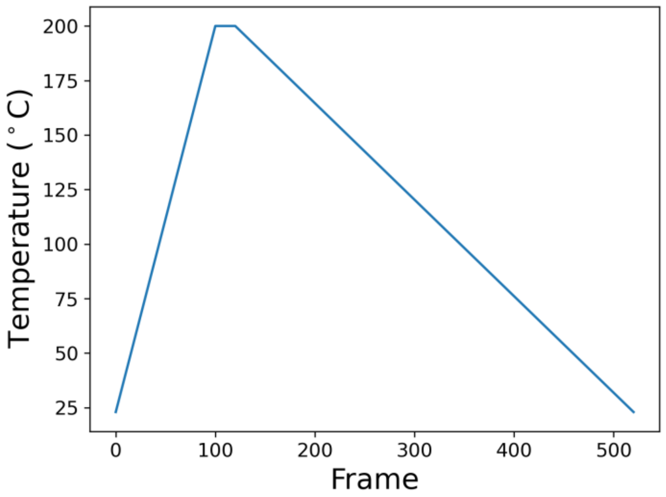
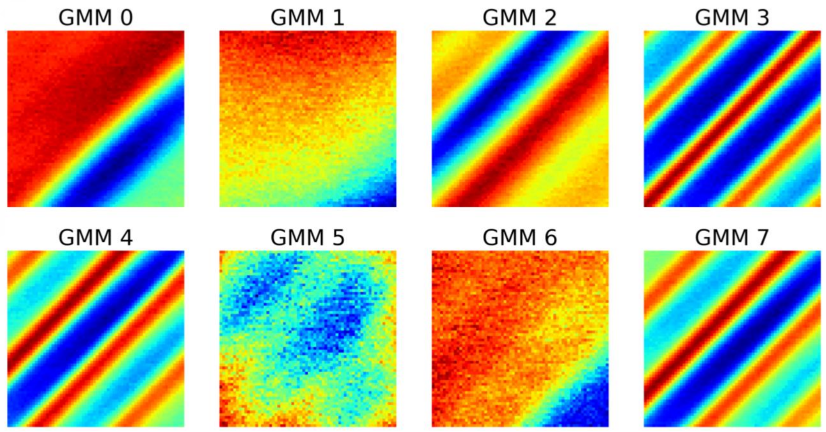


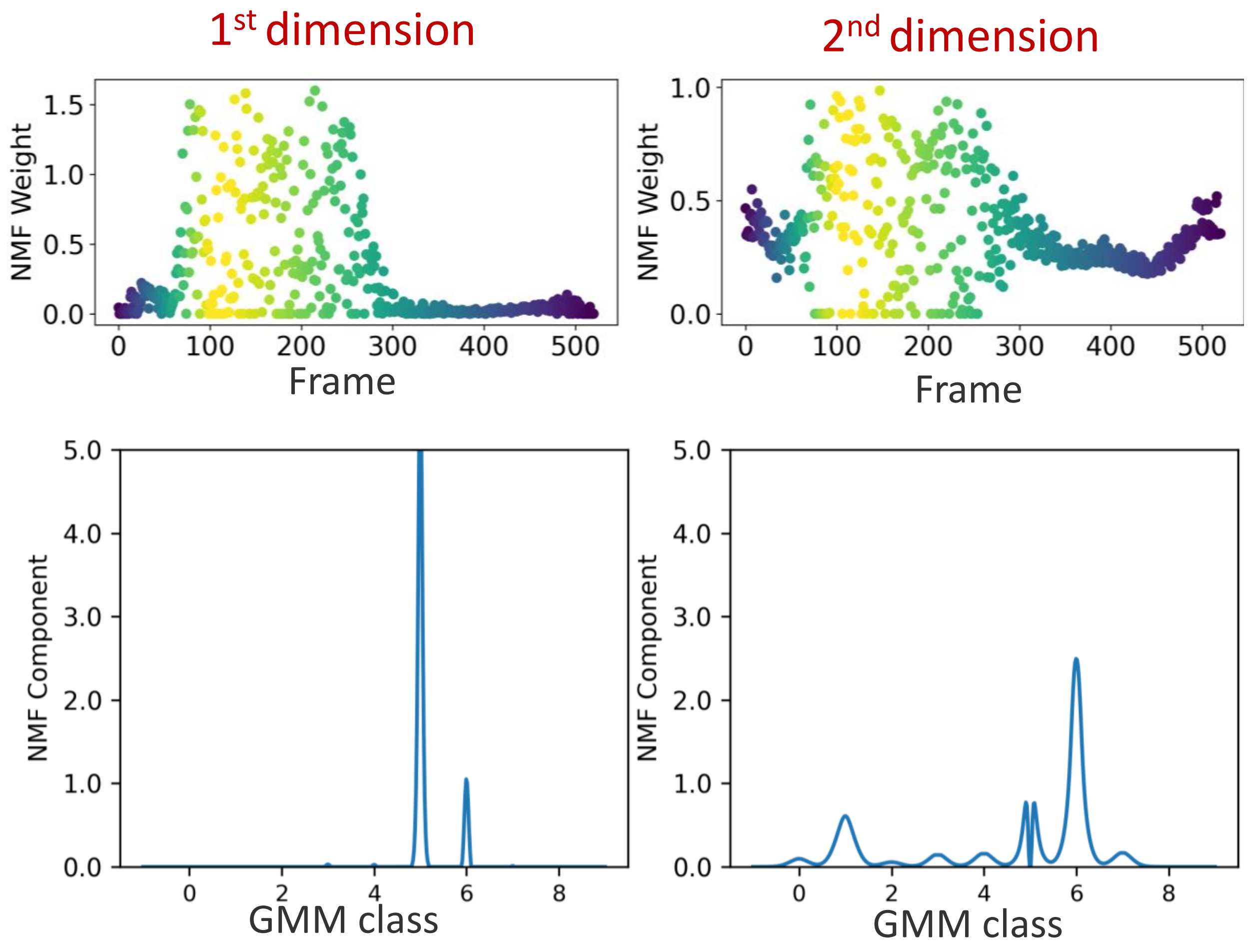




1) 180° domain walls -90°domain walls transition
2) Ferro to Paraelectric transition (Curie temperature)
3) Appearance of 90° domain walls
4) 90° domain - 180°domain walls transition

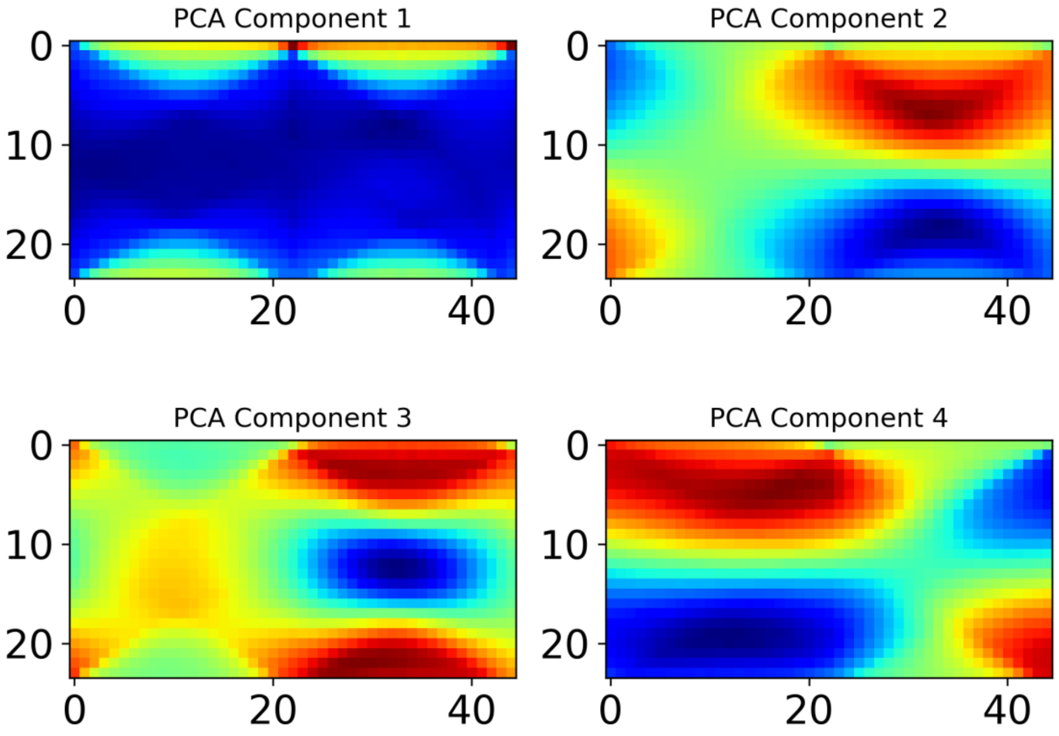
# Describing phase transition

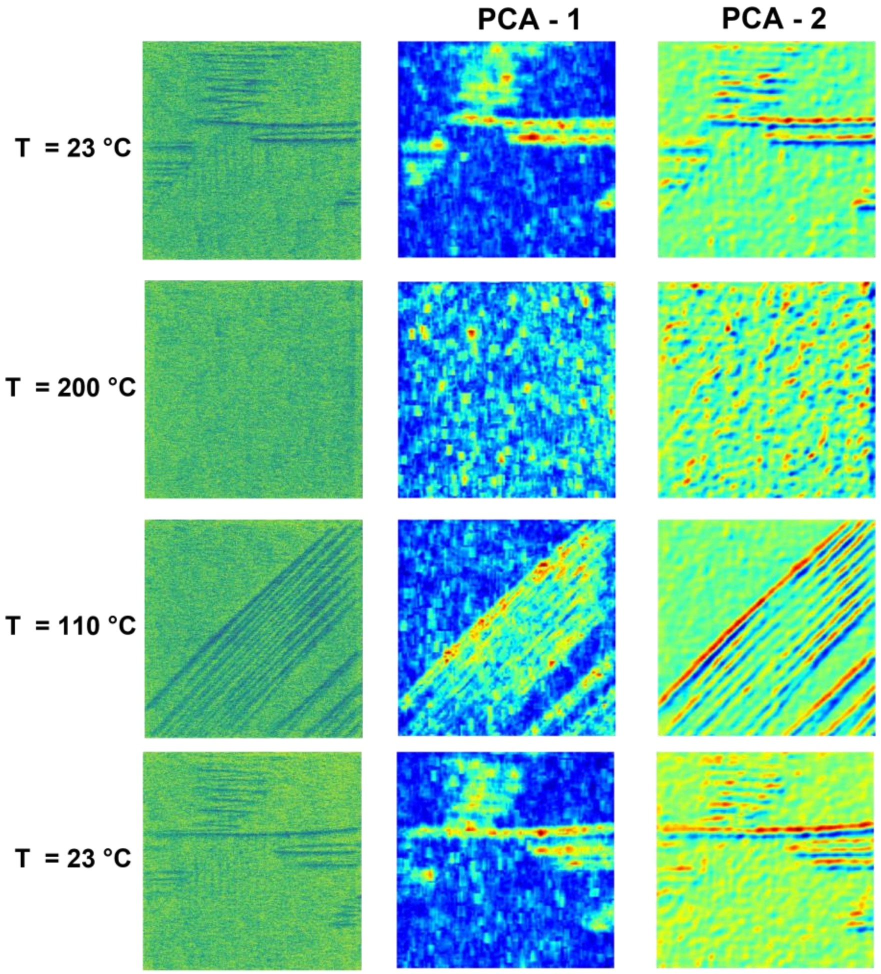
- The data representation can be furthe compressed using dimensionality reduction techniques on the feature vectors
- Applying NMF on the feature vectors matrix:



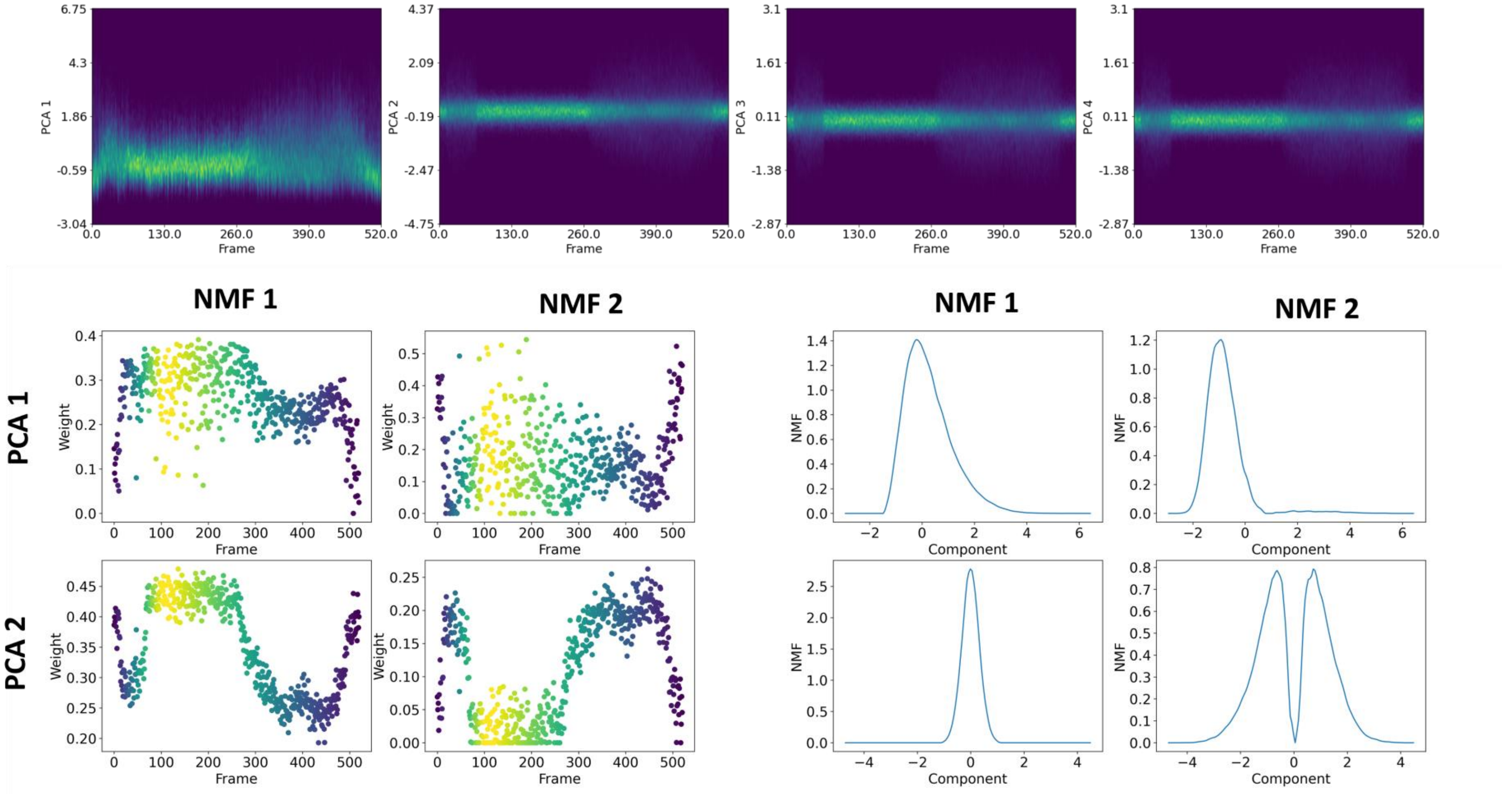



# Radon Transform - PCA



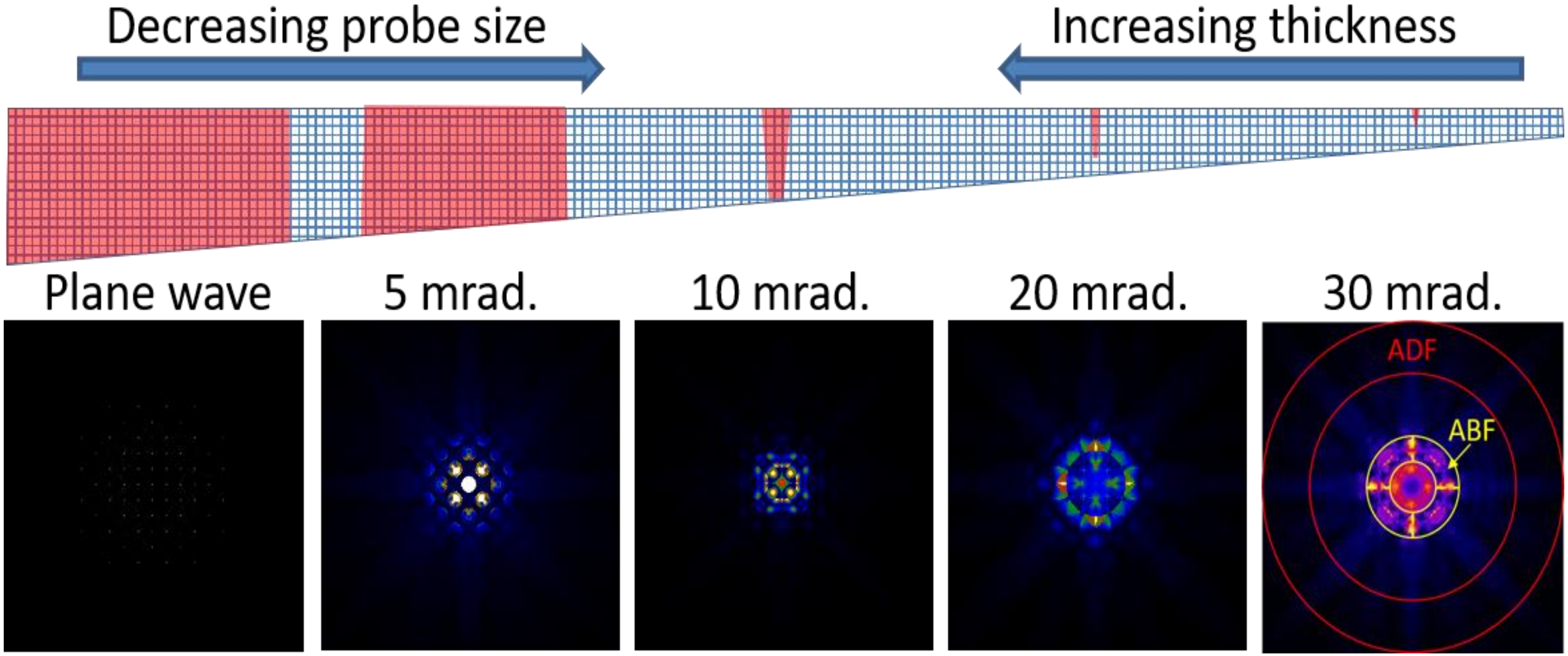
# Radon Transform - PCA




- We have mined the dataset at hand to produce low-dimensional representations for the images.
- The trends in this low-dimensional representations allowed us to study the physics of the system, in this case: phase transitions.

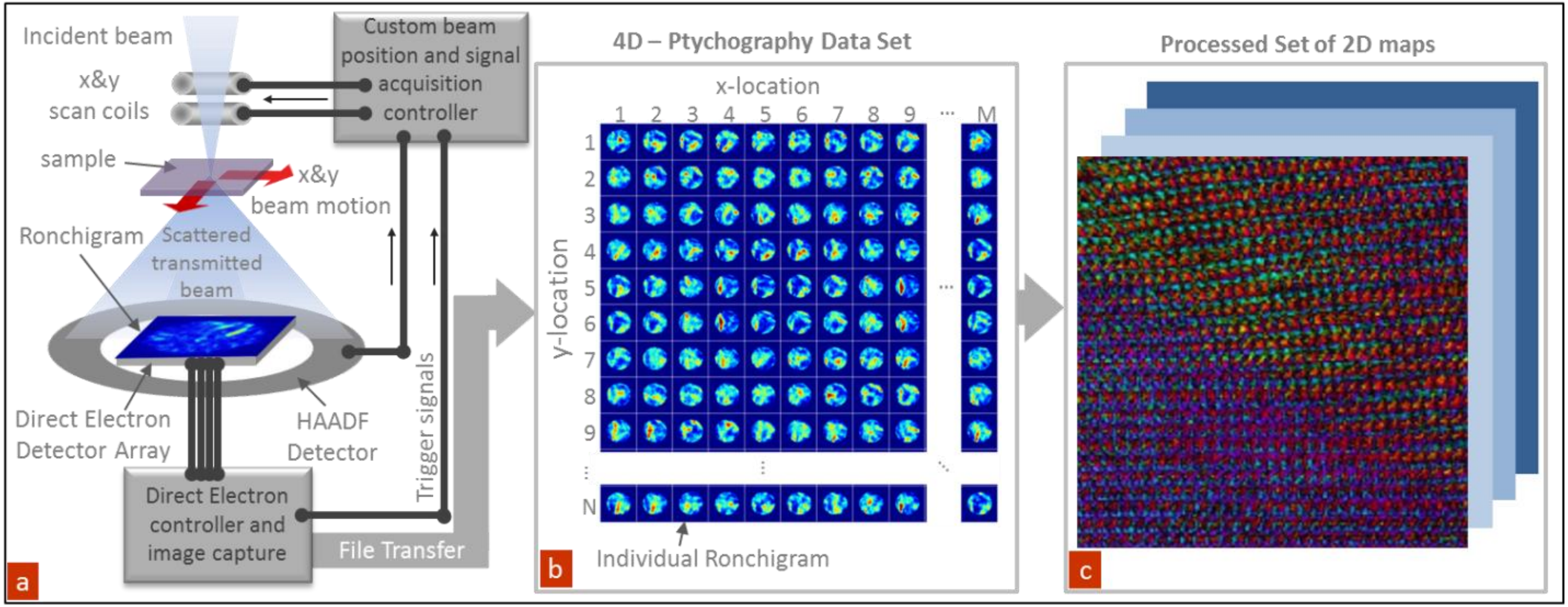
**Valleti, S. M. P.**; Ignatans, R.; Kalinin, S. V.; Tileli, V., Decoding the Mechanisms of Phase Transitions from In Situ Microscopy Observations. *Small* ***2022, 18 (40), 2104318.***

# PCA on 4D STEM



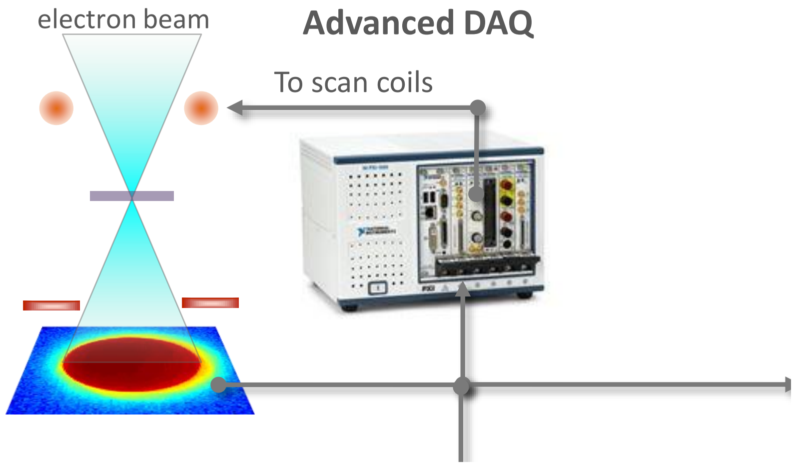

A.R. Lupini, M,P. Oxley, S,V. Kalinin, Pushing the limits of electron ptychography, Science 362, 399 (2018)

# 4D STEM Data

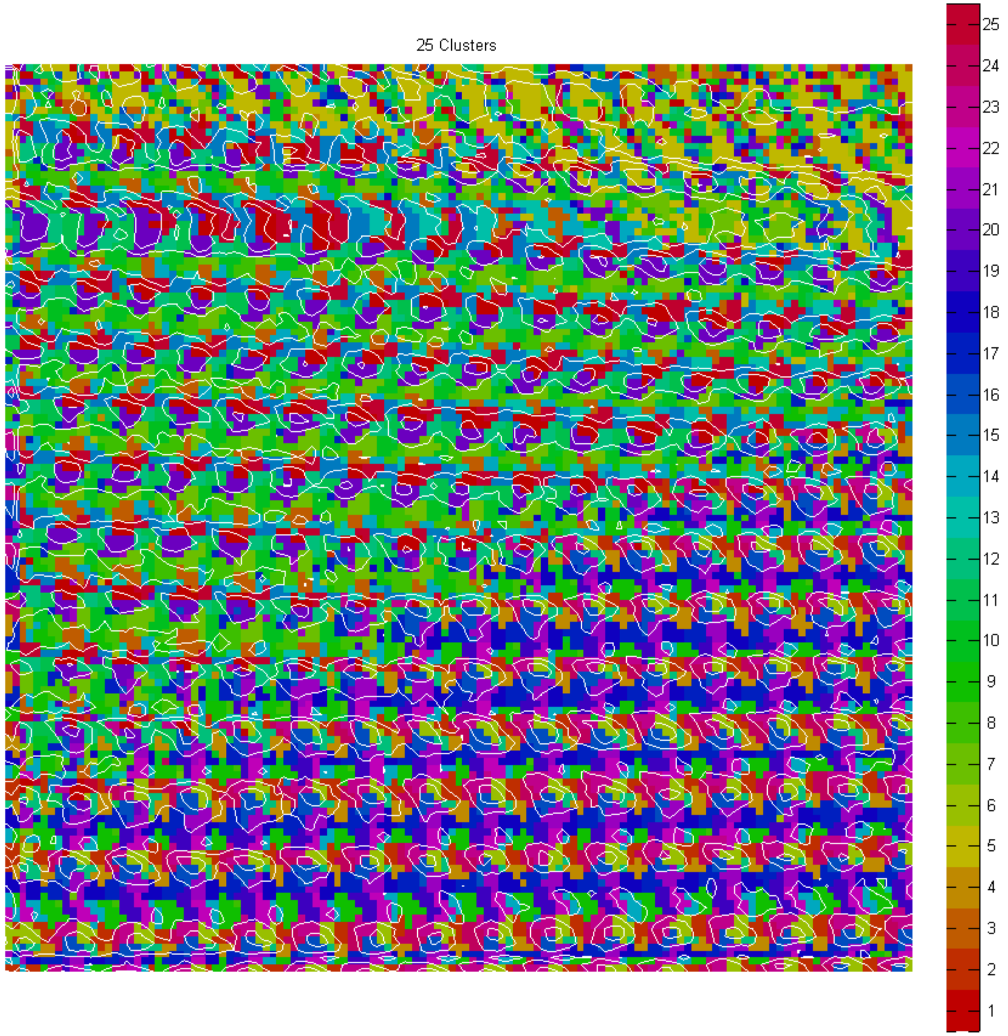



S. JESSE, M. CHI, A. BELIANINOV, C. BEEKMAN, S.V. KALININ, A.Y. BORISEVICH, and A.R. LUPINI, *Big Data Analytics for Scanning Transmission Electron Microscopy Ptychography*, Sci. Rep. **6**, 26348 (2016).

# Multivariate analysis of 4D STEM



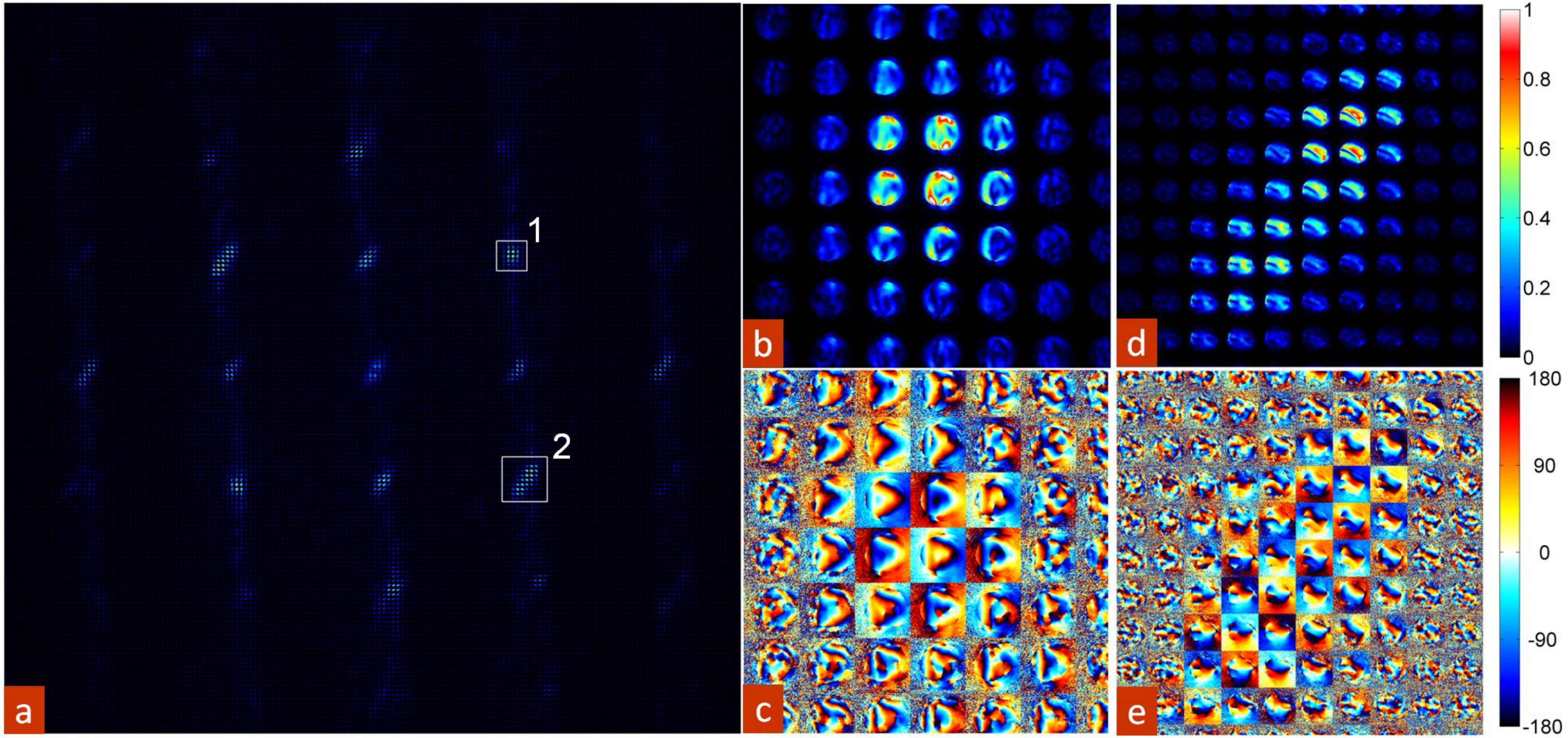

**Bismuth Ferrite Domain Boundary**




- Sub-Å imaging + Direct Electron Detection + Big data analysis enables effective STEM-ptychography.
- Next step: recover physical Information including strain, internal fields, orientation etc.

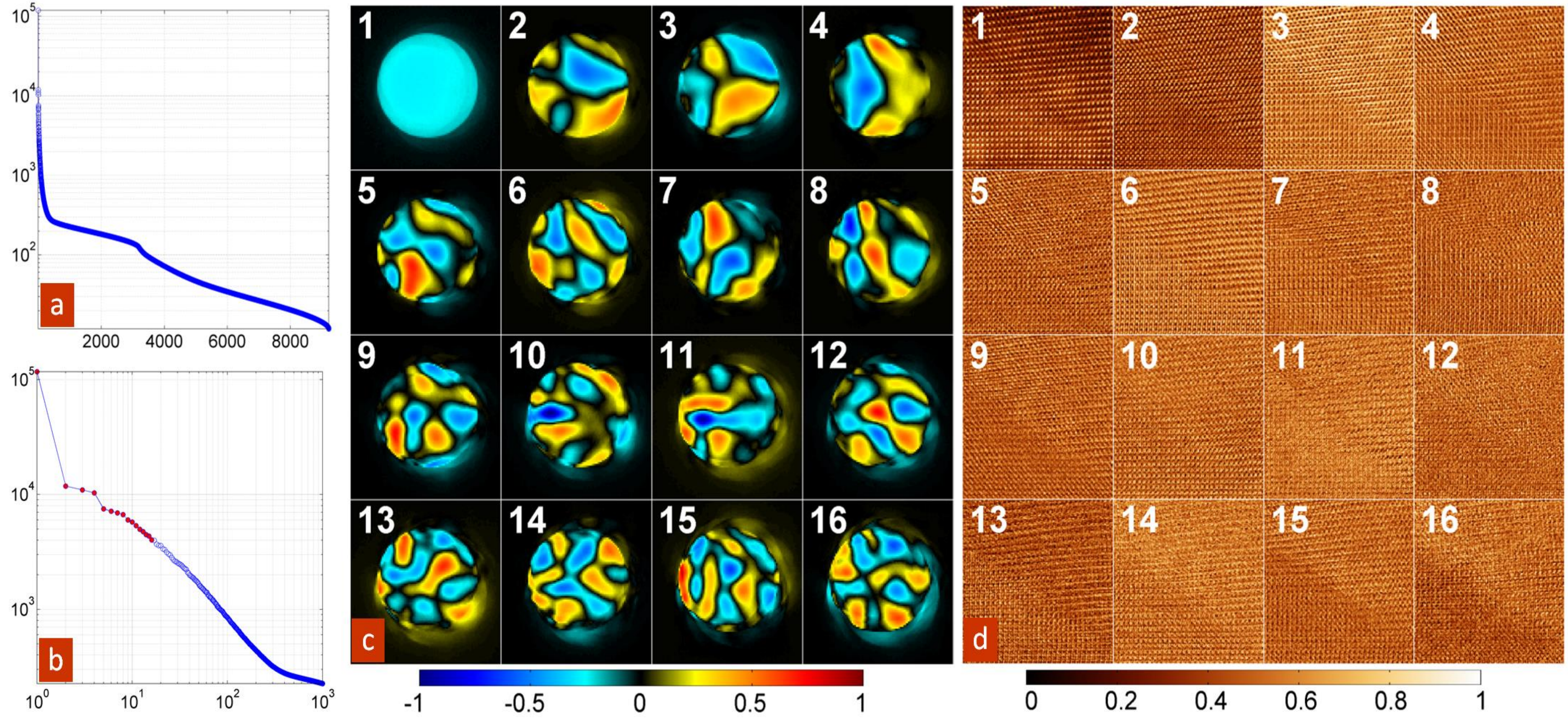
# FFT of 4D STEM






S. Jesse, M. Chi, A. Belianinov, C. Beekman, S.V. Kalinin, A.Y. Borisevich, and A.R. Lupini, *Big Data Analytics for Scanning Transmission Electron Microscopy Ptychography*, Sci. Rep. **6**, 26348 (2016).

# PCA on 4D STEM Data




S. JESSE, M. CHI, A. BELIANINOV, C. BEEKMAN, S.V. KALININ, A.Y. BORISEVICH, and A.R. LUPINI, *Big Data Analytics for Scanning Transmission Electron Microscopy Ptychography*, Sci. Rep. **6**, 26348 (2016).