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Machine Learning Techniques in Microscopic Characterization of Nanomaterials

Krakow, Poland 10/05/2023

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**University founded in 1364
by king Casimir the Great**



Nicolaus Copernicus (1491-1495)



New Campus for 650 anniversary of JU



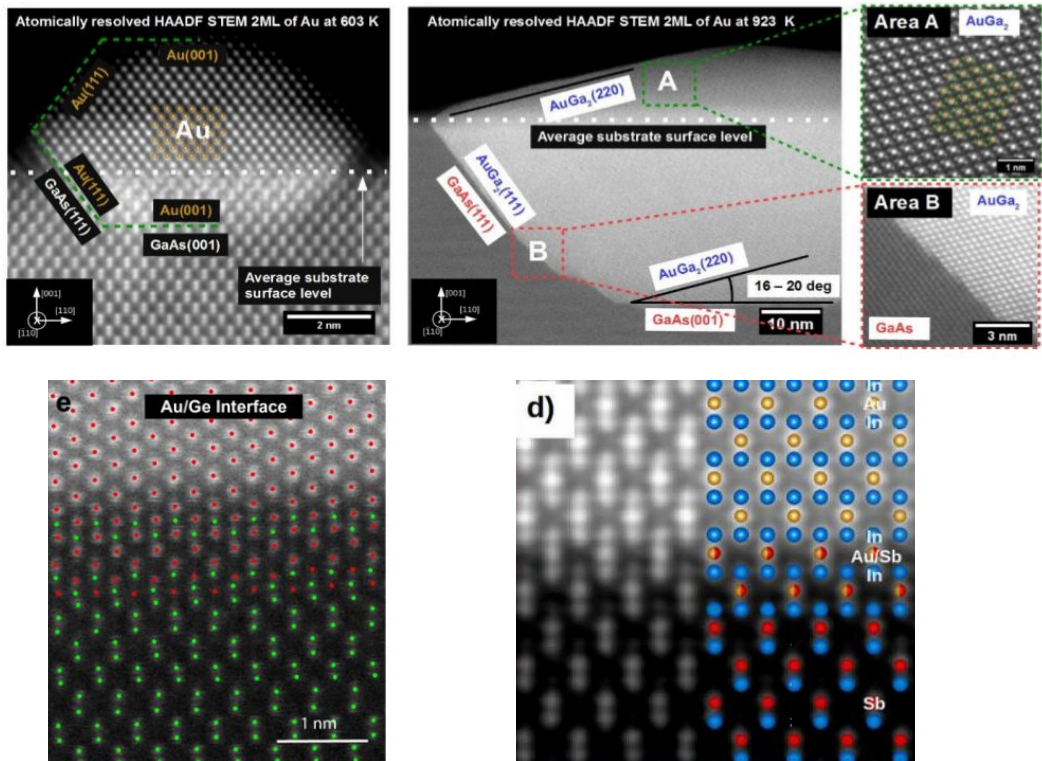
SOLARIS

NATIONAL SYNCHROTRON
RADIATION CENTRE

Institute of Physics

23 departments

Metal/Semiconductor systems at atomic scale



Double Beam SEM/FIB Quanta 3D FEG by FEI

- G.L. Szabo et al., *Small* 2023, 2207263 (2023)
- B.R. Jany et al., *Nanoscale* 12, 9067-9081 (2020)
- A. Janas et al., *Appl. Surf. Scie.*, Vol. 492, 703-710 (2019)
- B.R. Jany et al., *Nano Letters*, Vol. 17, Issue 11, 6507-7170 (2017)
- B.R. Jany et al., *Scientific Reports* 7, 42420 (2017)
- D. Wrana et al., *Nanoscale* 11, 89-97 (2019)

Studies of the light-matter interaction at micro/nano scale on the surface manifested as a color appearance.

The Colorimetric Microscopy and Machine Learning Group

Colorimetric Microscopy (C-Microscopy) coupled with different methods of surface characterization like Scanning Electron Microscopy (SEM) Electron Backscatter Diffraction (EBSD) and Atomic Force Microscopy (AFM).

Machine Learning and Data Analysis in Microscopy

- Automatic microscopic image analysis
- Hyperspectral Reflectance (R) data classification
- Machine Learning based HAADF STEM image quantification
- SEM EBSD 4D data analysis together with SEM EBSD Dictionary Based Indexing DI
- SEM EDX hyperspectral data analysis at nanoscale
- Atomically Resolved HAADF STEM Image Deconvolution
- Random Nucleation Modelling

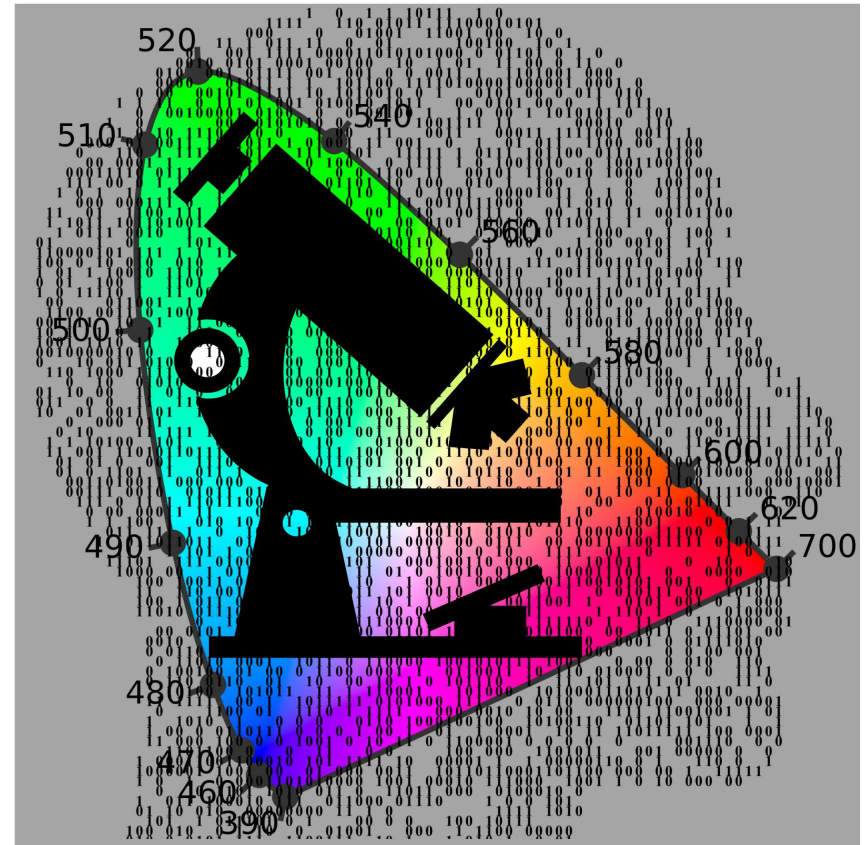
E. Cihan, *Rev. Lett.* **130**, 126205 (2023)

A. Janas et al., *Appl. Surf. Scie.* Volume **570**, 150958 (2021)

B.R. Jany et al., *Nanoscale* **12**, 9067-9081 (2020)

B.R. Jany et al., *Micron*, Vol. **130**, 102800 (2020)

B.R. Jany et al., *Nano Letters*, Vol. **17**, Issue **11**, 6507-7170 (2017)



<http://www.zfcs.if.uj.edu.pl/c-microscopy-ml>

- What is Machine Learning?
- Selected Machine Learning Techniques for:
 - SEM EDX Quantification at Nanoscale
 - Automatic Microscopic Image Analysis
 - Conductive-AFM (C-AFM) Data Clustering

Machine Learning



Algorithms Learning from Data

Statistics



Formation of Relations between Variables

scikit-learn
algorithm cheat-sheet

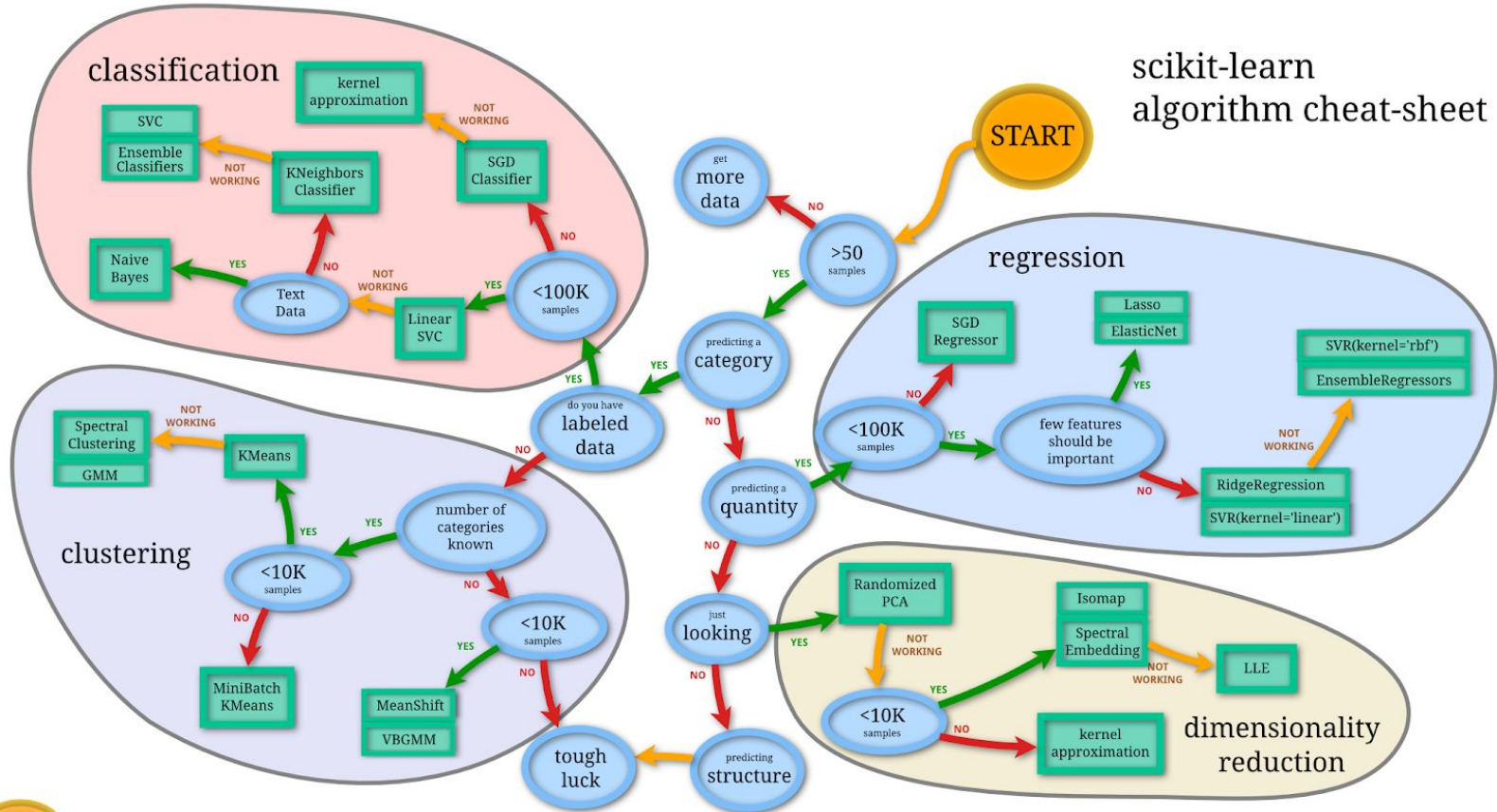
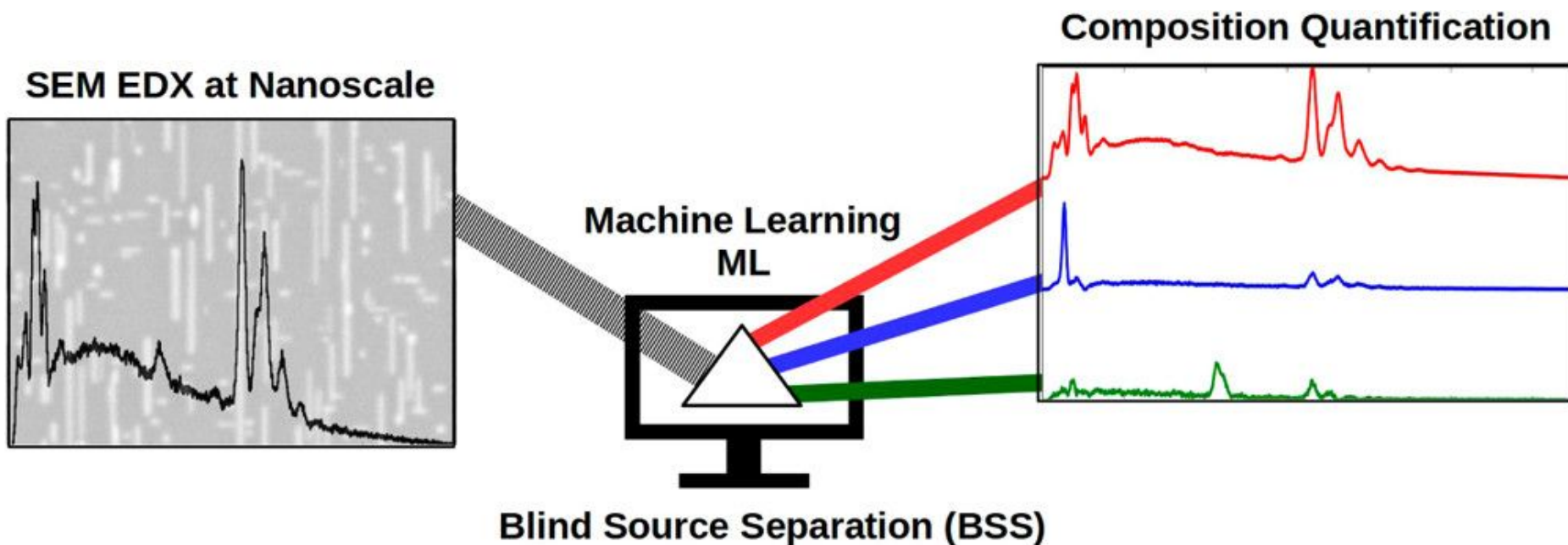


image source: scikit-learn.org

Linear Regression
K-Means Clustering (1967)

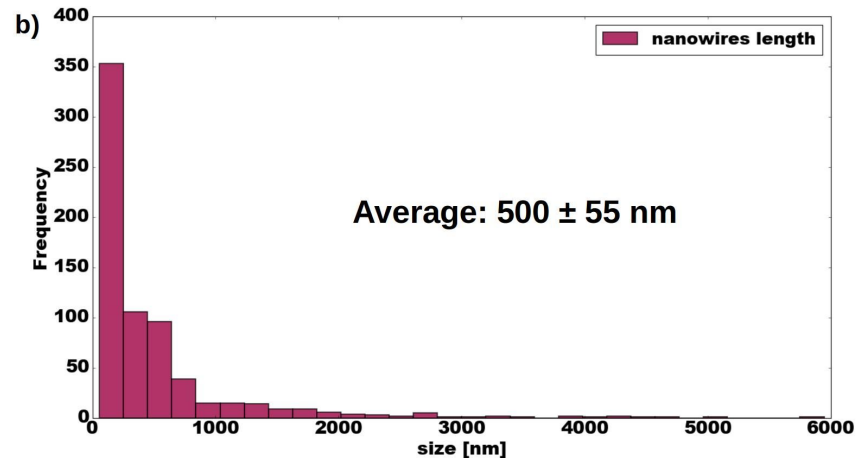
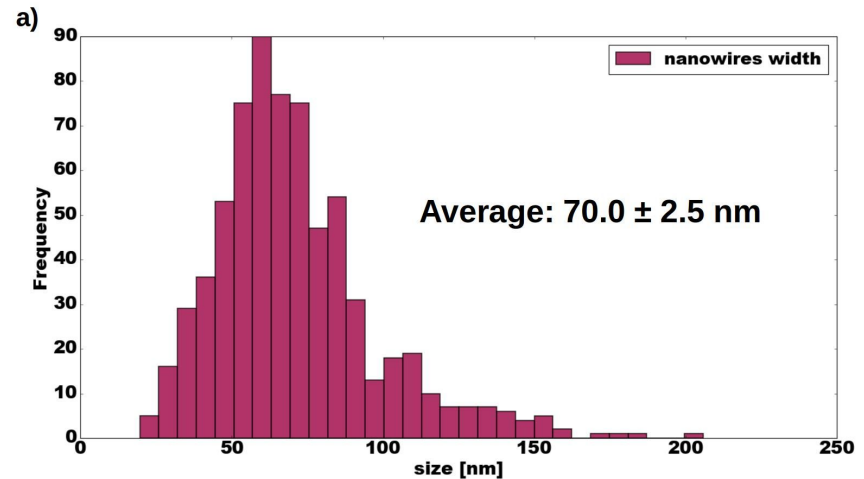
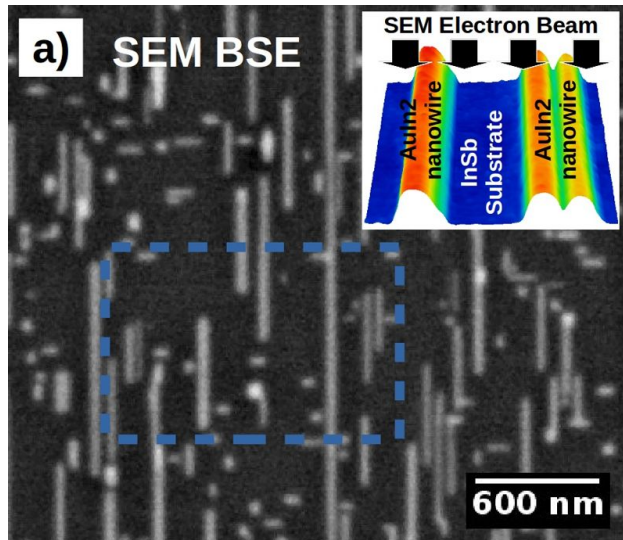
Machine Learning supported SEM EDX Quantitative Chemical Composition Determination of Nanostructures



B.R. Jany et al., Nano Lett. 2017, 17, 11, 6520-6525

SEM EDX Experimental data: <http://dx.doi.org/10.13140/RG.2.2.19558.93763>

2ML Au on InSb(001) surface → Au/In nanowires on InSb(001)



What is the chemical composition of nanowires?

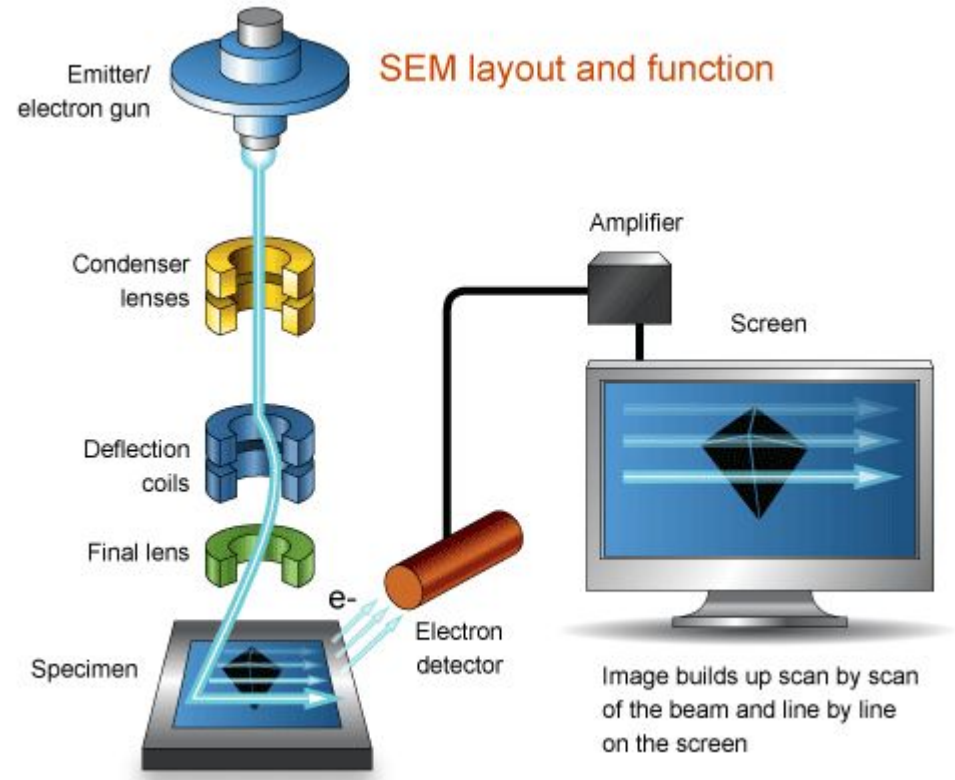
→ TEM (expensive, time consuming)

→ SEM (common, accessible, cheap)?

High Resolution Scanning Electron Microscopy (SEM)

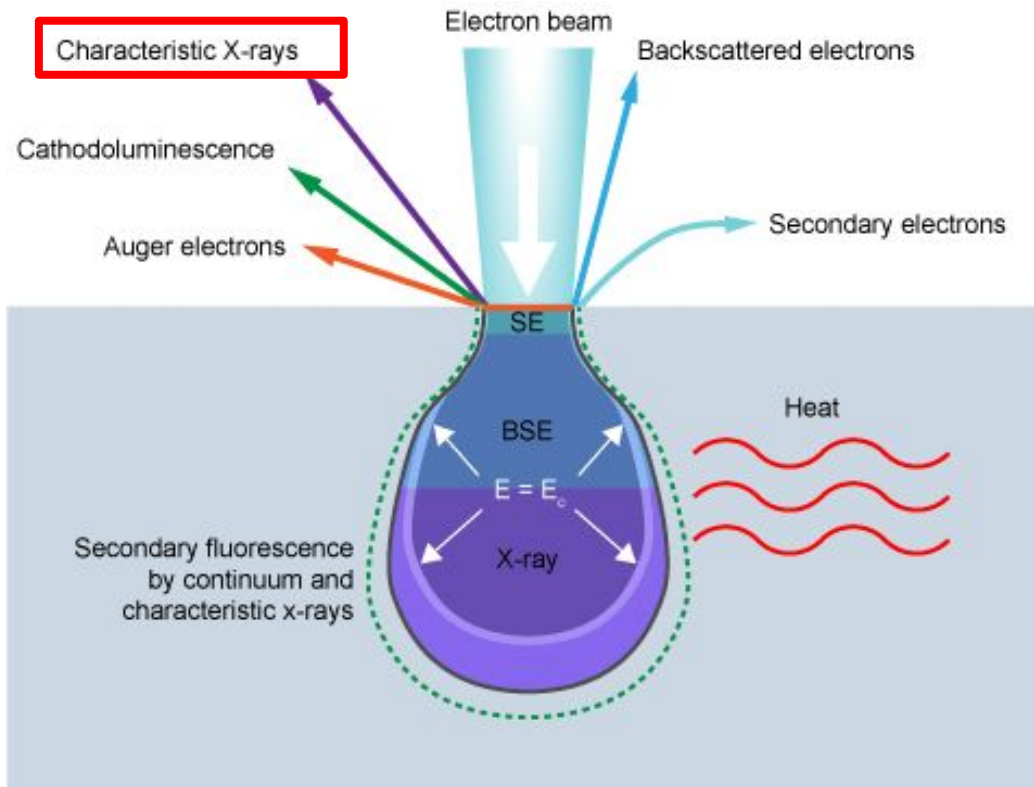


Double Beam SEM/FIB Quanta 3D FEG by FEI



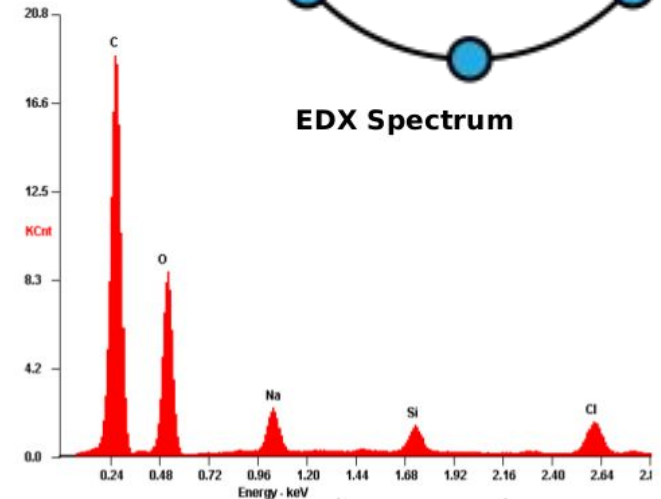
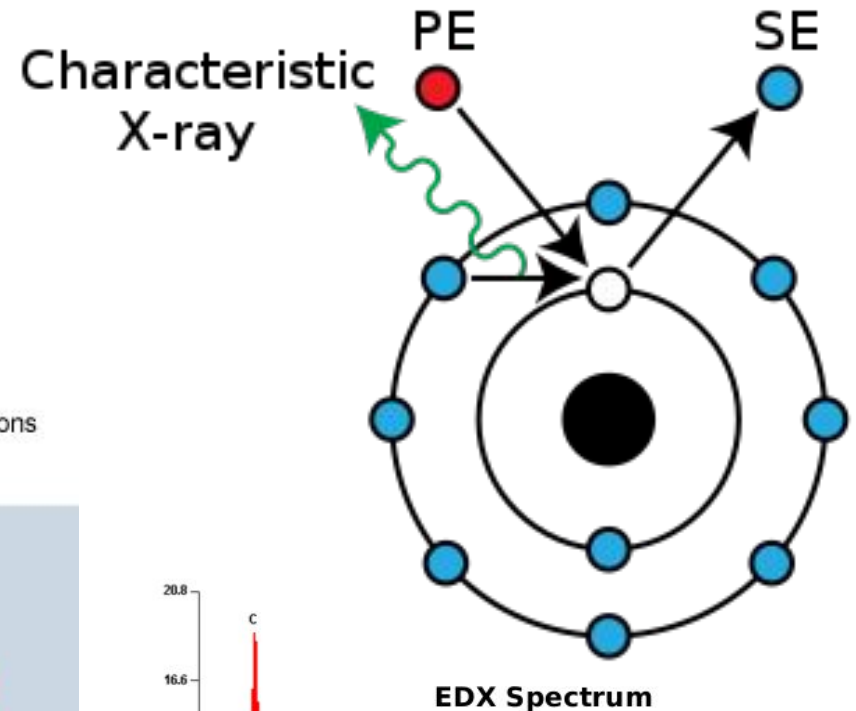
source: ammrf.org.au/myscope/sem/

Emission of Characteristic X-rays



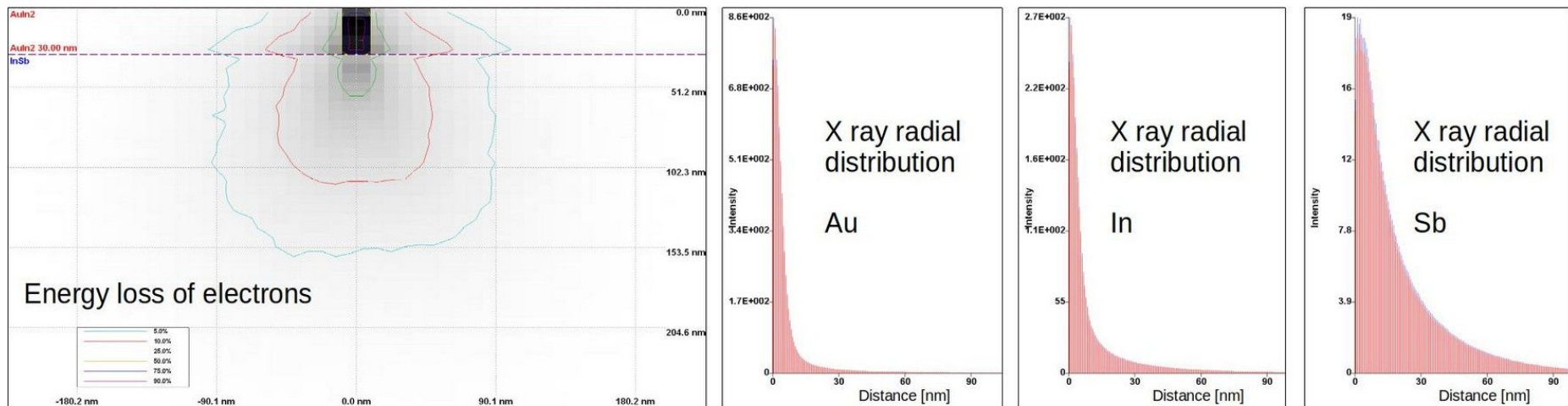
Resolution Limited by Interaction Volume size

source: ammrf.org.au/myscope/sem/



Measurements Optimization by CASINO Monte-Carlo Simulation

CASINO Monte-Carlo Simulation of AuIn2 nanowires on InSb @6.5keV



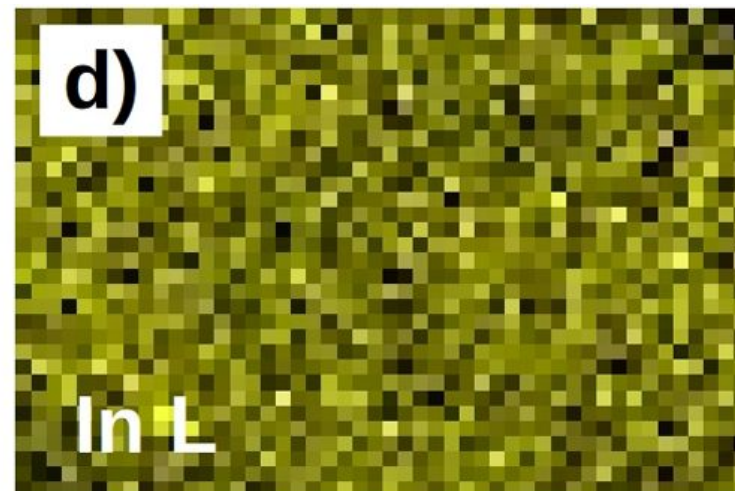
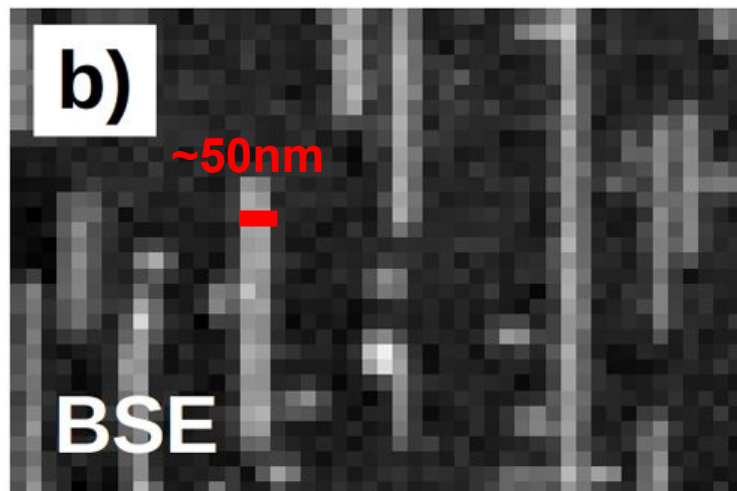
Acquisition of high spatial resolution X-ray maps at nanoscale

Goldstein, J. I.; Newbury, D. E. Scanning Electron Microscopy and X-ray Microanalysis; Springer: New York, 2003

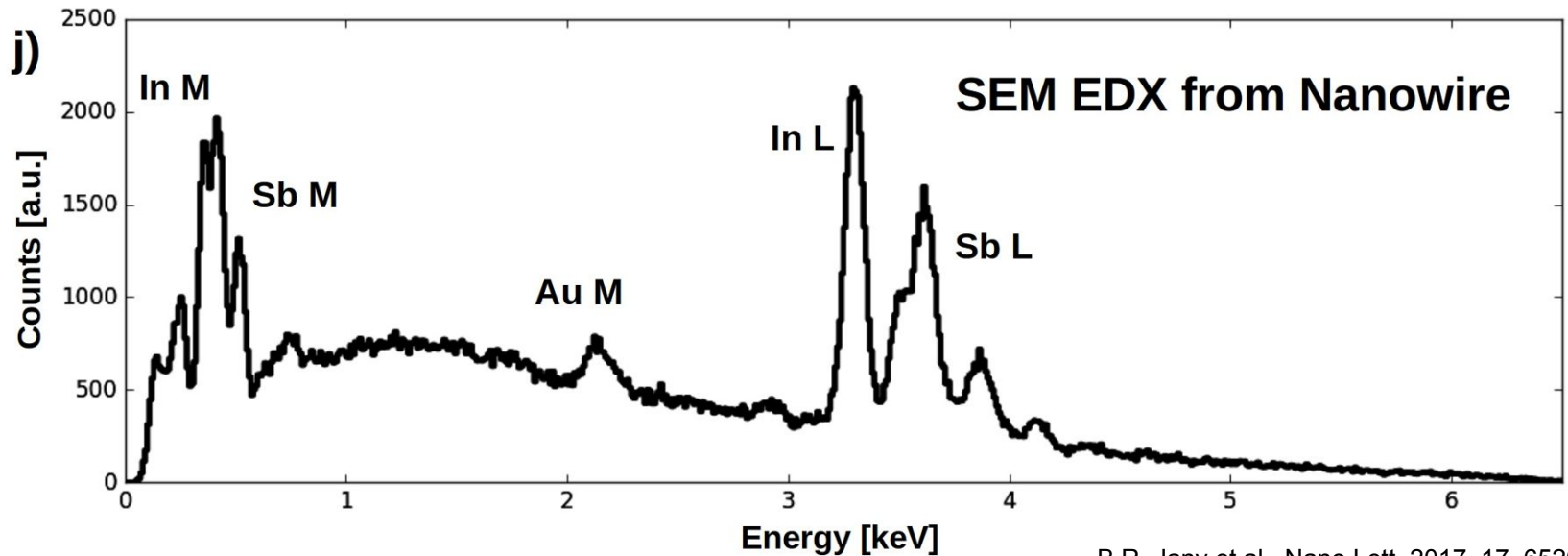
Rafaelson, J.; Nylese, T. High spatial resolution in X-ray mapping Microscopy and Analysis 2015, 29, 20– 24

B.R. Jany et al., Nano Lett. 2017, 17, 6520-6525

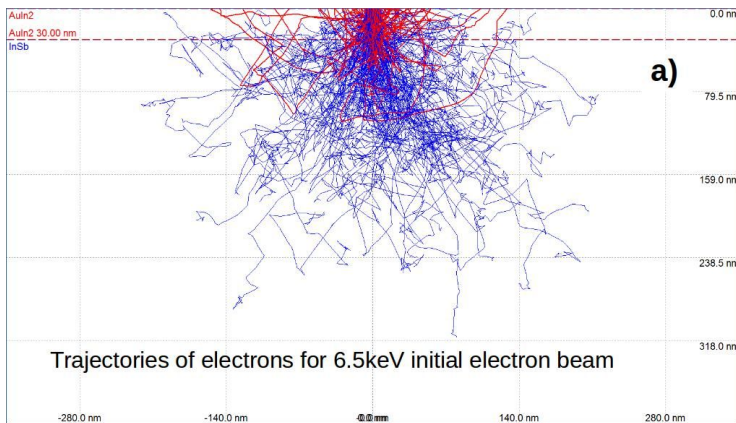
Au/In nanowires on InSb(001)



Au/In nanowires on InSb(001)



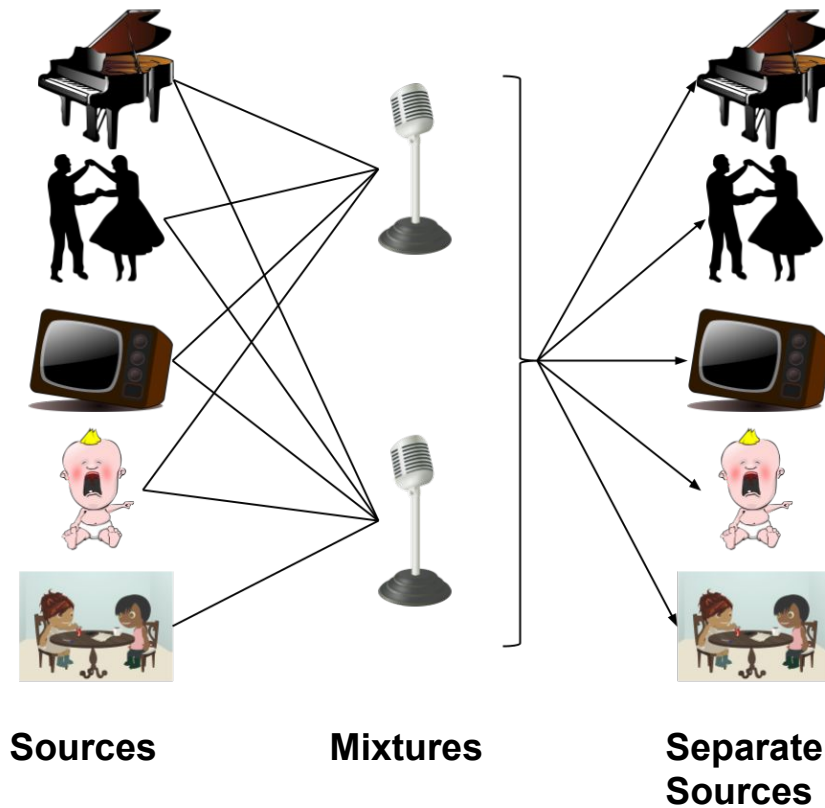
B.R. Jany et al., Nano Lett. 2017, 17, 6520-6525



How to perform quantification?

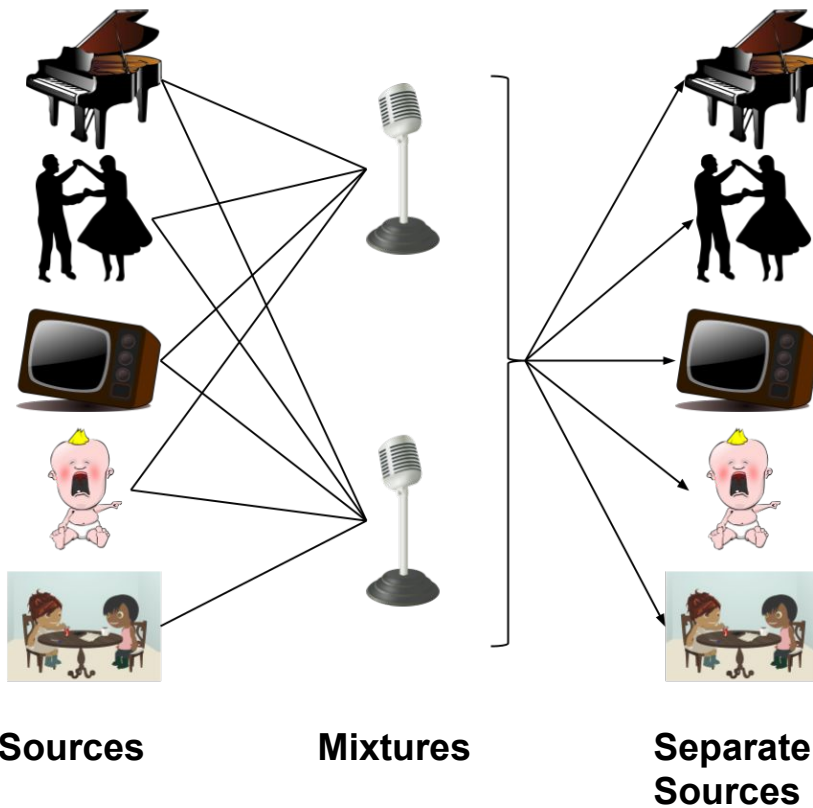
**Signal from Au/In wires is mixed with
signal from InSb substrate?**

Umixing signal by Machine Learning Blind Source Separation (BSS) using Non-negative Matrix Factorization (NMF)



The cocktail party problem - BSS problem

Umixing signal by Machine Learning Blind Source Separation (BSS) using Non-negative Matrix Factorization (NMF)



Possible Solution to the BSS problem using NMF

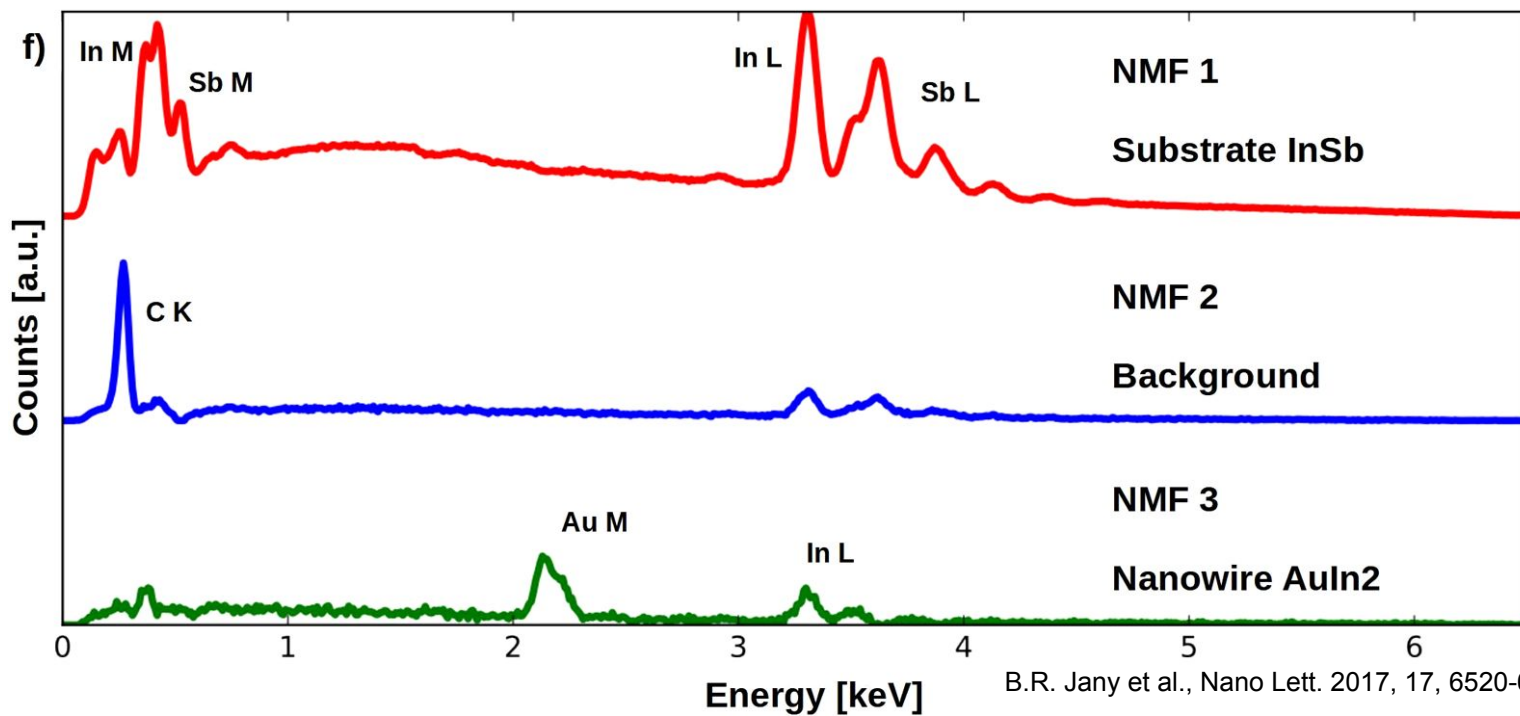
$$\begin{matrix} W \\ \begin{bmatrix} \square & \square & \square \\ \square & \square & \square \\ \square & \square & \square \end{bmatrix} \end{matrix} \times \begin{matrix} H \\ \begin{bmatrix} \square & \square & \square & \square & \square & \square \end{bmatrix} \end{matrix} \approx \begin{matrix} V \\ \begin{bmatrix} \square & \square & \square & \square & \square & \square \\ \square & \square & \square & \square & \square & \square \\ \square & \square & \square & \square & \square & \square \end{bmatrix} \end{matrix}$$

$$d_{\text{Fro}}(X, Y) = \frac{1}{2} \|X - Y\|_{\text{Fro}}^2 = \frac{1}{2} \sum_{i,j} (X_{ij} - Y_{ij})^2$$

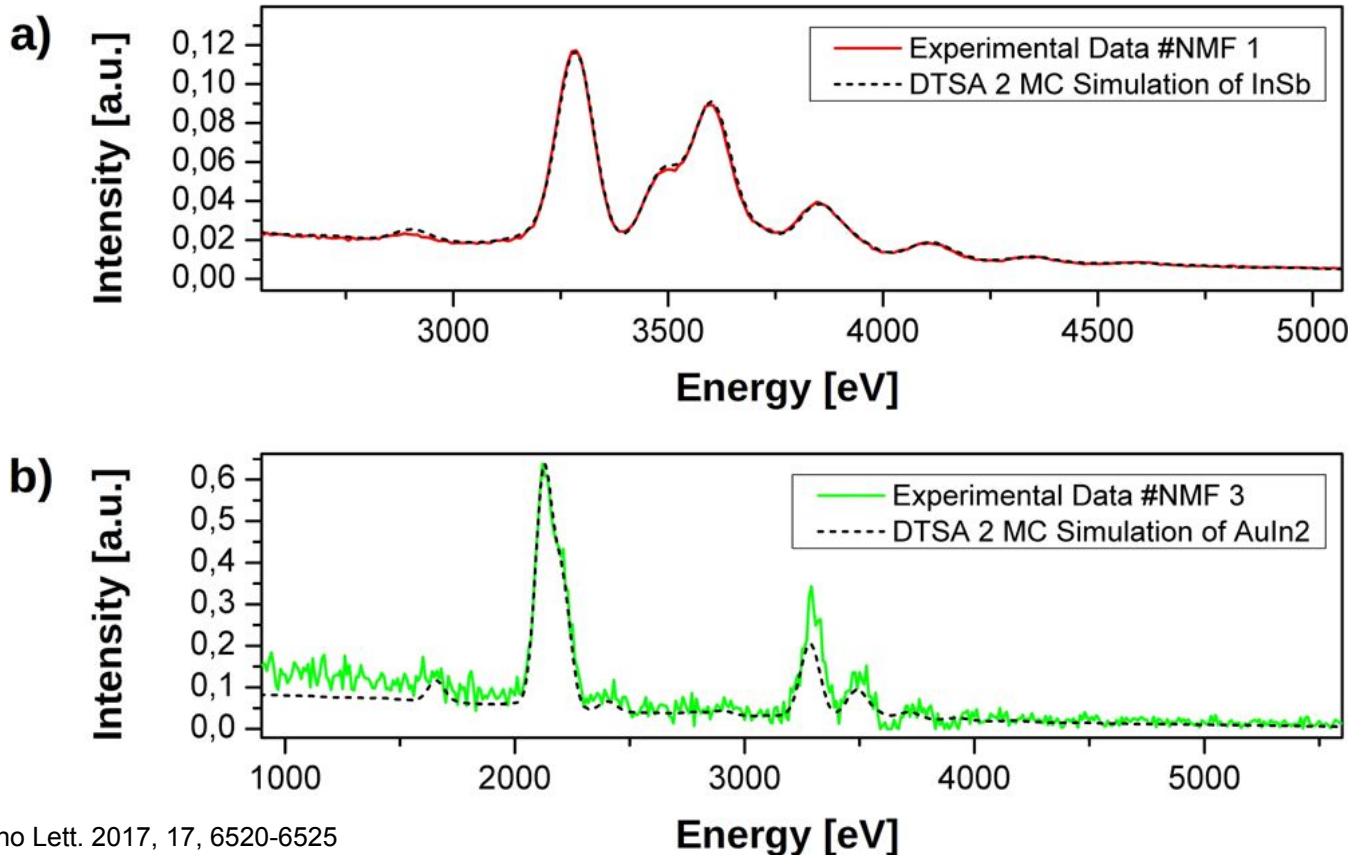
The cocktail party problem - BSS problem

optimization of the squared Frobenius norm
using Hyperspy Framework

Umixing signal by Machine Learning Blind Source Separation (BSS) using Non Negative Matrix Factorization (NMF)



Umixed Signal Quantification by EDX ZAF



B.R. Jany et al., Nano Lett. 2017, 17, 6520-6525

EDX ZAF quantification [atomic %]
true composition [atomic %]

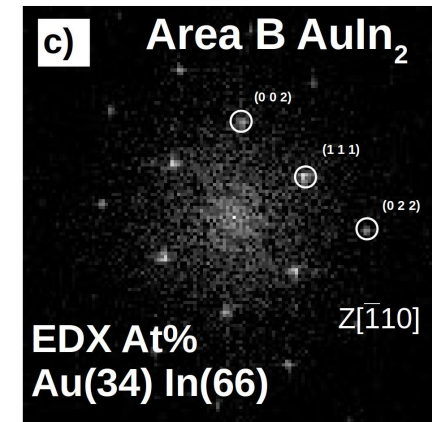
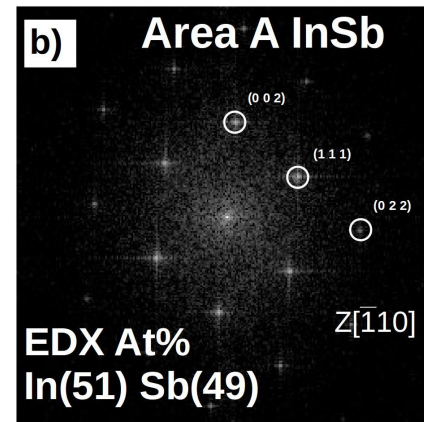
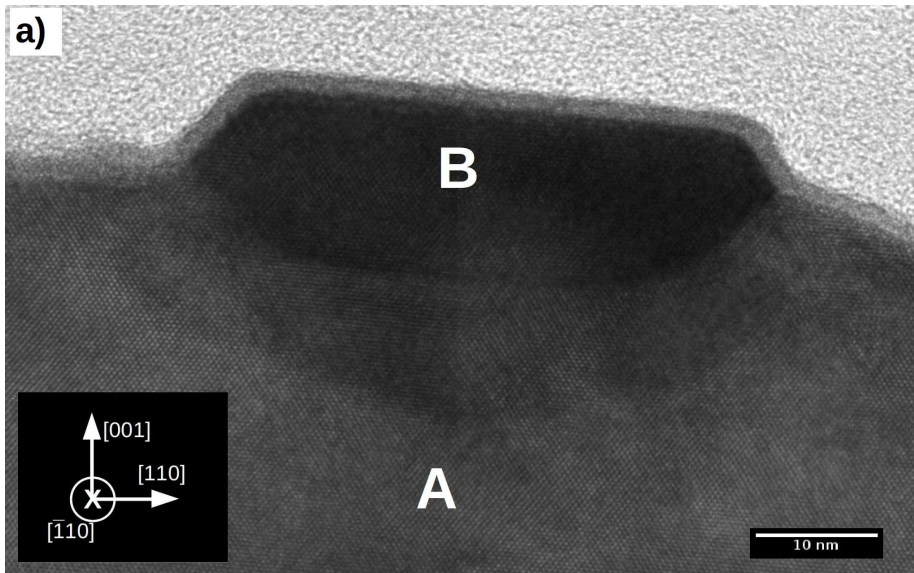
NMF 1 (InSb substrate)

In: 45.0 (4.7); Sb: 55.0 (5.7)
In: 50.00; Sb: 50.00

NMF 3 (AuIn₂ nanowires)

Au: 38.0 (5.9); In: 62.0 (5.9)
Au: 33.33; In: 66.67

Verification by TEM EDX and Diffraction measurements

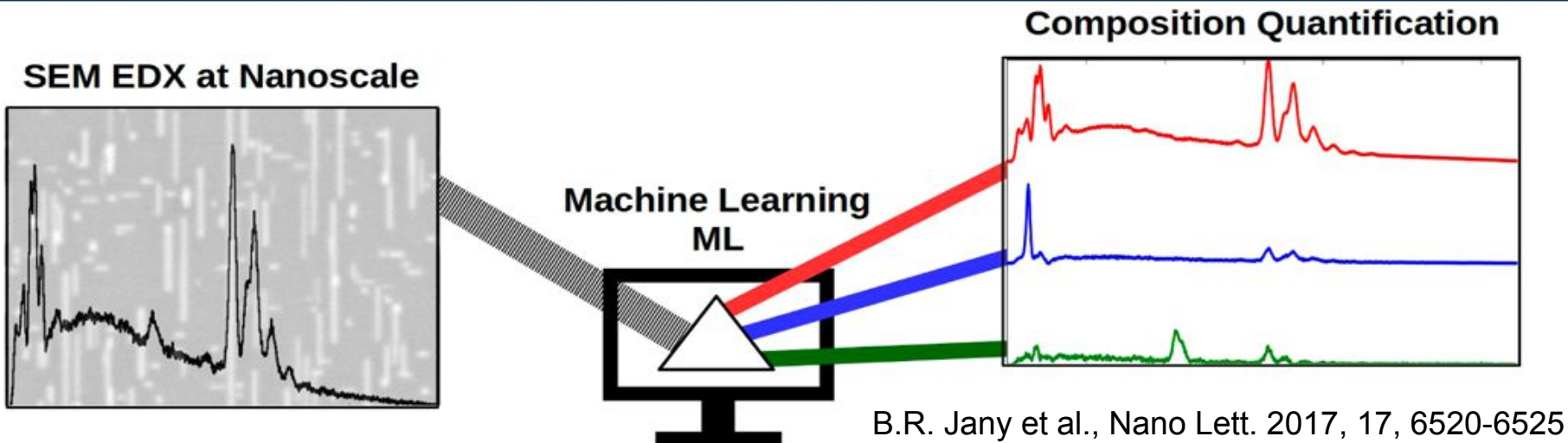


Composition Verified!

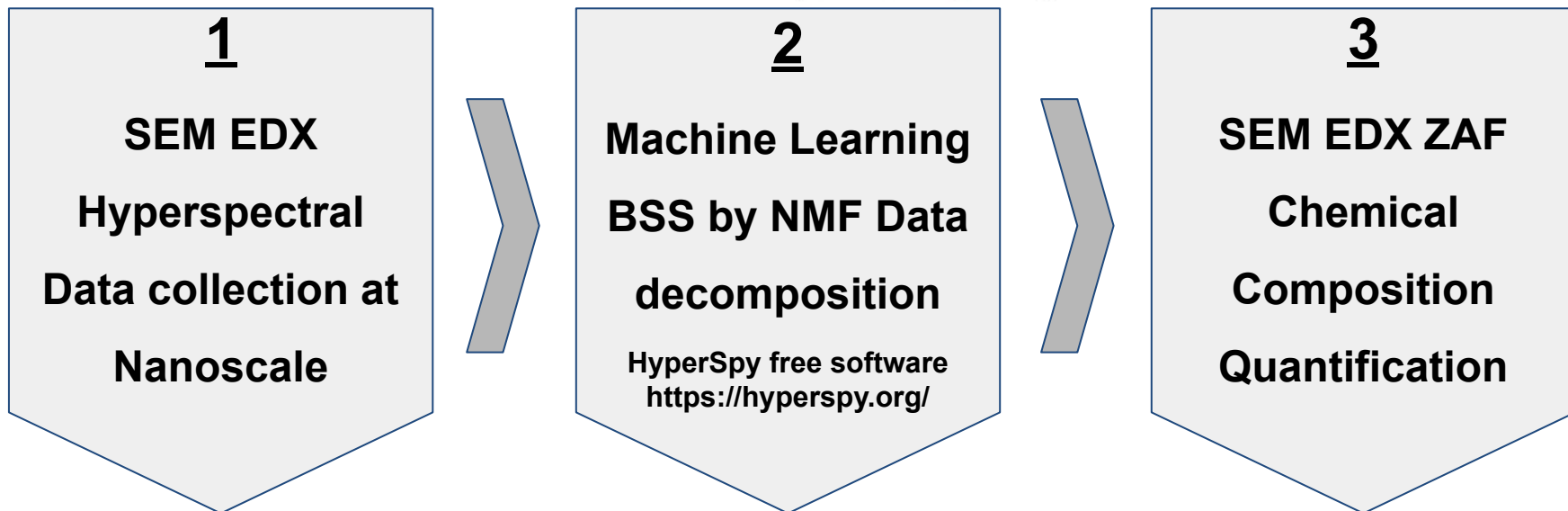
The BSS method by NMF successfully decomposed the Au/In nanowires signal from background, this allows simple and cheap Chemical Composition Quantification by SEM EDX at Nanoscale

B.R. Jany et al., Nano Lett. 2017, 17, 6520-6525

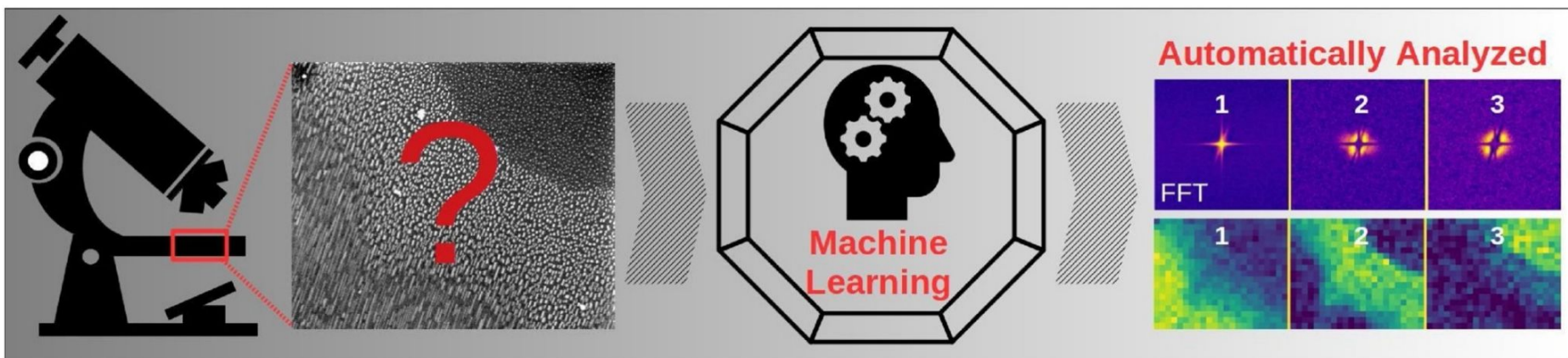
SEM EDX Quantification at Nanoscale The Method



Blind Source Separation (BSS)



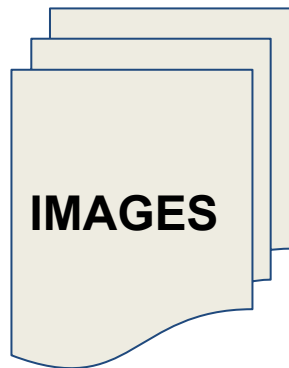
Automatic Microscopic Image Analysis by Machine Learning



B.R. Jany et al., *Micron*, Vol. 130, 102800 (2020)

Mendeley Data
<https://doi.org/10.17632/25x46xjyr5.2>

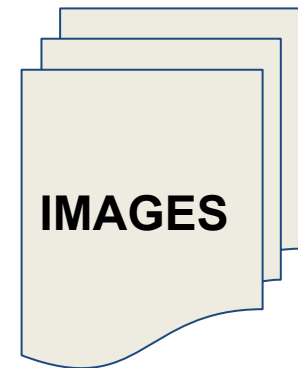
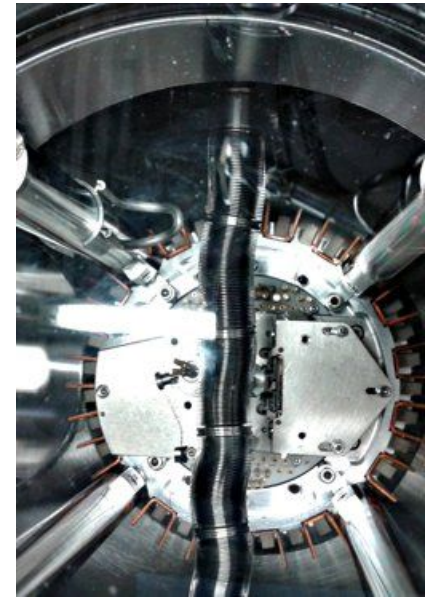
SEM/FIB Microscope

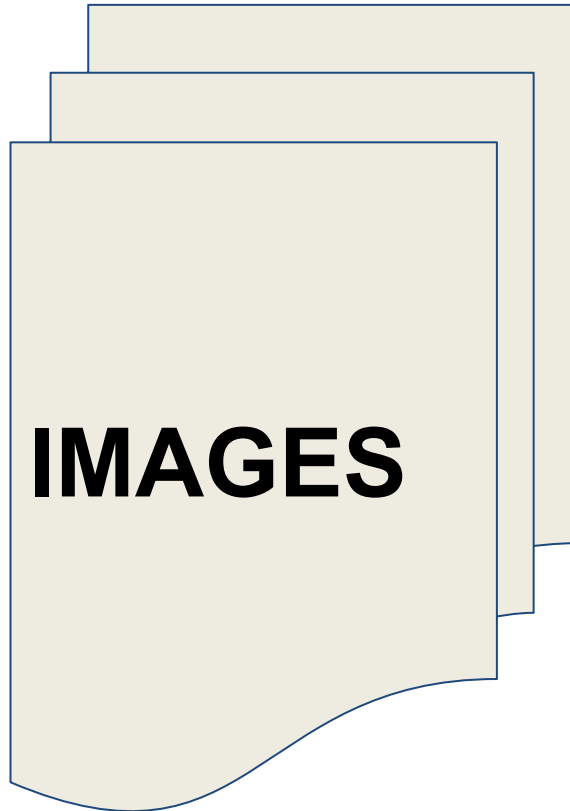


STEM/TEM Microscope



AFM/STM Microscope



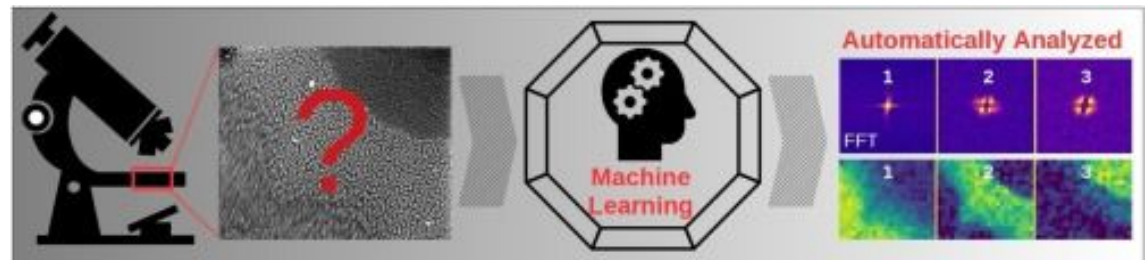


→ **Is there something interesting in the data (IMAGES)?**

→ Is there something interesting in the data (IMAGES)?

IMAGES

Automatic image analysis approach needed



There are many different approaches described but:

- **they are specialized in a very particular image type**
 - **the analysis program is usually not available**
 - **the program uses commercial software**
 - **requires training image data (Deep Learning)**

There are many different approaches described but:

- they are specialized in a very particular image type
 - the analysis program is usually not available
 - the program uses commercial software
 - requires training image data (Deep Learning)

Automatic analysis approach which works for images coming from different microscopes is proposed

The analysis program is written using Python (free)

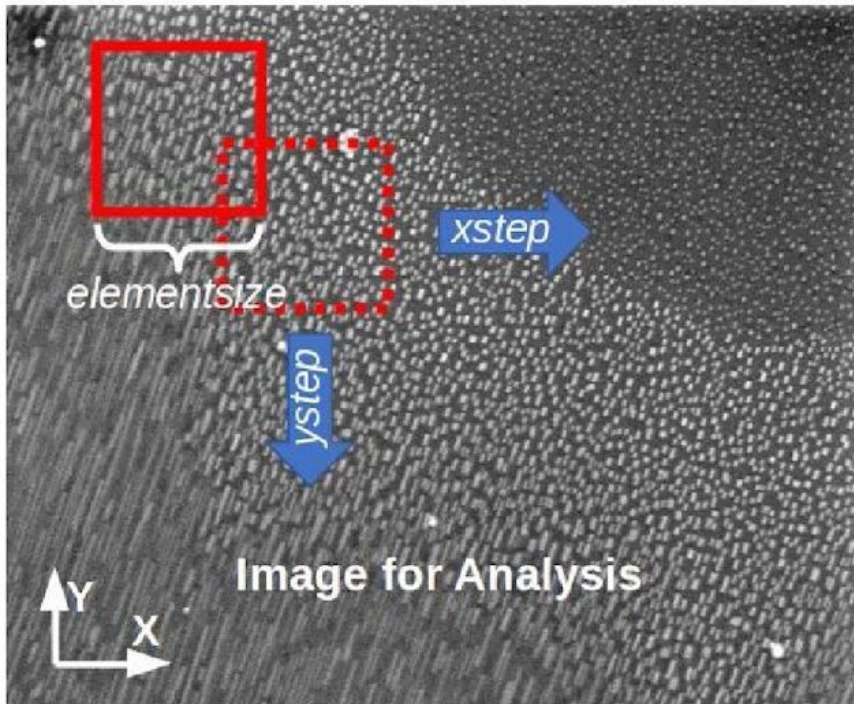


and is freely available from Mendeley Data <https://doi.org/10.17632/25x46xjyr5.2>



Moving Local Window

Selection of Window
Parameters



B.R. Jany et al., Micron, Vol. 130, 102800 (2020)

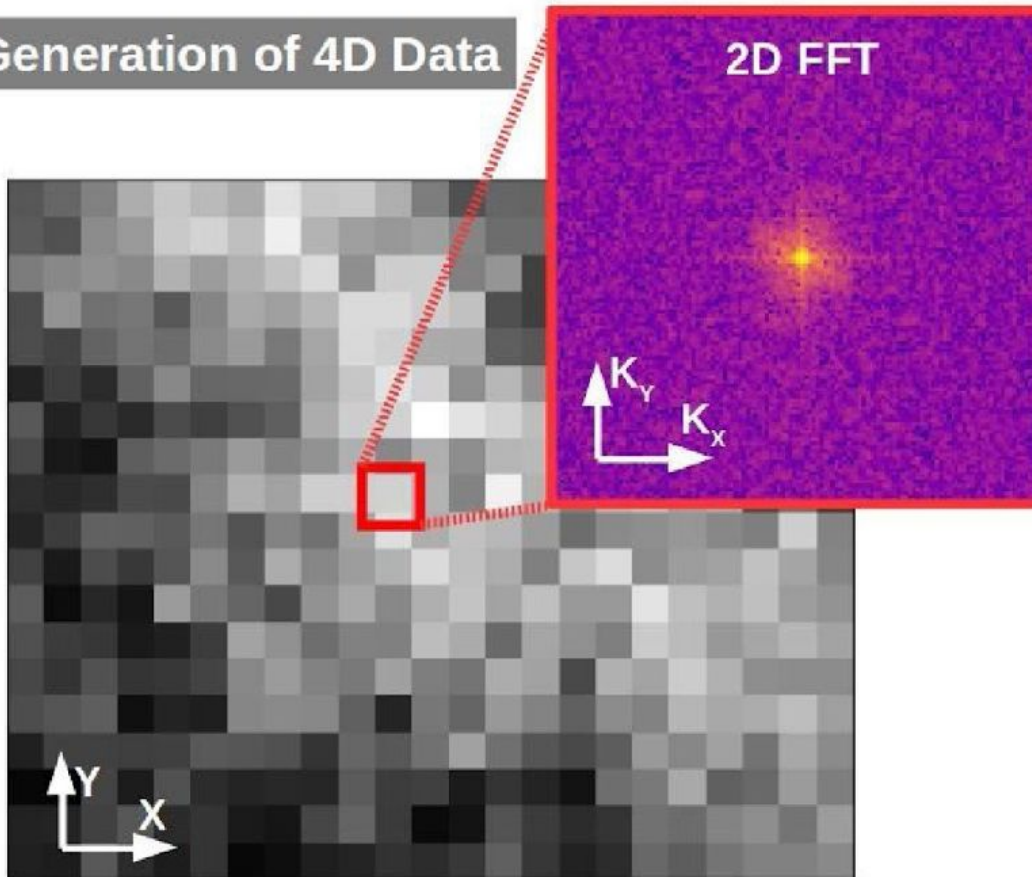
- Local window (ROI) with size *elementsize* (128pixels) is defined in the image
- the window is moved in x, y directions by $xstep$ (64pixels) and $ystep$ (64pixels), whole image is covered

$xstep, ystep$ - defines the resolution
elementsize - defines the local scale

default values are optimal for images of the size of $\sim 2000 \times 2000$ pixels

2D Fourier Transform for each Window

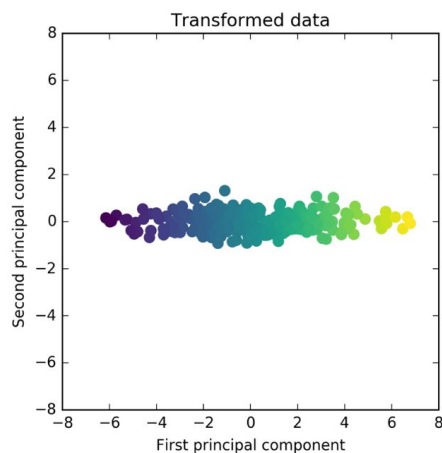
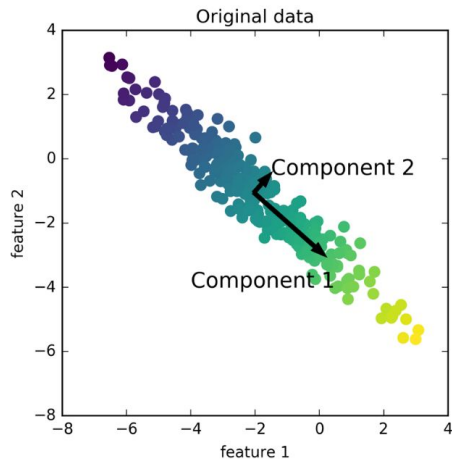
Generation of 4D Data



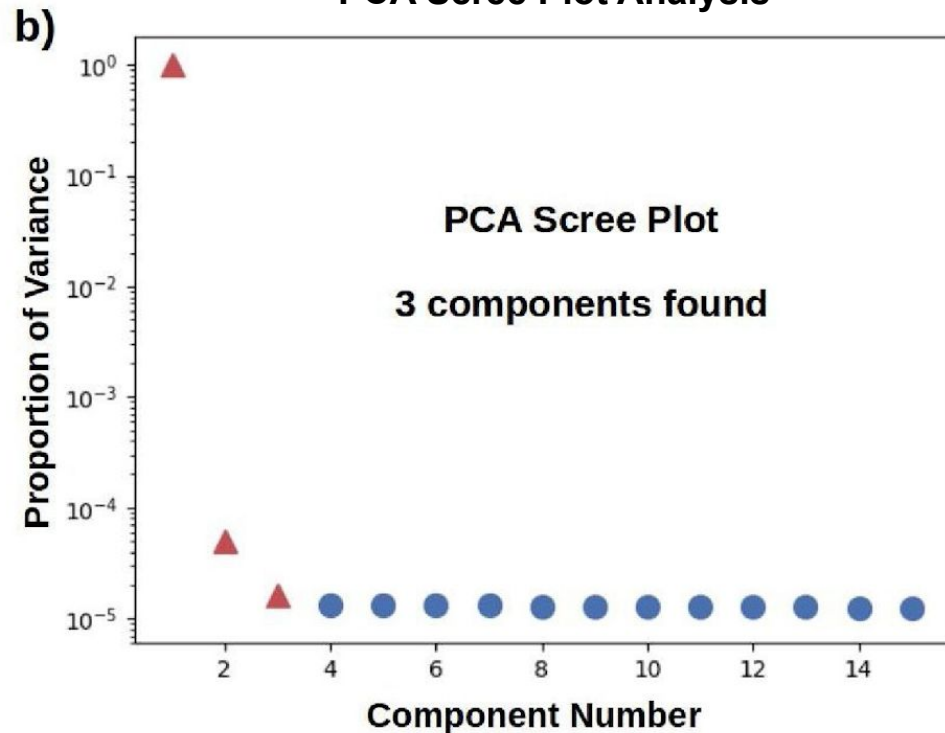
- For each local window (ROI) 2D FFT is calculated
- the single image is “converted” to **4D data** i.e. two spatial dimensions X, Y in real space and two dimension K_x, K_y in reciprocal space

How many components we have in the 4D data?

Principal Component Analysis (PCA)



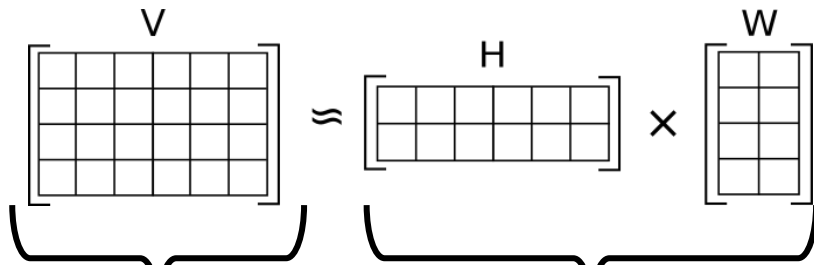
PCA Scree Plot Analysis



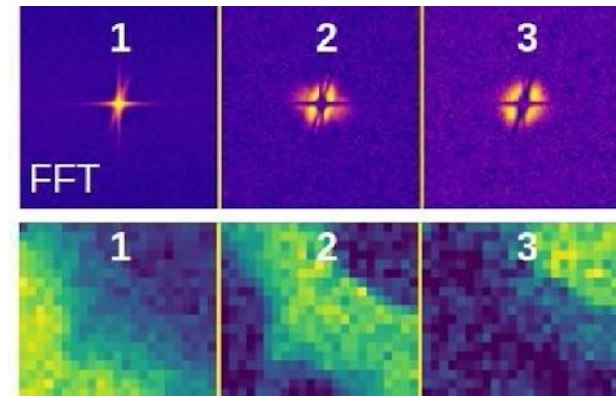
- The number of components in the 4D data is determined from PCA Scree Plot Analysis
- The program has the possibility to automatically analyze scree plot

4D data decomposition by Non-negative Matrix Factorization

data two non-negative
 matrices



- The NMF due to the non-negativity provides parts based representations of the data
- The NMF provides components which have physical meaning and are interpretable



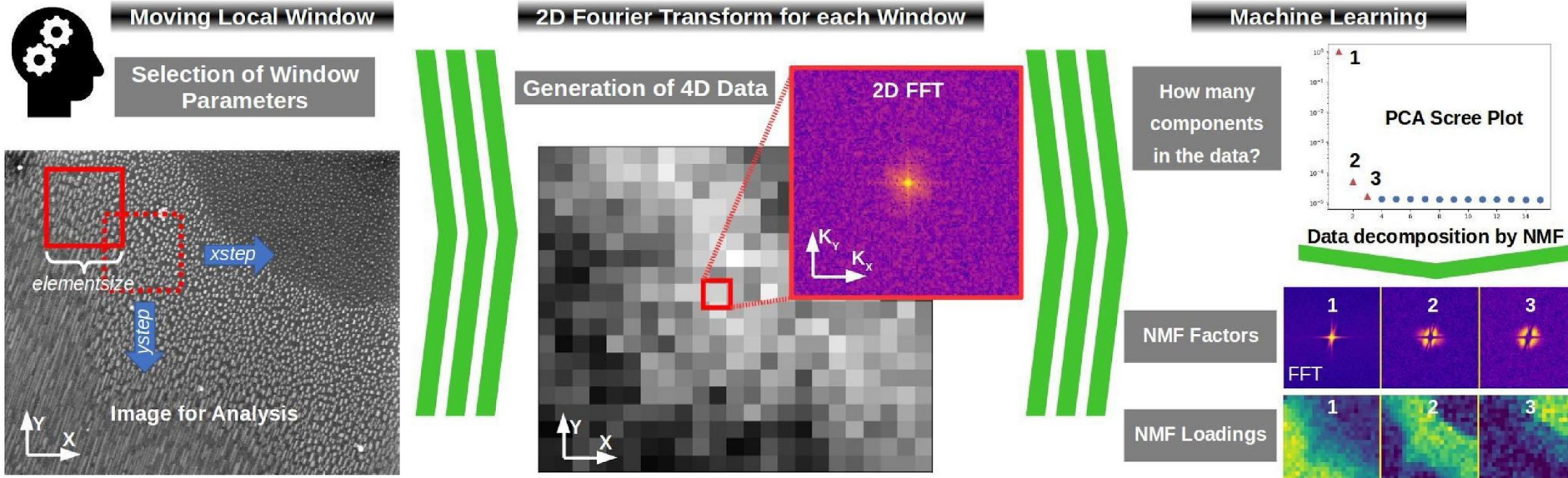
$$d_{\text{Fro}}(X, Y) = \frac{1}{2} \|X - Y\|_{\text{Fro}}^2 = \frac{1}{2} \sum_{i,j} (X_{ij} - Y_{ij})^2$$

optimization of the squared Frobenius norm
euclidean distance for matrices

NMF decomposition outputs interpretable
spatial maps (loadings) and corresponding 2D
FFT Power Spectra (factors)

B.R. Jany et al., Micron, Vol. 130, 102800 (2020)

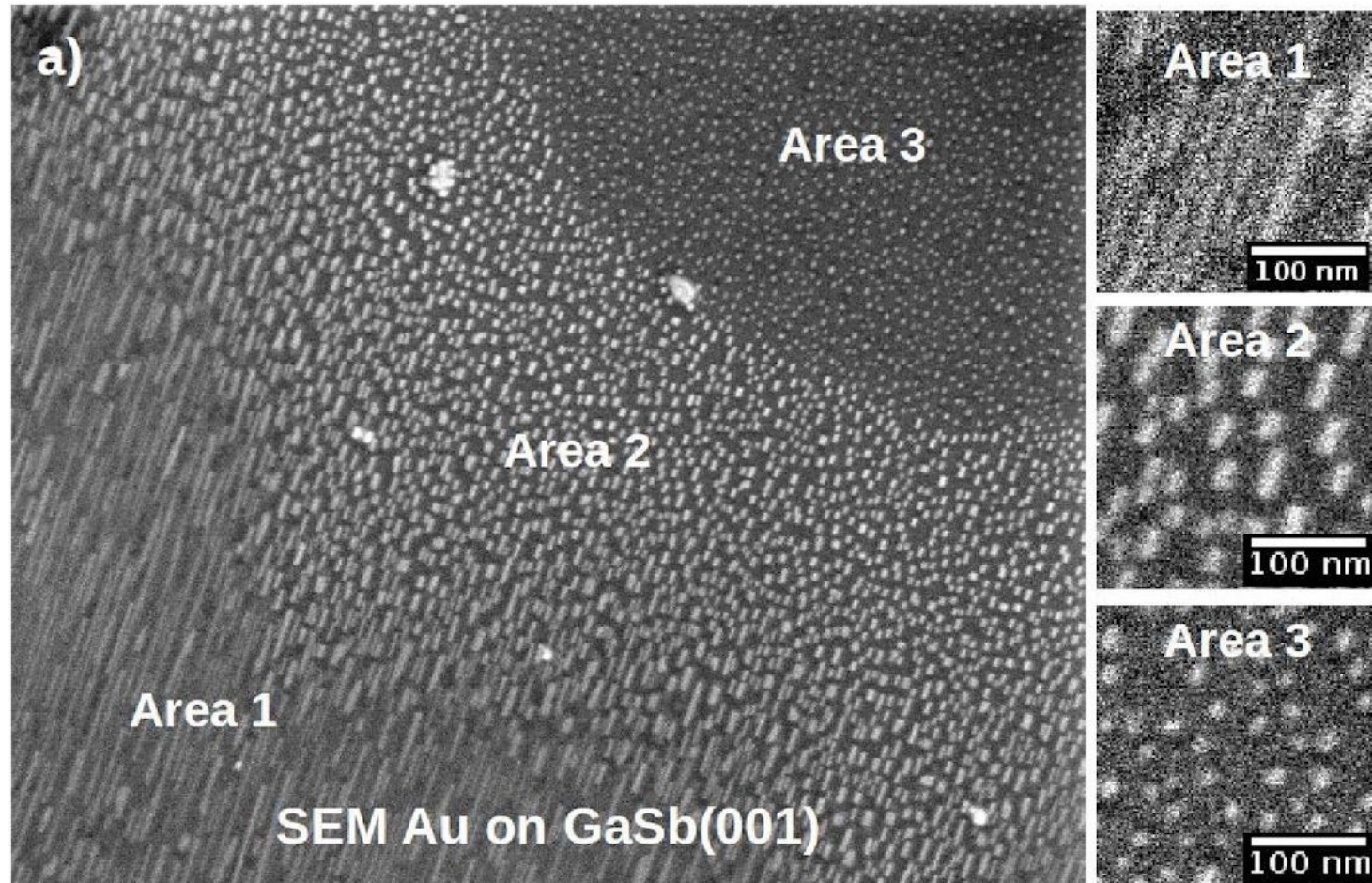
B.R. Jany et al., Micron, Vol. 130, 102800 (2020)



- The image is automatically analyzed i.e. the local features are automatically discovered based on FFT which is sensitive to the local orientation and spacing changes
- The program outputs a PDF file report with images and two tiff files which store original loadings and factors data. The files can be read and further analyze by free software ImageJ/FIJI (via import Bio-Formats)
- The automatic analysis of the microscopic images based on the presented method with a default parameters for the image of $\sim 2000 \times 2000$ pixels takes about a minute on a standard desktop or notebook computer

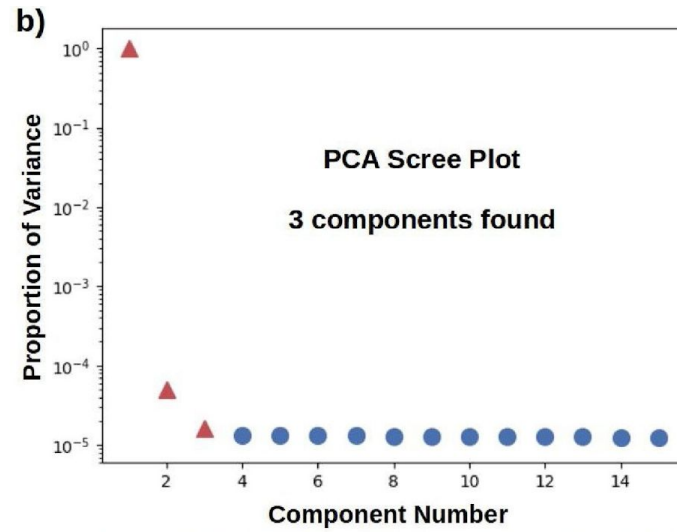
Applications to various microscopic images

Scanning Electron Microscopy Image of Au rich nanostructures on GaSb(001) surface

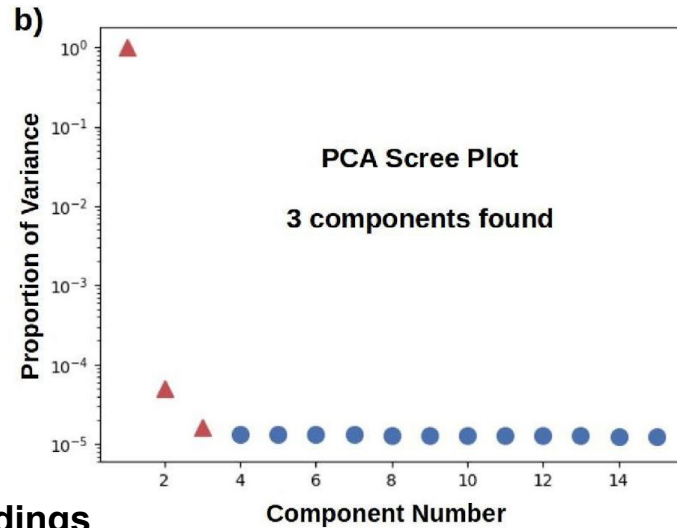


Is there something interesting in the image?

Scanning Electron Microscopy Image of Au rich nanostructures on GaSb(001) surface

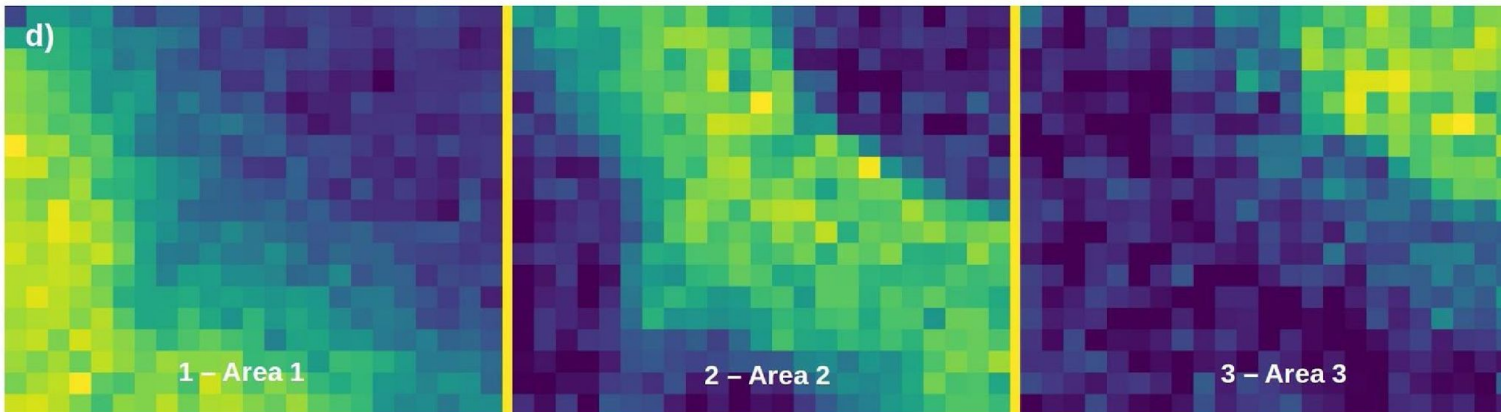
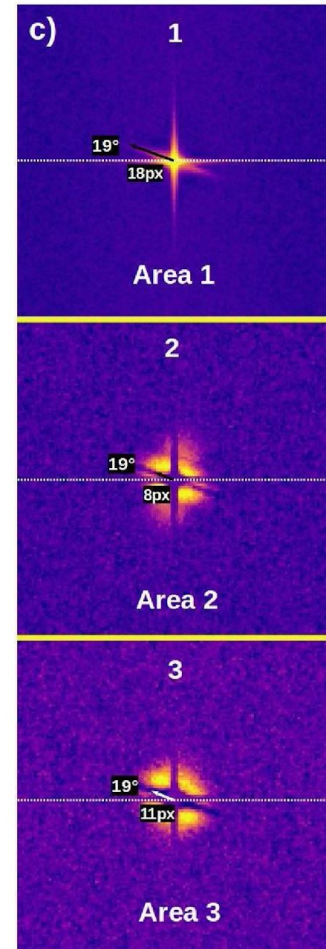


Scanning Electron Microscopy Image of Au rich nanostructures on GaSb(001) surface



NMF Loadings

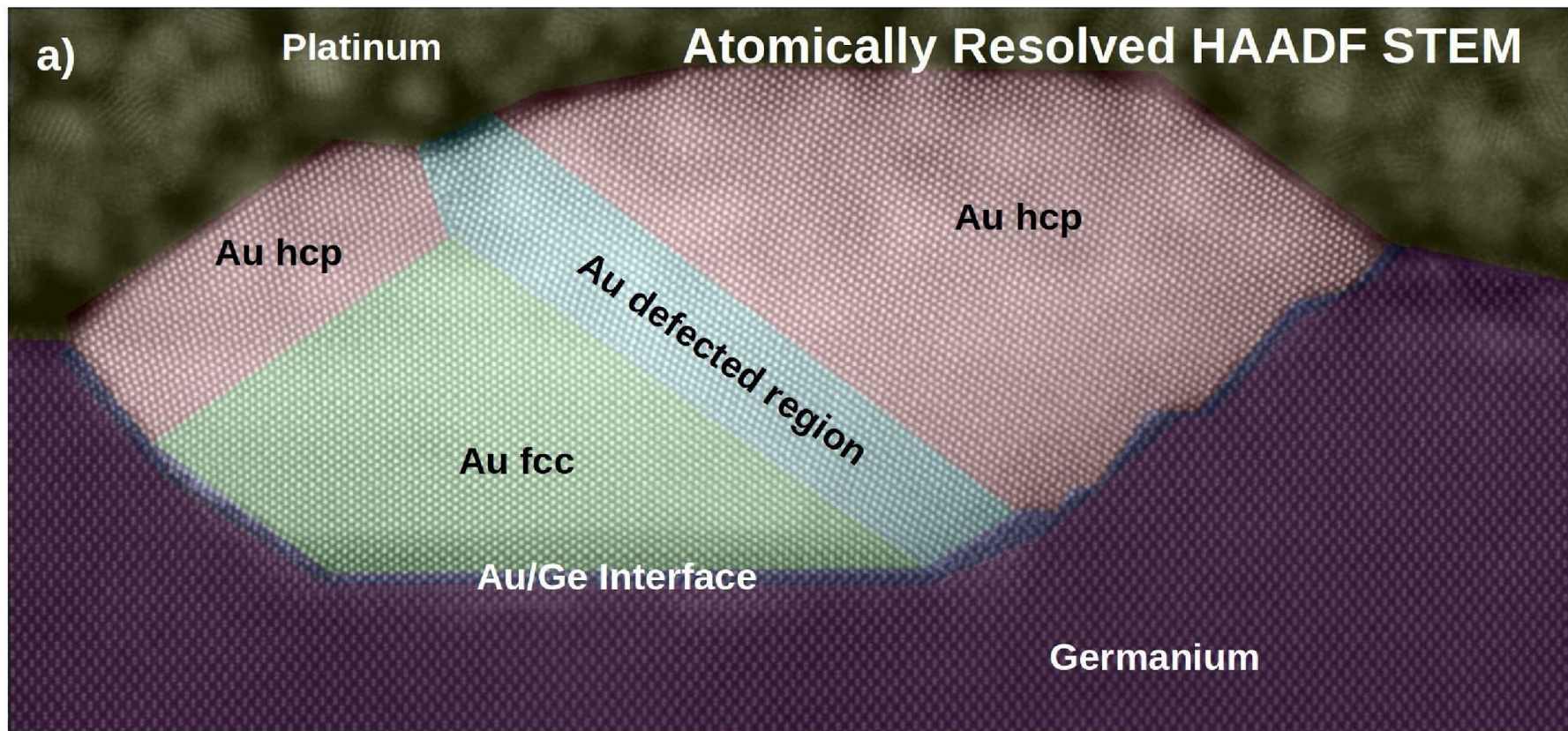
NMF Factors



→ correctly automatically analyzed

B.R. Jany et al., Micron, Vol. 130, 102800 (2020)

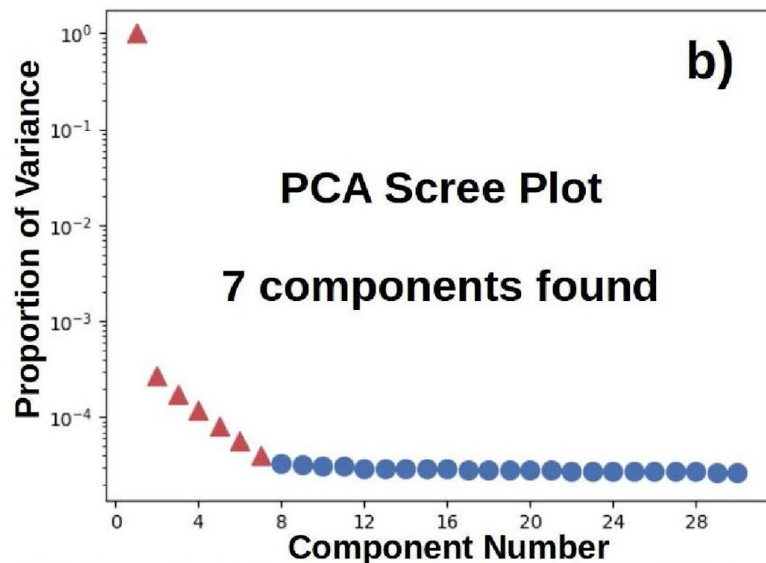
Atomically resolved STEM HAADF of nanostructures made of Au fcc and Au hcp



B.R. Jany et al., Scientific Reports 7, 42420 (2017)

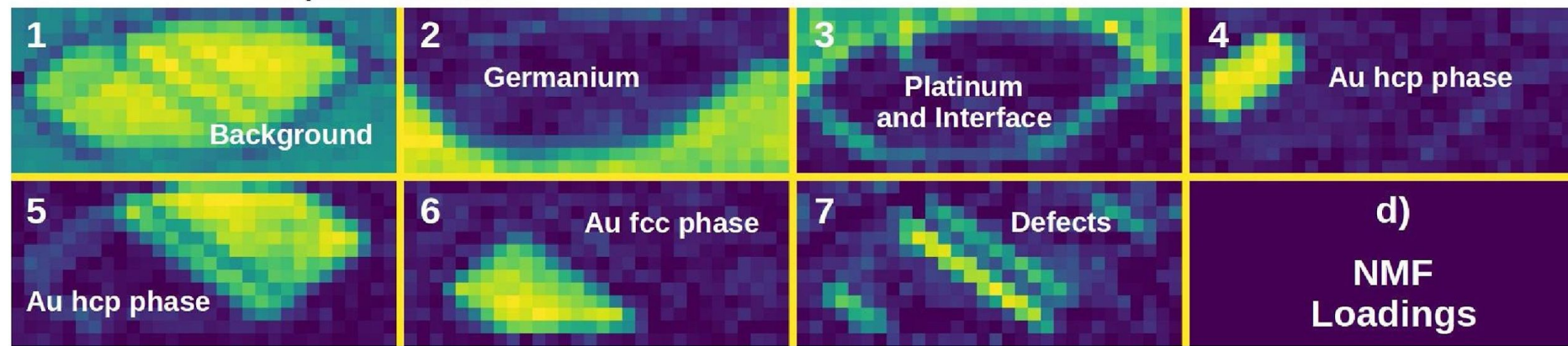
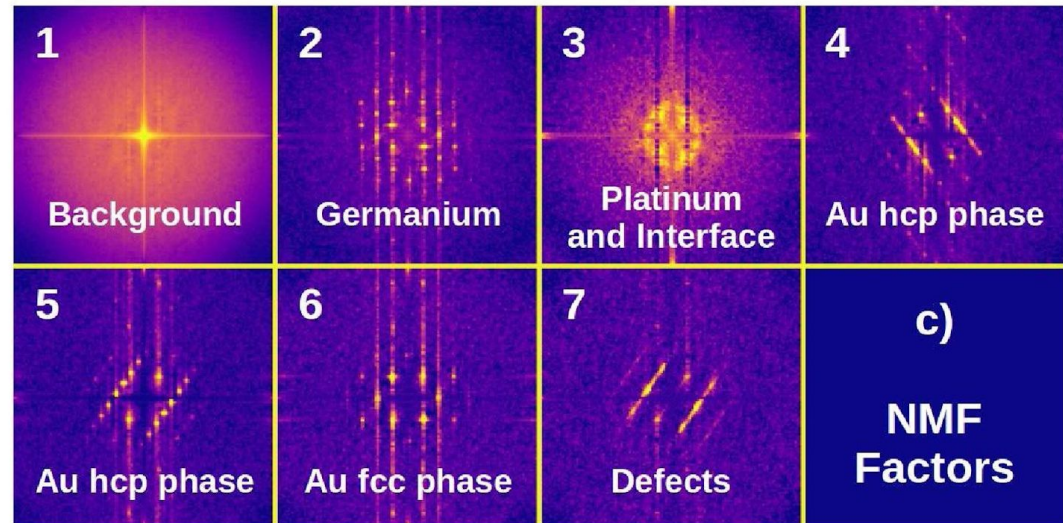
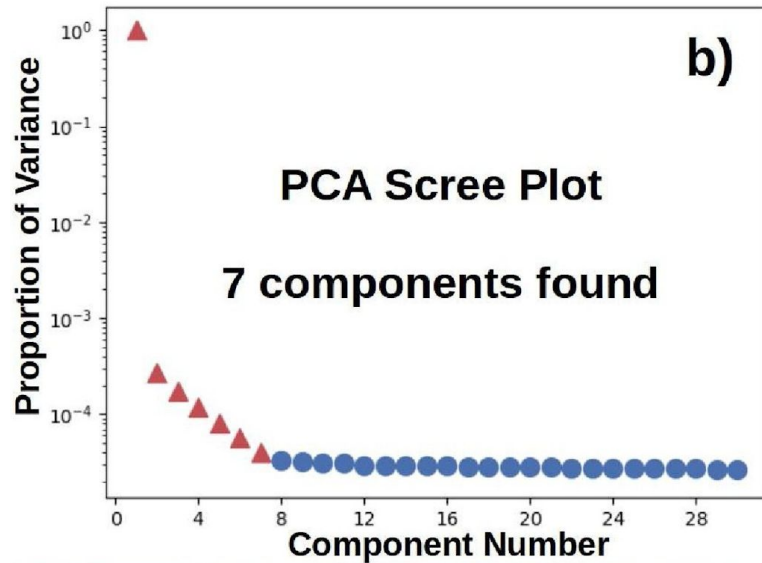
Is there something interesting in the image?

Atomically resolved STEM HAADF of nanostructures made of Au fcc and Au hcp



Applications to various microscopic images

Atomically resolved STEM HAADF of nanostructures made of Au fcc and Au hcp



→ correctly automatically analyzed

B.R. Jany et al., Micron, Vol. 130, 102800 (2020)

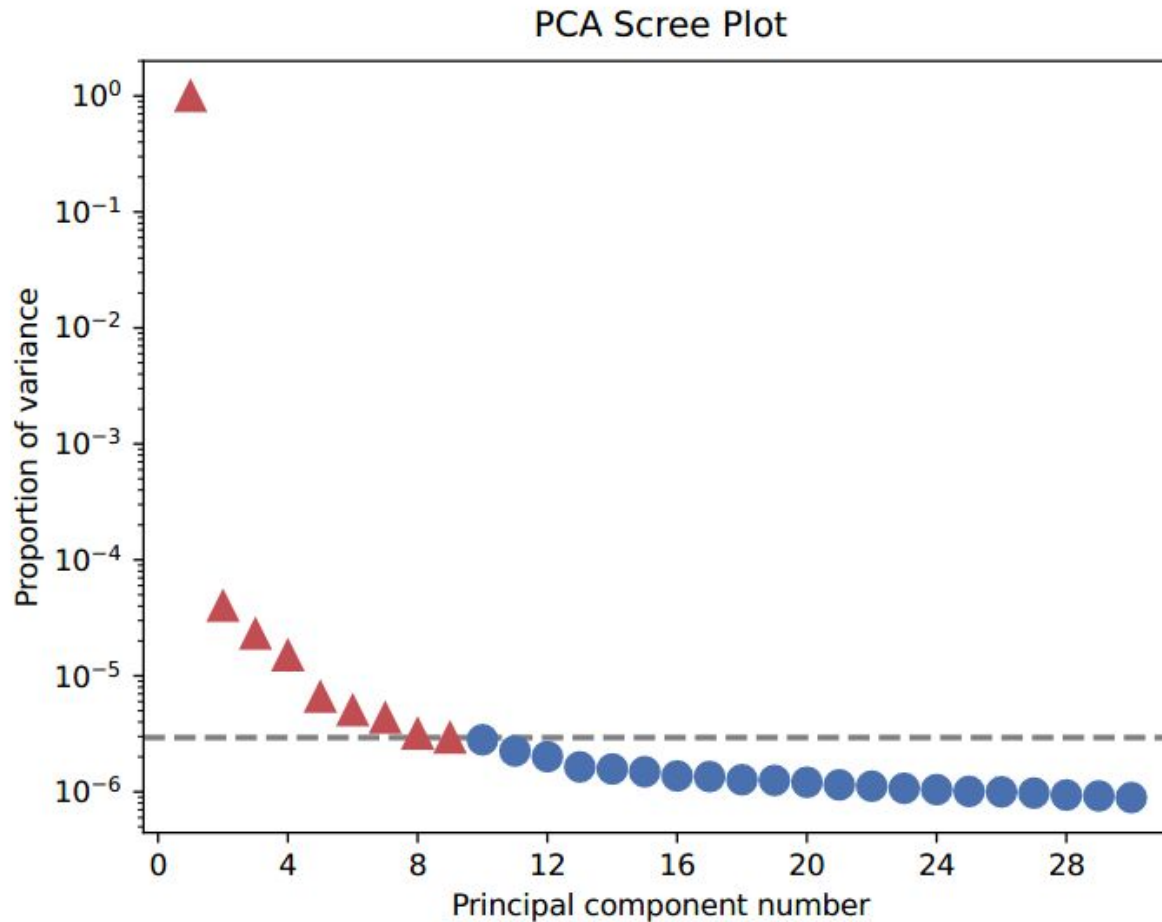
microscopic
(not only)

**Photo of the cat
(public domain)**

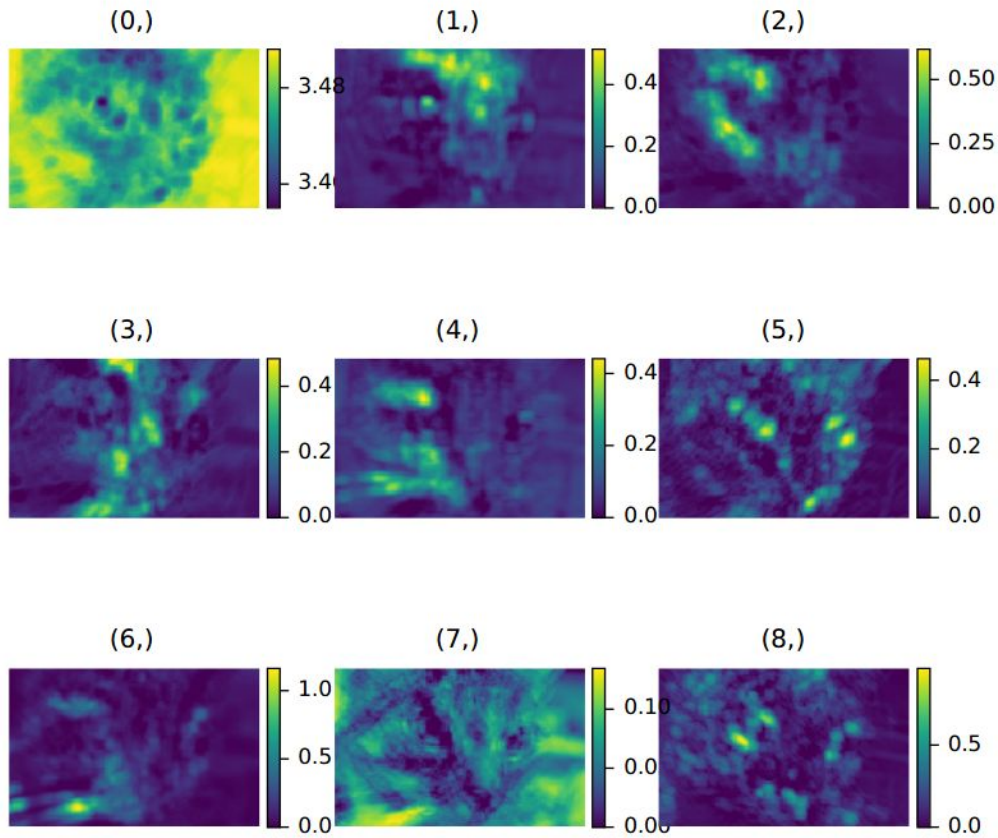


Is there something interesting in the image?

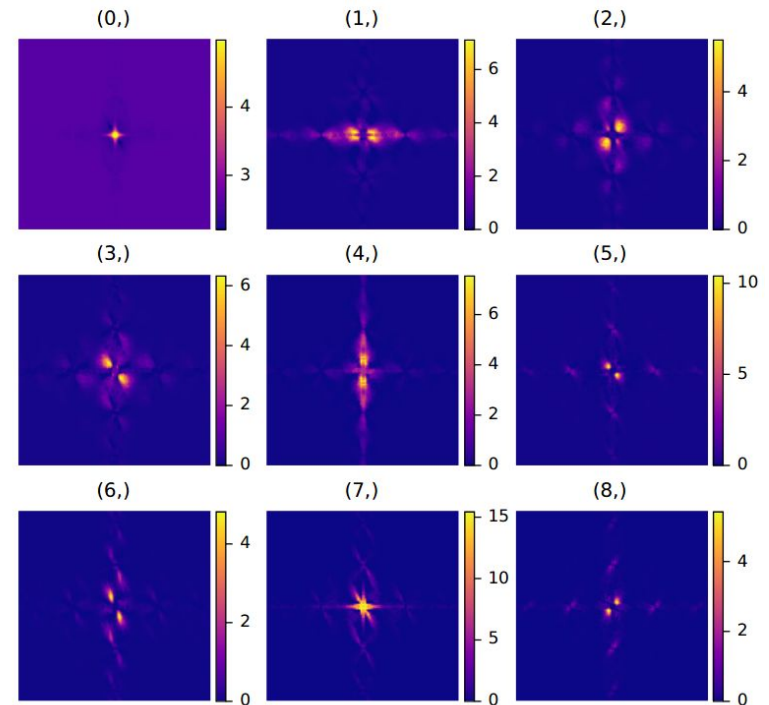
Photo of the cat
(public domain)



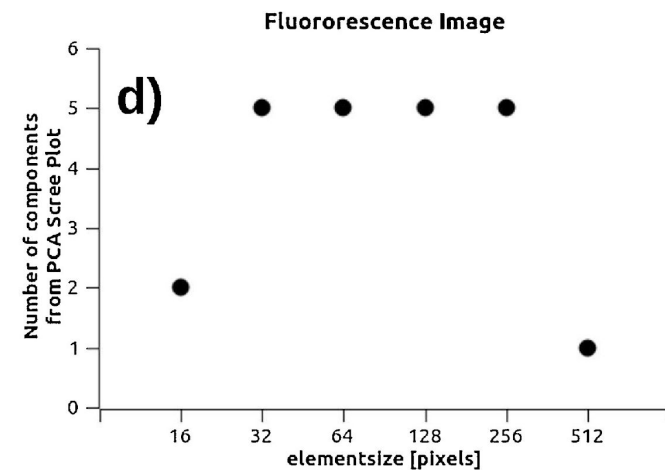
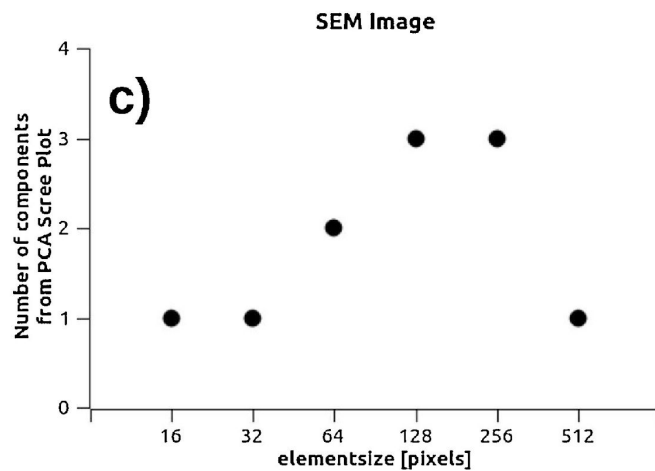
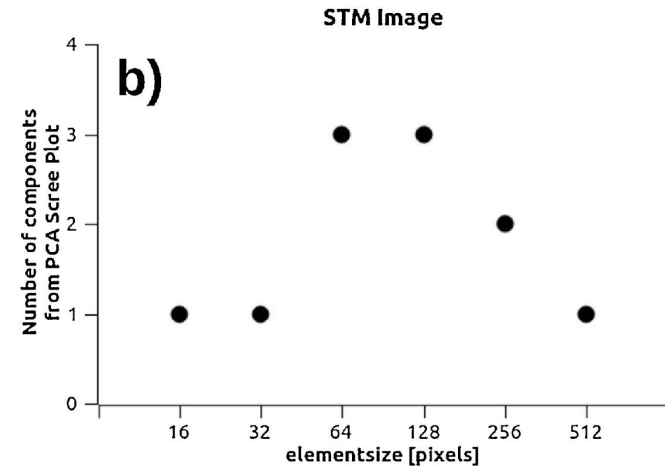
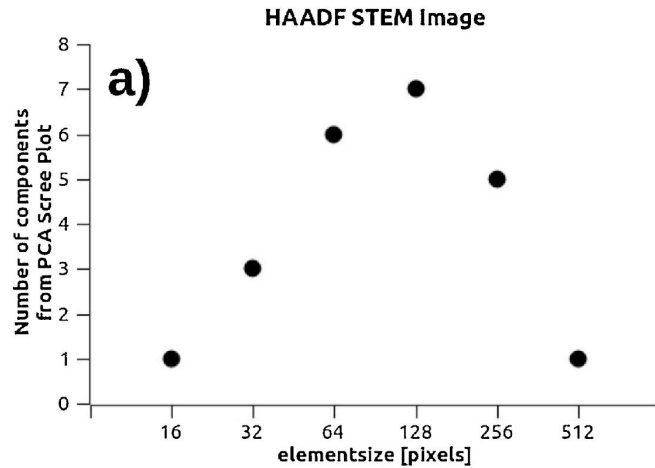
NMF Decomposition loadings



NMF Decomposition factors

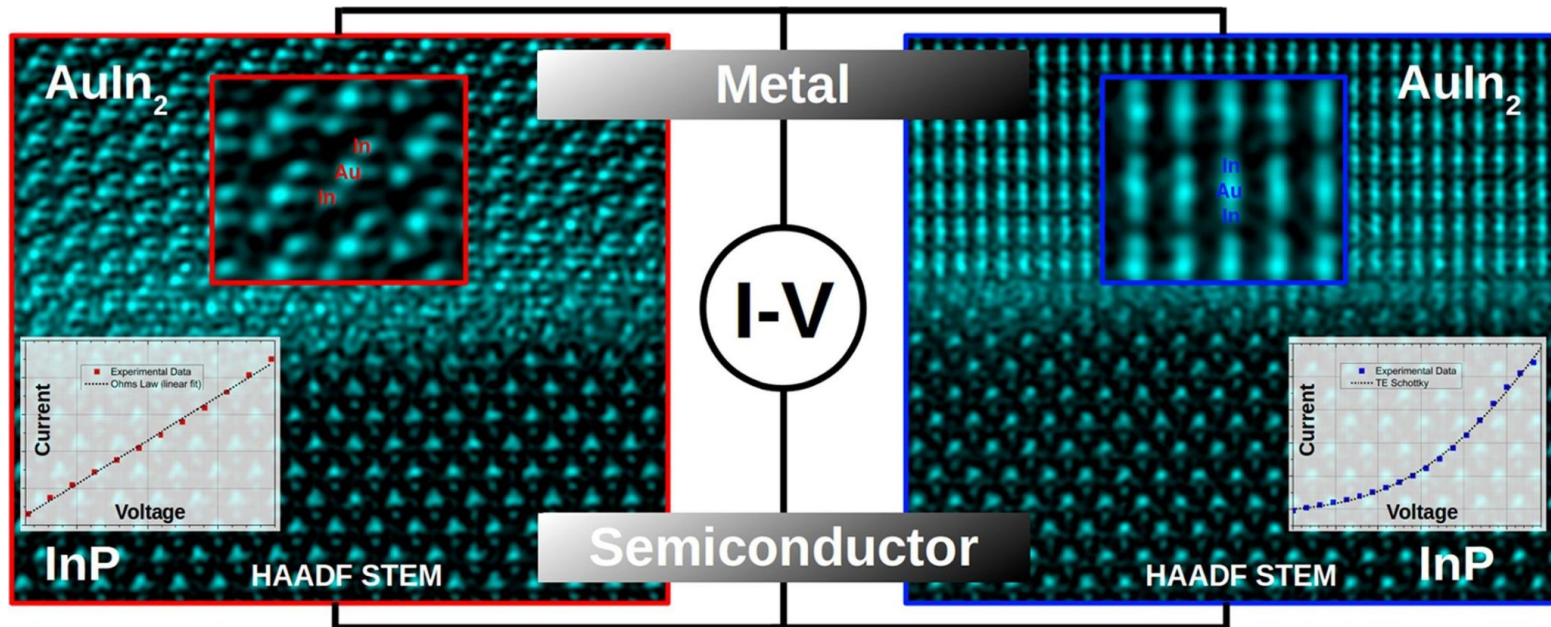


Optimizing Local Features detection error



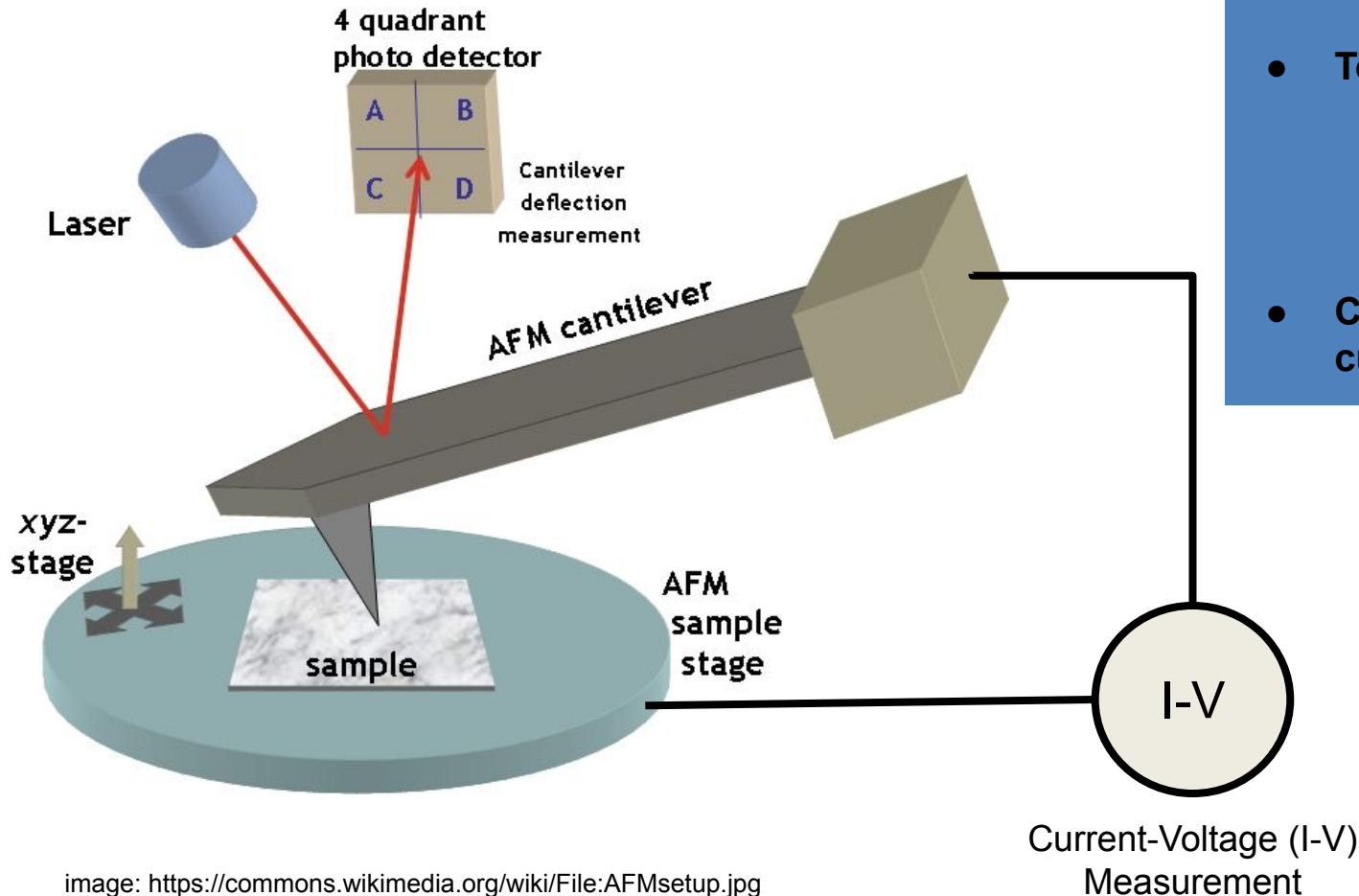
moving windows size 128px seems to optimal for all analyzed images ~2000x2000px

Analysis of Conductive Metal-Semiconductor (M-S) Junction at Nano and Atomic Level



A. Janas, W. Piskorz, A. Kryshstal, G. Cempura, W. Belza, A. Kruk, B.R. Jany, F. Krok,
Into the Origin of Electrical Conductivity for the Metal-Semiconductor Junction at the Atomic Level
Applied Surface Science Volume 570, 150958 (2021)

Conductive AFM (C-AFM)



C-AFM

- Topography

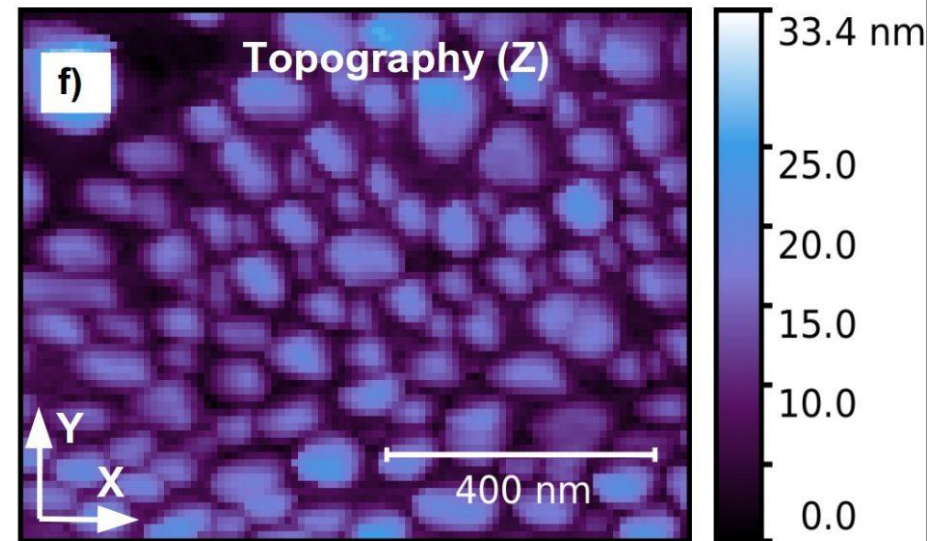
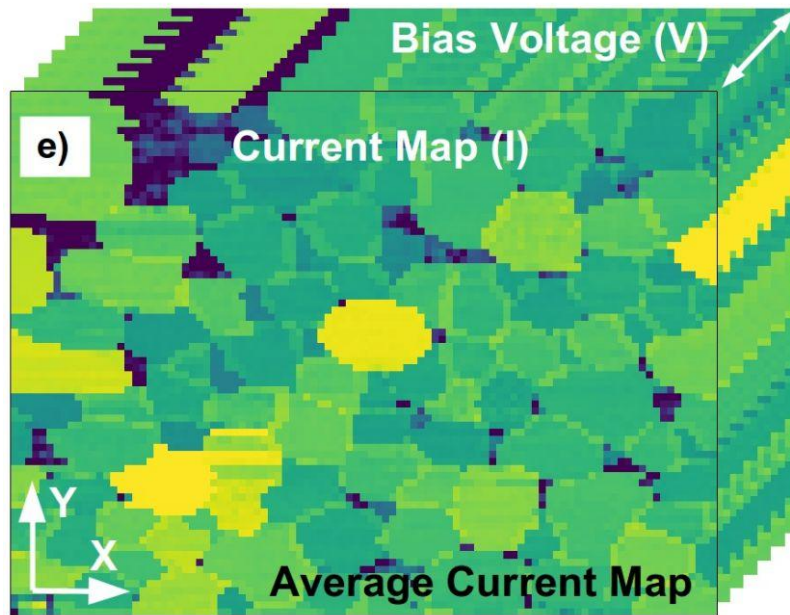
+

- Current-Voltage (I-V) curve at each point

image: <https://commons.wikimedia.org/wiki/File:AFMsetup.jpg>

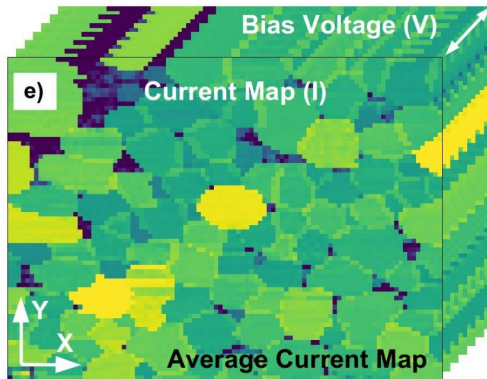
AuIn_2 Nanoelectrodes on $\text{InP}(001)$

Conductive AFM Current-Voltage (I-V) Hyperspectral Data



Conductive AFM shows regions of different conductivity

Grouping Conductive AFM (C-AFM) hyperspectral data by k-means



- Hyperspectral C-AFM data (I-V curve in each pixel ~7200 curves)
- Automatic grouping/clustering by k-means

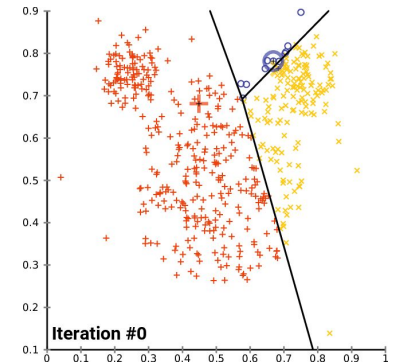
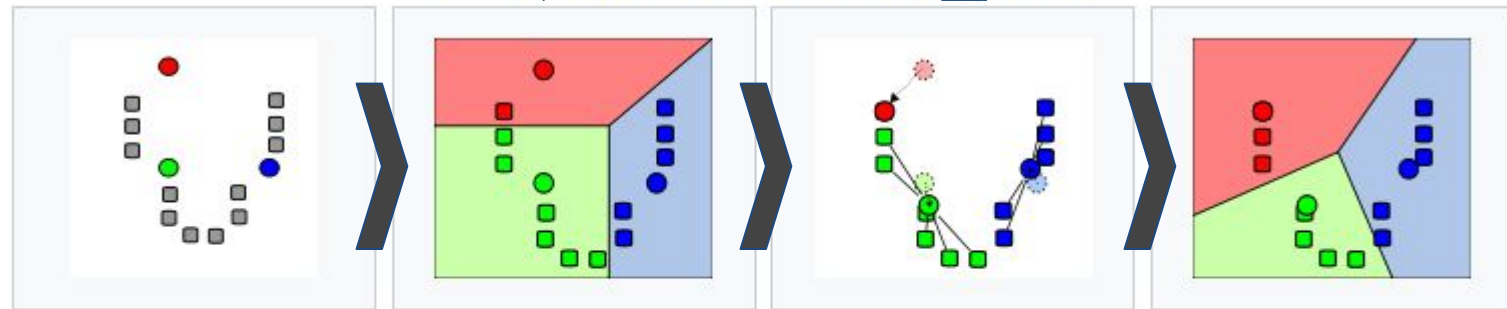


image: https://commons.wikimedia.org/wiki/File:K-means_convergence.gif



1. k initial "means" (in this case $k=3$) are randomly generated within the data domain (shown in color).

2. k clusters are created by associating every observation with the nearest mean. The partitions here represent the Voronoi diagram generated by the means.

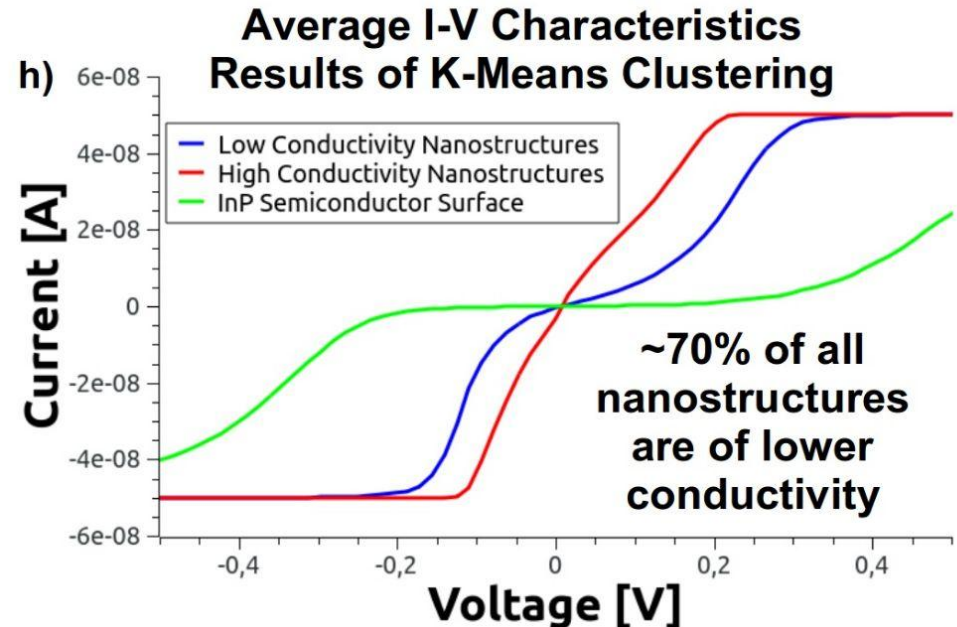
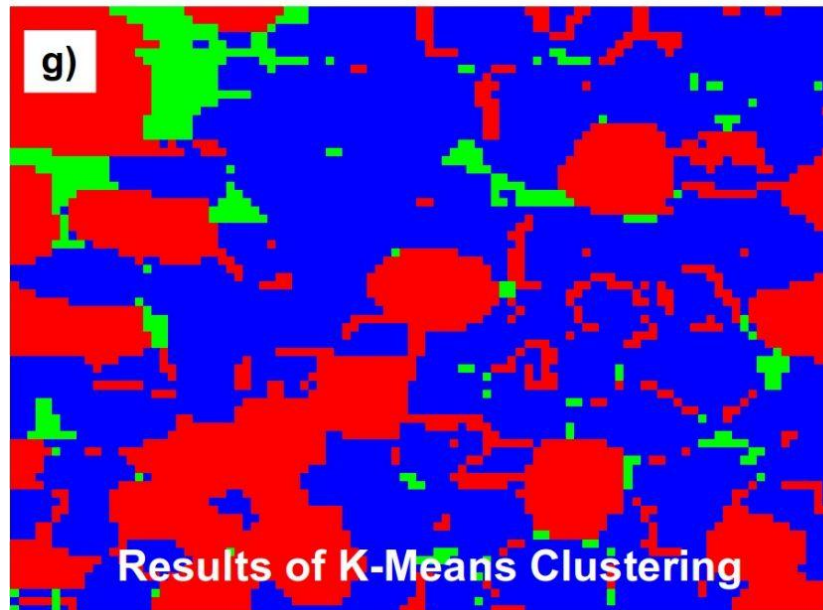
3. The centroid of each of the k clusters becomes the new mean.

4. Steps 2 and 3 are repeated until convergence has been reached.

image: https://en.wikipedia.org/wiki/K-means_clustering

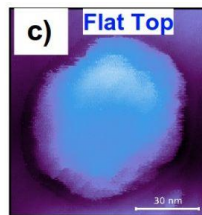
Applied Surface Science Volume 570, 150958 (2021)

Grouping (clustering) Current-Voltage (I-V) Hyperspectral Data

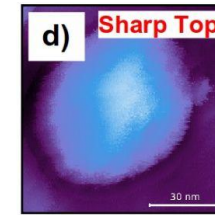


Discovery of Two different electrical types of nanoelectrodes

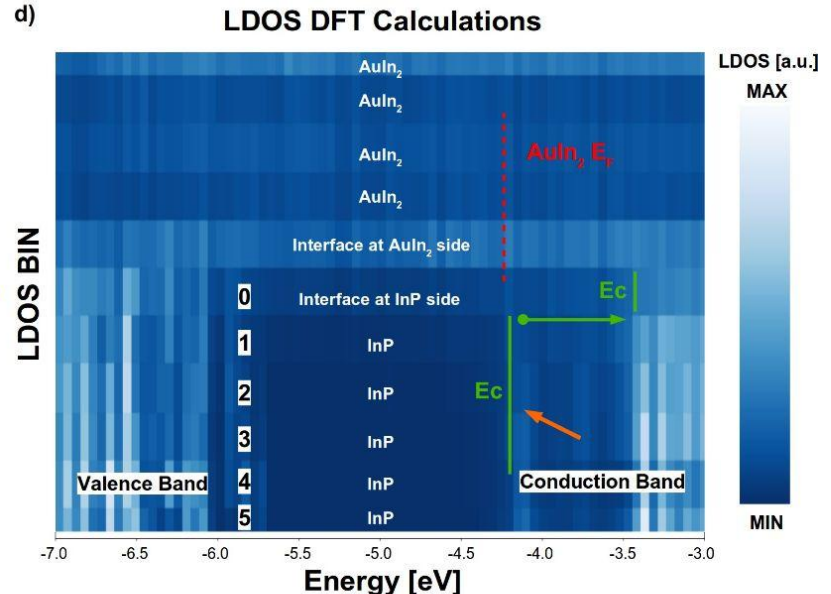
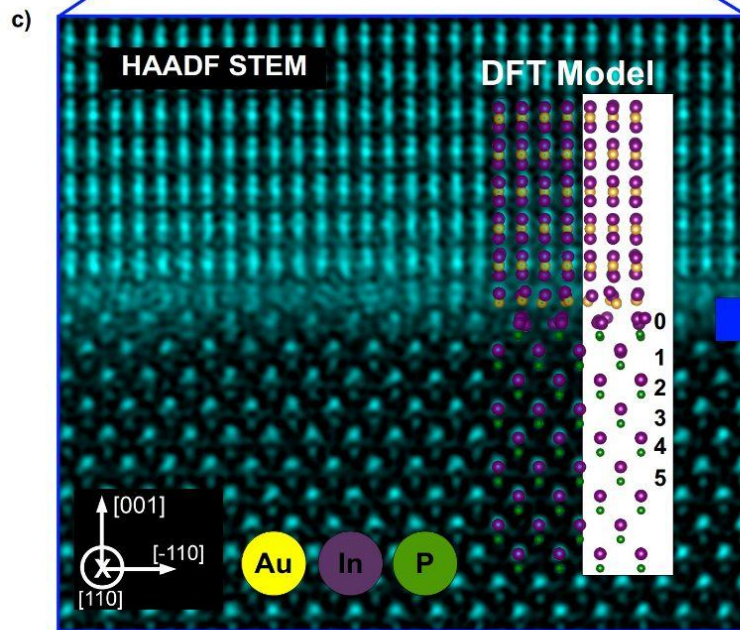
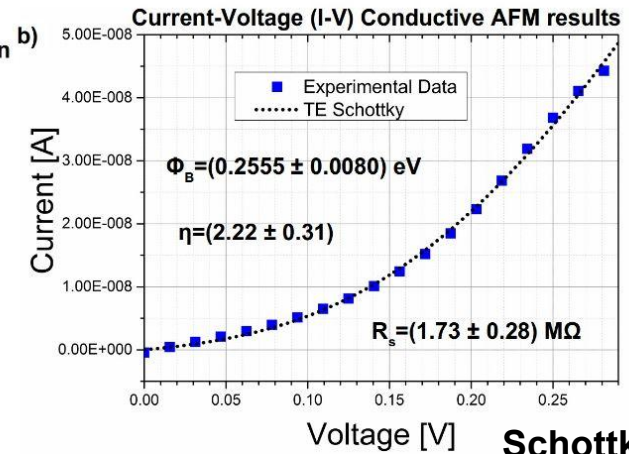
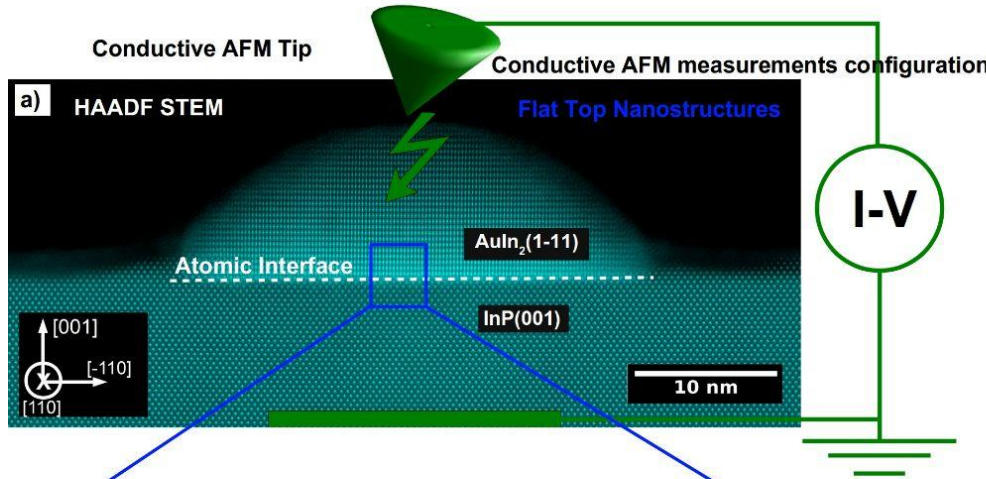
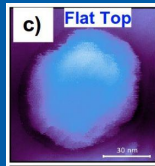
Low Conductivity



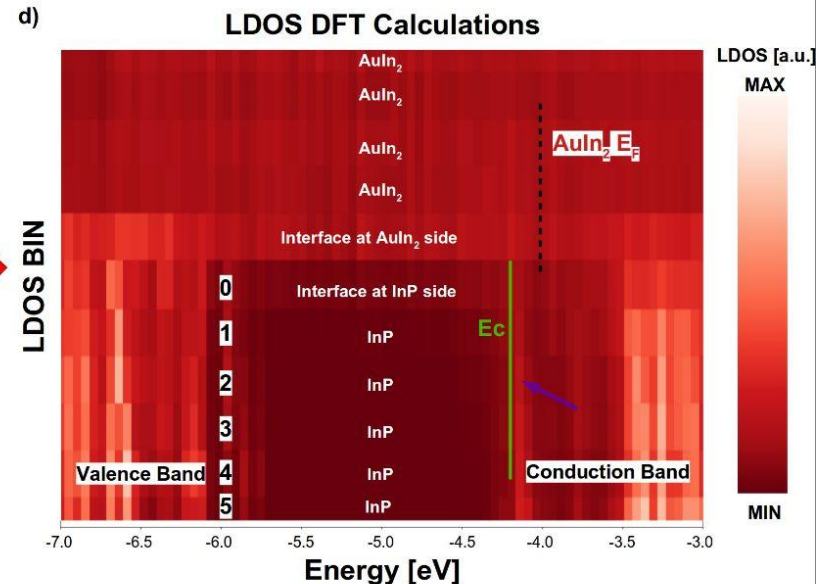
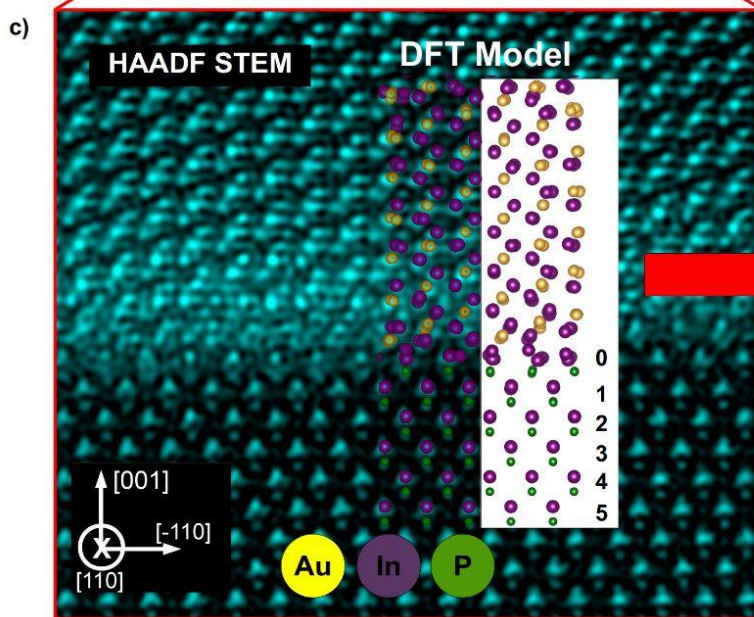
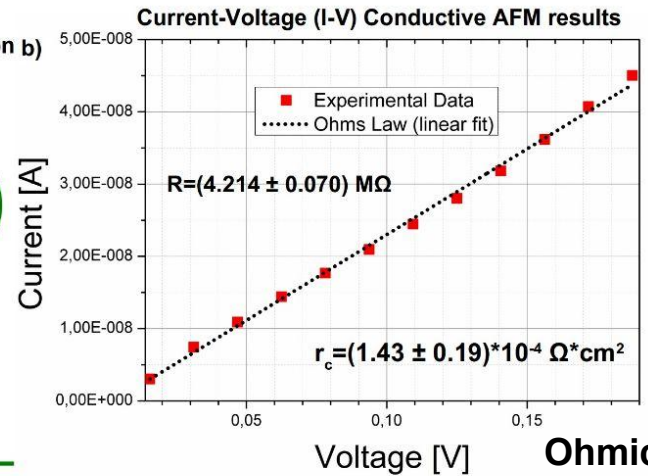
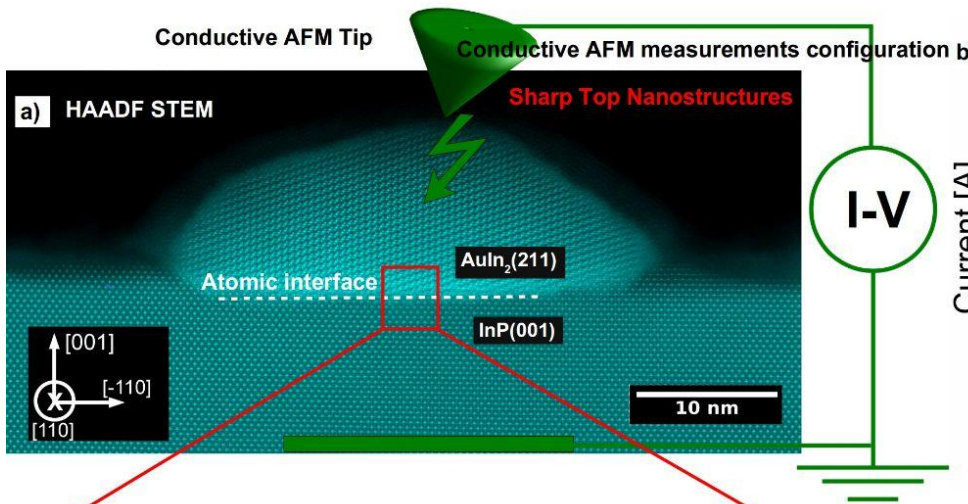
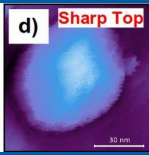
High Conductivity



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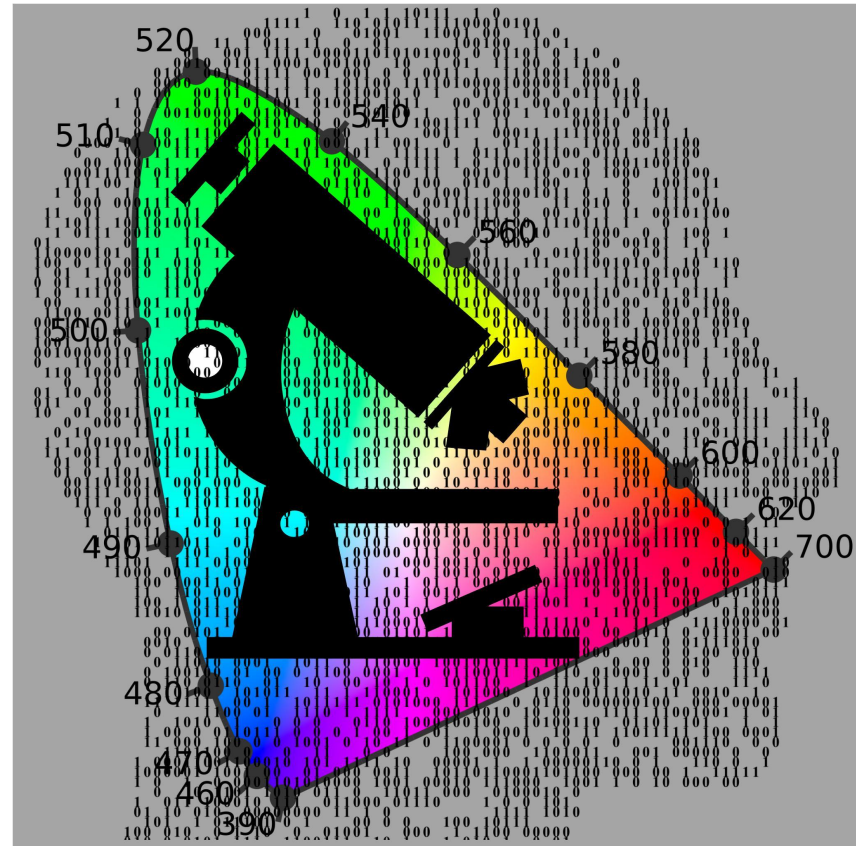
Applied Surface Science Volume 570, 150958 (2021)

- Machine Learning is very useful in Nanomaterials Characterization
- It allows for reliable extraction of materials properties at the nanoscale level (without human bias)
- It is not limited only to the presented examples (could be used also for HAADF image quantification, or in EBSD)

The synergetic coupling between microscopic and Machine Learning techniques provides new quality and perspectives in materials characterization.



Thank You

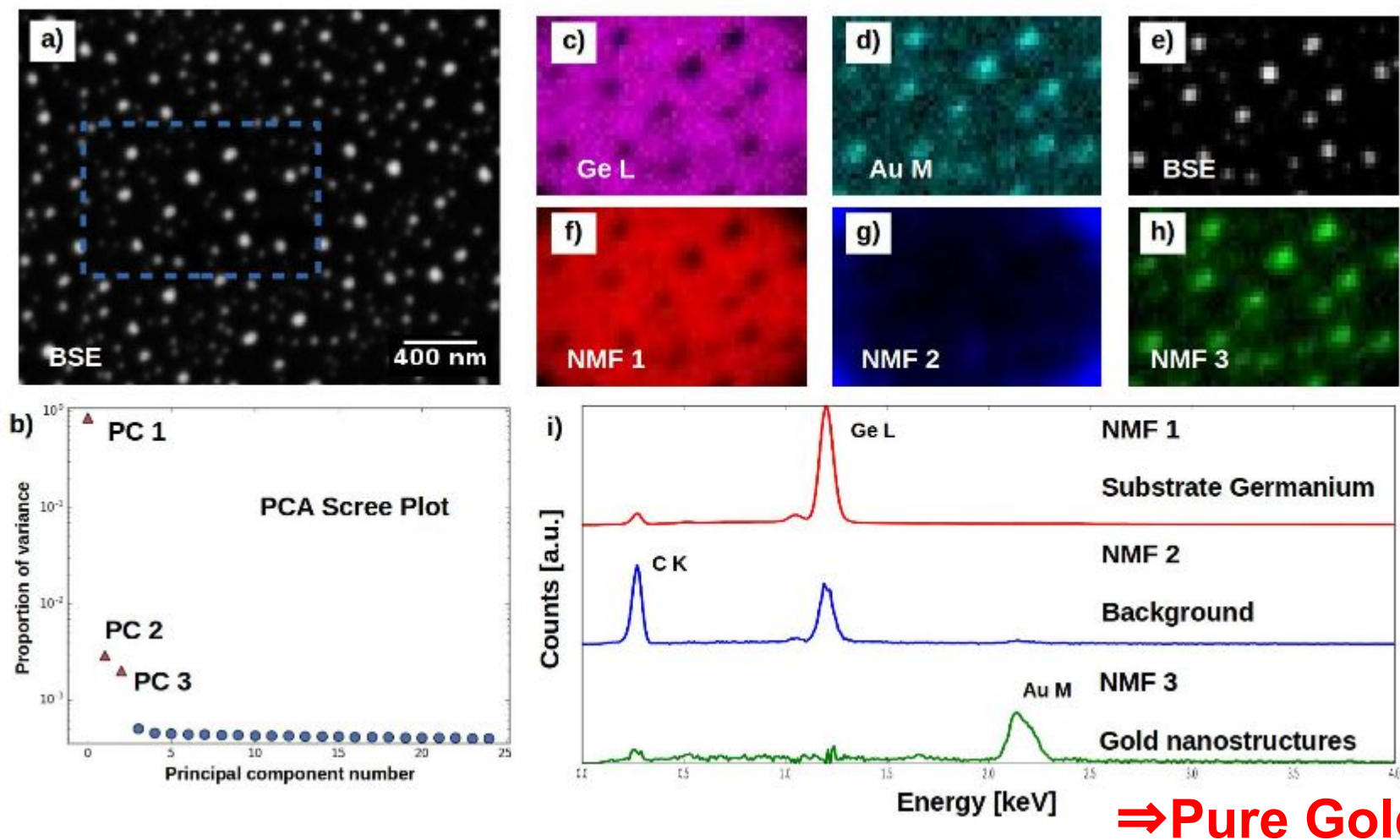


e-mail: benedykt.jany@uj.edu.pl

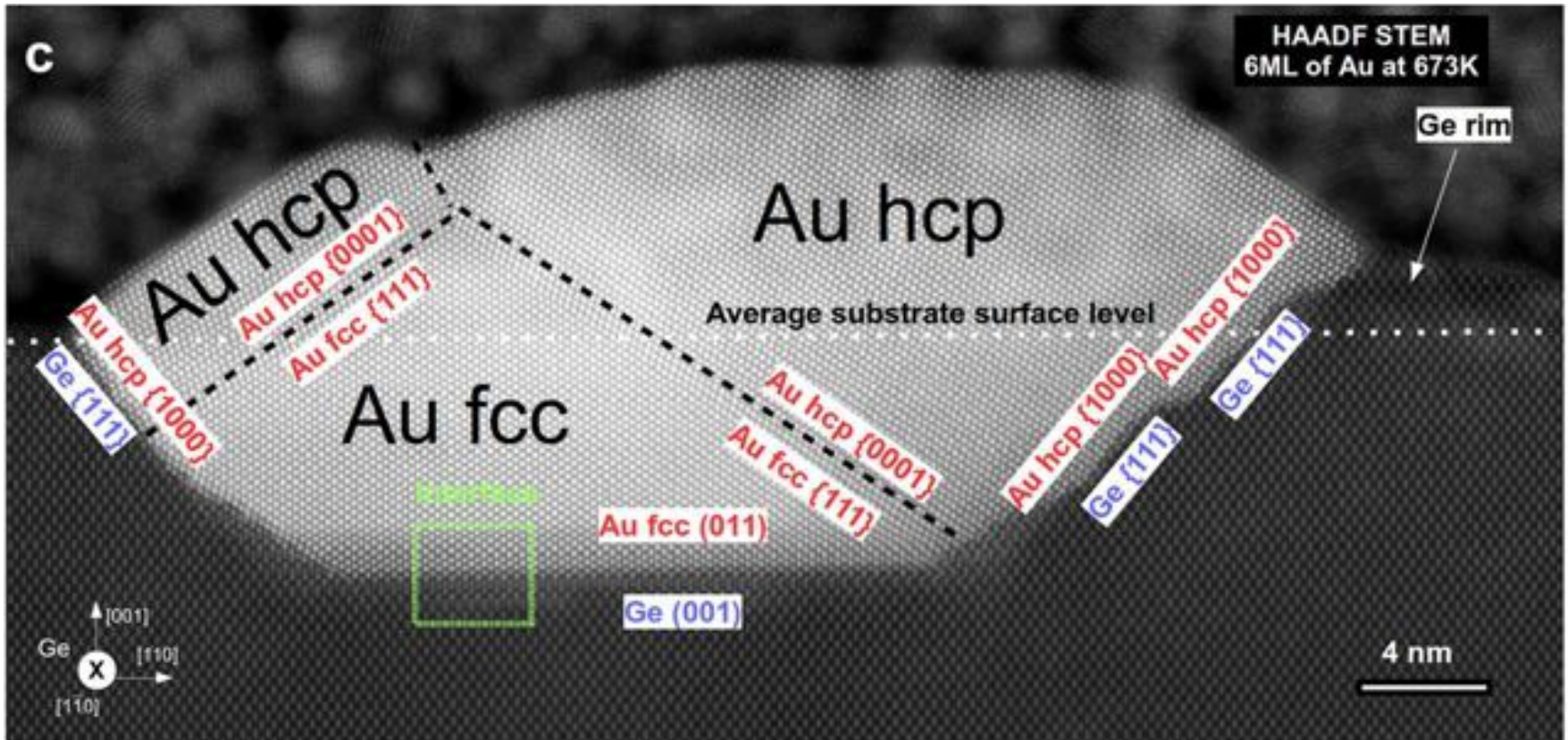
<http://www.fcfs.if.uj.edu.pl/c-microscopy-ml>

More Examples

6ML Au on Ge(001): Pure Gold or Gold/Germanium alloy?

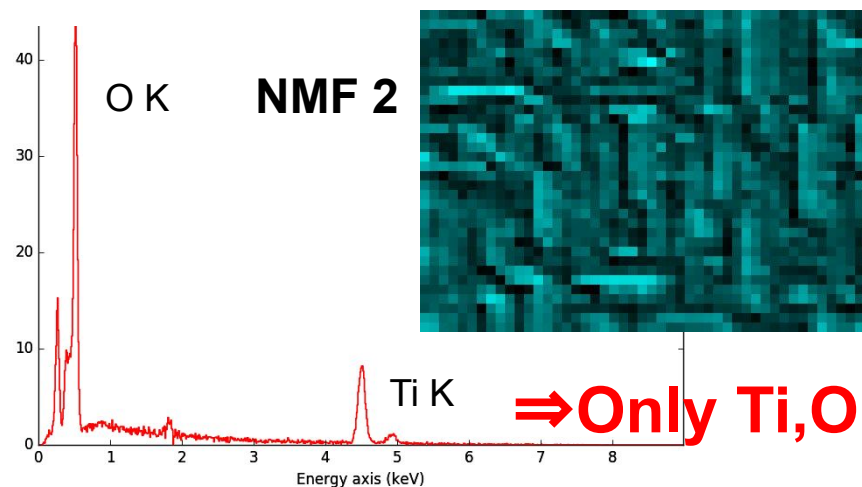
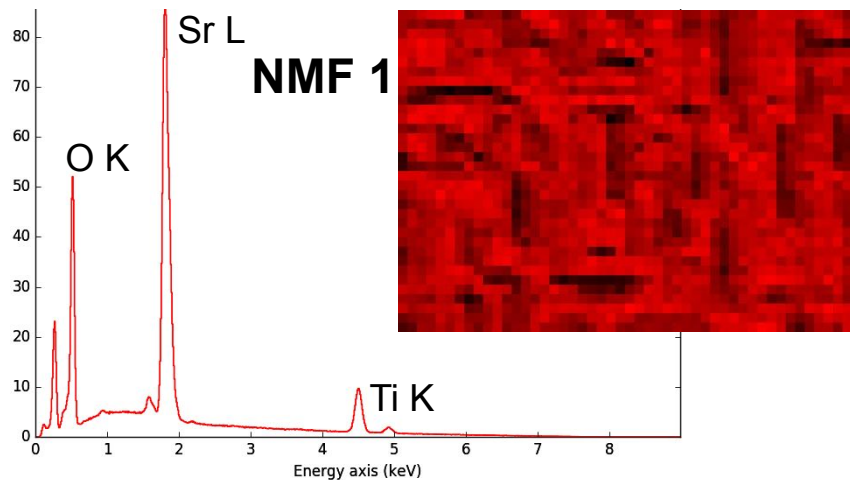
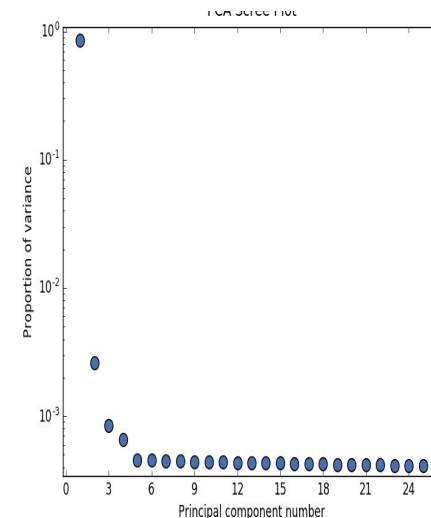
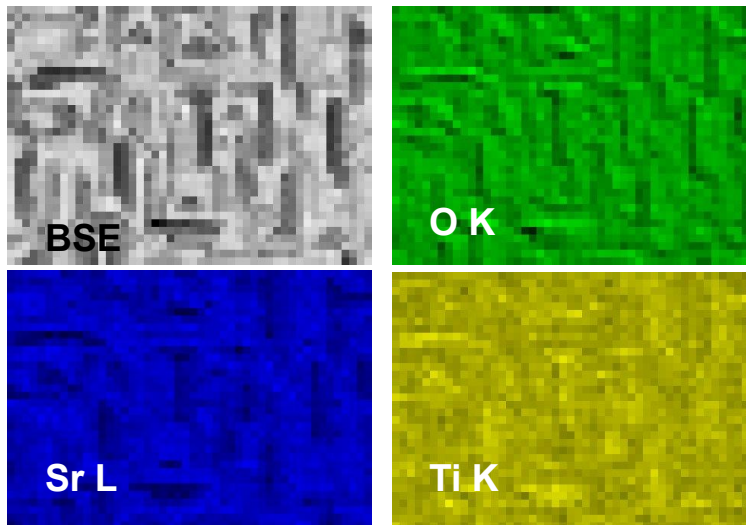
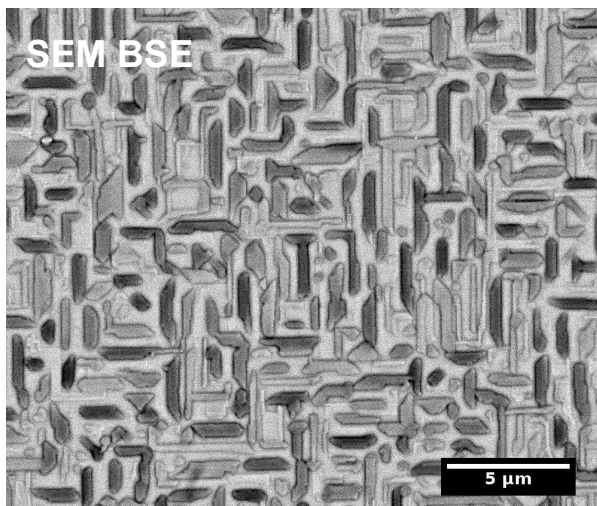


6ML Au on Ge(001): Au fcc and Au hcp Nanostructures

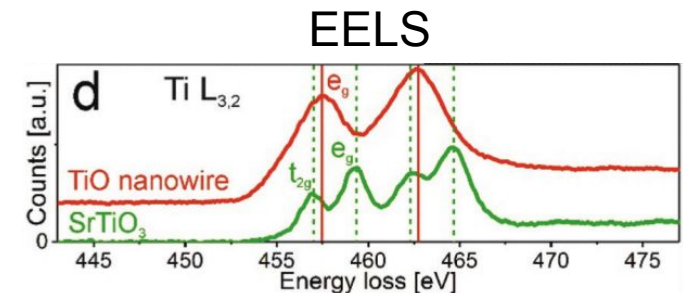
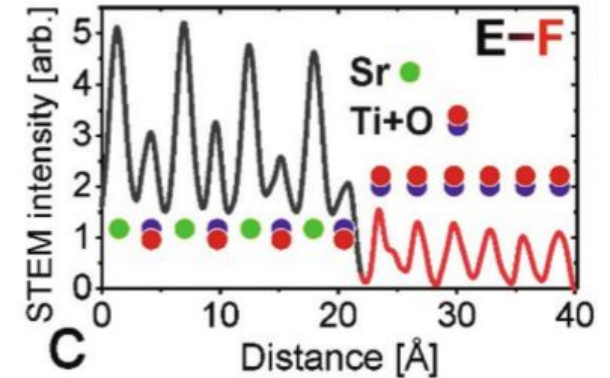
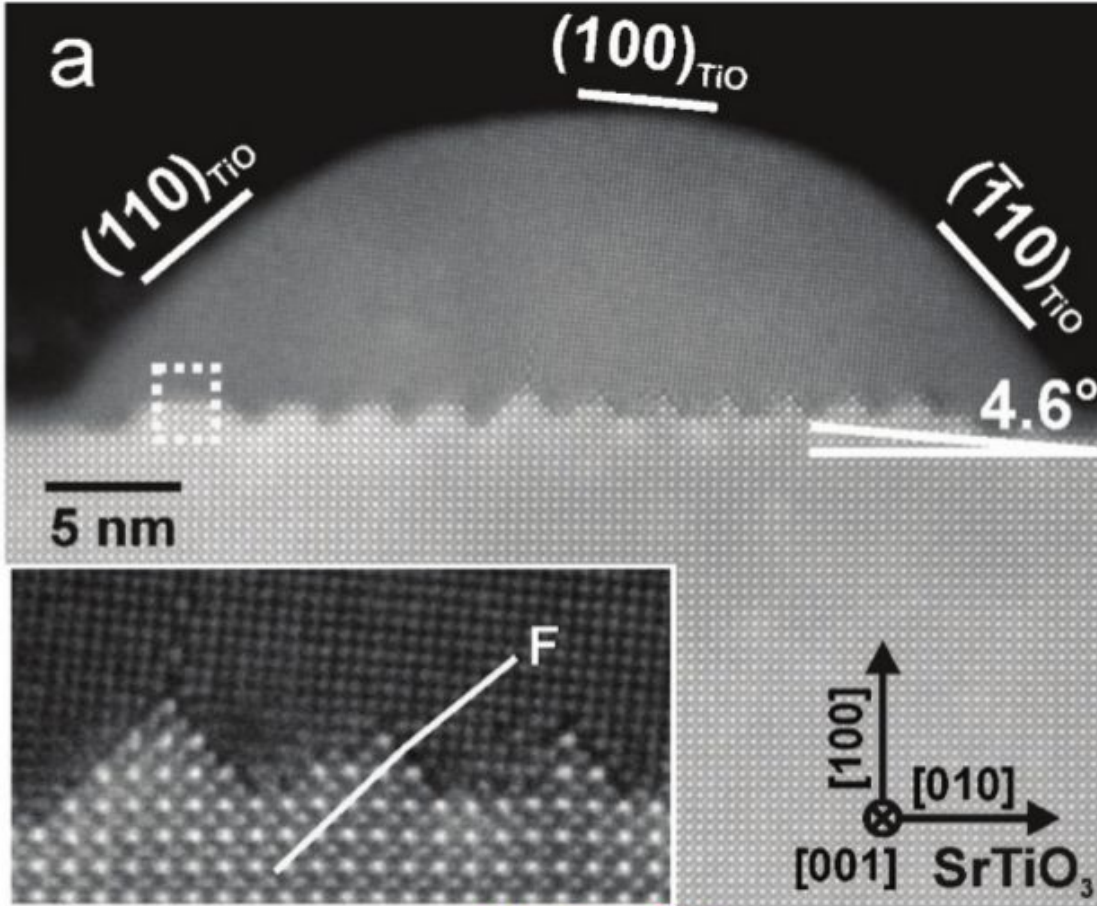


B.R. Jany et al., Scientific Reports volume 7, Article number: 42420 (2017)

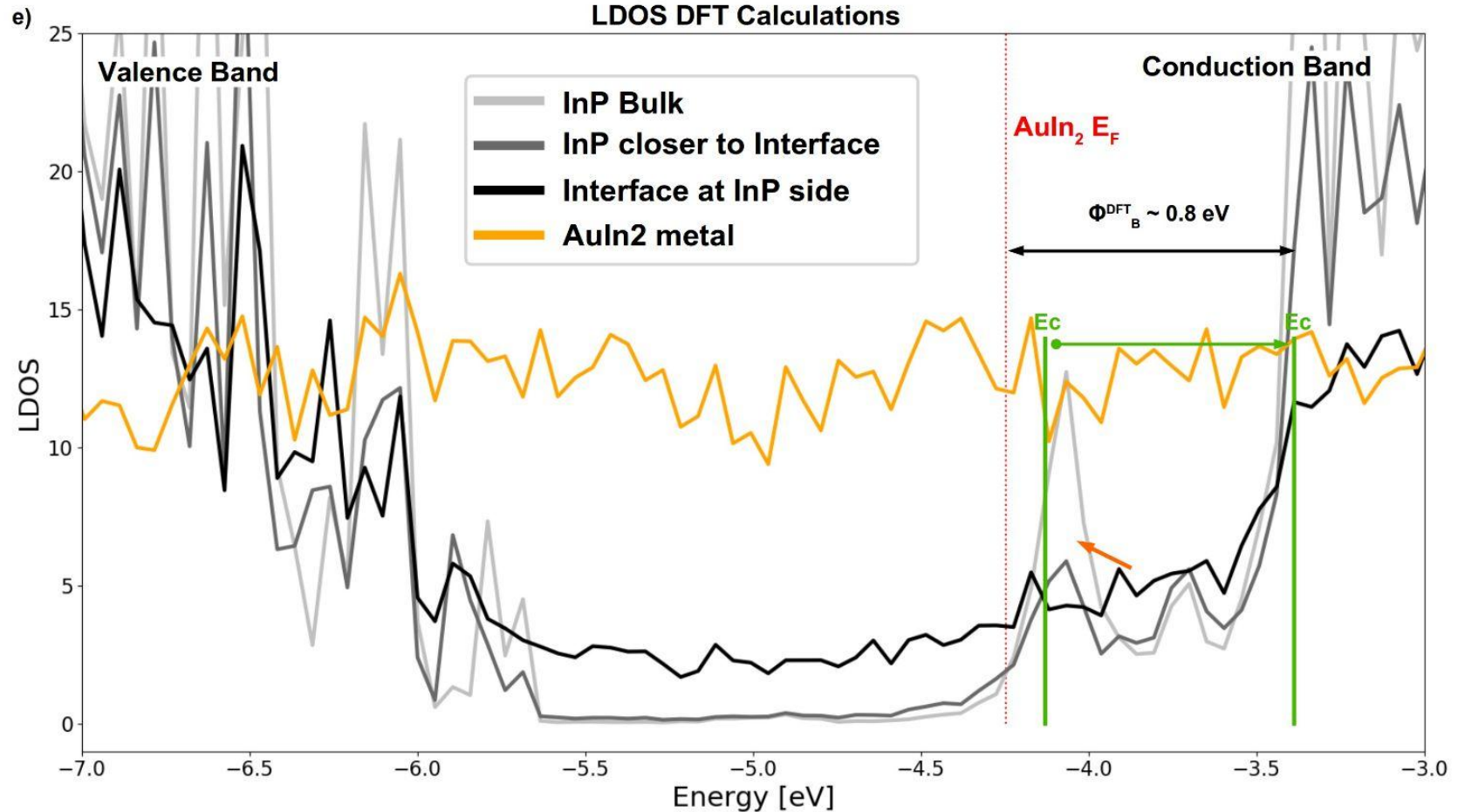
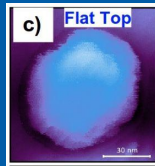
Reduced SrTiO₃: What is the chemical composition?



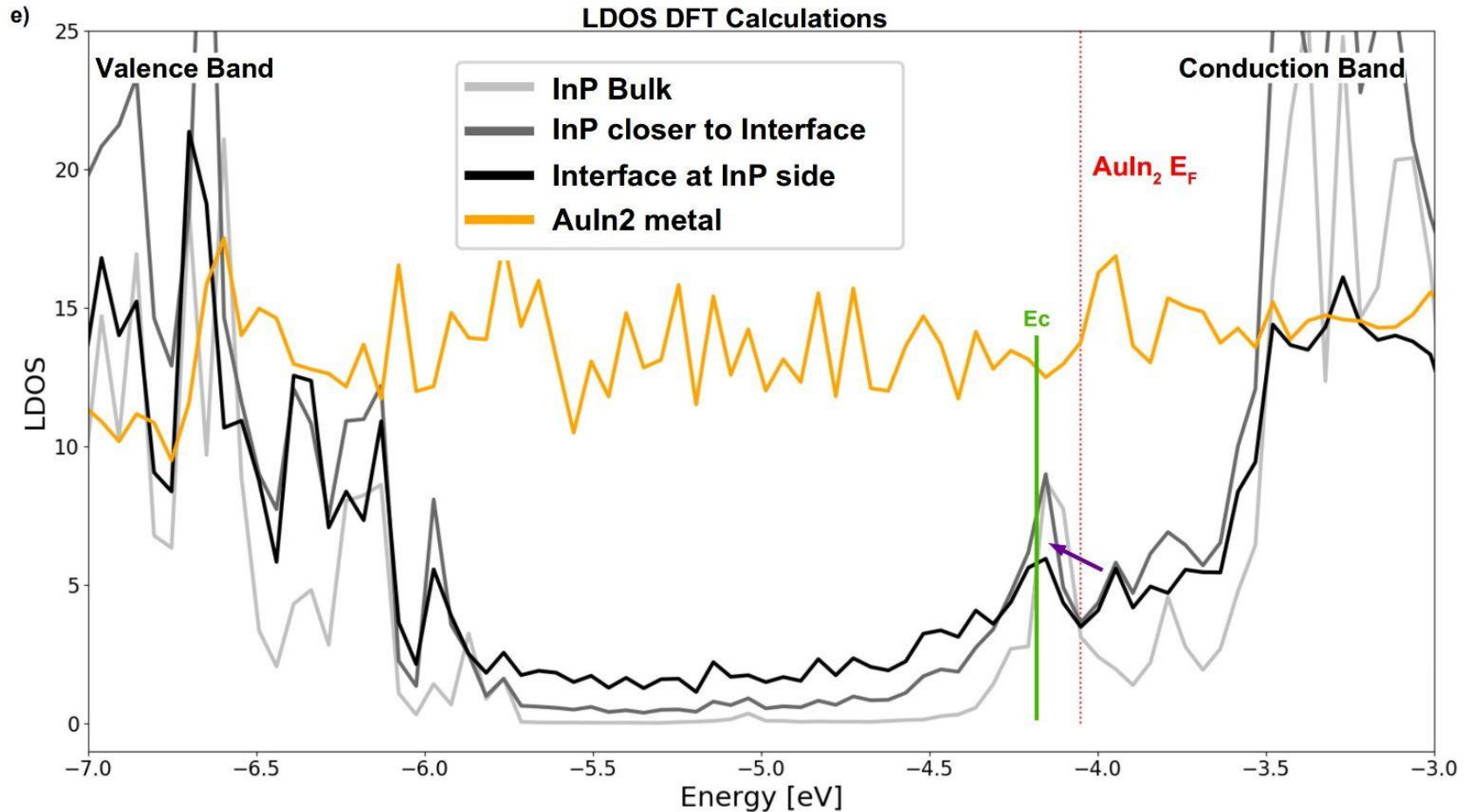
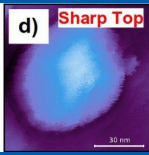
Reduced SrTiO₃: TiO nanowires on SrTiO₃



D. Wrana et al., *Nanoscale*, 11, 89-97 (2019)

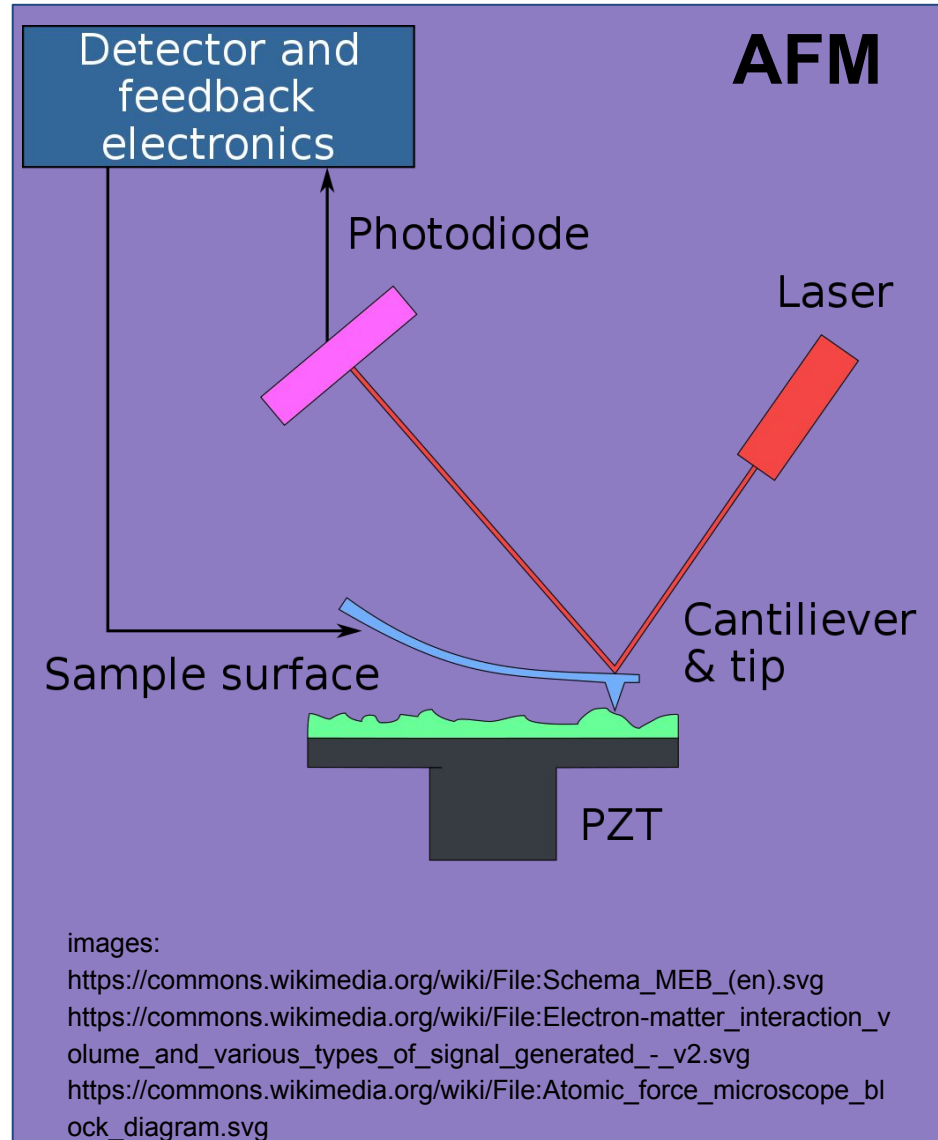
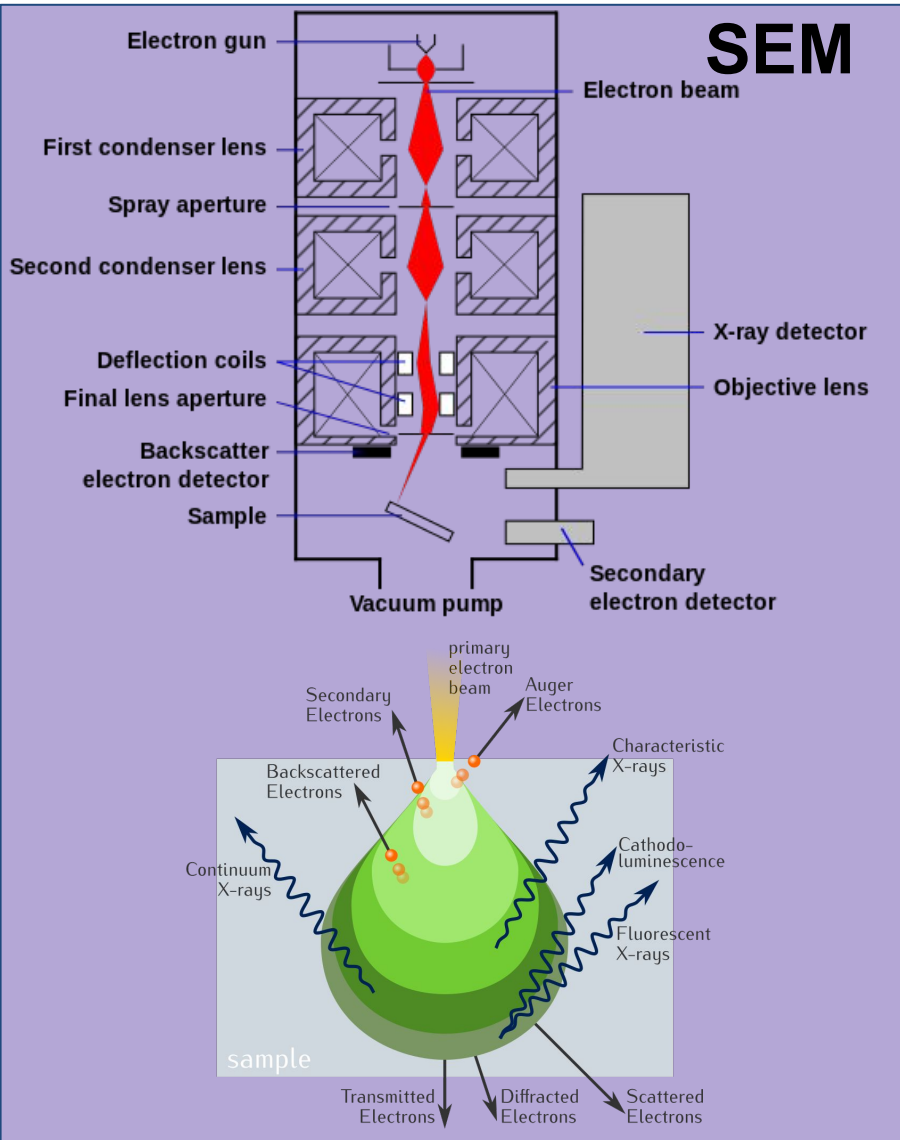


- Conduction Band edge moving at the interface
- Schottky barrier formation (comparing AuIn2 Fermi Energy with Conduction Band Edge)

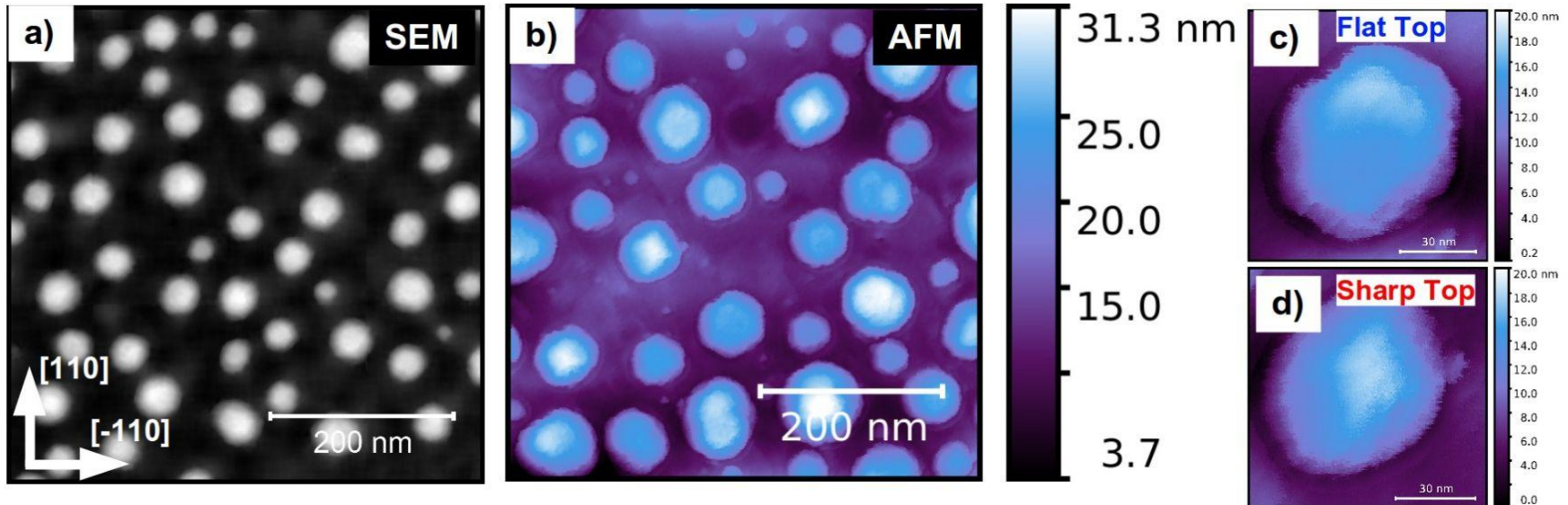


- No Conduction Band edge moving at the interface
- No Barrier - Ohmic contact (comparing AuIn2 Fermi Energy with Conduction Band Edge)

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SEM and AFM Measurements - AuIn₂ on InP(001)



Two morphological types of nanoelectrodes

Flat Top and Sharp Top