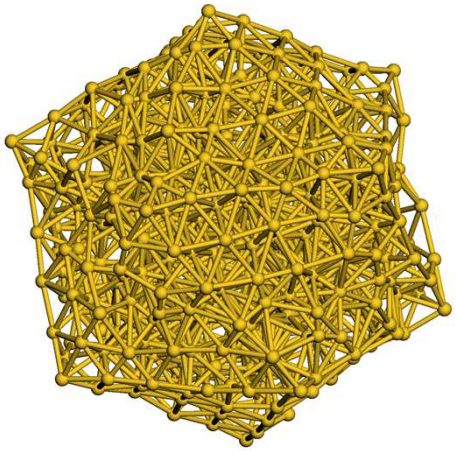
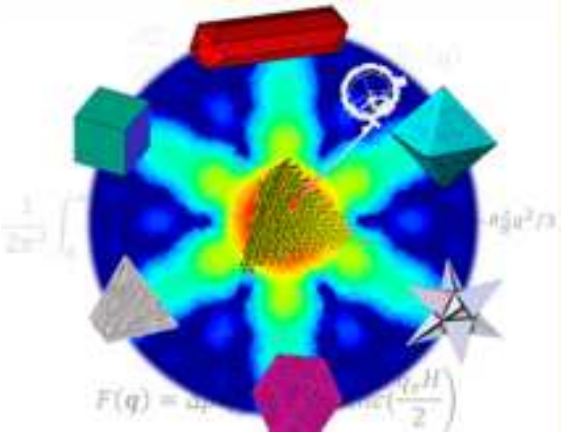
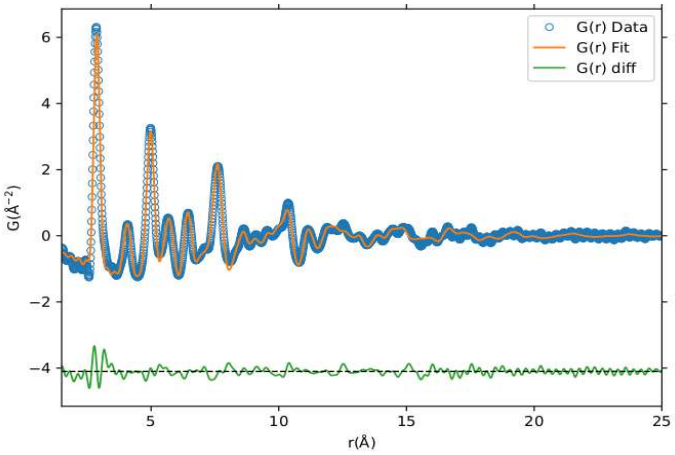
Machine learning assisted structural characterization of nanoclusters using X Ray scattering







N. Ratel-Ramond – S. Cayez

X-Ray scattering techniques

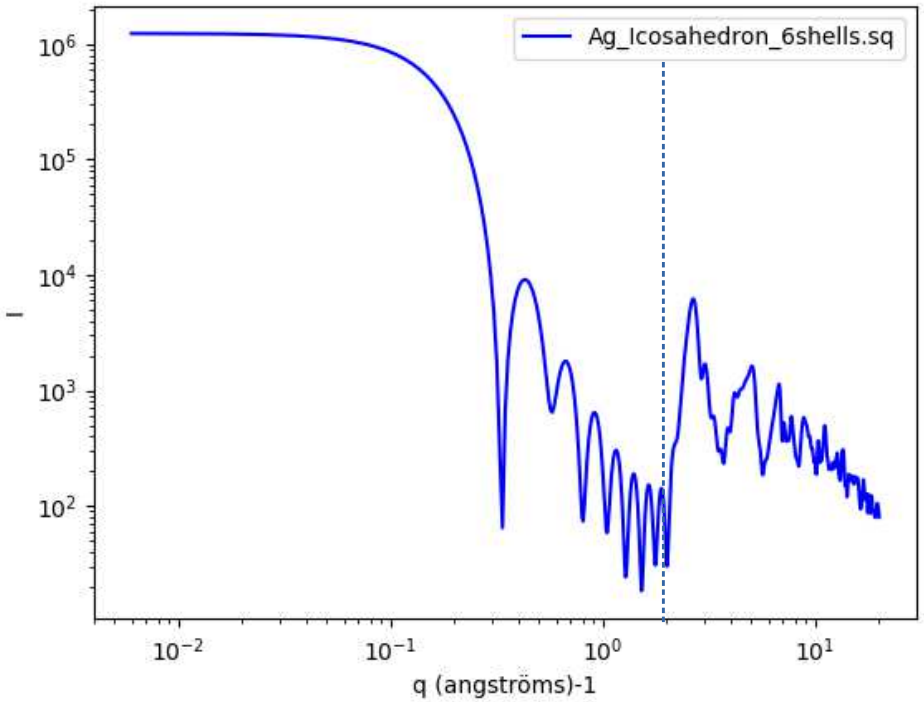
Q<1Å^{-1} : Small Angle Region (SAXS)

Q>1Å-1 : Wide Angle Region (WAXS`

- particle size

- particle shape

- arrangement of particles



X-Ray scattering techniques

Q<1Å^{-1} : Small Angle Region (SAXS)

1 : Wide Angle Region (WAXS)

- particle size

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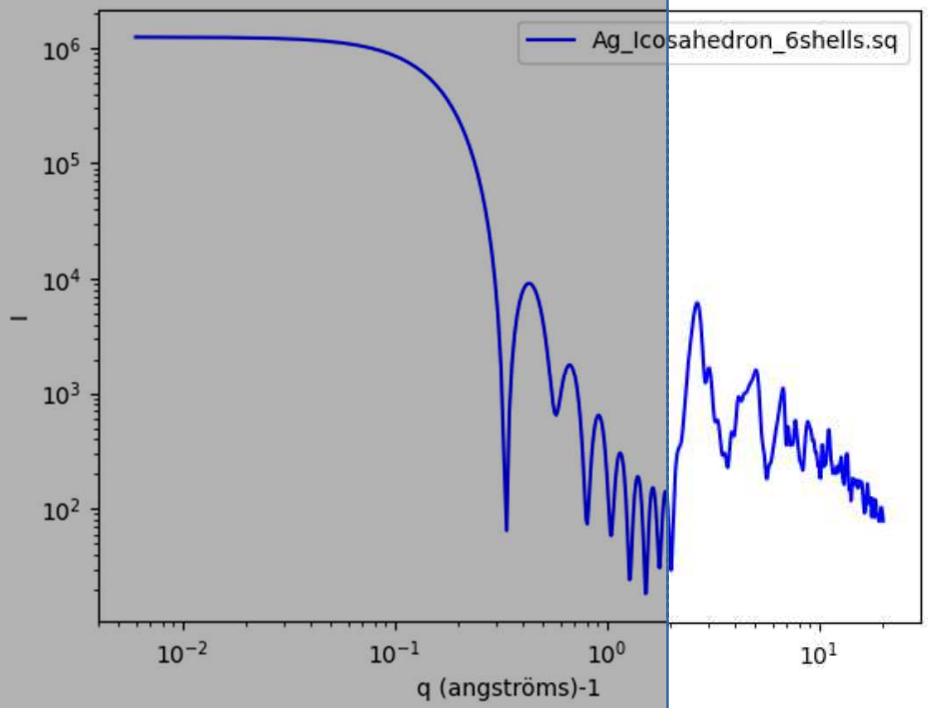


Q>1Å-

X-Ray scattering techniques: WAXS

RECIPROCAL SPACE

REAL SPACE

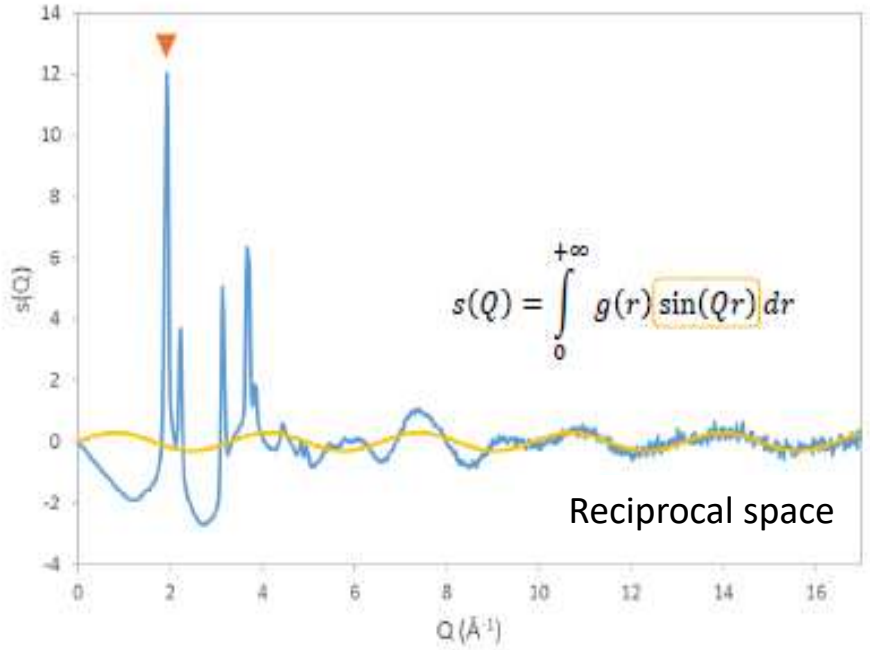
$I(Q)$
$\rightarrow S(Q)$
$F(Q)$

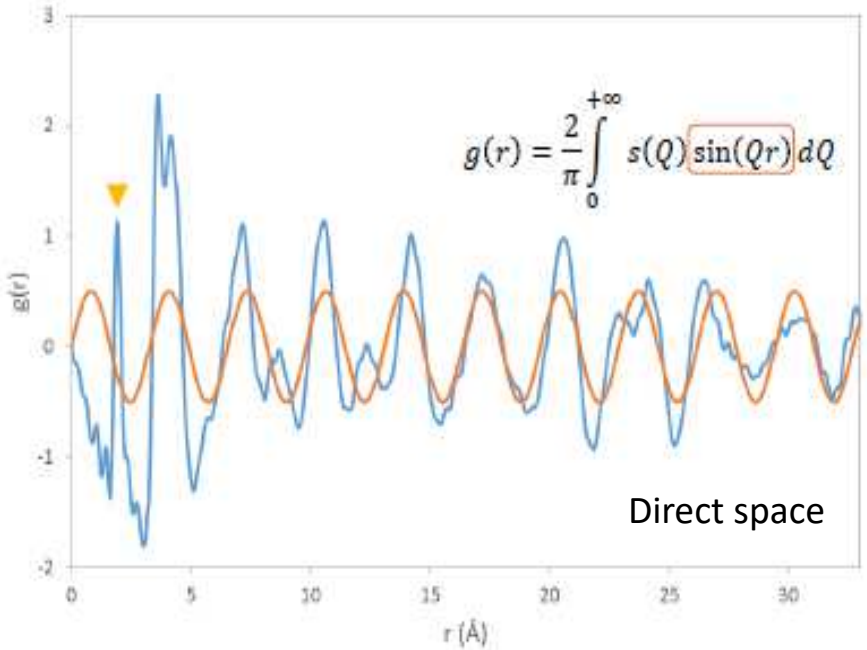
$Q = \frac{4\pi sin(\theta)}{\lambda}$

Fourier transform

$g(r)$

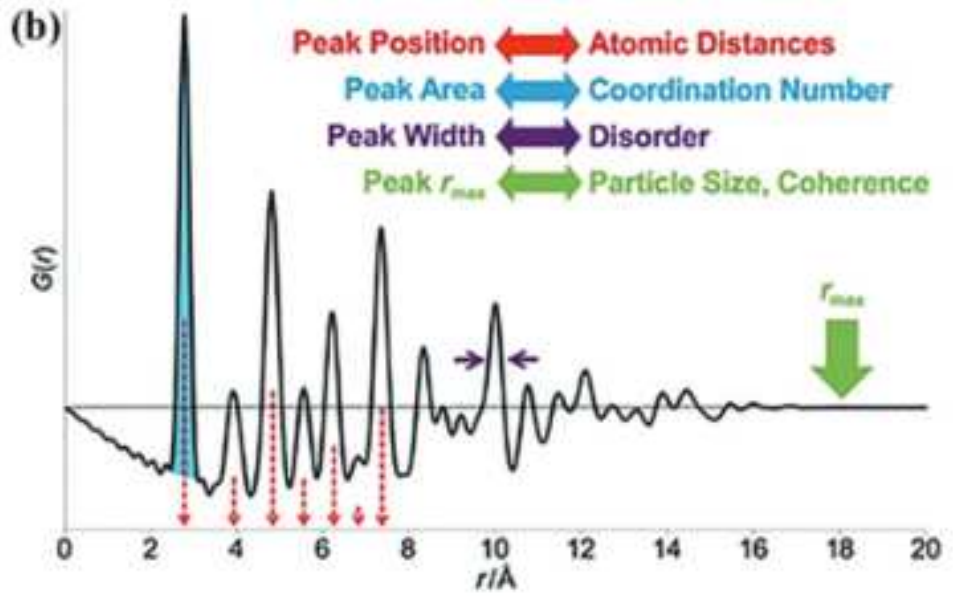
$G(r)$





X-Ray scattering techniques: WAXS

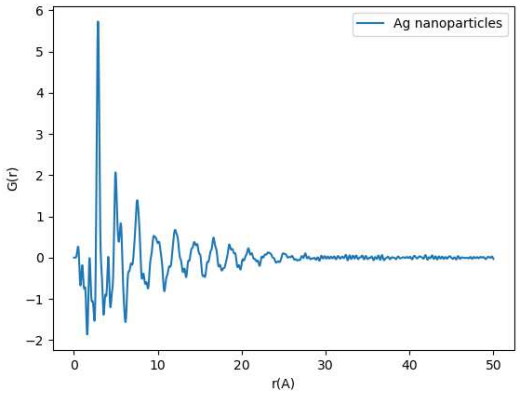
- What type of information in a PDF ?
  - Atomic distances
  - Coordination number
  - Disorder
  - Particle size



Structural information at the atomic scale

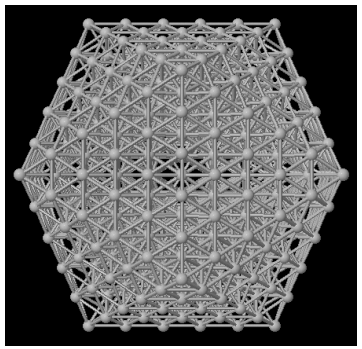
X-Ray scattering techniques: WAXS

◆ <u>Example:</u> Ag nanoparticles (R. Parmar, L.M. Lacroix, G. Viau)



Experimental PDF does not match with cubic structure

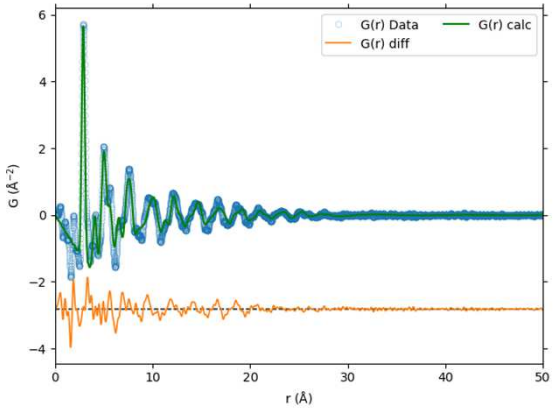
Build model using ASE Python library



Icosahedral model with 6 layers

Refine model using diffpy-cmi python library

Adjust the parameters of the model



Best Rw=0,23

X-Ray scattering techniques: SAXS

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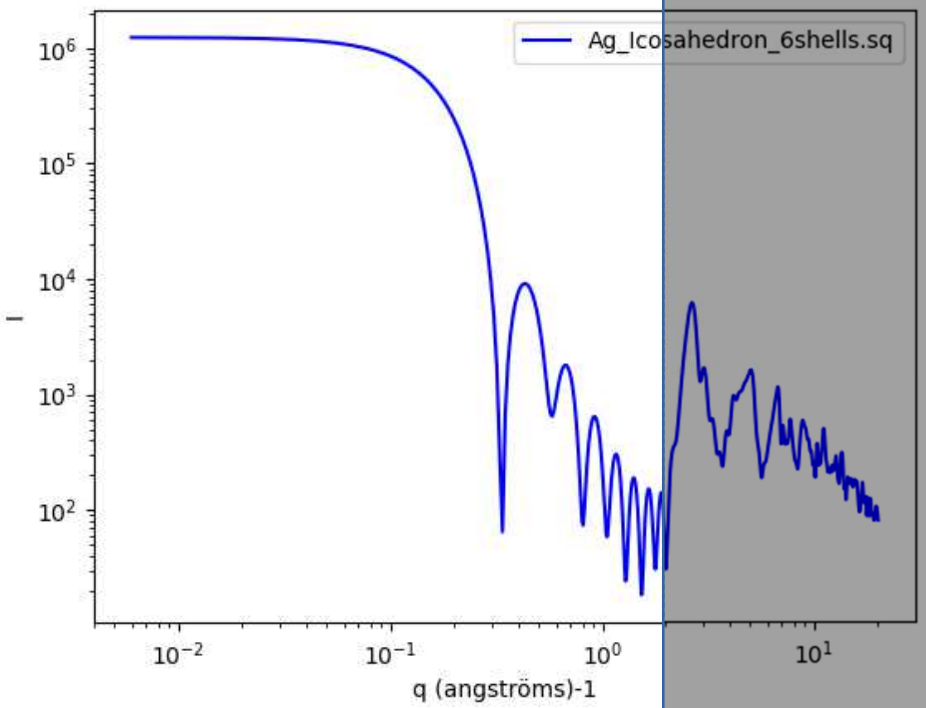
- particle size

- particle shape

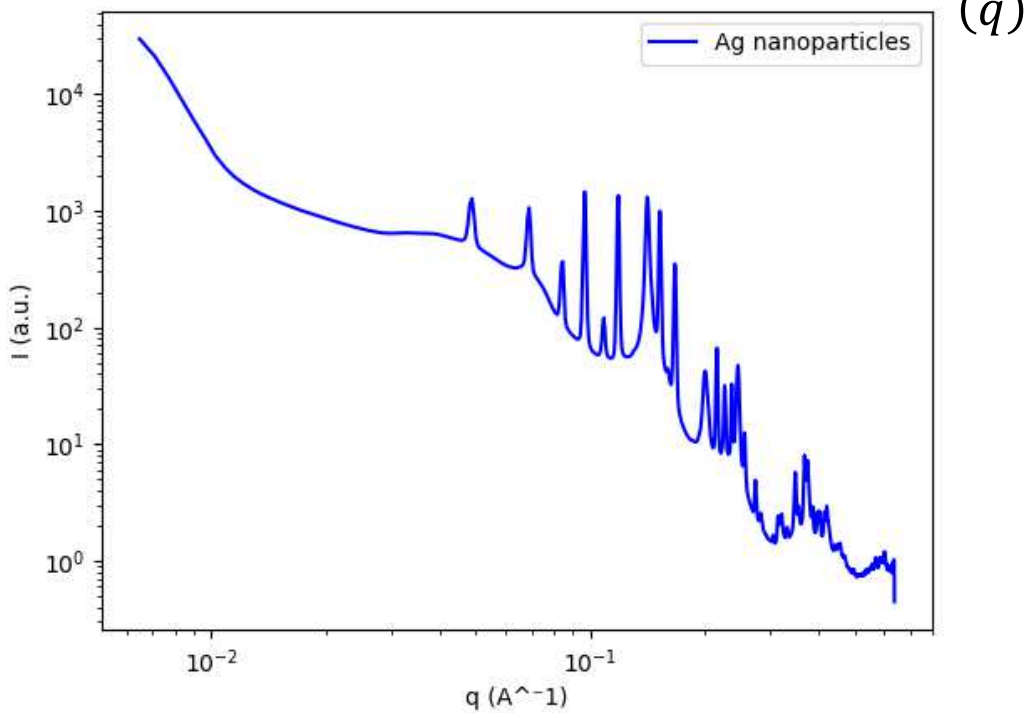
- arrangement of particles



X-Ray scattering techniques: SAXS

- Theoretical Aspects : the general equation of SAXS has the following expression
-

$$I(q)$$



- Information about:
  - Size
  - Size distribution
  - Shape
  - Surface structure
  - Arragement of particles

DOI: 10.1021/acs.chemrev.5b00690

X-Ray scattering techniques: SAXS

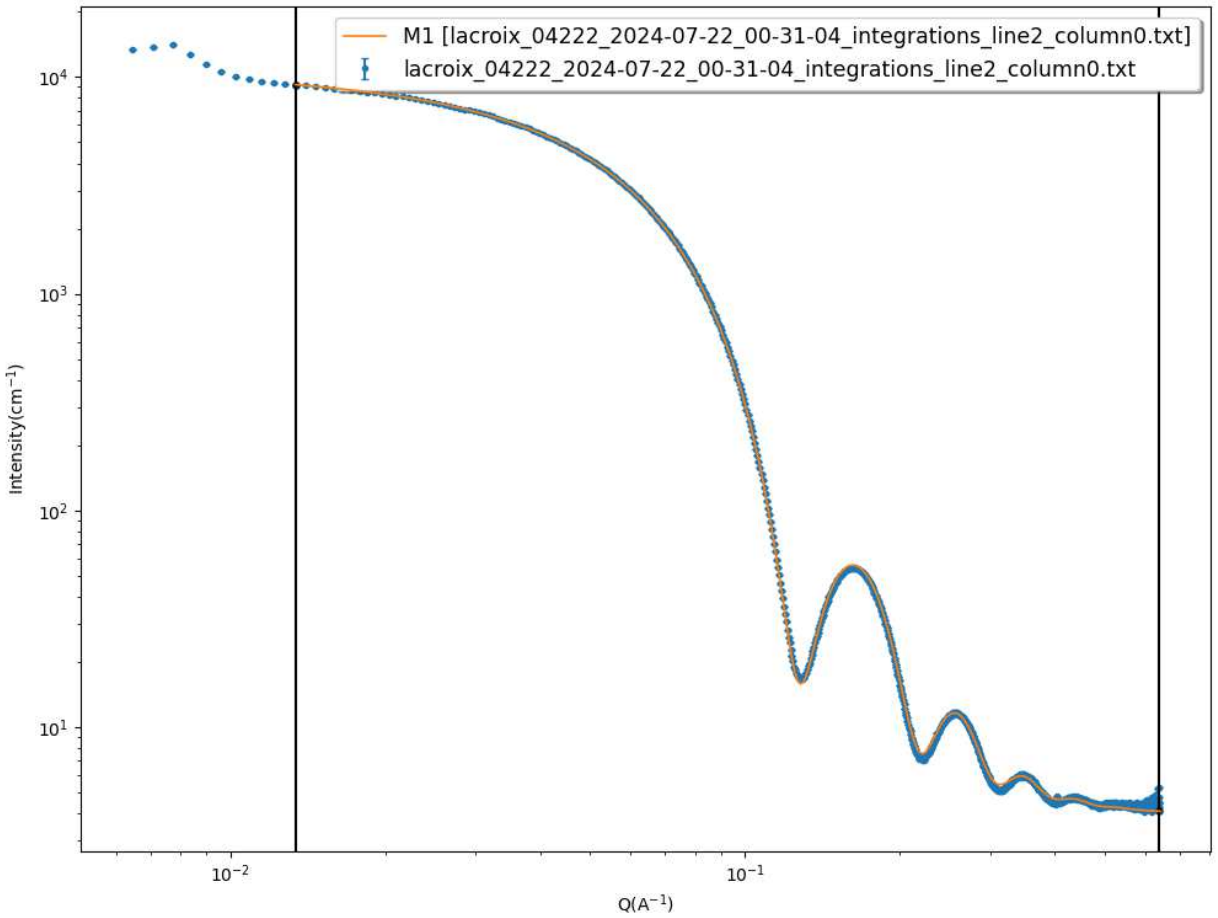
1. Form factor analysis

For dilute systems

Example of Au nanoparticles:

Model: sphere

Radius=34.907 ± 0.071 Å,
polydispersity ratio= 0.062 ± 0.004
(gaussian)



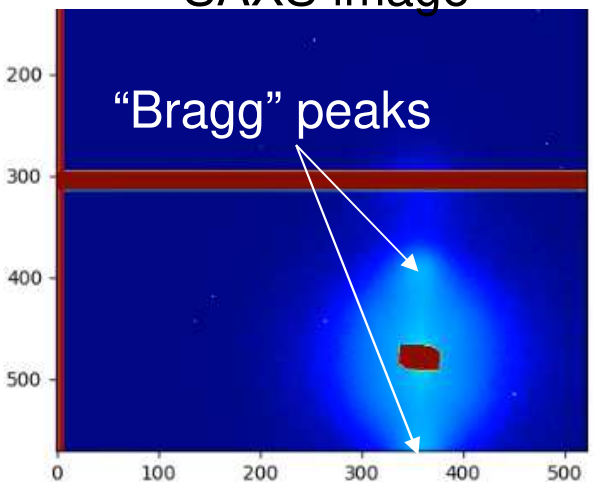
A. Pham, C. Chinaud-Chaix, S. Tricard

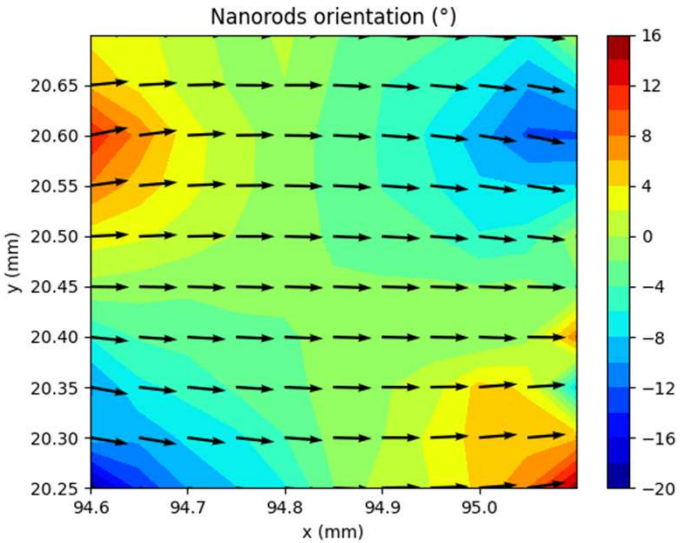
X-Ray scattering techniques: SAXS

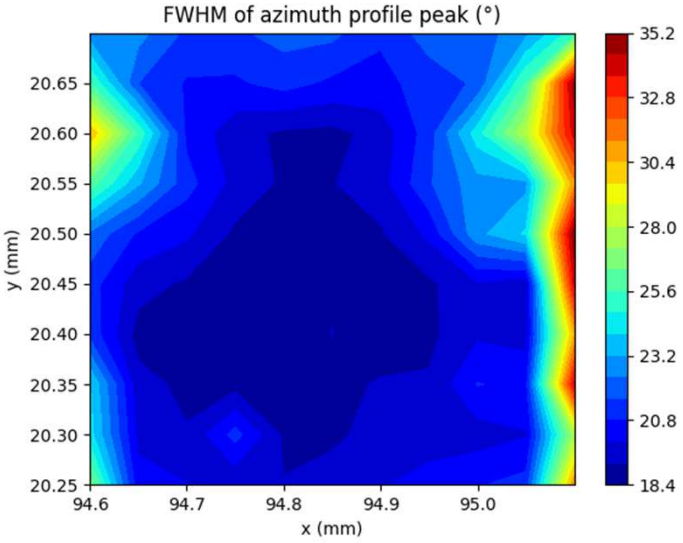
2. Structure factor analysis: Crystalline assemblies – lattice indexing

Example of µSAXS investigation of micromagnets built by magnetophoresis from Co nanorods (L.M. Lacroix, G. Viau)

SAXS image







X-Ray scattering techniques: SAXS

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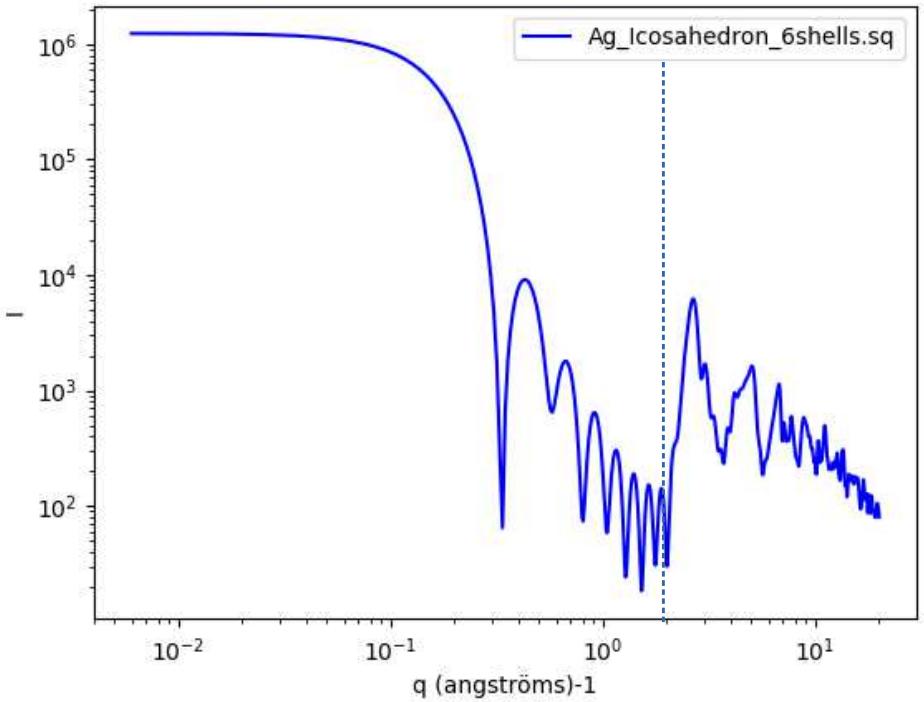

SWAXS techniques are well suited for nano and atomic scale structural characterization of nanoclusters

X-Ray scattering techniques: SAXS

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Q>1Å-

- particle size

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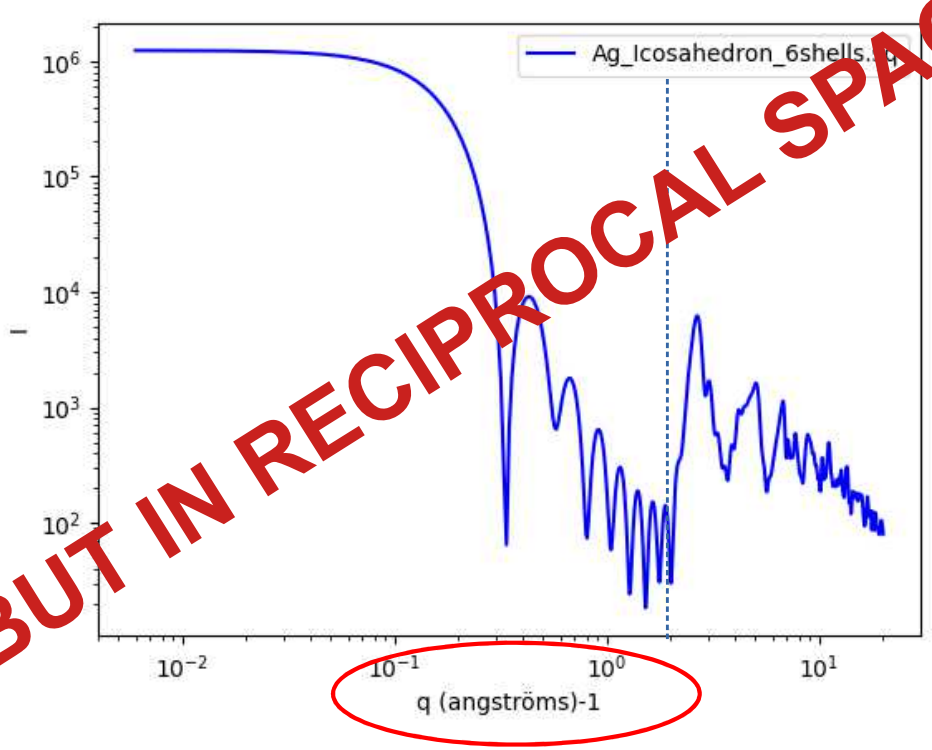
- arrangement of particles



BUT IN RECIPROCAL SPACE!!

SWAXS techniques are well suited for nano and atomic scale structural characterization of nanoclusters

Machine learning assisted SWAXS analysis

Purpose: algorithm for the prediction of the size and shape of monometallic nanoparticles from SAXS/WAXS scattering data, particularly for non crystalline structures

Methodology:

- Create dataset
  - build structural models
  - calculate the corresponding scattering curve
  - store the results with the corresponding labels (shape, size, atom type)
- Choose ML model

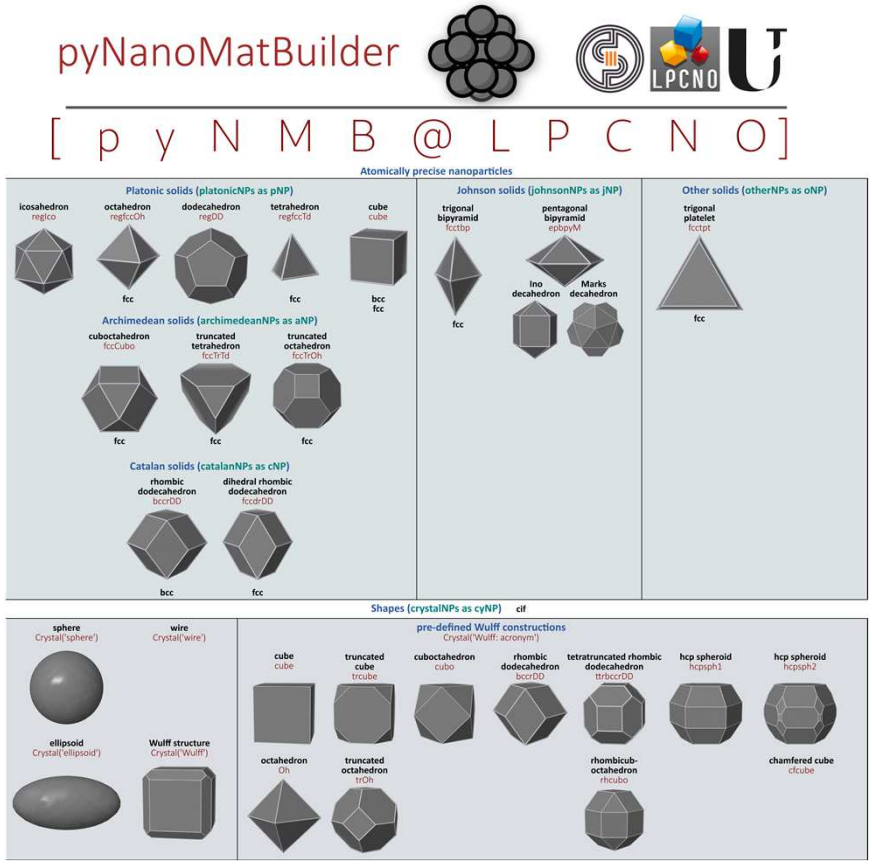
- Training

- Testing

Machine learning assisted SWAXS analysis

- Create dataset
  - build structural models
  - calculate the corresponding scattering curve
  - store the results with the corresponding labels (shape, size, atom type)

R. Poteau – LPCNO - MCP



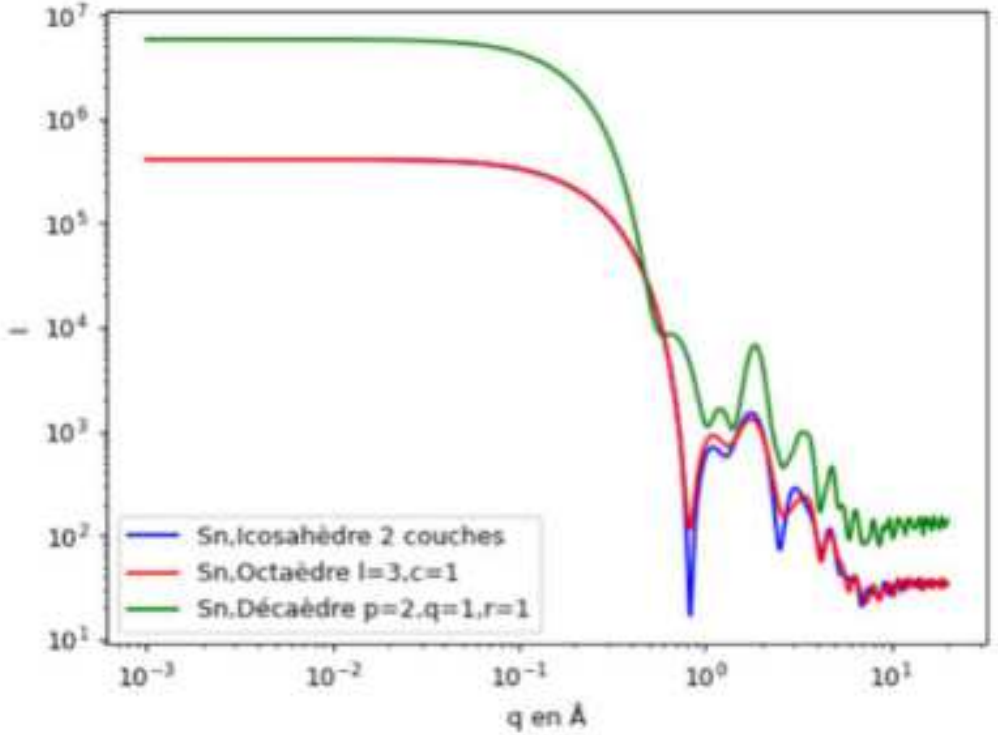
Machine learning assisted SWAXS analysis

- Create dataset
  - build structural models
  - calculate the corresponding scattering curve
  - store the results with the corresponding labels (shape, size, atom type)

$$I(Q) = \sum_i \sum_j f_i f_j \frac{\sin(Q r_{ij})}{Q r_{ij}}$$

where

- Q is the scattering vector, called also momentum transfer vector ($Q = |\boldsymbol{Q}| = 4\pi \sin\theta/\lambda$, where θ is diffraction half-angle and λ is the wavelength),
- $r_{ij} = |\boldsymbol{r}_i - \boldsymbol{r}_j|$ is the distance between atoms i and j,
- and f_i is the atomic scattering factor of i-th atom; in general it depends on Q and we should write it as $f(Q)$, but we don't, to keep the notation simple.



Machine learning assisted SWAXS analysis

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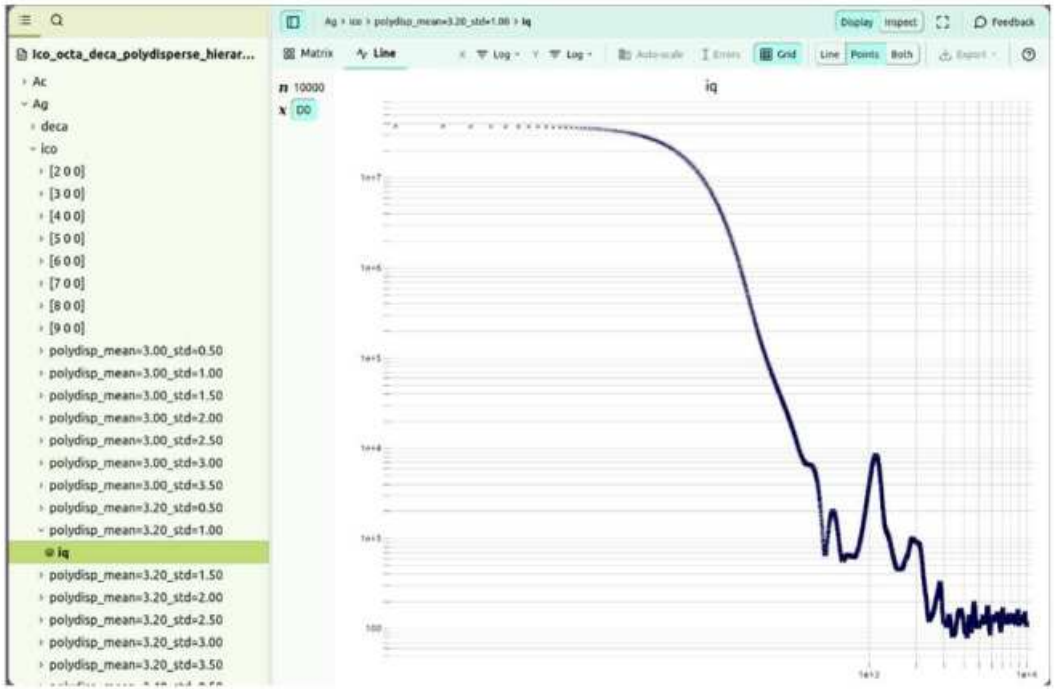
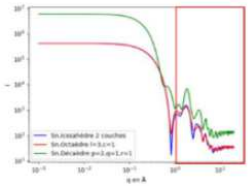


Creation of a large database of scattering data tagged with structural features (labels)

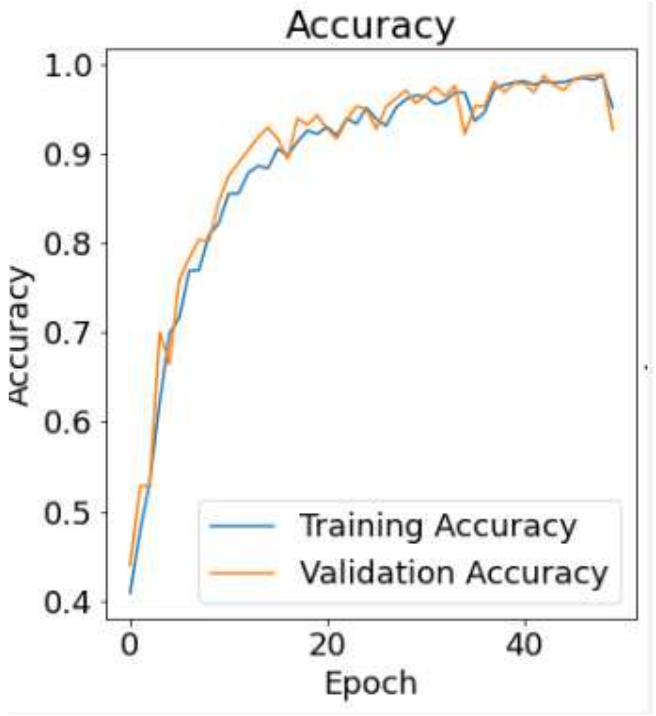
Machine learning assisted SWAXS analysis

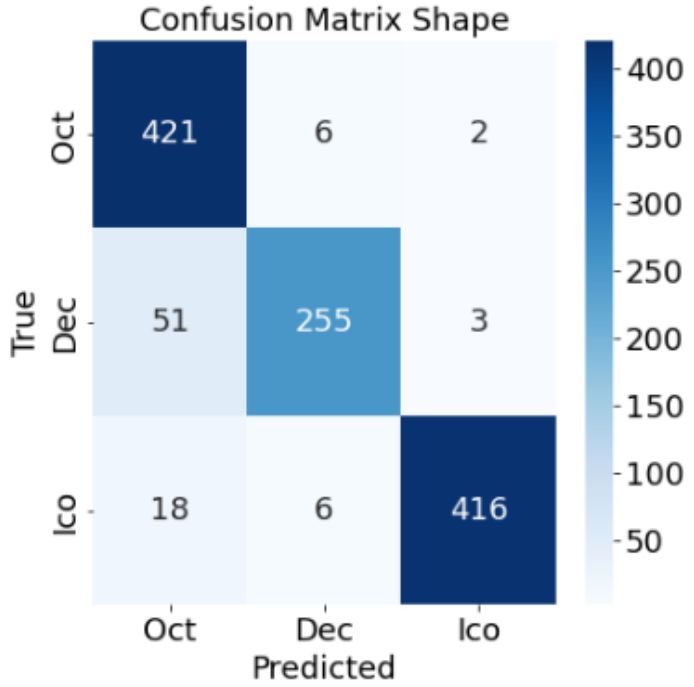
Shape Prediction

Predictions made in the WAXS domain



92,6 % accuracy

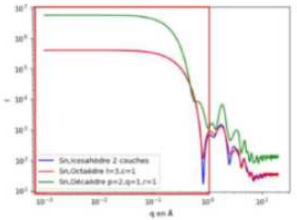




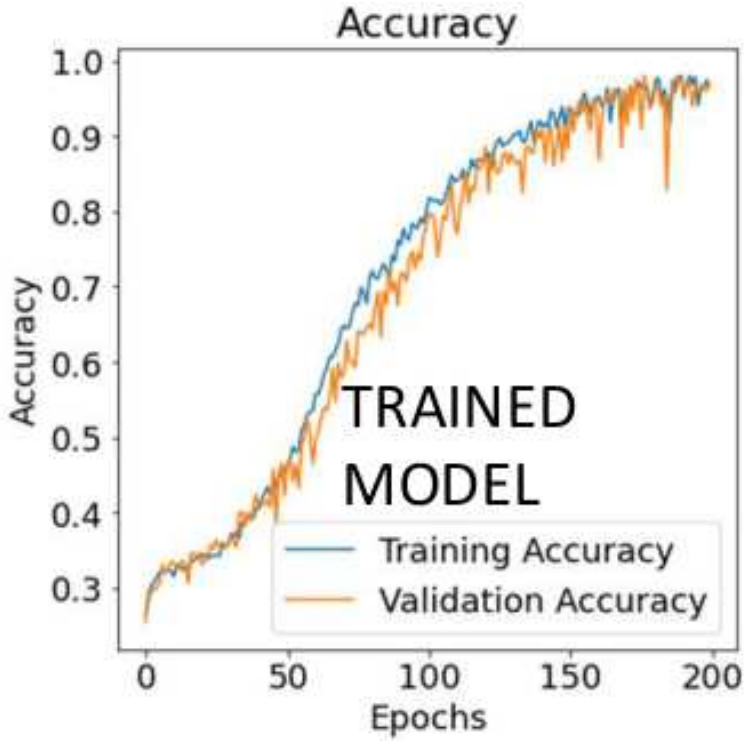
Machine learning assisted SWAXS analysis

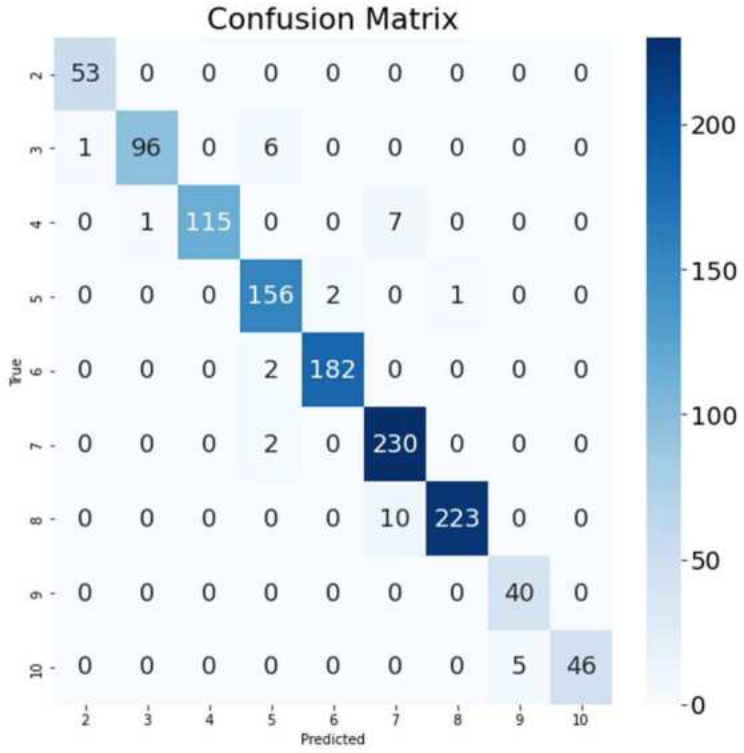
Size Prediction

- Predictions made in the SAXS domain (Guinier plateau)



96.8 % accuracy



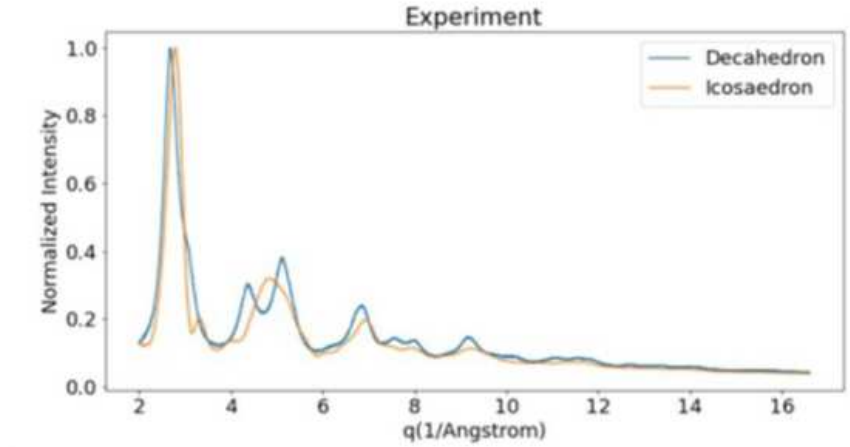


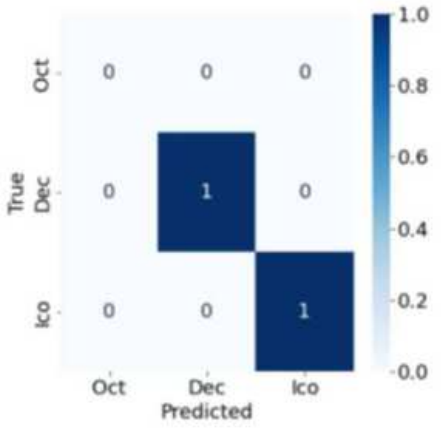
Machine learning assisted SWAXS analysis

Shape Prediction

Real systems: Au decahedron - Au icosahedron NPs

I(q) data were corrected from solvent contribution



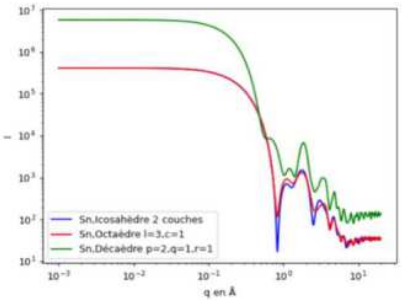


Accuracy 100%
on … 2 experimental samples

Machine learning assisted SWAXS analysis

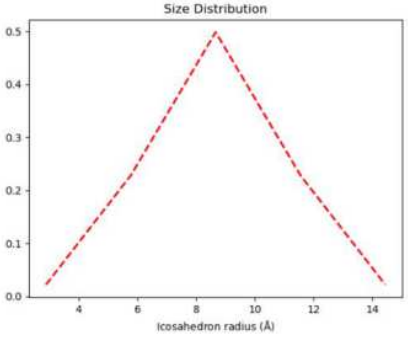
Perspectives

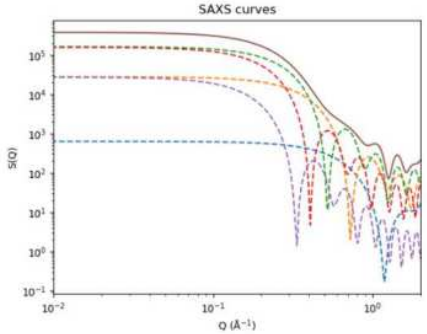
- Size and shape predictions from combined SAXS and WAXS domains (rather than separate SAXS for size, WAXS for shape)

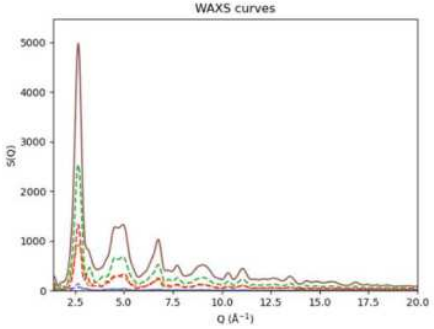


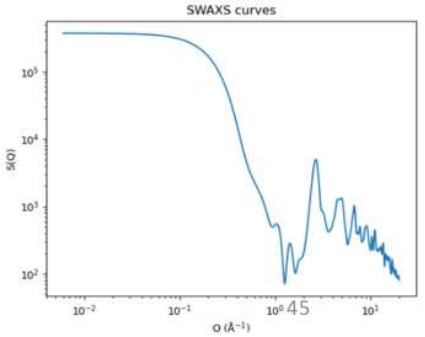
- Simultaneous size and shape predictions?

- Introduction of polydispersity in the dataset to mimic real systems (e.g. icosahedral, mean =4 layers, 20% polydisp.)







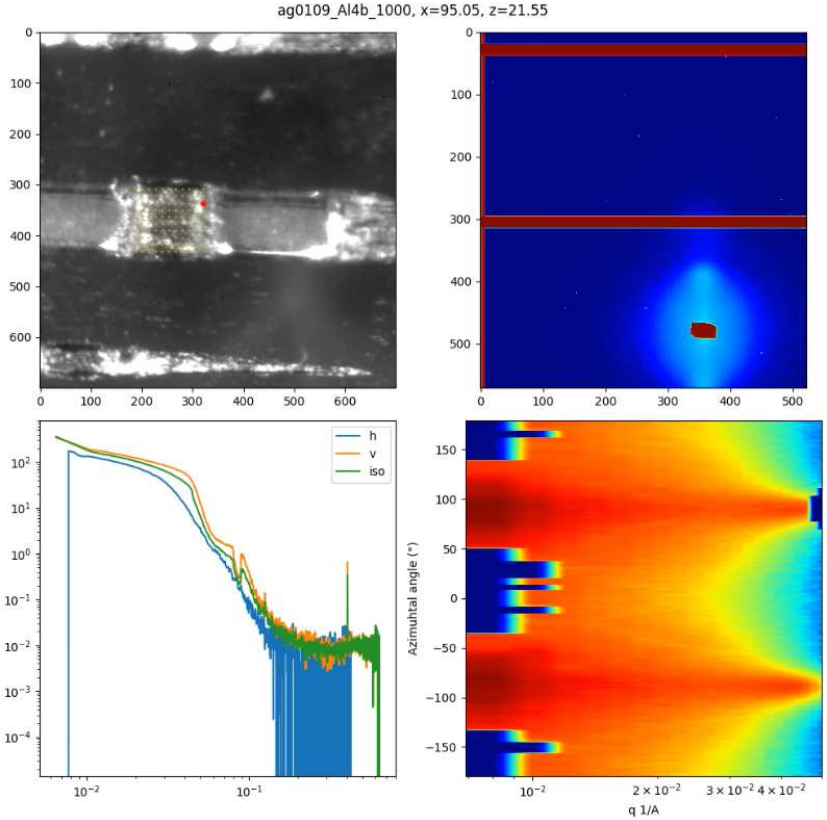


Thank you for your attention!

Applications in SAXS

2. Structure factor analysis: Crystalline assemblies – lattice indexing

Example of µSAXS investigation of micromagnets (L.M. Lacroix, G. Viau)

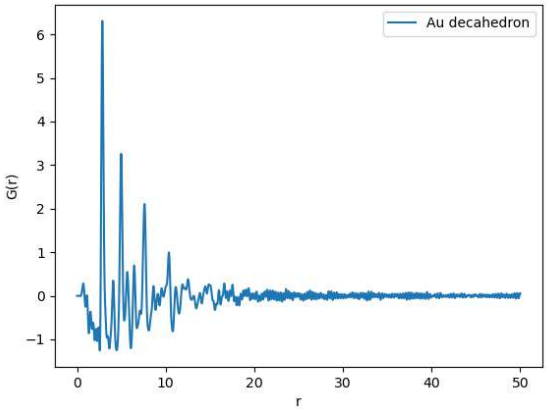


video

Applications in WAXS

M. Imperor- LPS Orsay

◆ <u>Example 3:</u> Decahedron Au : explore decahedron parameters and find best model



Experimental PDF does not match with cubic structure

Build models using ASE Python library

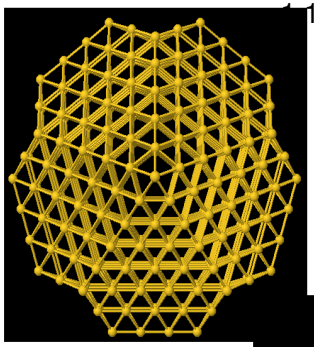
ASE : 3 parameters are used to describe a decahedron :

p: Number of atoms on the (100) facets perpendicular to the five fold axis.

q: Number of atoms on the (100) facets parallel to the five fold axis. q = 1 corresponds to no visible (100) facets.

r: Depth of the Marks re-entrence at the pentagon corners. r = 0 corresponds to no re-entrence

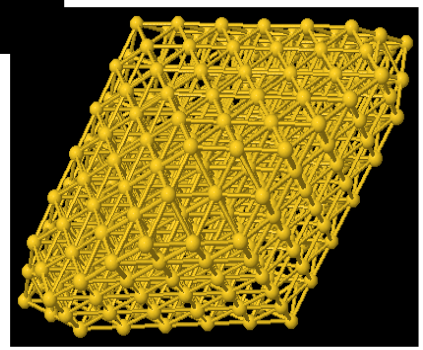
The following (p,q,r) were tested :
(3,1,2),(3,1,3),(4,1,0),(4,1,1),(4,1,2),(4,1,3),(5,1,0),(5,1,1),(5,1,2),(5,1,3),(6,1,0),(6,1,1),(6,1,2),(7,1,0),(7,1,1),(7,1,2)

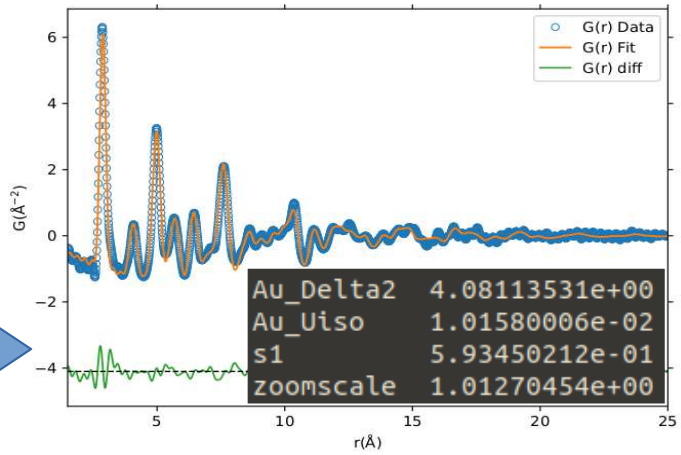


Refine model using diffpy-cmi python library

Adjust the parameters of the model

(4,1,2)





Best refinement obtained for (6,1,1) decahedron : Rw=0,167

Applications in WAXS

- Example 3: Decahedron Au : optimize structure using ASE

ASE library contains optimizers for atomic structures

BFGS Optimization : (video)

-

Applications in WAXS

- Example 3: Decahedron Au - optimize structure using ASE

ASE library contains optimizers for atomic structures

BFGS Optimization :

-

The structure shrinks. Would that improve the PDF refinement ?

→ Model refinement at each step of the optimization

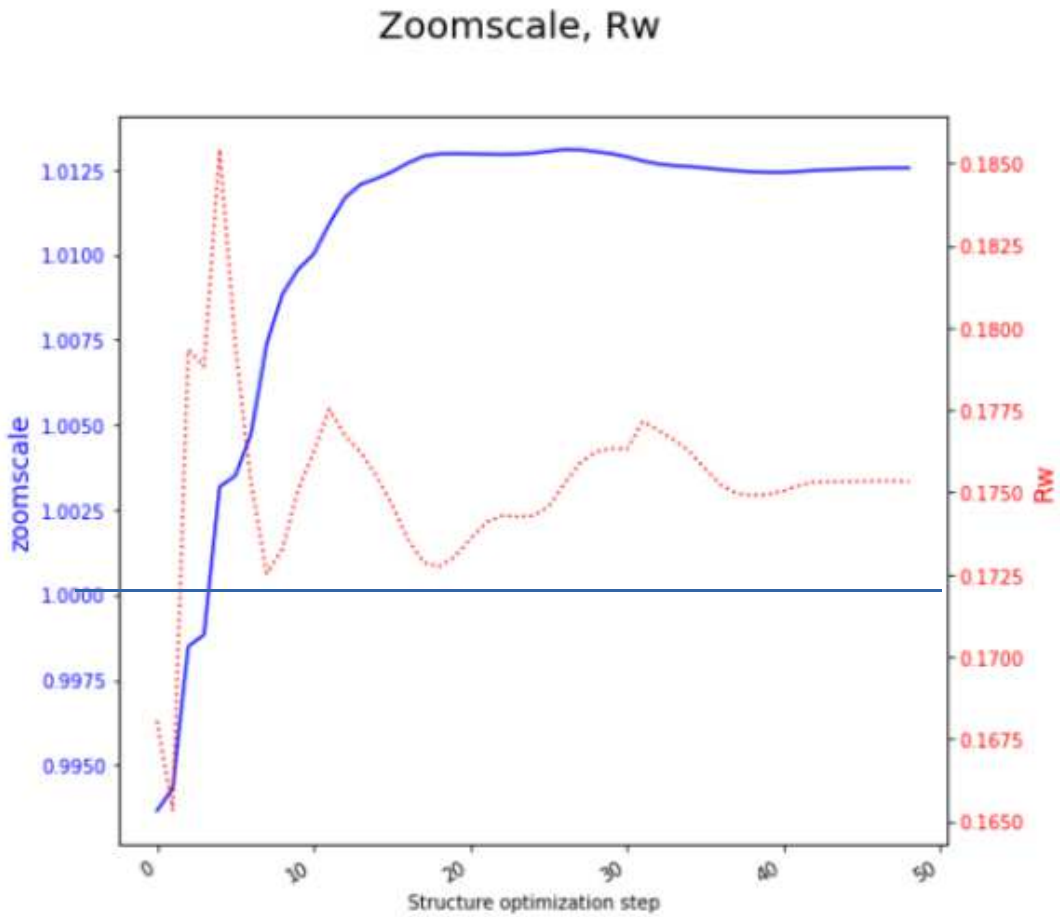
Applications in WAXS

◆ Example 3: Decahedron Au : optimize structure using ASE

ASE library contains optimizers for atomic structures

◆ BFGS Optimization

Improvement of refinement ?



➡ Optimization did not improve the refinement