X-ray and Neutron Scattering Data for Machine Learning of Invertible Neural Networks

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Outline

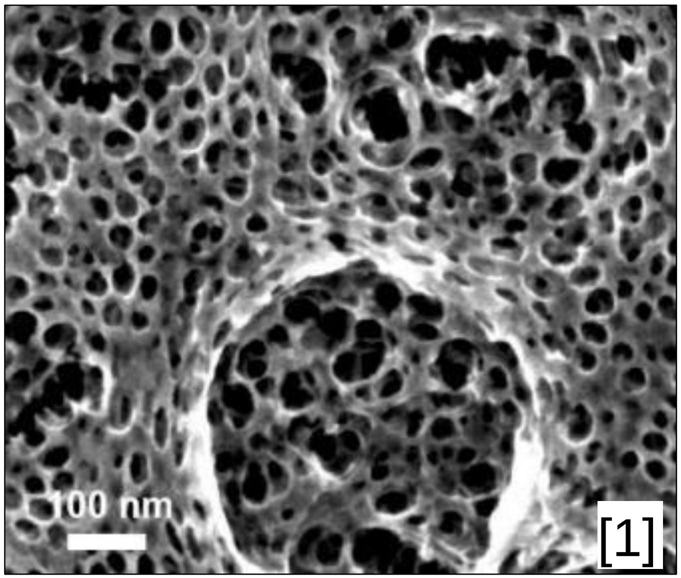
Background
Low-temperature and water-based biotemplating of titania films

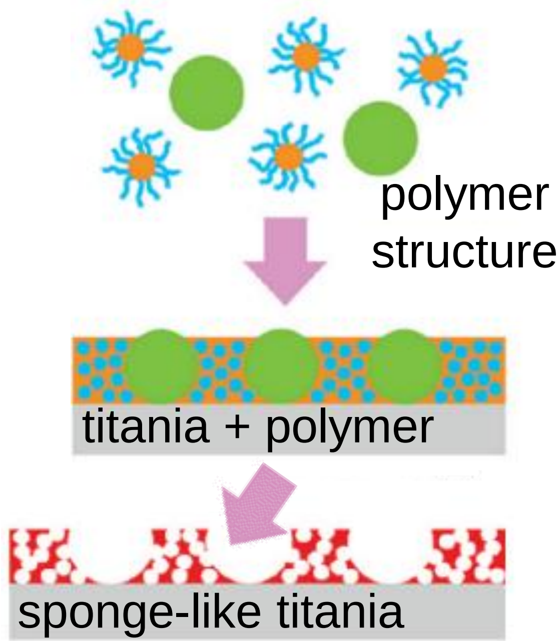
Experimental
Structure-function-relationship in the formation of biohybrid films revealed by in-situ X-ray scattering and neutron scattering

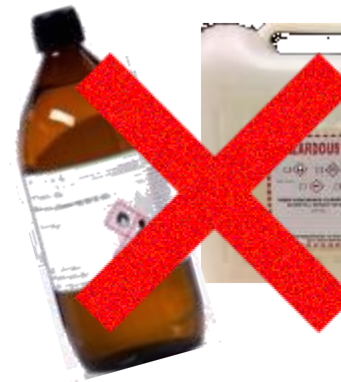
Classification of GISAXS features
2D line cut plots of scattering data as training set for machine learning of invertible neural networks

How to achieve nanostructured titania?

idea: polymer-directed sol-gel synthesis of titania







problem: synthetic polymers include harmful organic solvents

use environmentally friendly biopolymers instead

[1] G. Kaune et al., *ACS Appl. Mater. Interfaces*, **1**, 2862-2869 (2009)

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ß-lactoglobulin

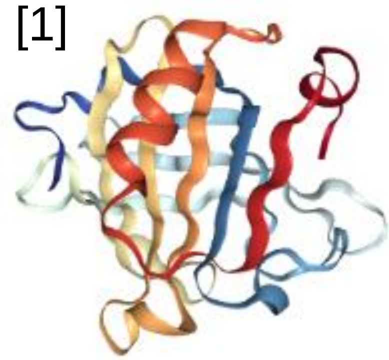
globular transport protein

foaming and structure agent in food industry

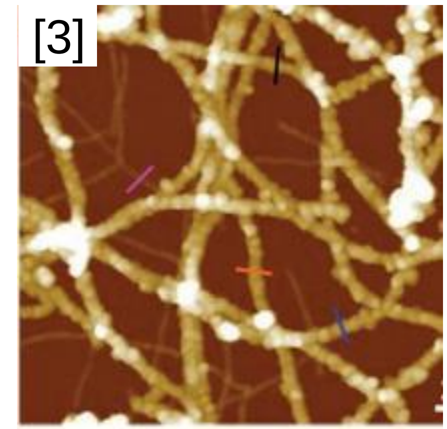
water-soluble and non-toxic

changes structure with pH [2]

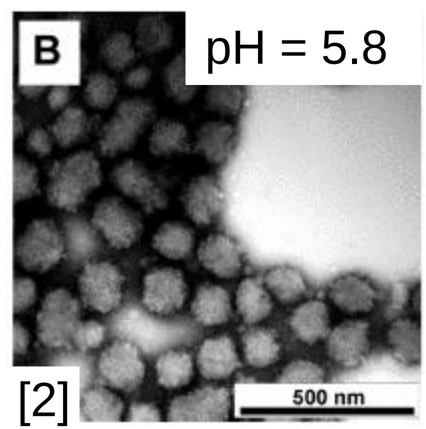
amyloid-directed synthesis of nanowires for application in hybrid solar cells [3]



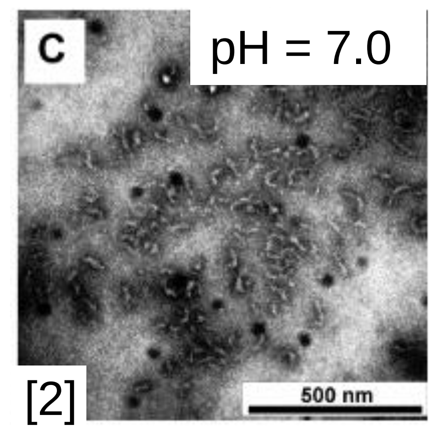
titania nanowires



spheres



worm-like



[1] S. Brownlow et al., *Structure*, **5**, 481-495 (1997)
[2] J.-M. Jung et al., *Biomacromolecules*, **9**, 2477-2486 (2008)
[3] S. Bolisetty et al., *Adv. Funct. Mater.*, **22**, 3424-3428 (2012)

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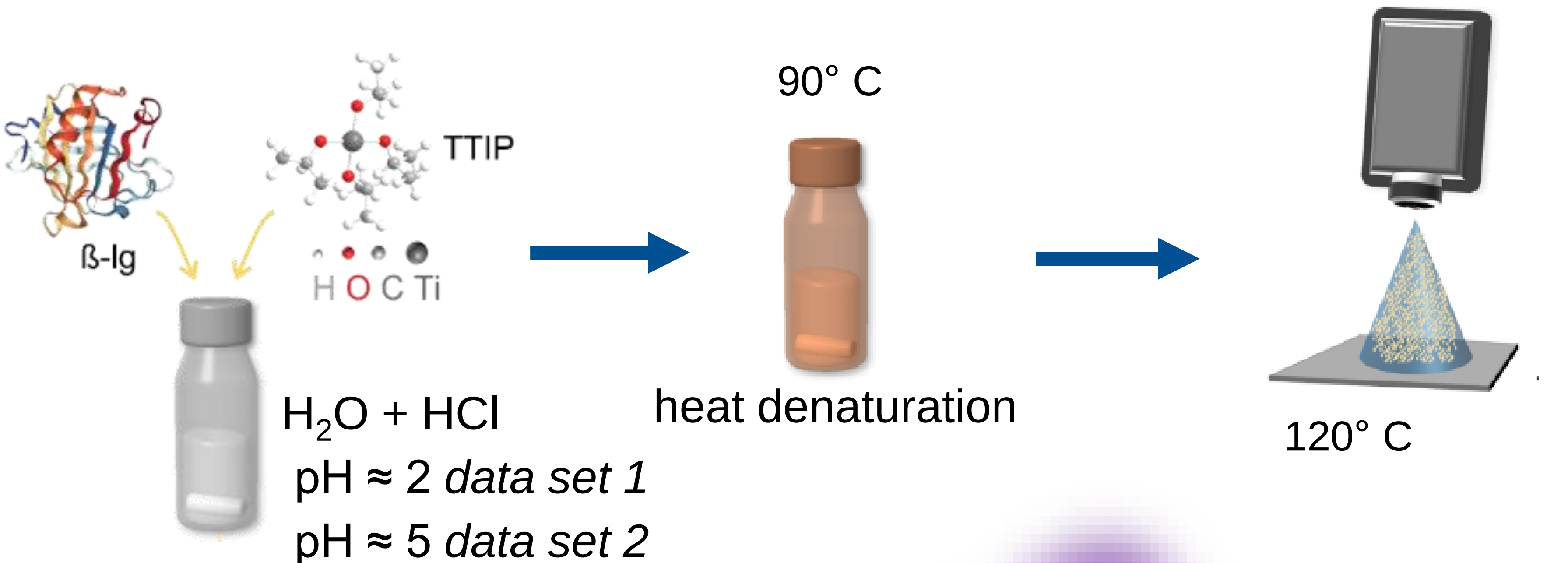
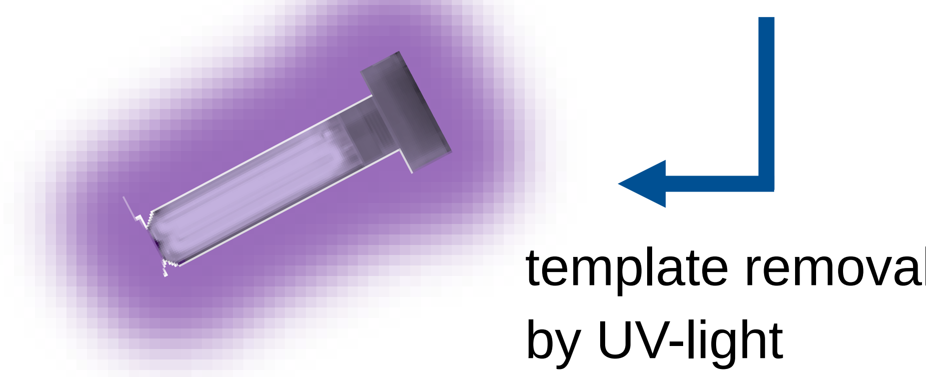
Sample fabrication

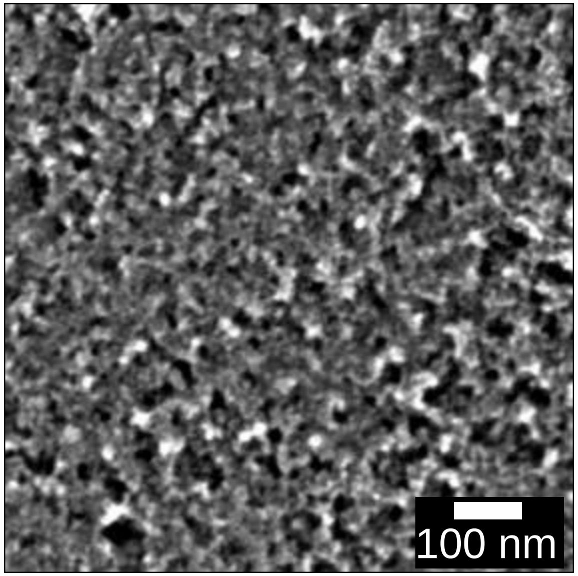




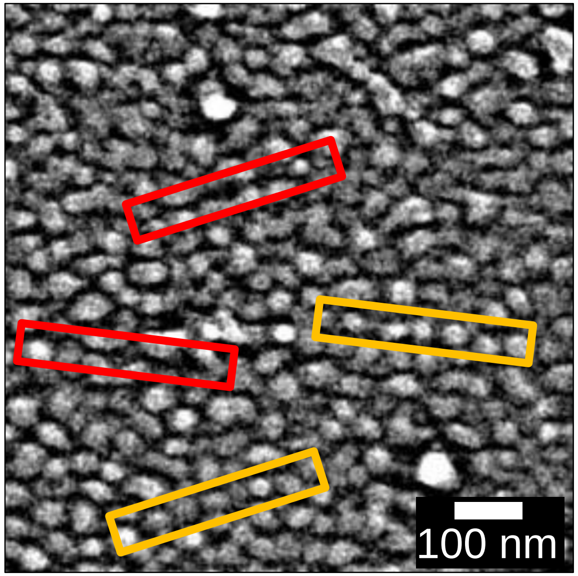
low-temperature and large-scale fabrication process

Surface morphology

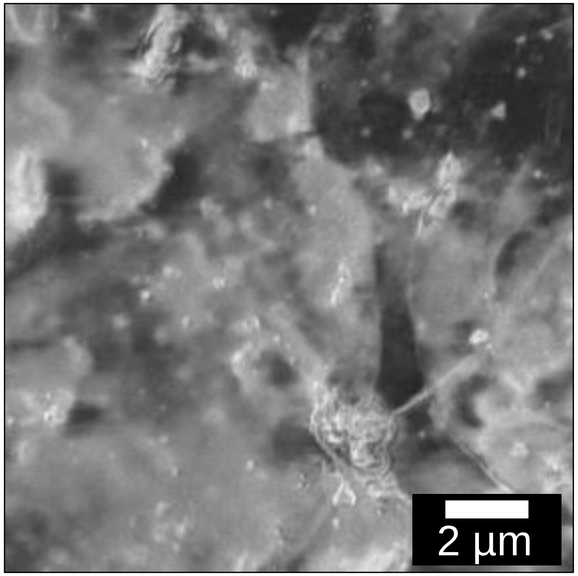
scanning electron microscopy comparison of sample surfaces



pristine titania



UV-treated,
biotemplated titania



biohybrid ß-lg:titania

pearl necklace shaped titania structures in fibrillae matrix

how is morphology forming **inside** the biohybrid sample **during** spray deposition?

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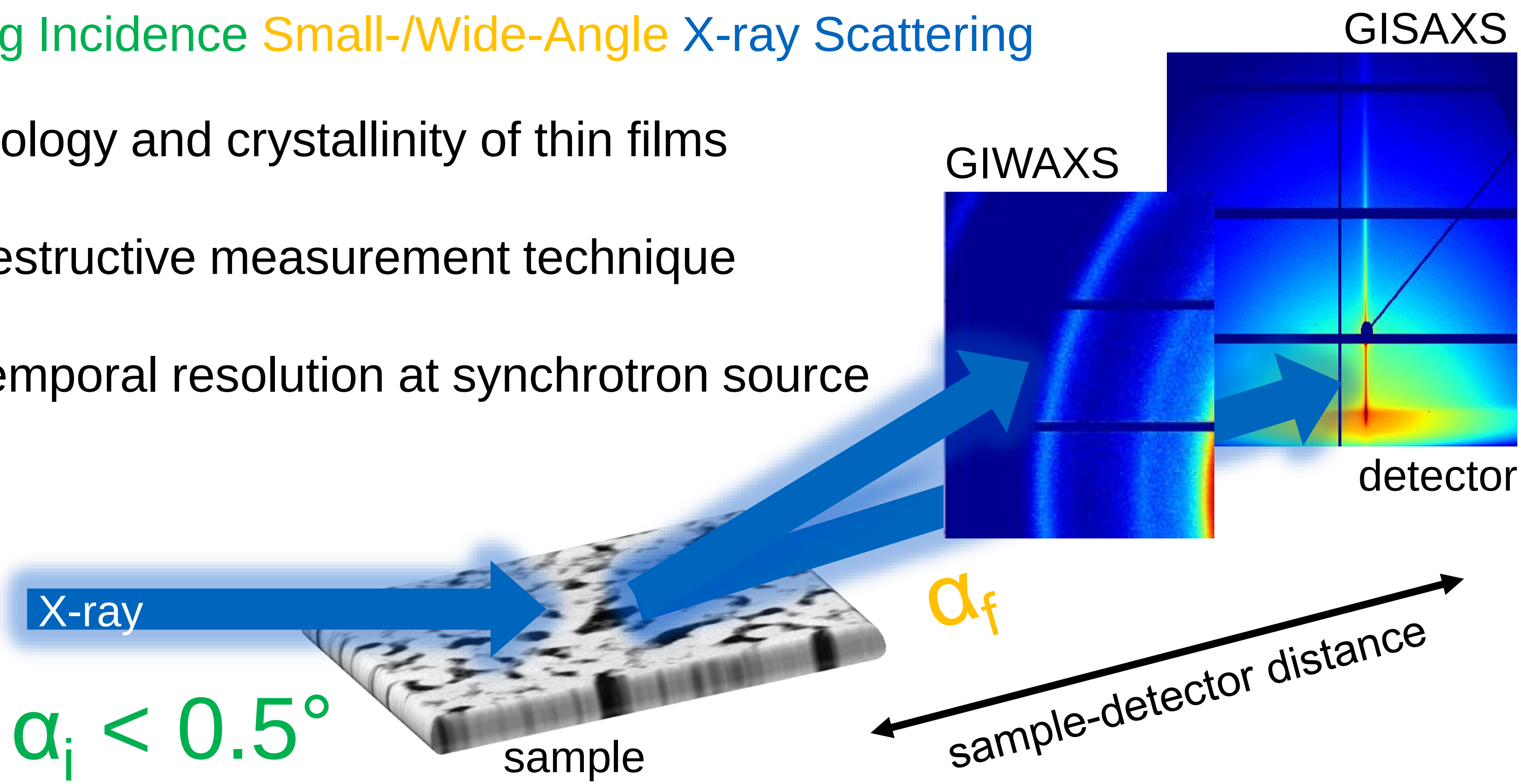
Structure investigation with X-rays (and neutrons)

Grazing Incidence Small-/Wide-Angle X-ray Scattering

morphology and crystallinity of thin films

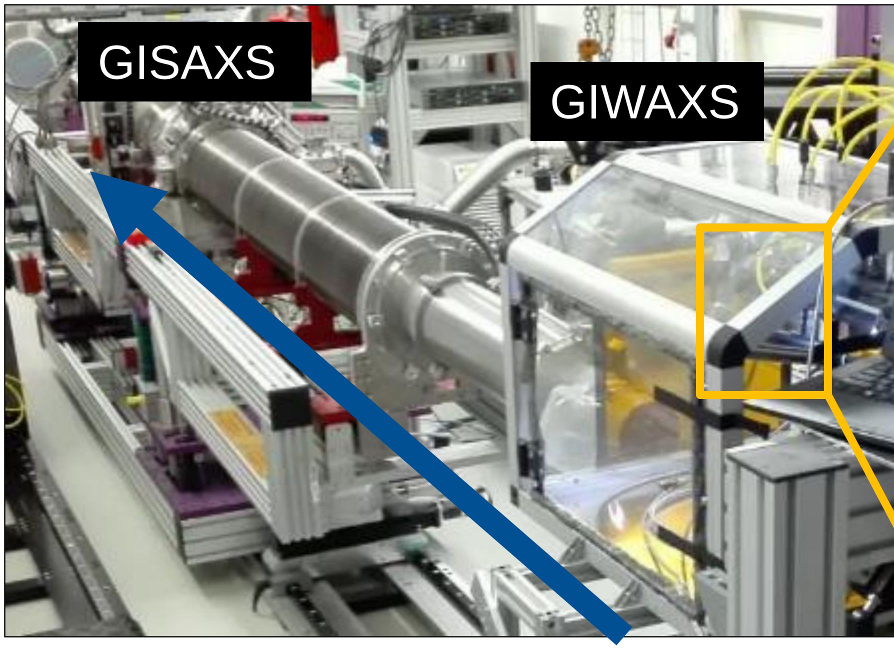
non-destructive measurement technique

high temporal resolution at synchrotron source

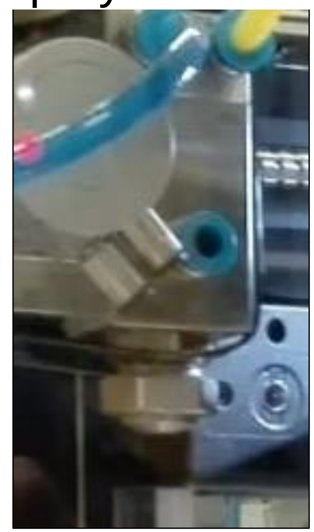


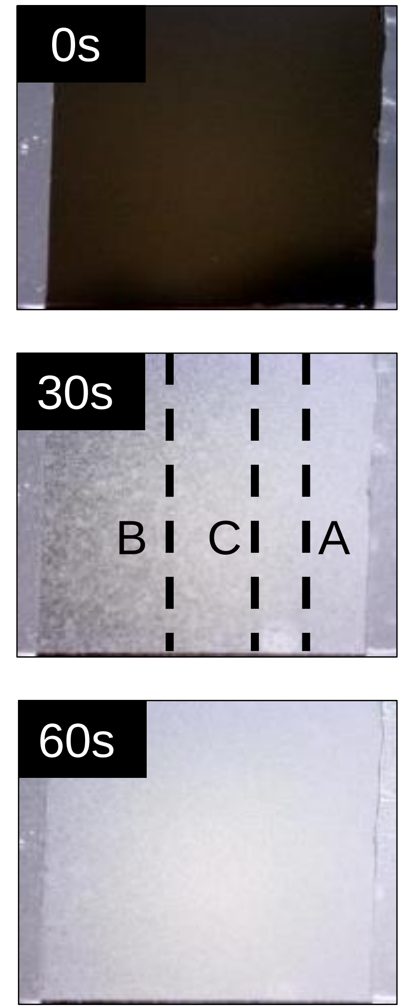
***in-situ* observation of film formation during spray deposition**

In-situ monitoring the biohybrid film formation



industrial spray nozzle





sample detector distance:

GISAXS 4900 mm

GIWAXS 326 mm

exposure time 0.1 s

pulsed spray deposition:

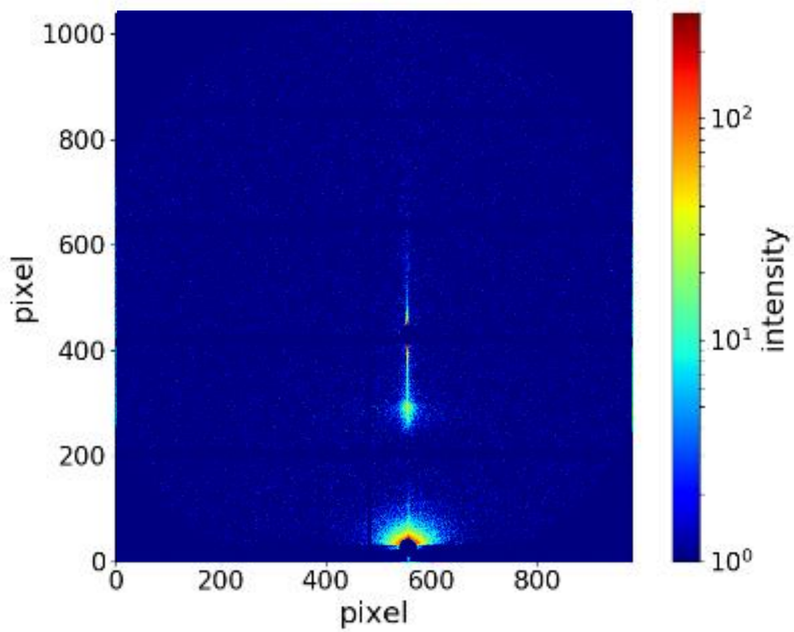
20 x 0.2 s / 2.8 s + 10 s annealing

→ 700 images per detector

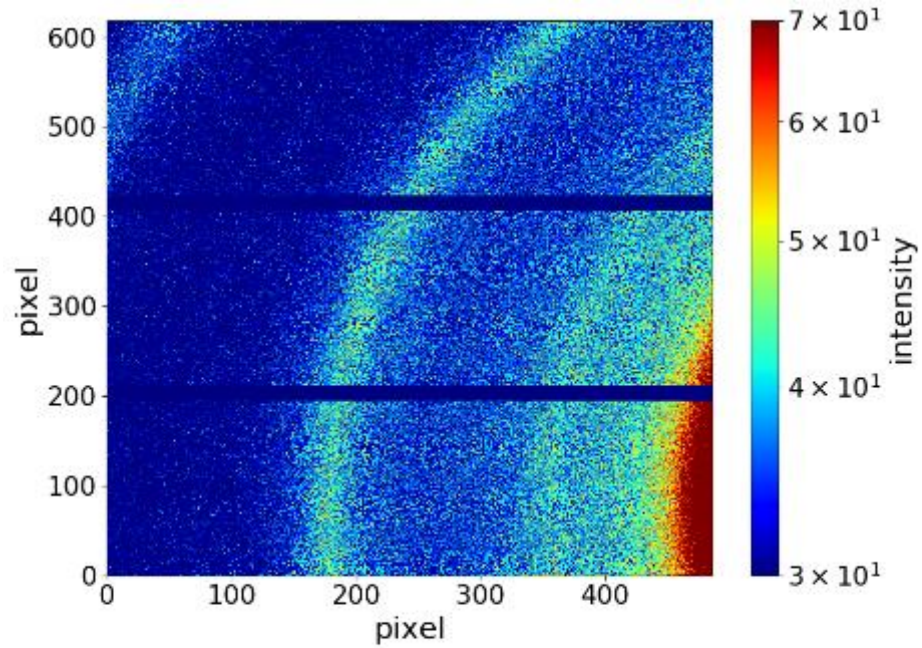
simultaneous GISAXS/GIWAXS measurements

2D detector data evolution

GISAXS



GIWAXS



Pilatus 1M
172 µm x 172 µm

Pilatus 300k
172 µm x 172 µm

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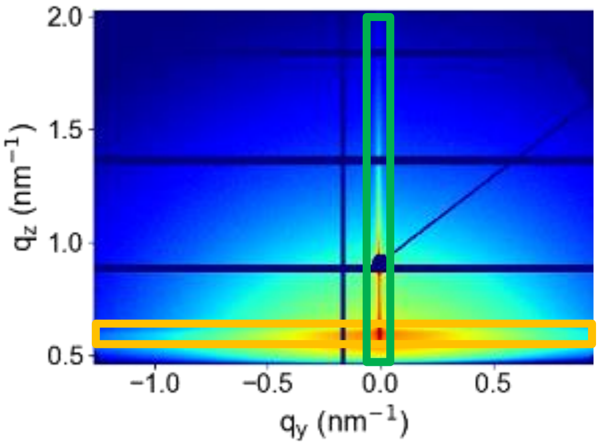
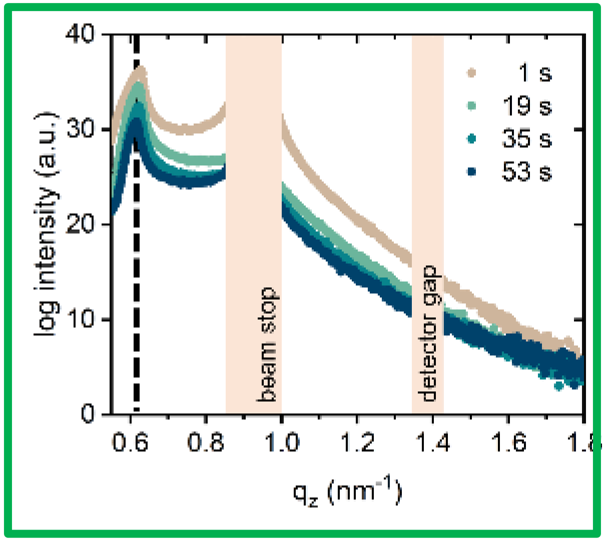
GISAXS evaluation

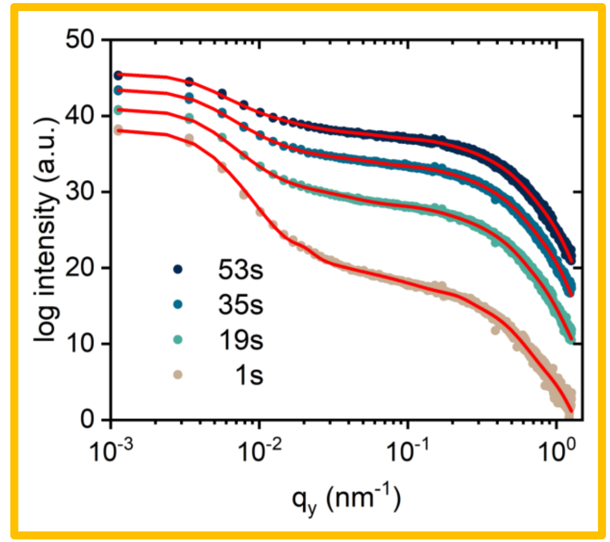
2D raw detector data:

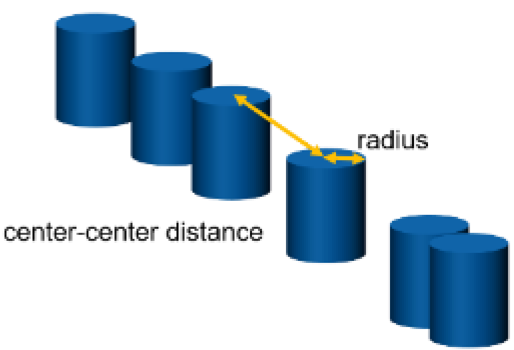


vertical line cut:



horizontal line cut:





$$I(q) \propto \sum_i N_i \left\langle \left|F(q, R_i)\right|^2 \right\rangle S(q, D_i)$$

$$\left|F_{cyl}(q_y)\right|^2 = \left(R \frac{J_1(q_y, R)}{q_y}\right)^2$$

$$S(q_y) = \frac{1 - \exp\left(\pi \sigma_D^2 D^2 q_y^2\right)^2}{1 + \exp\left(\pi \sigma_D^2 D^2 q_y^2\right)^2 - 2\exp\left(\pi \sigma_D^2 D^2 q_y^2\right)\cos(q_y D)}$$

morphology modeled with cylinders on 1D paracrystal lattice

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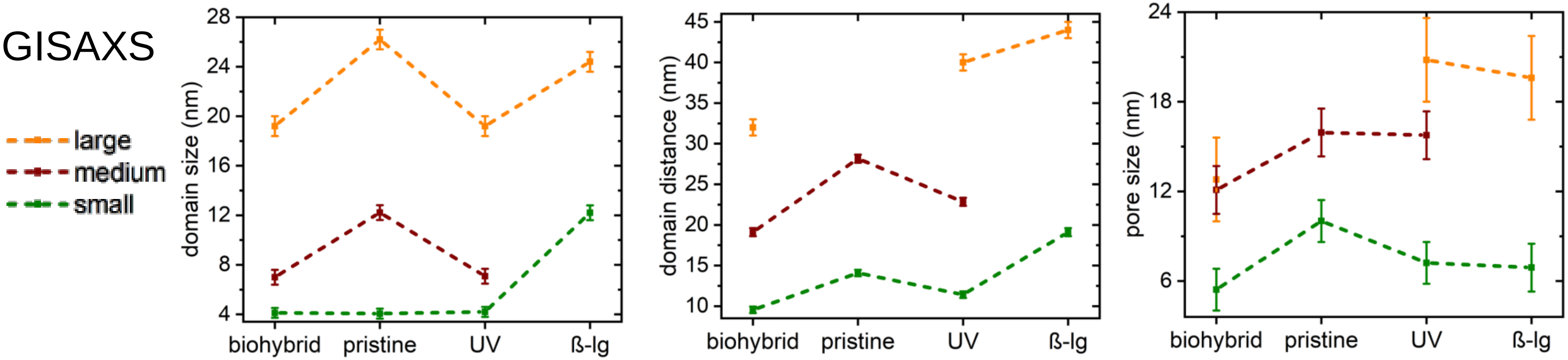
Comparing the inner film structure

GISAXS

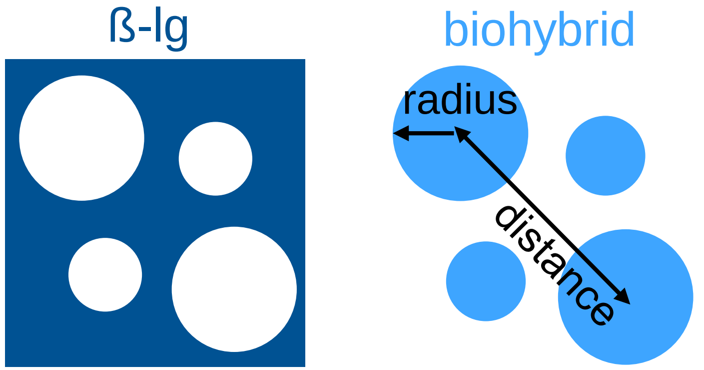


biohybrid domains equal to UV-treated domains and smaller than pristine titania

ß-lg pores equal to biohybrid domains and vice versa

biohybrid distances related to ß-lg matrix

$$domain\ distance - domain\ size = pore\ size$$



titania inside pores of ß-lg matrix

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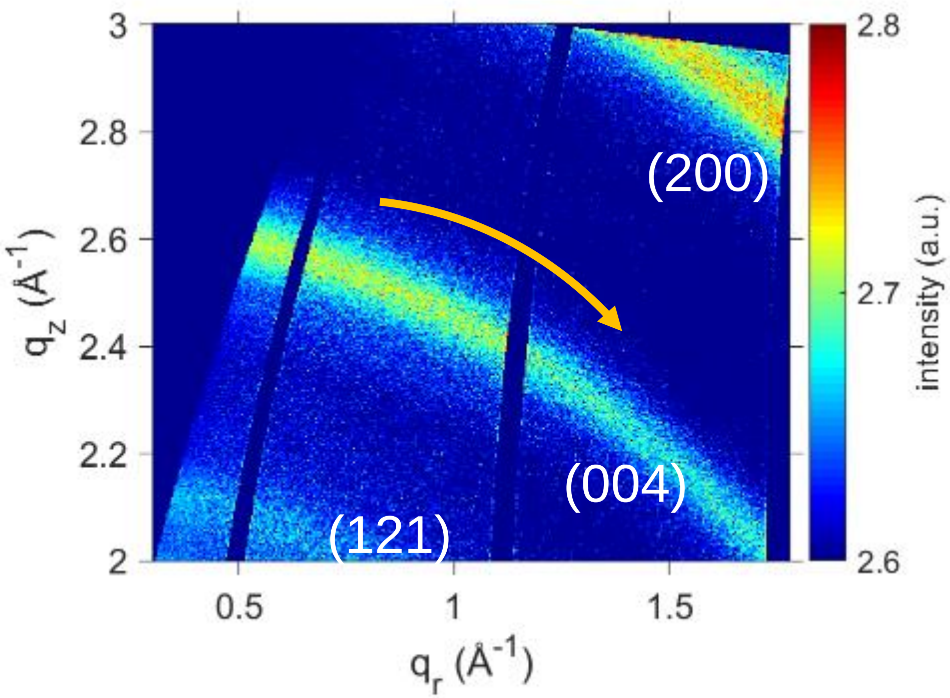
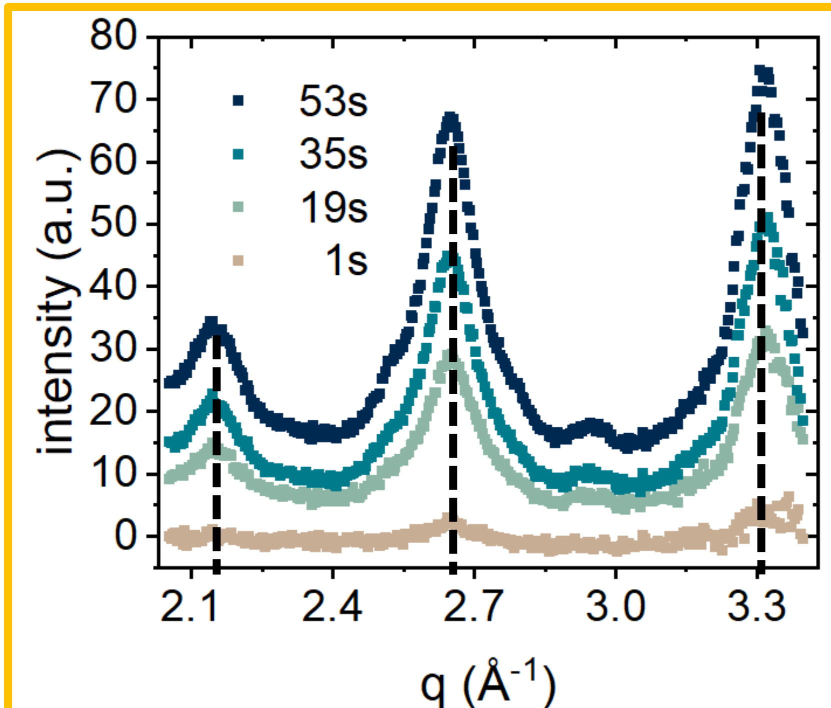
GIWAXS evaluation

2D detector data:



anatase and brookite crystal phases

azimuthal integration of rings:



(121) brookite

(004) anatase

(200) anatase

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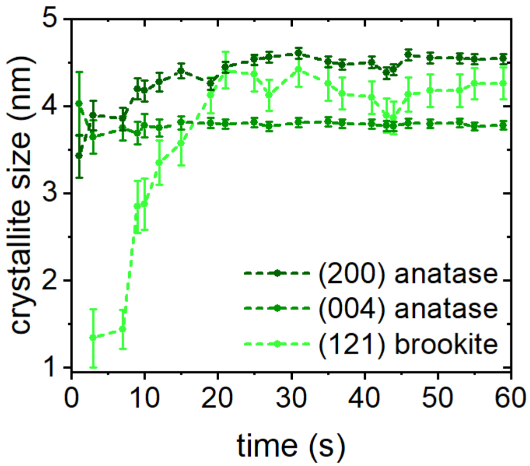
Film formation during spray deposition

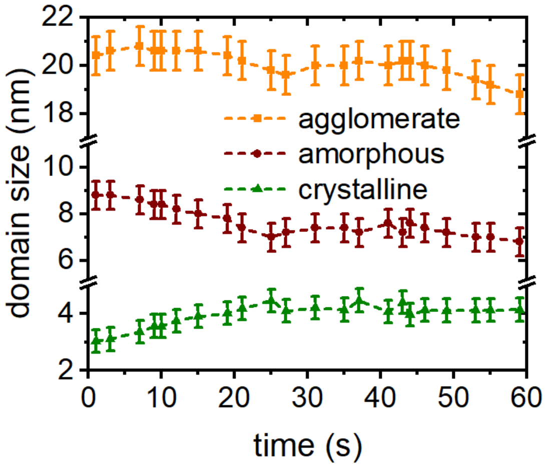
GIWAXS: mixed anatase and brookite crystal phase

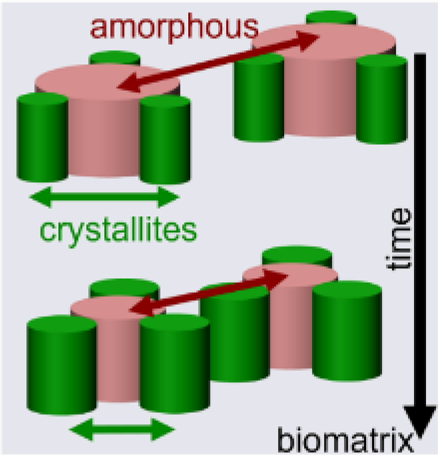
GISAXS: domains in good agreement with crystallite sizes

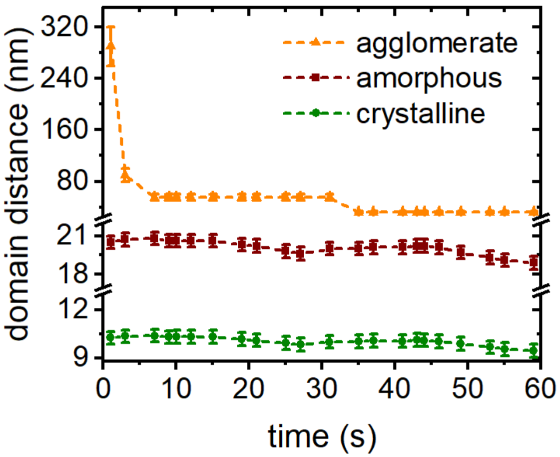
symmetric evolution of growing crystallites and shrinking amorphous domains

triangular arrangement of crystallites around amorphous domains









$$radius_{amorphous} = distance_{crystalline}/\sqrt{3} - radius_{crystalline}$$

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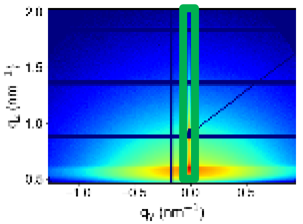
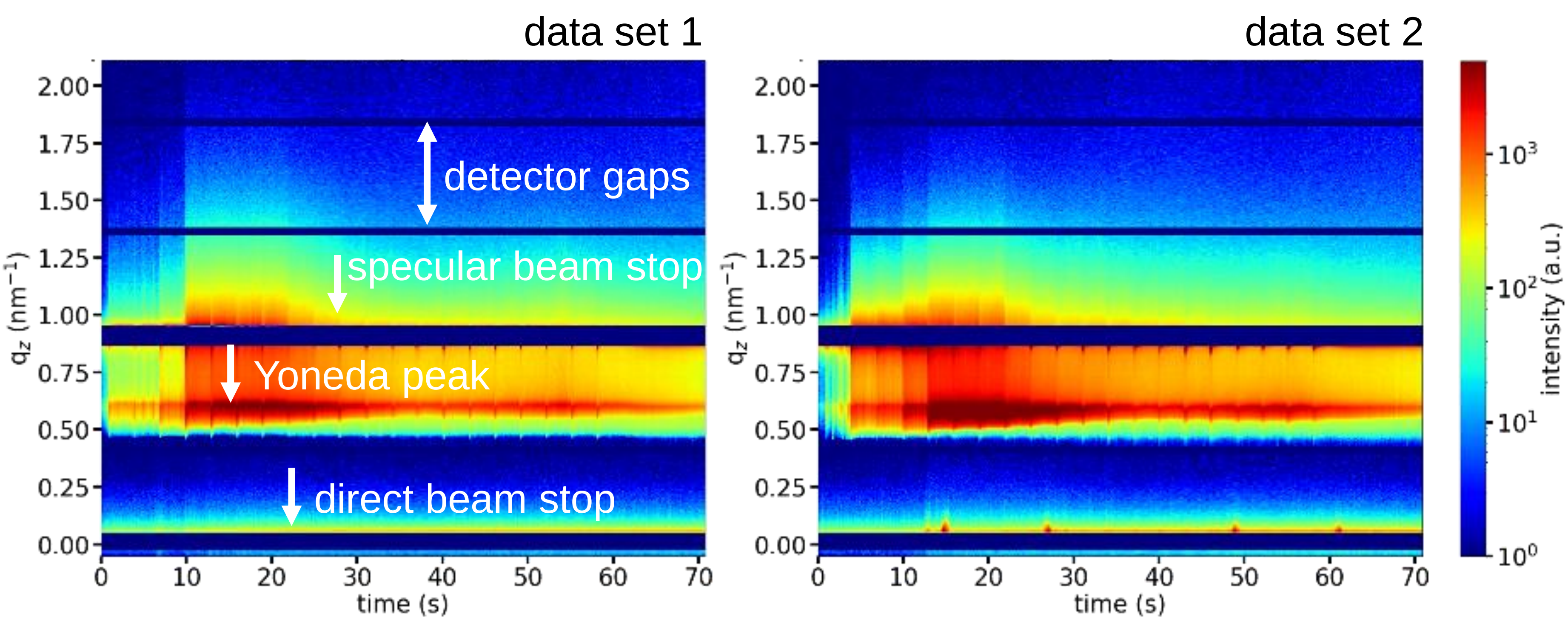
Vertical classification of the intensity pattern

- tag characteristic GISAXS features in 2D line cut plot
- use data set 1 as training set 1
- use data set 2 for evaluation





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Horizontal classification of the intensity pattern

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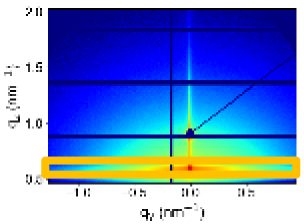

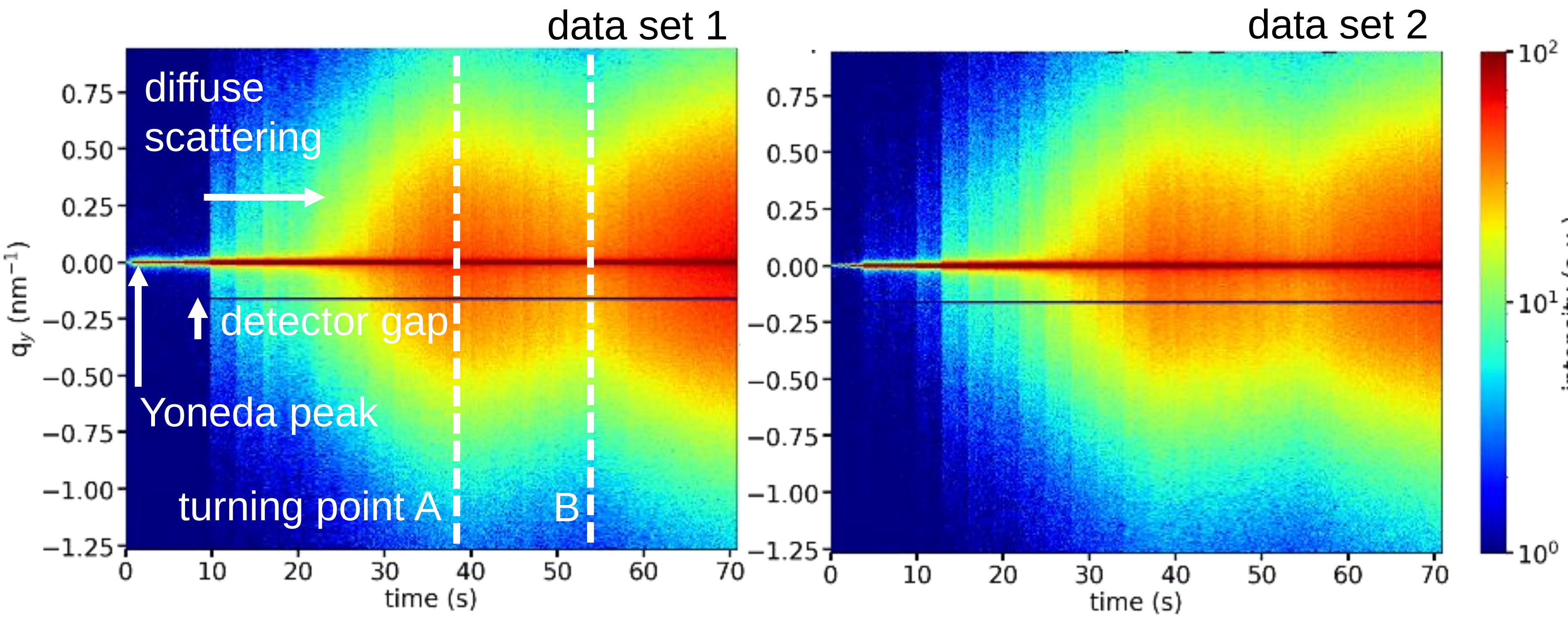

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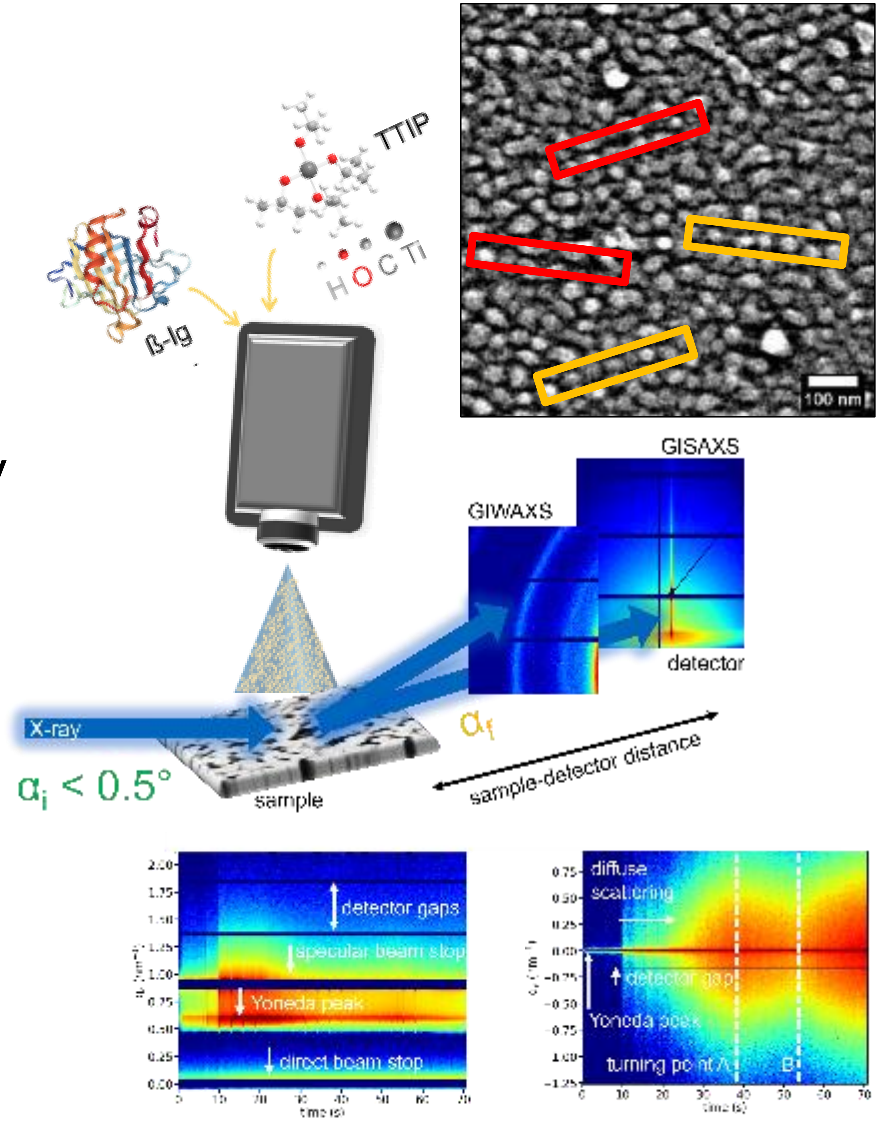
Conclusion & outlook

ß-lactoglobulin templated titania shows pearl-necklace-like nanostructure

film formation revealed by in-situ GISAXS/GIWAXS: sterically directed by biomatrix during spray deposition

apply data anaylsis for classification of 2D line cut plots to use as training set for INN

use INN to evaluate complementary set of in-situ GISAXS/GIWAXS data



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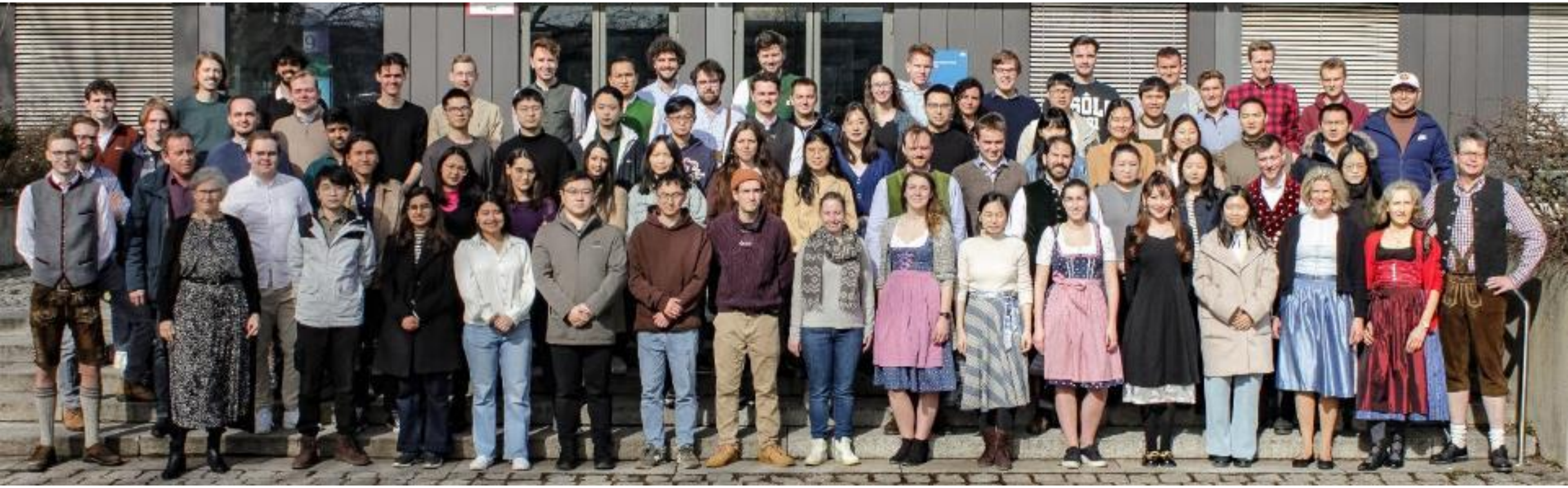

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