Advancing Nuclear Microprobe Analysis:

from 2D Elemental Maps to 3D Visualization with Machine Learning

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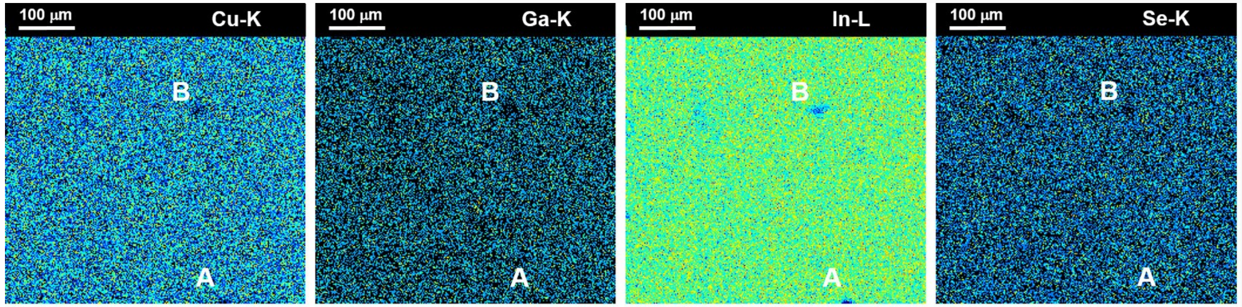
ICNMTA 2024 19th INTERNATIONAL CONFERENCE ON NUCLEAR MICROPROBE TECHNOLOGY AND APPLICATIONS

Outline

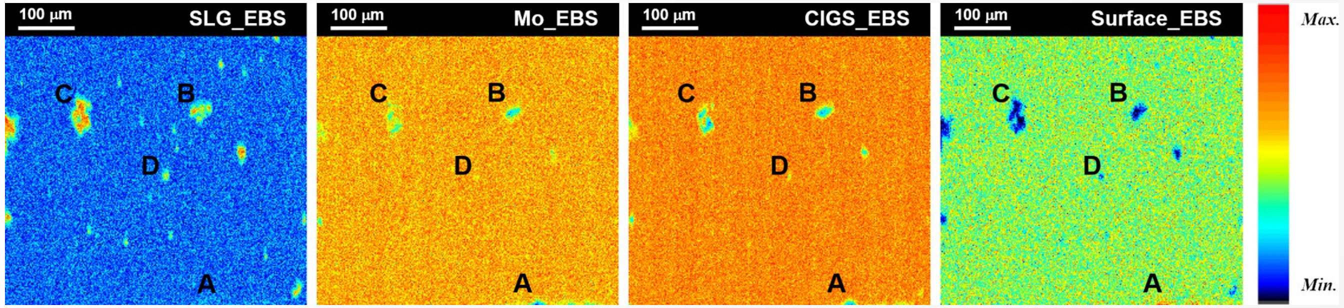
- Motivation
- State of the art
  - MORIA
  - Artificial Neural Networks
- Artificial Neural Networks and data from nuclear microprobe, challenges.
- Example: GaSb thermophotovoltaic cell
- Future and next developments

Motivation

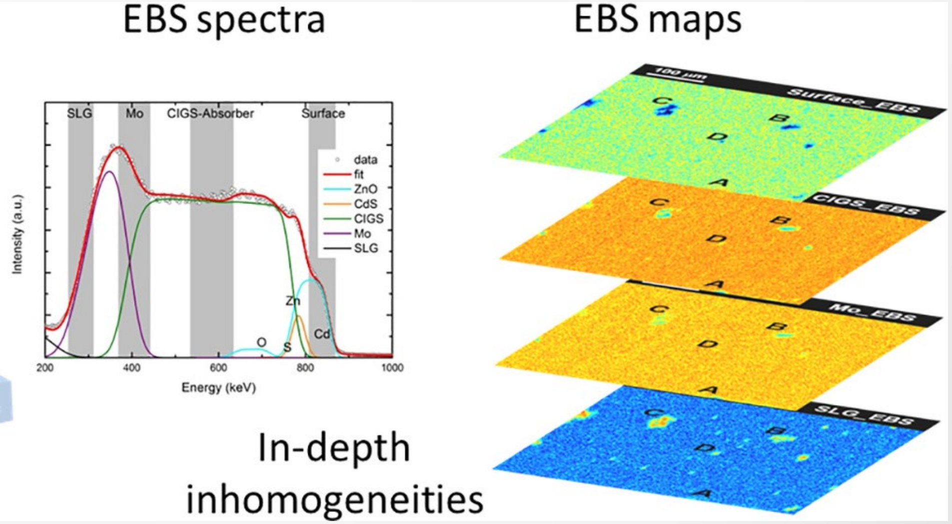
The nuclear microprobe allows the creation of 2D elemental distribution maps from regions of interest defined in the multiple spectra recorded during the experiment, such as RBS, PIXE, STIM, etc.



2D elemental distribution maps (530 × 530 µm2) from PIXE spectra for the elements present in a **CIGS** absorber layer (**Cu, In, Ga, and Se**)



2D maps (530 × 530 µm2) from **EBS spectra** (900 keV proton beam)



3D elemental distribution maps ???

In-Depth Inhomogeneities in CIGS Solar Cells
V. Corregidor, et al., *J. Phys. Chem. C* 2021, 125, 29, 16155–1616

State of the Art

Biophysical Journal Volume 104 April 2013 1419–1425 1419

High-Resolution 3D Imaging and Quantification of Gold Nanoparticles in a Whole Cell Using Scanning Transmission Ion Microscopy

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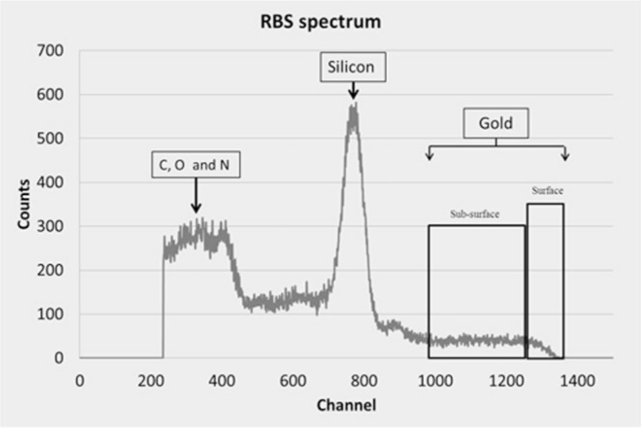
ABSTRACT Increasing interest in the use of nanoparticles (NPs) to elucidate the function of nanometer-sized assemblies of macromolecules and organelles within cells, and to develop biomedical applications such as drug delivery, labeling, diagnostic sensing, and heat treatment of cancer cells has prompted investigations into novel techniques that can image NPs within whole cells and tissue at high resolution. Using fast ions focused to nanodimensions, we show that gold NPs (AuNPs) inside whole cells can be imaged at high resolution, and the precise location of the particles and the number of particles can be quantified. High-resolution density information of the cell can be generated using scanning transmission ion microscopy, enhanced contrast for AuNPs can be achieved using forward scattering transmission ion microscopy, and depth information can be generated from elastically backscattered ions (Rutherford backscattering spectrometry). These techniques and associated instrumentation are at an early stage of technical development, but we believe there are no physical constraints that will prevent whole-cell three-dimensional imaging at <10 nm resolution.



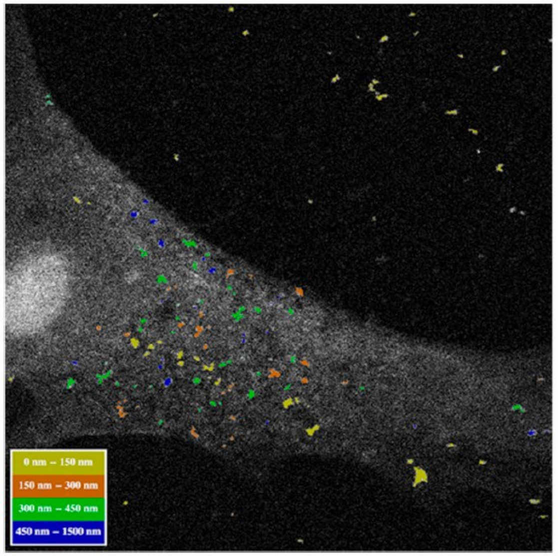


FIGURE 8 FSTIM image of the NP cell, using RBS depth information to color code the depth of the NPs and NP clusters within the cell; 0–150 nm represents the surface NPs.

State of the Art, MORIA

Journal of Microscopy

Journal of Microscopy, Vol. 267, Issue 2 2017, pp. 227–236 doi: 10.1111/jmi.12561
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3D map distribution of metallic nanoparticles in whole cells using MeV ion microscopy

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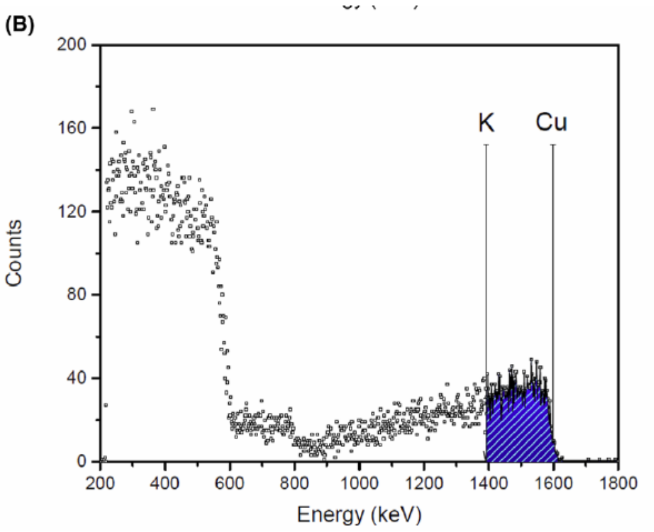
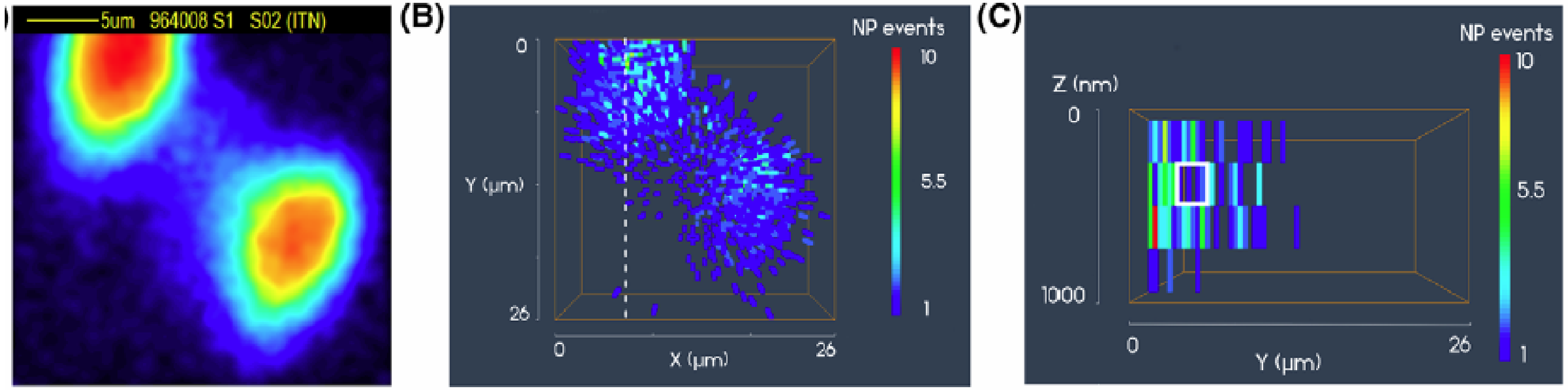
Summary

In this work, a new tool was developed, the MORIA program that readily translates Rutherford backscattering spectrometry (RBS) output data into visual information, creating a display of the distribution of elements in a true three-dimensional (3D) environment.

The program methodology is illustrated with the analysis of yeast *Saccharomyces cerevisiae* cells, exposed to copper oxide nanoparticles (CuO-NP) and HeLa cells in the presence of gold nanoparticles (Au-NP), using different beam species, energies and nuclear microscopy systems. Results demonstrate that for both cell types, the NP internalization can be clearly perceived. The 3D models of the distribution of CuO-NP in *S. cerevisiae* cells indicate the nonuniform distribution of NP in the cellular environment and a relevant confinement of CuO-NP to the cell wall. This suggests the impenetrability of certain cellular organelles or compartments for NP. By contrast, using a high-resolution ion beam system, discretized agglomerates of Au-NP were visualized inside the HeLa cell. This is consistent with the mechanism of entry of these NPs in the cellular space by endocytosis enclosed in endosomal vesicles. This approach shows RBS to be a powerful imaging technique assigning to nuclear microscopy unparalleled potential to assess nanoparticle distribution inside the cellular volume.

State of the Art, MORIA





State of the Art, Artificial Neural Networks

- N.P. Barradas and A. Vieira are pioneers in use Neural Networks to classify RBS spectra.

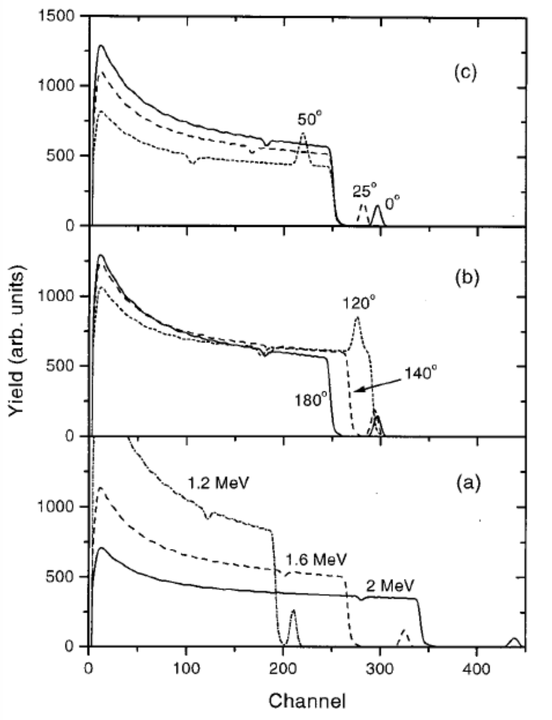


FIG. 2. Spectra calculated for different experimental conditions for a 25-Å-thick Ge δ layer located under a 400-nm-thick Si layer: (a) Beam energy E_0=1.2, 1.6, and 2 MeV. (b) Scattering angle α_{scatt}=120°, 140°, and 180°. (c) Angle of incidence θ_{inc}=0° (normal incidence), 25°, and 50°.

PHYSICAL REVIEW E VOLUME 62, NUMBER 4 OCTOBER 2000

Artificial neural network algorithm for analysis of Rutherford backscattering data

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(Received 5 May 2000)

Rutherford backscattering (RBS) is a nondestructive, fully quantitative technique for accurately determining the compositional depth profile of thin films. The inverse RBS problem, which is to determine from the data the corresponding sample structure, is, however, in general ill posed. Skilled analysts use their knowledge and experience to recognize recurring features in the data and relate them to features in the sample structure. This is then followed by a detailed quantitative analysis. We have developed an artificial neural network (ANN) for the same purpose, applied to the specific case of Ge-implanted Si. The ANN was trained with thousands of constructed spectra of samples for which the structure is known. It thus learns how to interpret the spectrum of a given sample, without any knowledge of the physics involved. The ANN was then applied to experimental data from samples of unknown structure. The quantitative results obtained were compared with those given by traditional analysis methods and are excellent. The major advantage of ANNs over those other methods is that, after the time-consuming training phase, the analysis is instantaneous, which opens the door to automated on-line data analysis. Furthermore, the ANN was able to distinguish two different classes of data which are experimentally difficult to analyze. This opens the door to automated on-line optimization of the experimental conditions.

PACS number(s): 07.05.Mh, 82.80.Yc, 07.05.Kf, 68.55.Nq

Just one of the multiple publications…

State of the Art, Artificial Neural Networks

- Also in the IBA&PIXE-SIMS 2021 conference, the use and efficacy of Neural Networks were also discussed. ...

IBA&PIXE-SIMS 2021
11-15 October 2021

An artificial neural network algorithm for the simultaneous analysis of multi-detector RBS depth profiling
Goele Magchiels, KU Leuven, Germany

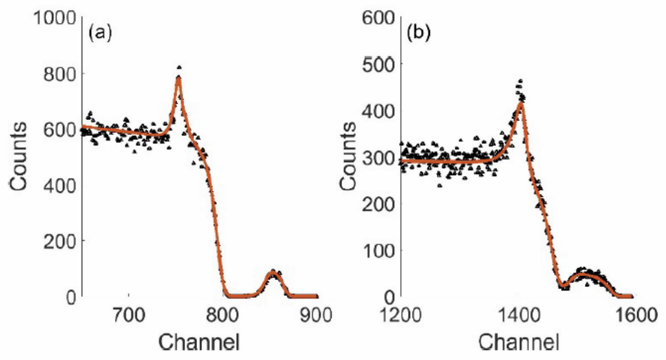
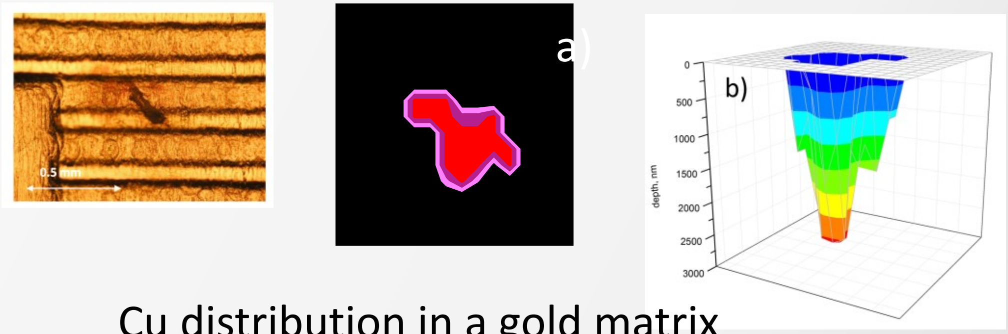


Fig. 1: RBS spectra of Ni/$Ge_{0.914}Sn_{0.086}$/Ge after deposition measured in (a) backscattering geometry and (b) glancing geometry. The experi- mental data are represented by the black triangles, the simulations based on the ANN output by the red solid line.

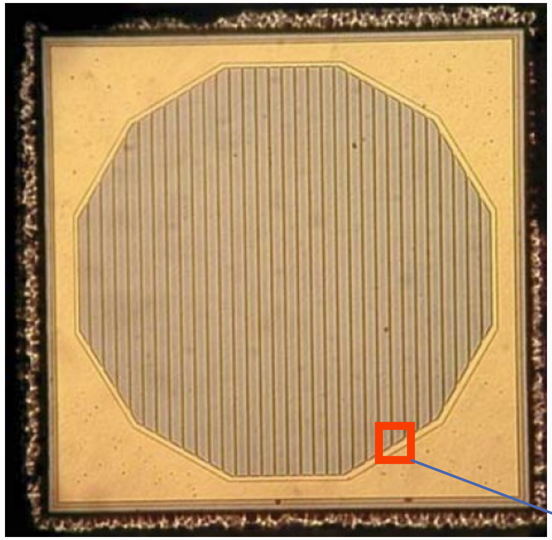
P24 **Deep Convolutional Neural Networks applied to nuclear microprobe data**
Victoria Corregidor, C2TN /DECN, IST-Ulisboa, Portugal



Cu distribution in a gold matrix

In both cases, again, only RBS data were considered.

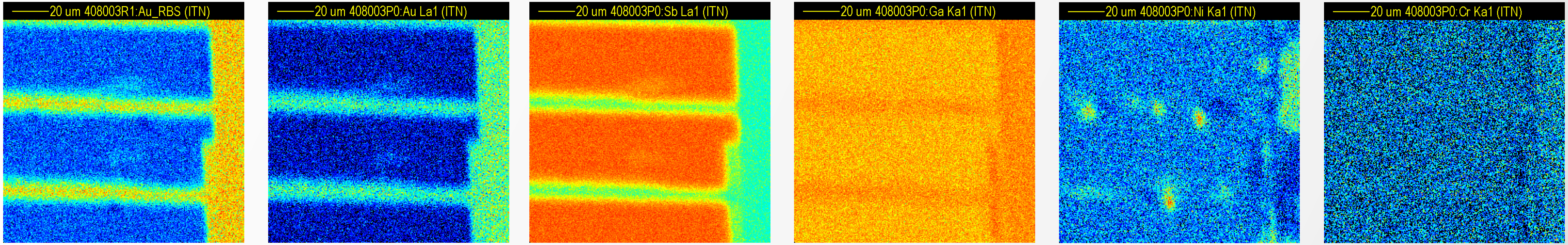
Results: Thermophotovoltaic GaSb cell



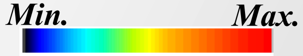
TPV cell: 2 x 2 mm^2
The front grid metallization:
finger width: ~ 10 μm
Evaporation of: 5 nm Cr/ 25nm Au/ 60 nm Ni/ 1 μm Au

3D map for Au?

2D elemental maps, 130x130 μm^2 from RBS and PIXE spectra recorded during 6 hours.

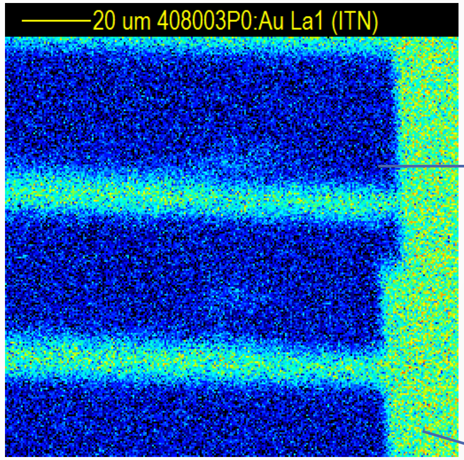


Au-RBS | Au-L | Sb-L | Ga-K | Ni-K | Cr-K



Artificial Neural Networks and nuclear microprobe

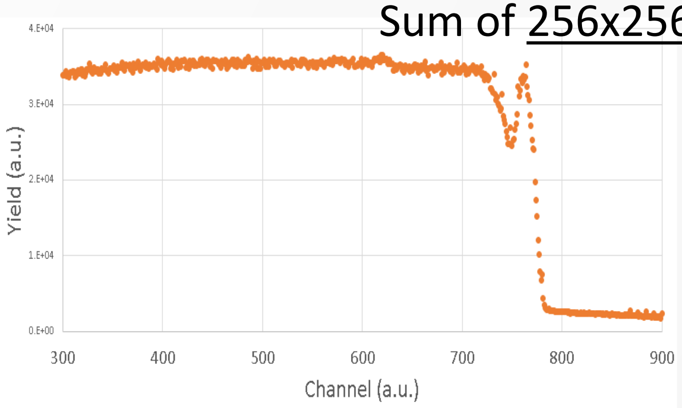
Although the overall RBS spectra (the sum of all pixel spectra) may show good counting statistics, when each pixel is considered individually, the corresponding single spectra usually have rather poor counting statistics.

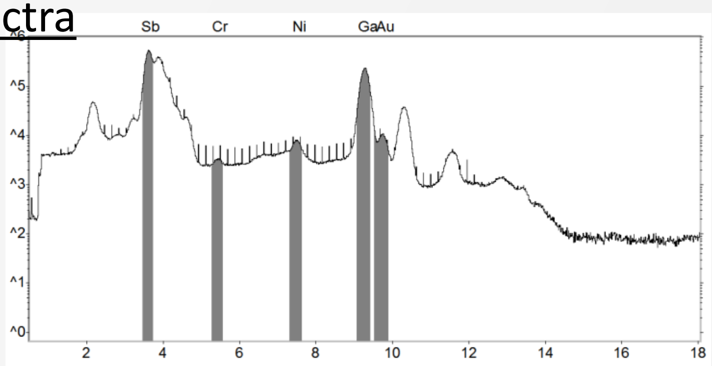


Run time: 6 hours

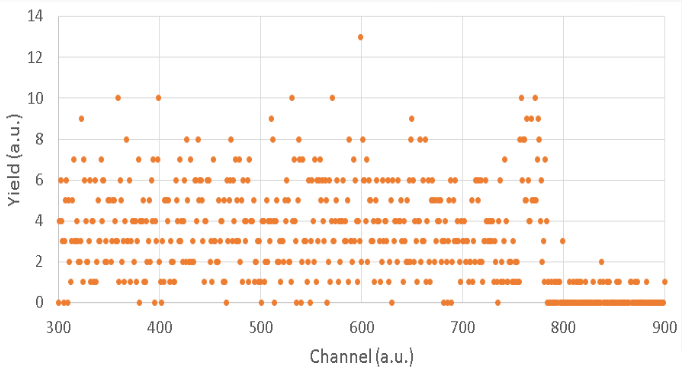
256x256=65 536 spectra

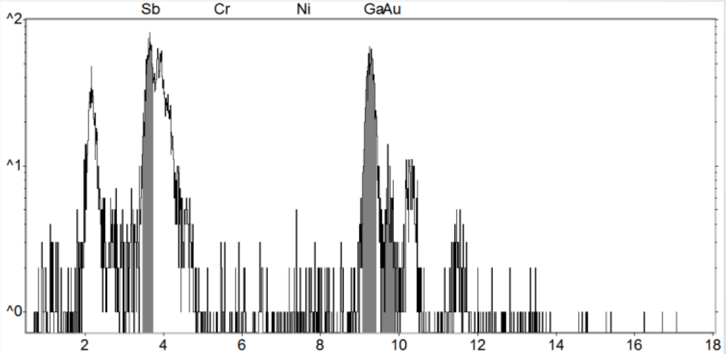
Sum of <u>256x256 RBS and PIXE spectra</u>





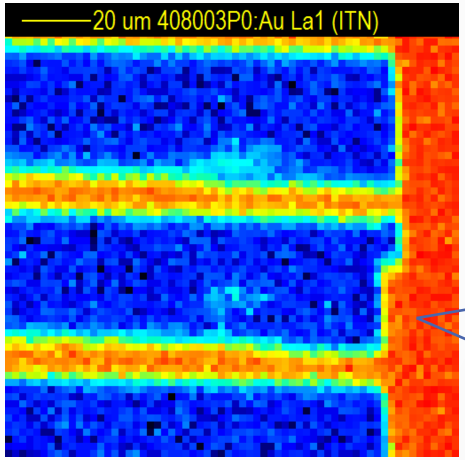
Sum of <u>2 pixels RBS and PIXE spectra</u>



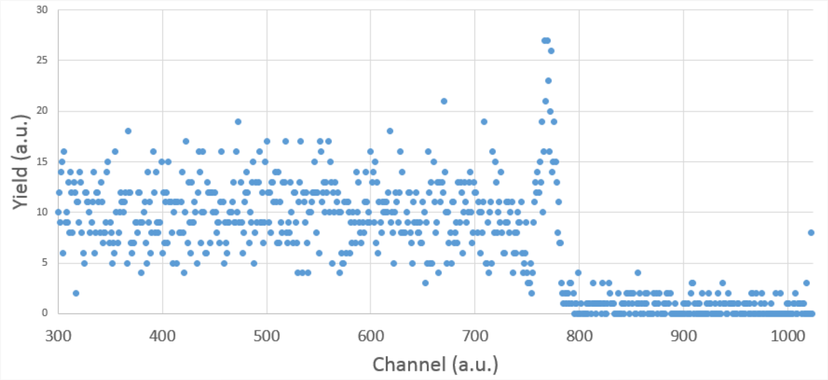


Artificial Neural Networks and nuclear microprobe

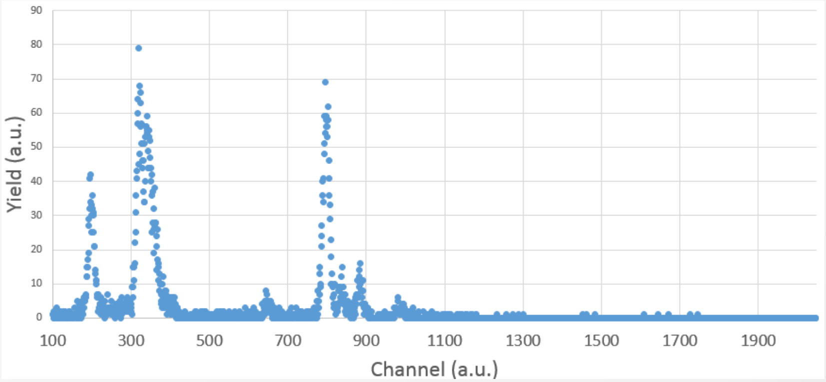
To increase the statistic of the spectra, the raw data should be pre-processed. The data were 4x4 compressed.



Sum of 4x4 pixels RBS spectra

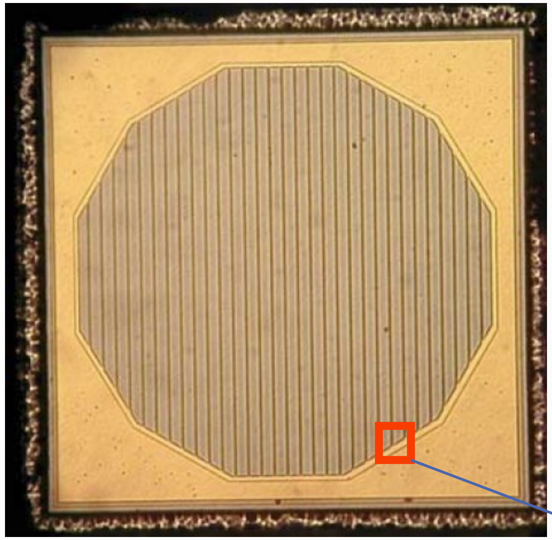


Sum of 4x4 pixels PIXE spectra



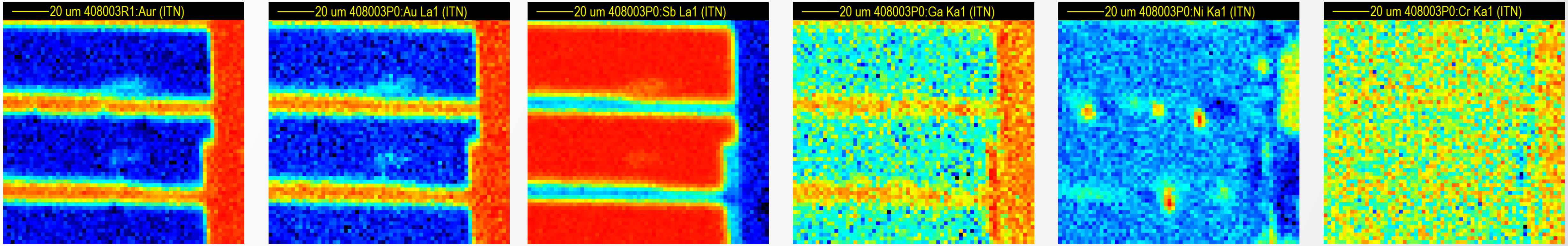
64x64=4 096 spectra

Results: Thermophotovoltaic GaSb cell



TPV cell: 2 x 2 mm2
The front grid metallization:
finger width: ~ 10 um
Evaporation of: 5 nm Cr/ 25nm Au/ 60 nm Ni/ 1 um Au

2D elemental maps, 130x130 μm^2 from RBS and PIXE spectra, 4x4 compressed.

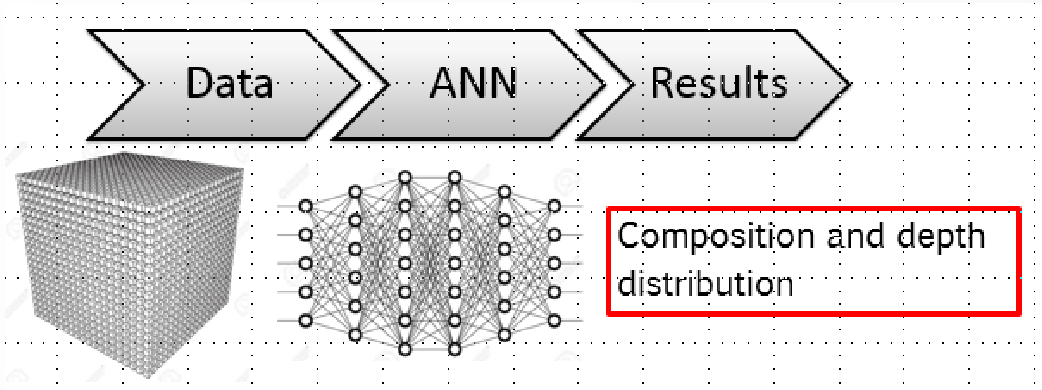


Au-RBS | Au-L | Sb-L | Ga-K | Ni-K | Cr-K

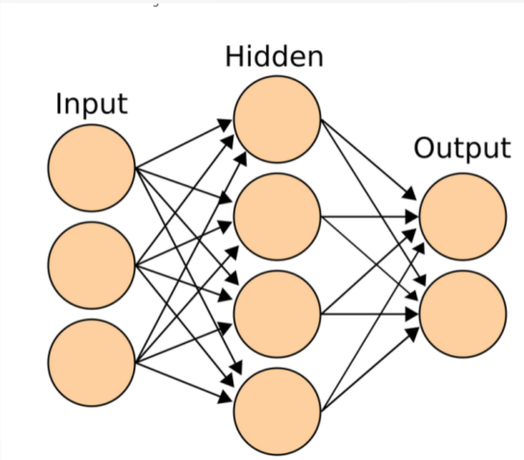
Min. *Max.*

Artificial Neural Networks and nuclear microprobe

Typically, data from each area scanned by a nuclear microprobe is acquired as a 256 x 256 x *n* pixel matrix, each pixel containing *n* of the IBA spectra recorded during the experiment.



Neural networks

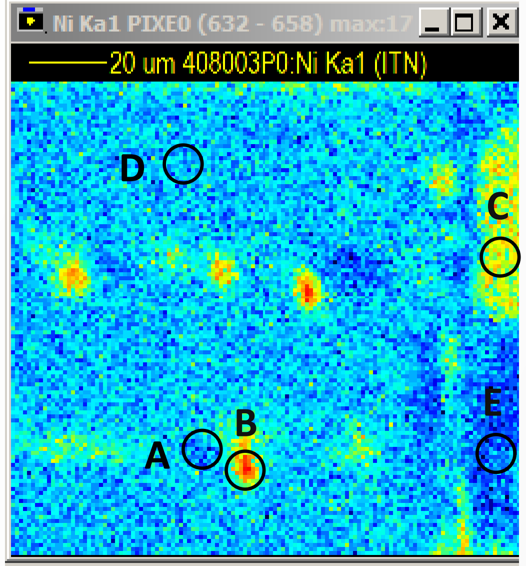


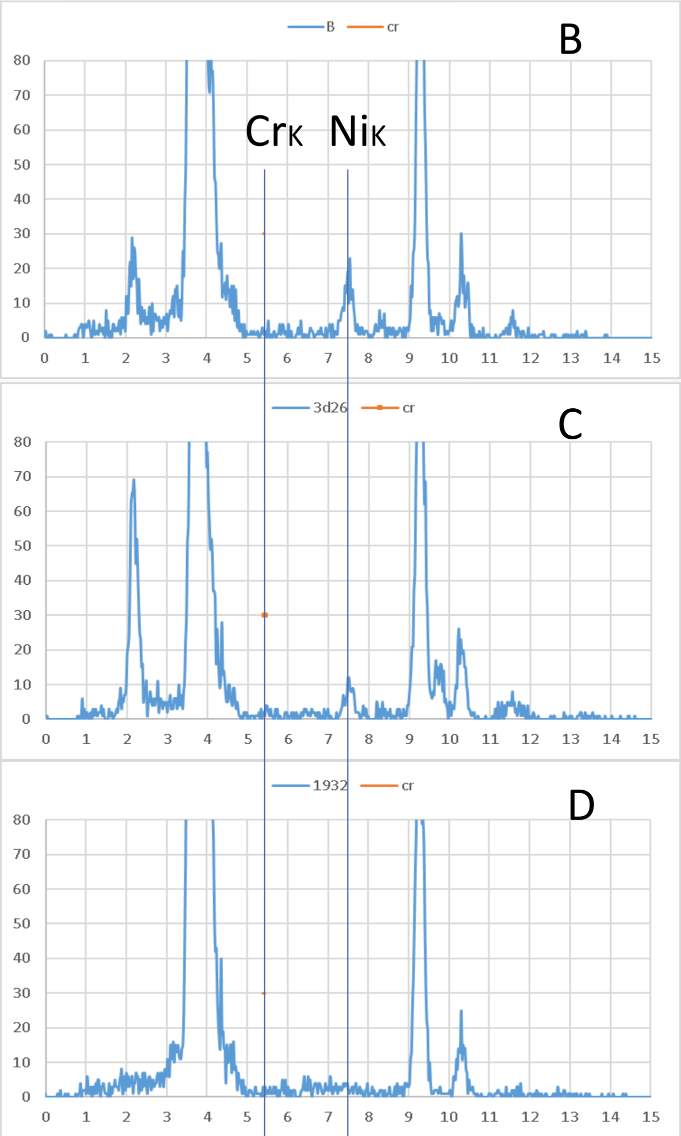
Input data: simulated RBS spectra (WiNDF) + noise; Real PIXE spectra (5) + noise (thousands)

Output data: Au thickness of the Au layer and Ni+Cr distribution.

Hidden layers, number of input data….. Parameters to be adjusted.

Artificial Neural Networks: PIXE input data

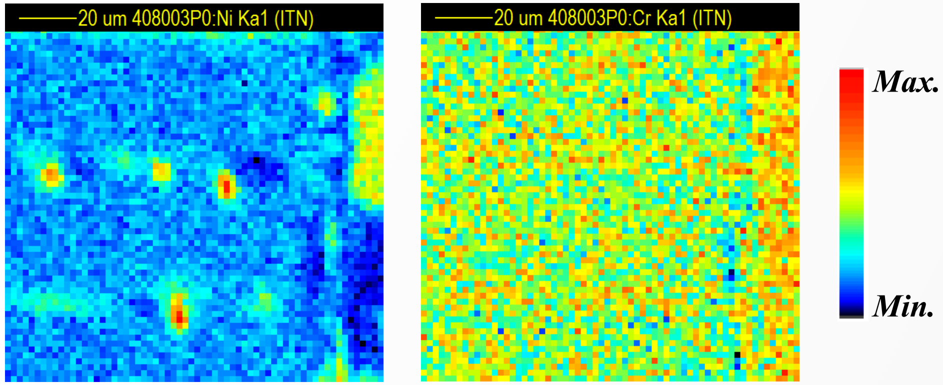




input data:
5 real PIXE spectra, which are “representative”, add noise to generate thousands of PIXE spectra to train the networks

Output data:
Qualitative 2D maps for **Cr** and **Ni** distribution

Artificial Neural Networks: 2D maps using PIXE spectra

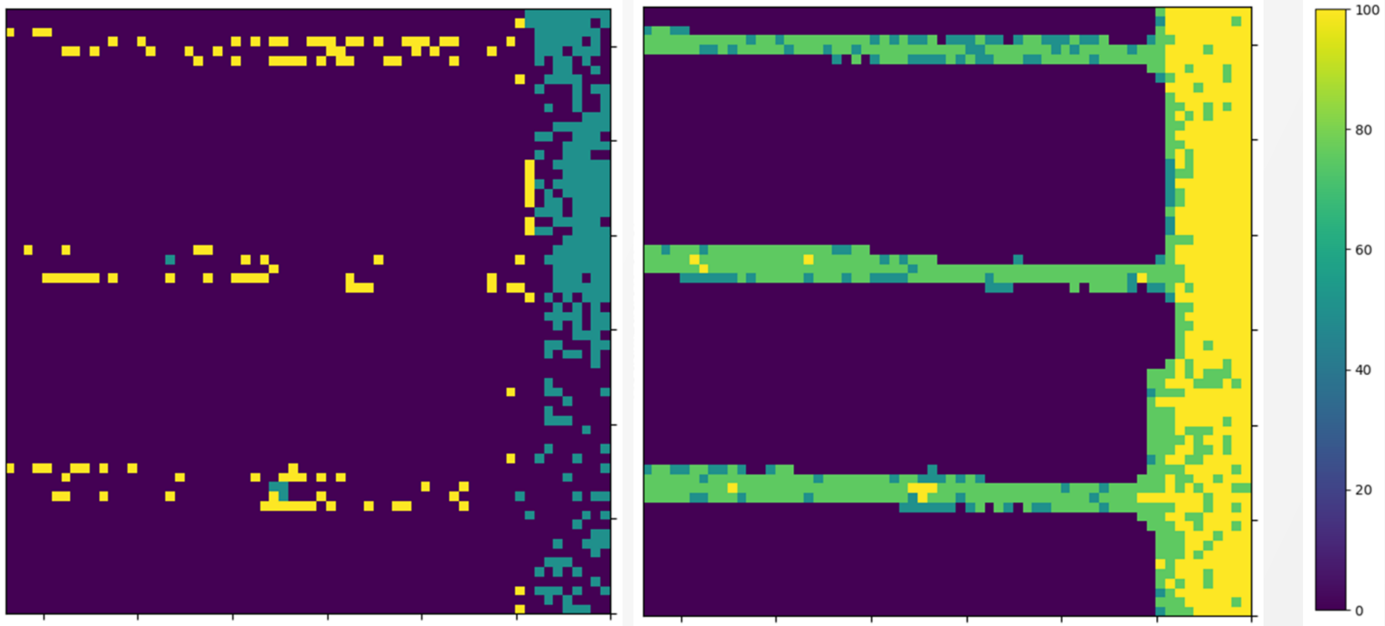


Ni-K Cr-K

Using OMDAQ sotware

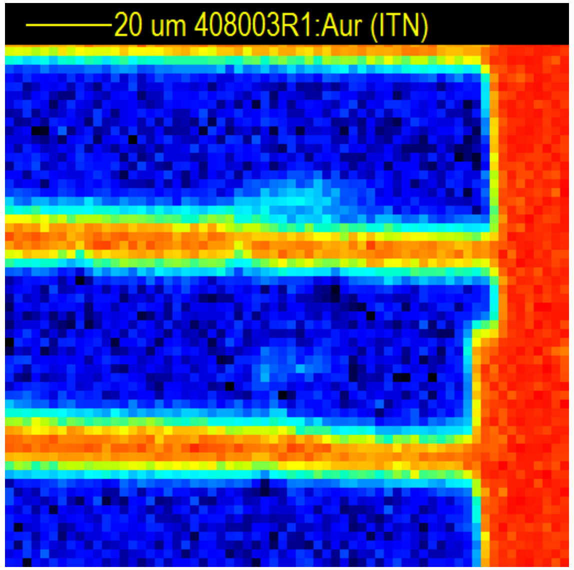
The neural networks are able to reproduce 2D elemental maps and, in some cases, provide better visualization, as in the case of Cr distribution.

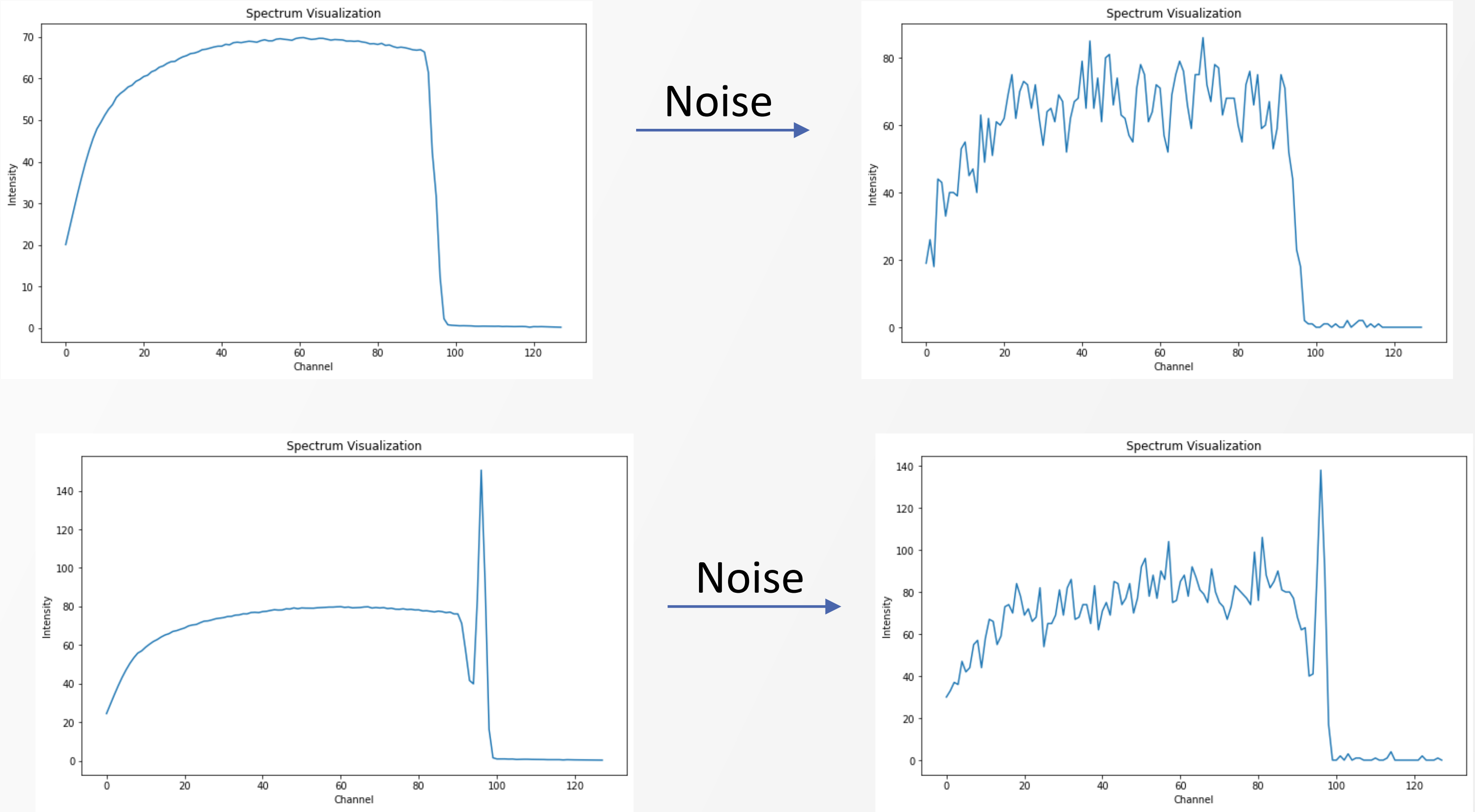
Ni-K Cr-K



130x130 μm^2

Artificial Neural Networks: RBS input data

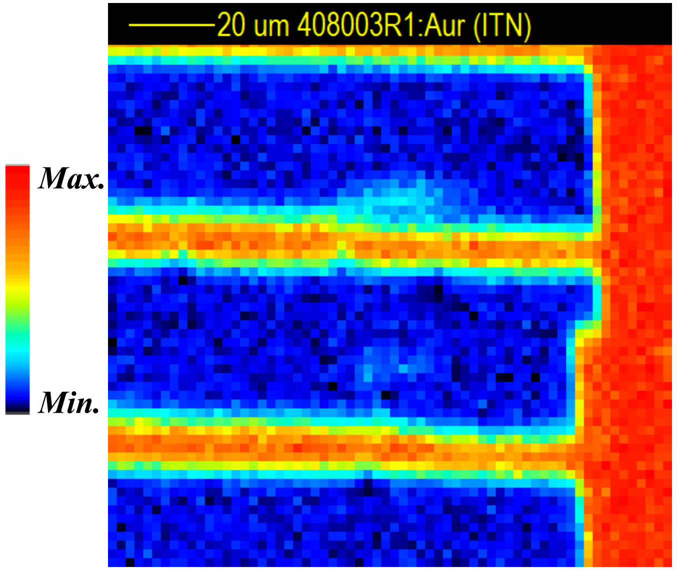




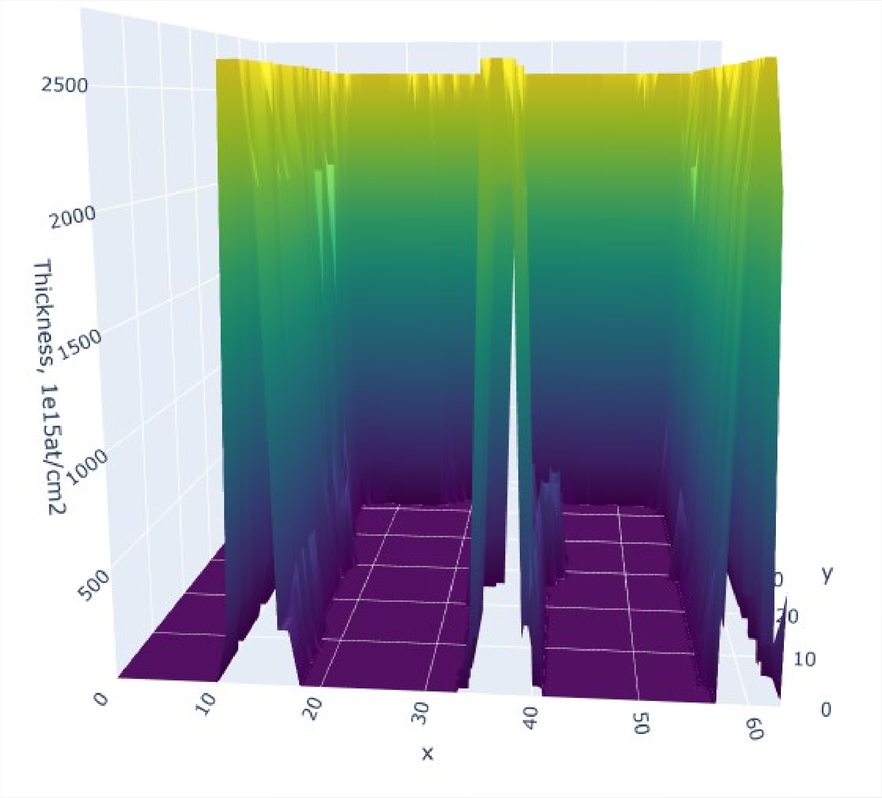
input data:
10 simulated RBS spectra, which consider different gold thickness values, add noise to generate thousands of RBS spectra to train the networks

Output data:
Quantitative 3D maps for **Au** distribution

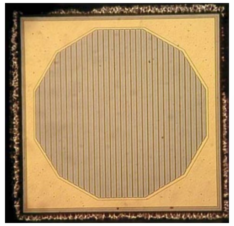
Artificial Neural Networks: 3D map using RBS spectra



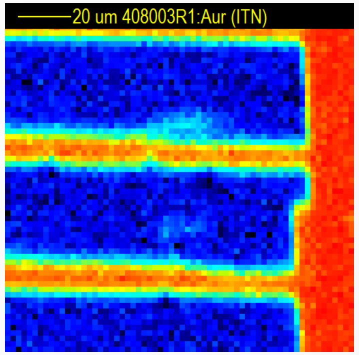
Output data:
Quantitative 3D maps for **Au** distribution

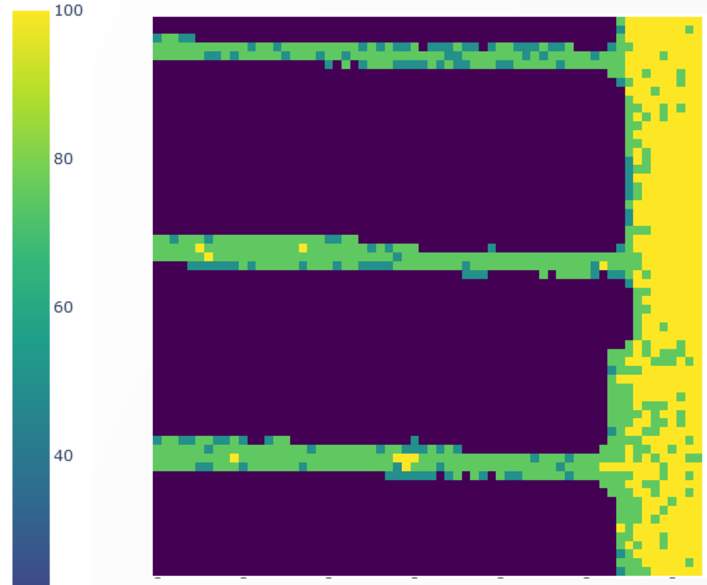


Artificial Neural Networks: 3D map using PIXE and RBS spectra



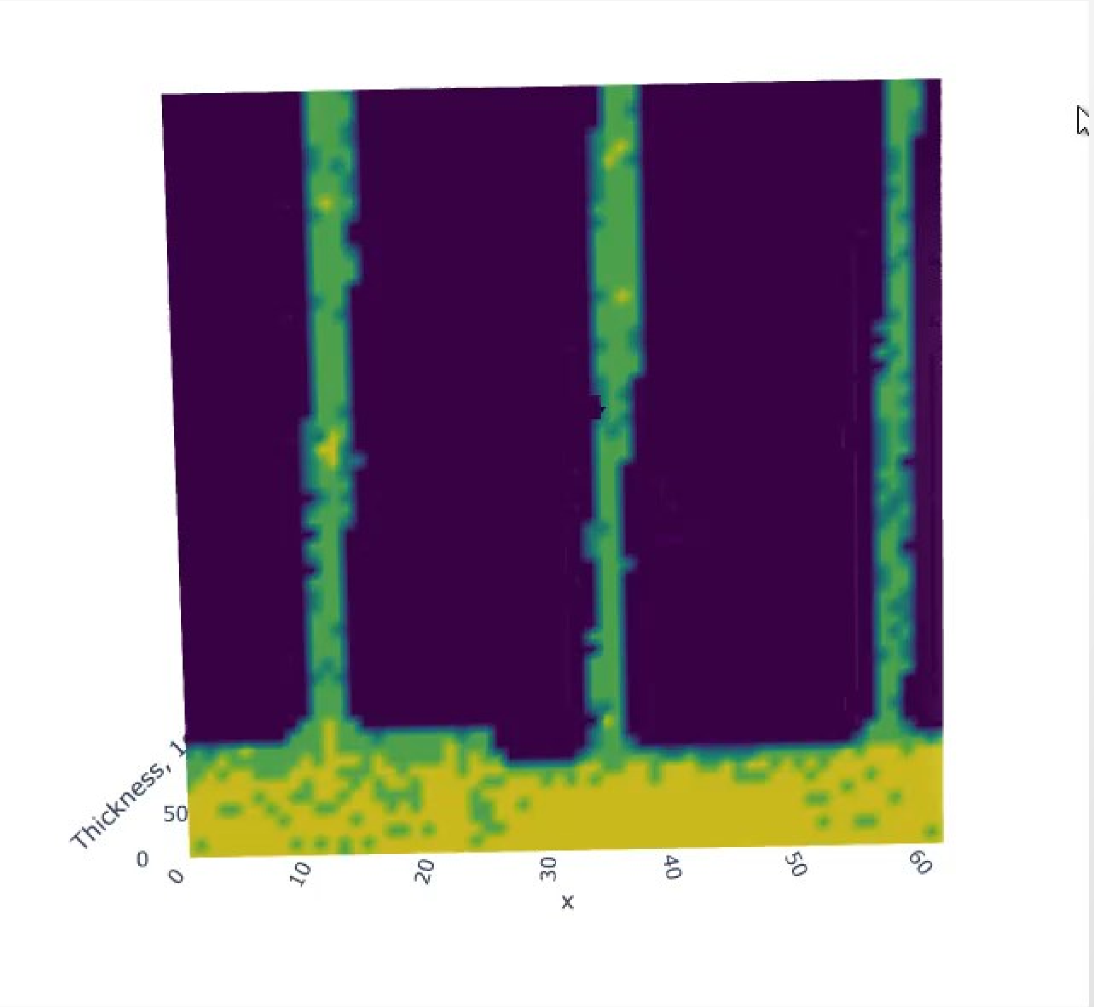
Front grid metallization:
finger width: ~ 10 μm
Evaporation of: Cr/ Au/ Ni/ Au





Cr + Ni distribution

$130x130\ \mu m^2$



Future, next work

- Explore methods to create artificial PIXE spectra to avoid using real PIXE spectra for training the networks. All ideas are welcome!

- Compress the RBS and PIXE spectra to increase statistics.

- Try Convolutional Neural Networks (which are ideal to classify images) to analyse the spectra

- Reduce the time needed to obtain the experimental data (hours)

- Consider that each case may require a dedicated neural network

- It is possible to have a general neural network?

Computational resources: FCT project: 2023.10769.CPCA

INCD National Distributed Computing Infrastructure

Thanks for your attention

V. Corregidor, N.P. Barradas, L.C. Alves

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