Machine-learning-enhanced analysis of surface scattering data: Fundamentals and applications

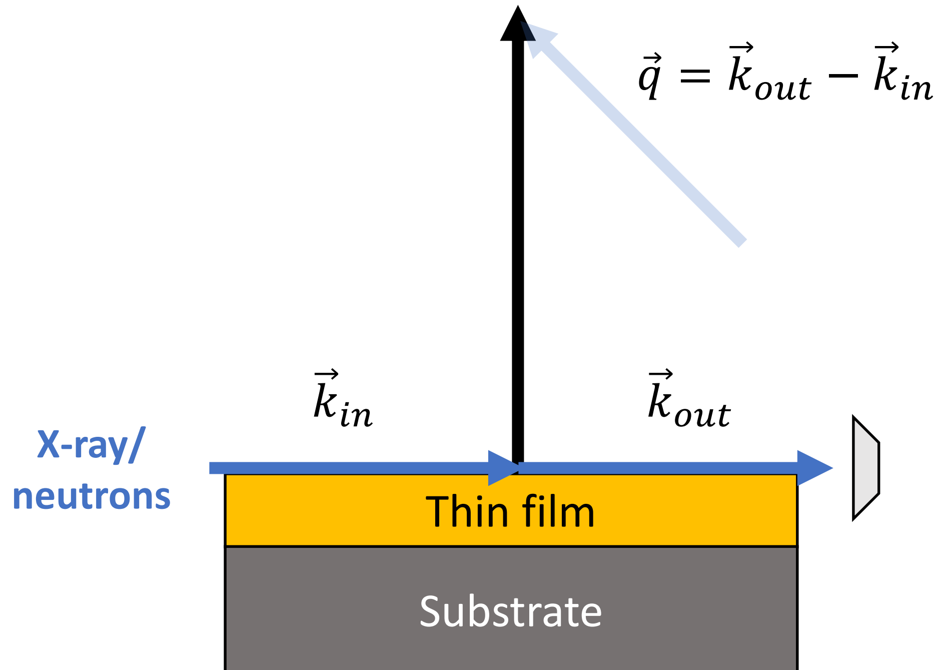
A. Hinderhofer, L. Pithan, V. Starostin, L. Petersdorf, S. Hövelmann, B. Murphy, F. Schreiber

1. ML for reflectivity data analysis
2. Training Data
3. Open Reflectivity Data
4. GIWAXS peak detection
5. The *mlreflect* package as tool for screening XRR data

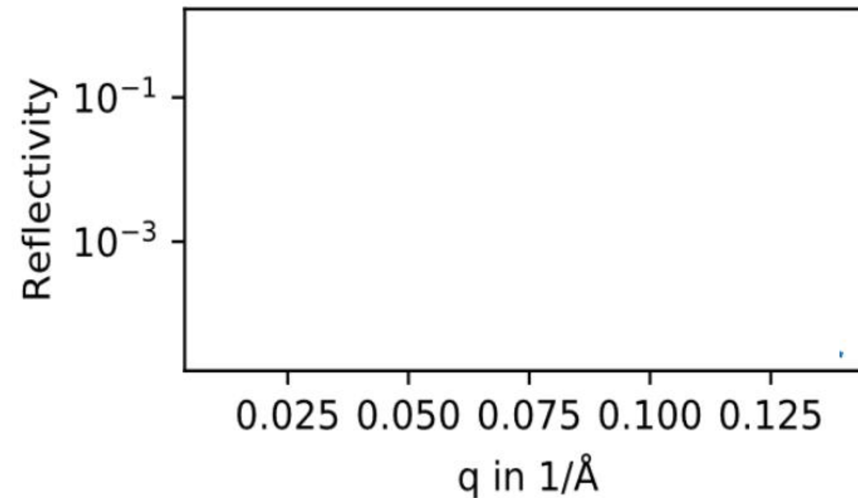
EBERHARD KARLS UNIVERSITÄT TÜBINGEN

X-ray and neutron reflectivity measurements

IN:
X-ray beam at certain discrete angles



OUT:
Reflected beam intensity for each angle



Shape of reflectivity curve provides information about thin film properties

Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

Characterizing thin film samples

SLD profile

Height z [Å]

roughness$_2$

thickness

SLD$_2$

roughness$_1$

SLD$_1$

Scattering length density ρ [10^{-6} 1/A^{-2}]

Thin film

Substrate

Scattering:
FT with phase loss

Neural Net

Measured reflectivity curves $R(q;\boldsymbol{\theta})$

Reflectivity

q in 1/Å

TR edge
(SLD)

Kiessig oscillations
(thickness)

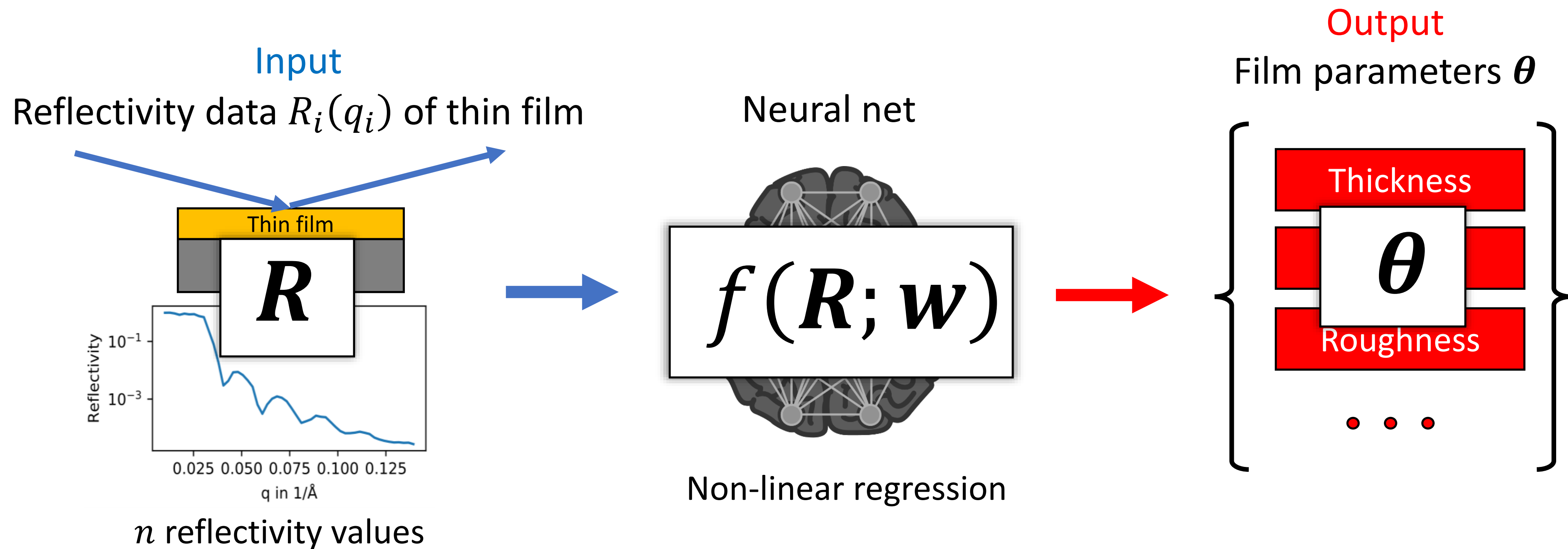
- SLD profile $\rho(z;\boldsymbol{\theta})$ is parameterized by a set of film properties $\boldsymbol{\theta}$
- Reflectivity $R(q;\boldsymbol{\theta})$ can be simulated with $\rho(z;\boldsymbol{\theta})$ via recursive algorithms (but not uniquely!)
- Usual solution is an iterative fitting algorithm

Iterative fitting is slow and strongly depends on human expertise!

Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

Using neural networks for “fitting”

Build a neural network that takes reflectivity curves as *input* and yields the correct film properties as *output*



Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

Sample model: thin film layer on Si/SiOx

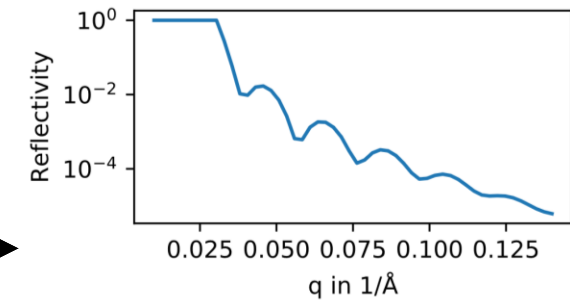
Thin film model for training:

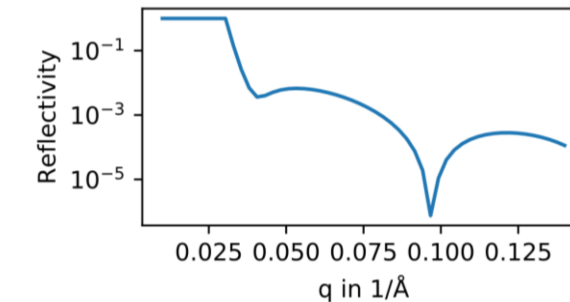
Layer	Parameter	Value
Air	SLD	0 Å^{-2}
Thin film	Thickness	20 – 1000 Å
	Roughness	0 – 40 Å
	SLD	1 – 14 10^{-6}Å^{-2}
SiOx	Thickness	10 Å
	Roughness	2.5 Å
	SLD	17.77+*i*0.40 10^{-6}Å^{-2}
Si	Roughness	1 Å
	SLD	20.07+*i*0.46 10^{-6}Å^{-2}

= open

= fixed

Generate random parameter sets and simulate curves



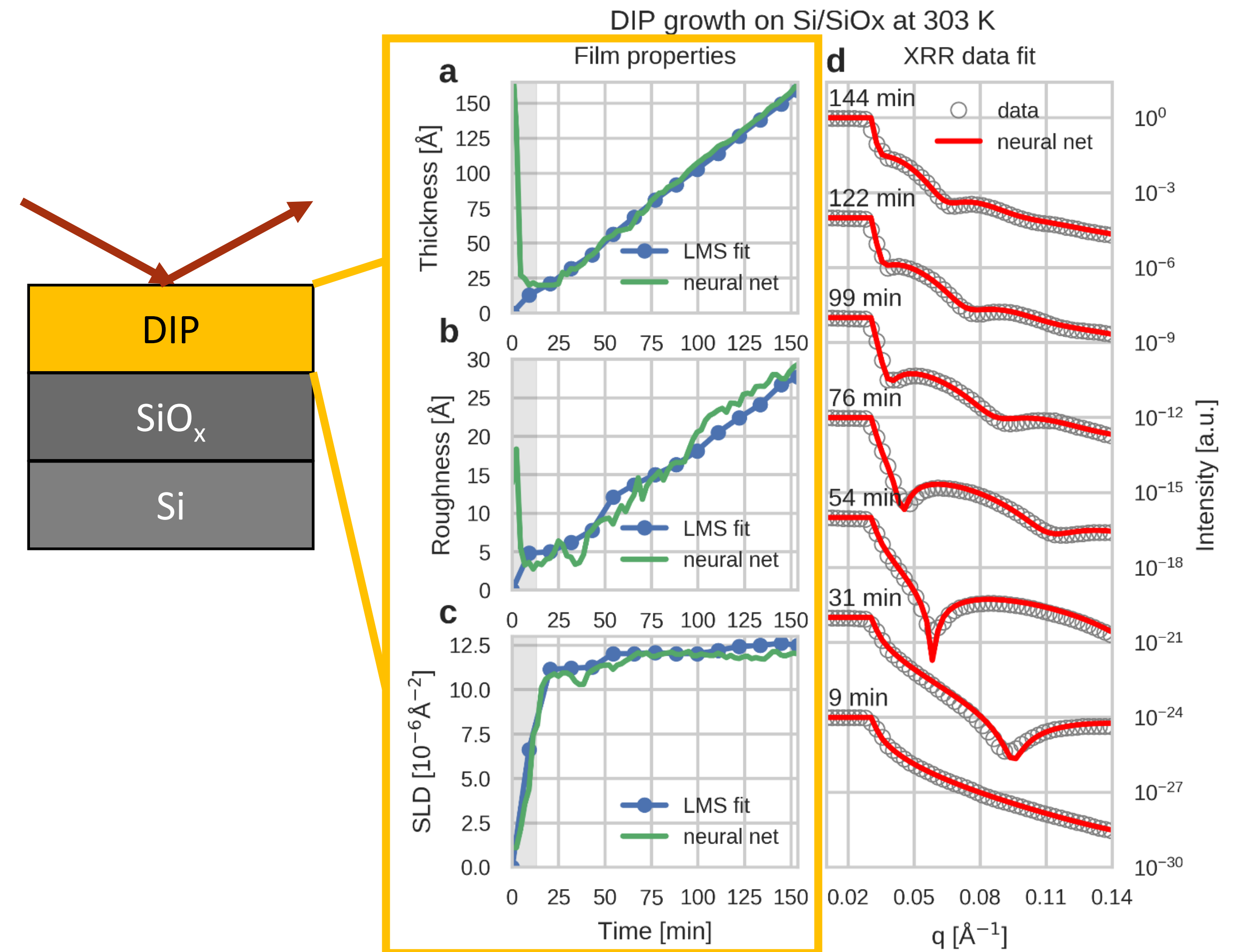


Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

Fitting real-time XRR of film growth

- Tested on 5 real-time datasets of film growth: 3 x DIP, 1 x CuPc, 1 x 6T (370 curves)
- Result obtained less than 10ms/curve
- Results compared with GenX fits “by hand”
- Output was used to simulate “red” curve
- Results are good!

	Thickness	Roughness	SLD
Average relative error	11%	18%	8%



Greco et al., *J. Appl. Cryst.*, **52**, 1342 (2019)
Greco et al., *Mach. Learn.: Sci. Technol.* **2,** 045003 (2021)
Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

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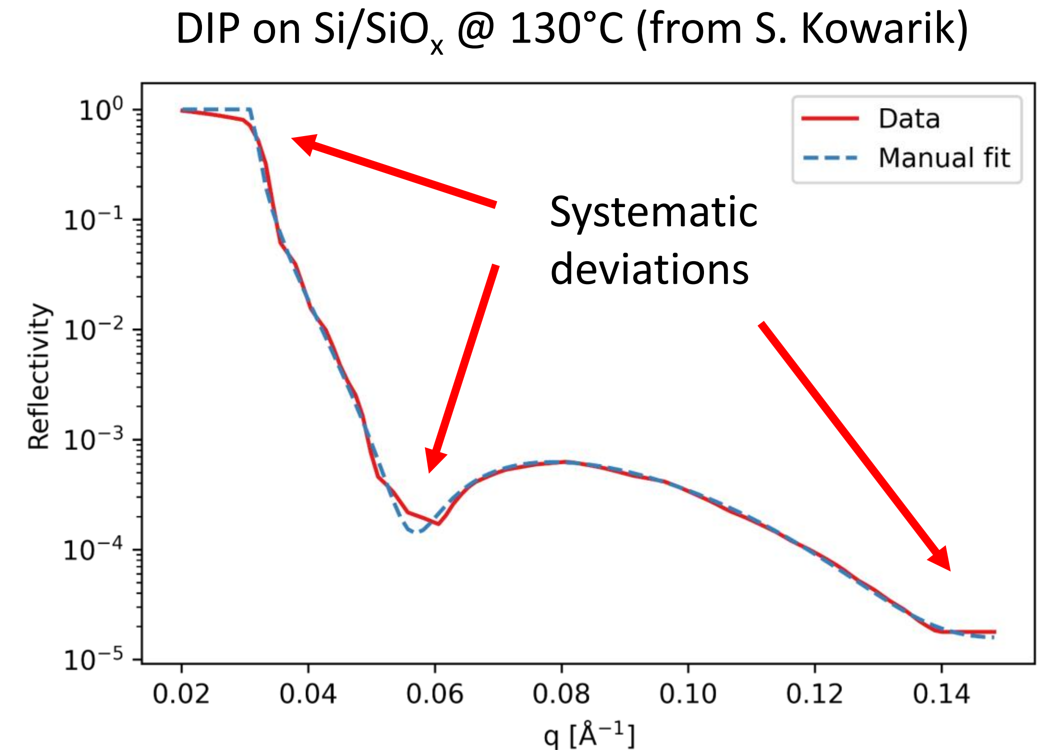
Simulated vs. experimental data

- Performance on simulation is much better than on experimental data
- Reason are experimental deviations from theoretical model, e.g.
  - Alignment
  - Slit function
  - Instrument noise
  - Wrong film model
  - …

Need to include some kind of noise to the training data!

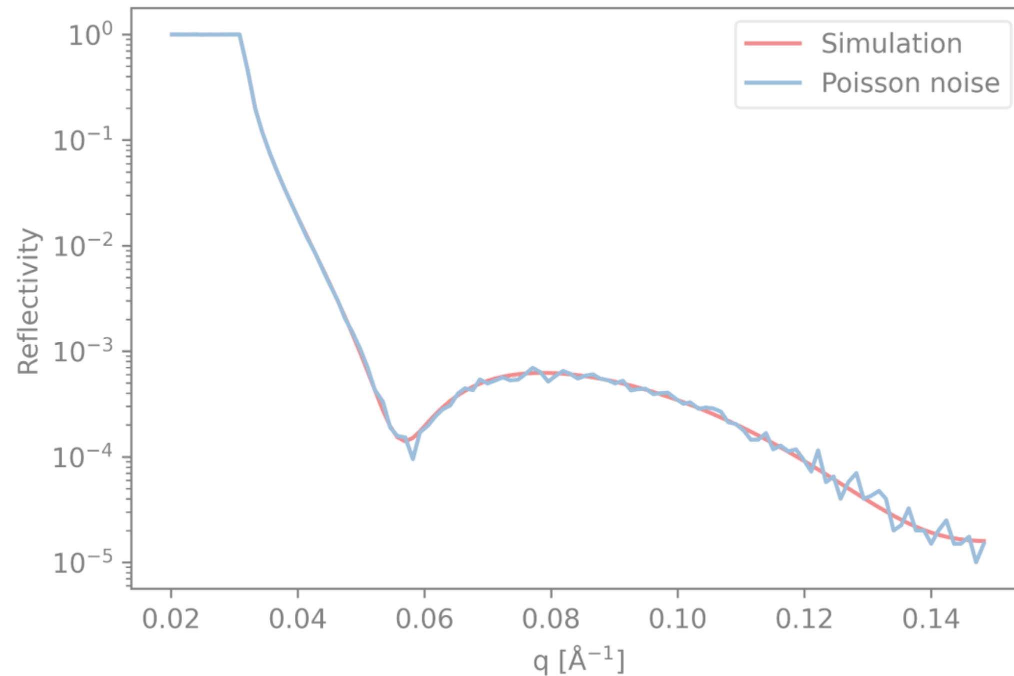
DIP on Si/SiO_x @ 130°C (from S. Kowarik)



Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

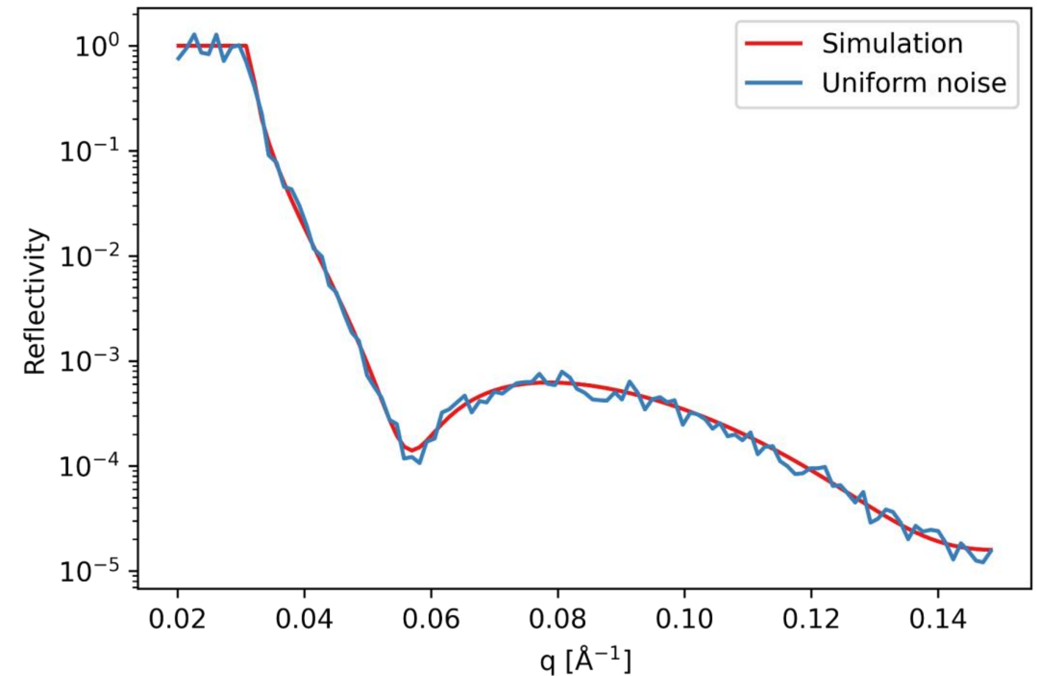
What type of noise should be added?

Poisson noise (e.g. low SNR)



Scales with intensity $\sim \frac{1}{\sqrt{I}}$

Uniform noise



Each point multiplied with a random value between $1 - n$ and $1 + n$

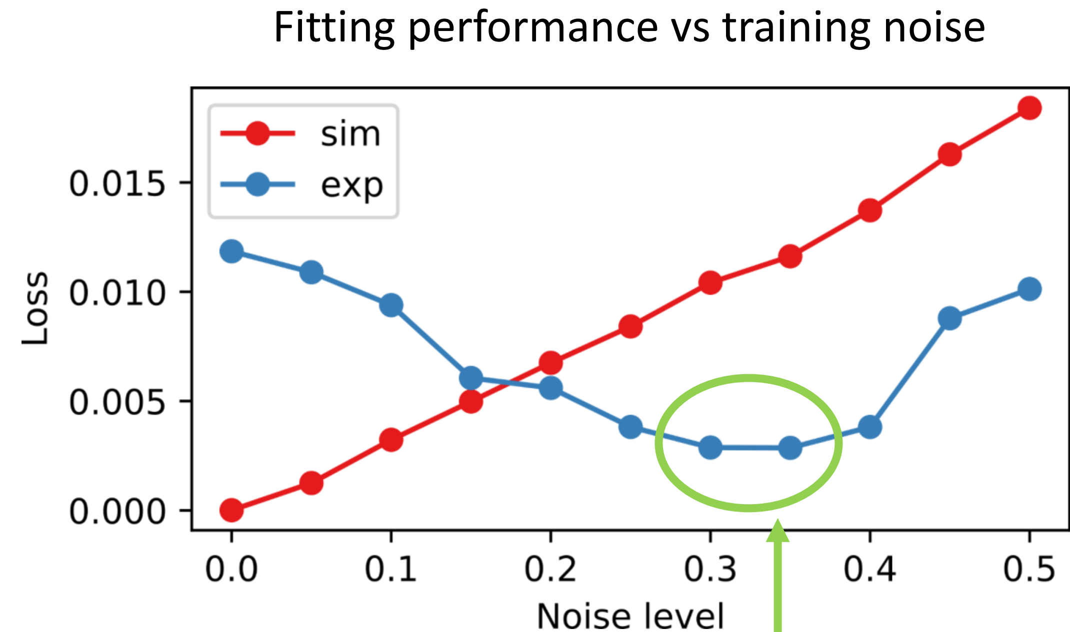
Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

How much noise should be added?

- Train 11 different neural networks with increasing noise on training data
- By applying noise, performance is increased by a factor of up to 3!

Loss:

- Mean squared error across all test data and all parameters
- Low loss means high accuracy



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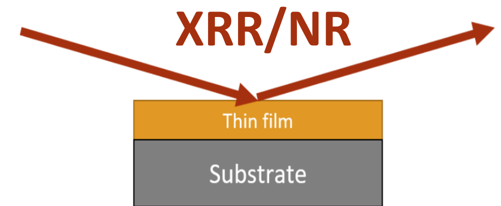
Open Reflectivity Data



- There are ~15-20 publications on ML analysis of reflectivity data
- Training was always done on simulated data
- ML Model evaluation was mostly done on either simulated data or very few reflectivity curves the authors had available.

=> Model evaluation without large labelled dataset is difficult

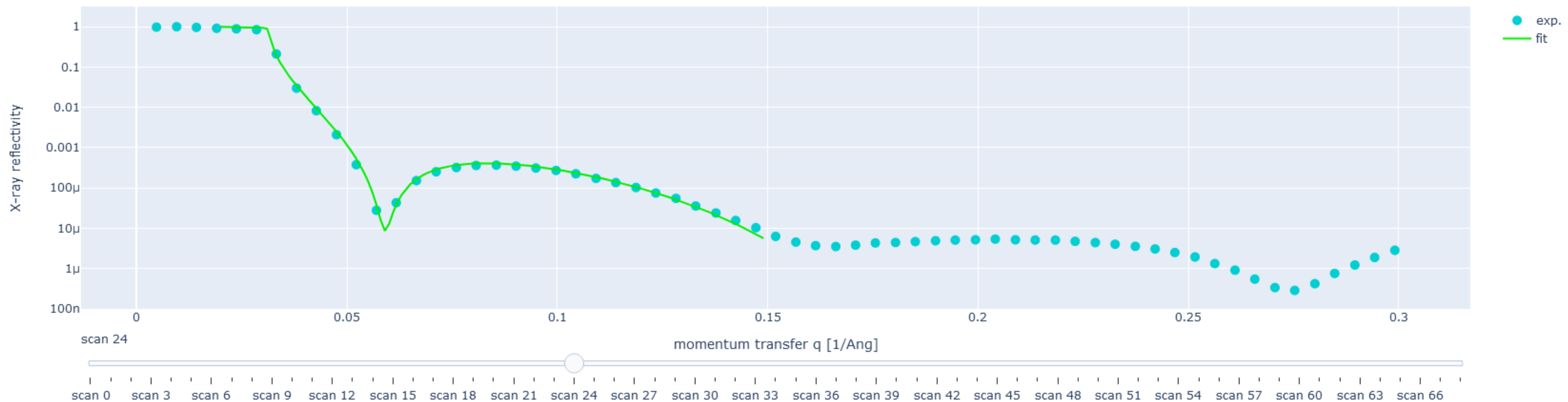
A. Hinderhofer, A. Greco, V. Starostin, V. Munteanu, L. Pithan, A. Gerlach, and F. Schreiber.
Machine learning for scattering data: strategies, perspectives, and applications to surface scattering
J. Appl. Cryst. **56** (2023) 3
https://doi.org/10.5281/zenodo.6497438

Open Reflectivity Data

Dataset: DIP_2
Experimentalists: ['Hinderhofer, Alexander']

	Dataset	Layer_CAS	Layer_formula	Layer_material	Substrate_temperature	Substrate_temperature@unit	instrument	q_max_fit	q_max_fit@unit	year_experiment
0	DIP_2	188-94-3	C32H16	Diindenoperylene	303	K	ESRF, ID10b	0.15	1/Ang	2010

Diindenoperylene on SiOx: 54.05 Ang.



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Median error of improved neural network

Test neural network on a test dataset of **242 curves**

Dataset contains thin films of:
- DIP
- PEN
- PDI-C8
- DNTT:PDIF
- PDI-C8

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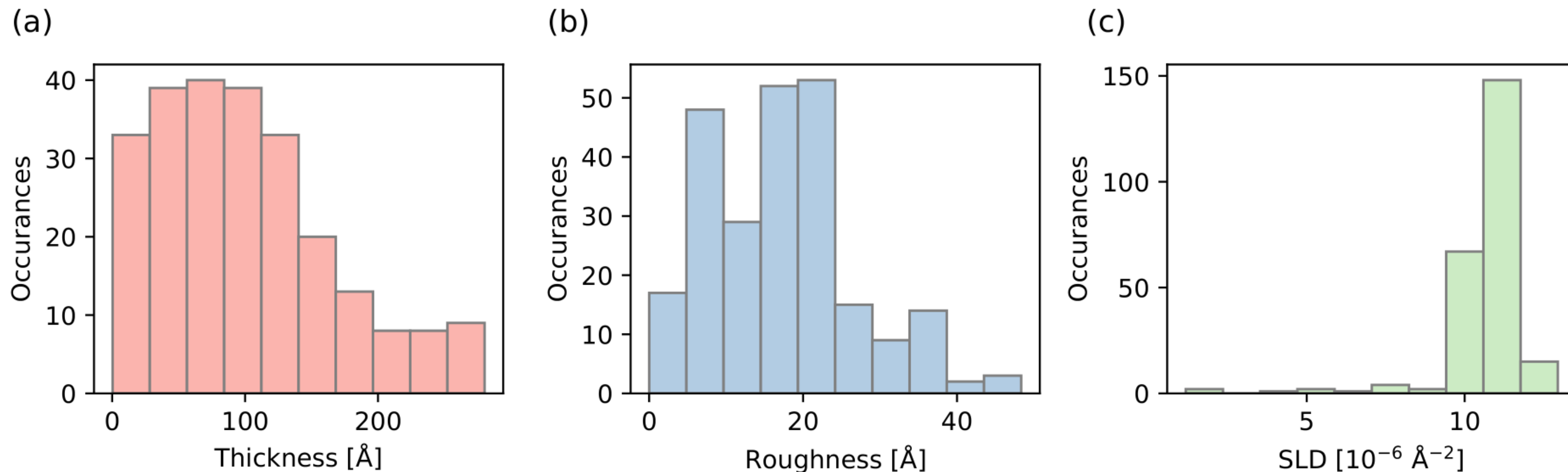
Open Reflectivity Data

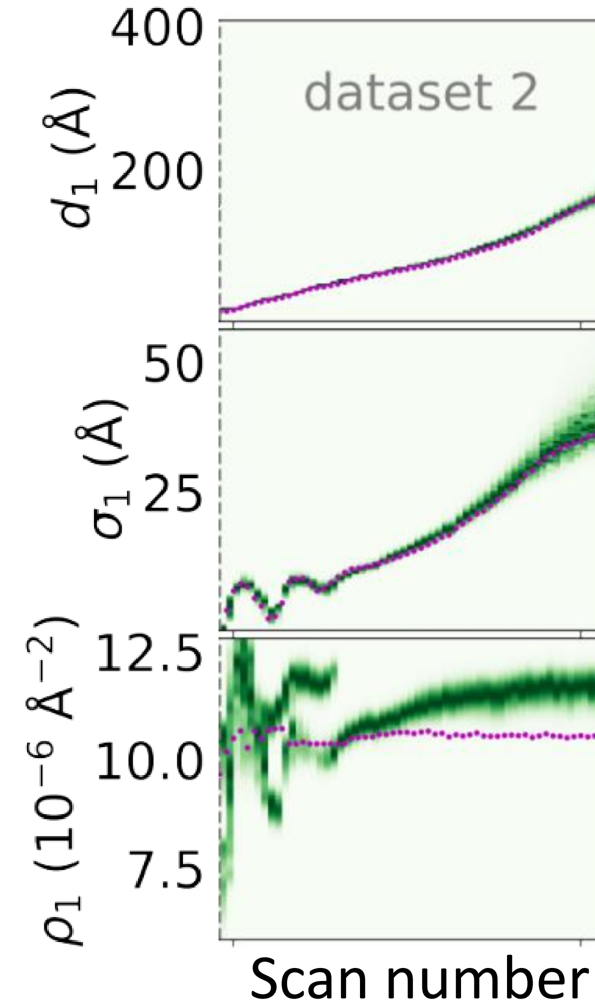
- All films are molecular thin films on a silicon substrate.
- Datasets should have a more diverse parameter variety.



Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

Open Reflectivity Data

- To use experimental data for testing or training, the labelling is critical.
- Labels in our datasets were done by fitting with GenX or RefNX (Differential evolution)
- Importance sampling (IS) shows that SLD labels can still be improved.
- There can be non-unique fitting solutions even in simple models.



Starostin et al., in preparation

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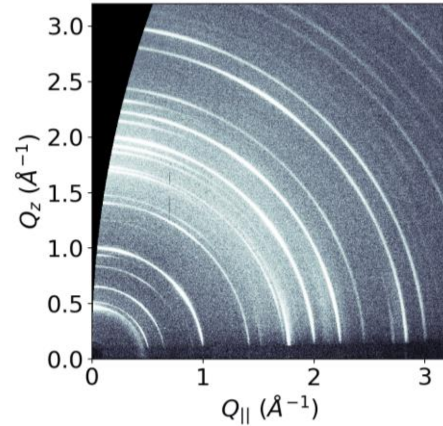
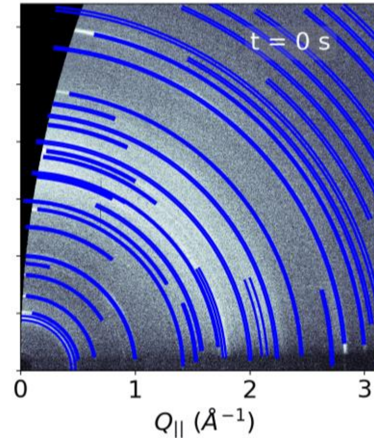
GIWAXS data analysis

In situ measurements



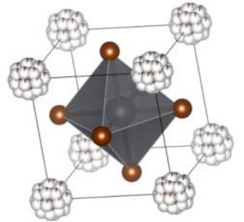
ML Peak Detection

Peak positions & sizes & intensities



Automated analysis

Information about the sample:



- crystal structures
- unit cell orientations
- fractions of coexisting phases
- lattice parameters
- …

≈100k images / day - automated analysis is essential

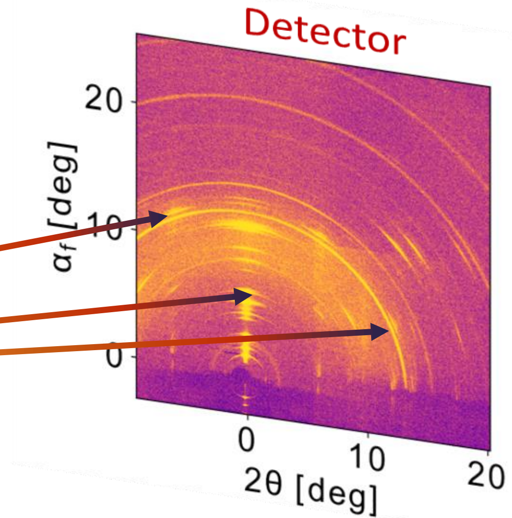
X-ray beam

Grazing-incidence geometry

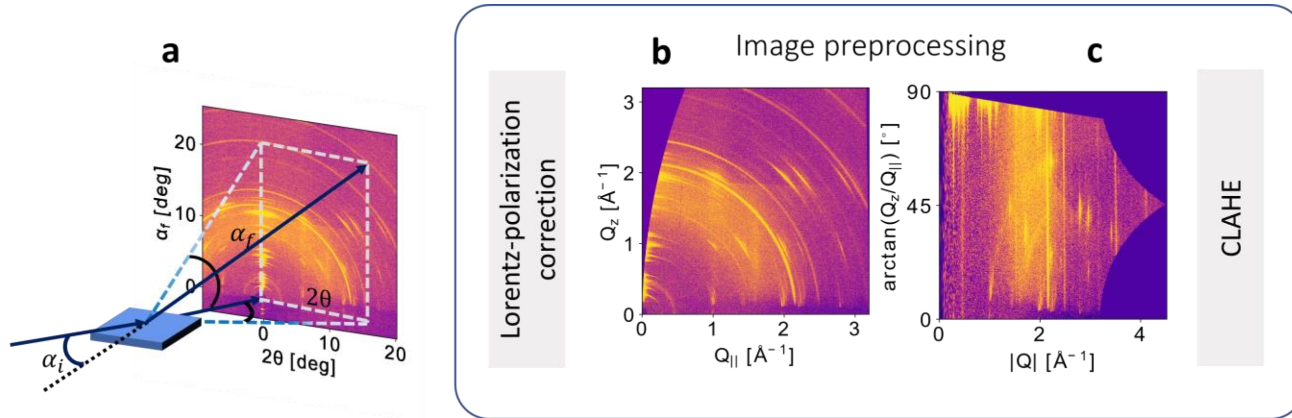
$\approx 0.1^\circ$

Evanescent field

Sample surface



Deep learning peak detection pipeline



- Images converted to polar coordinates
- Optimized lightweight two-stage peak detection architecture
- Asymmetric feature maps to address the specifics of the data geometry

Starostin, V. *et al. npj Comput Mater* **8**, 101 (2022)

Deep learning peak detection pipeline

Compressed image for prolonged peaks

Large resolution for small segments

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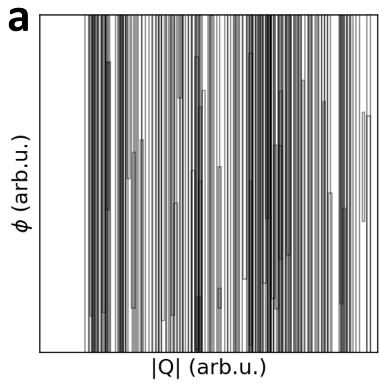
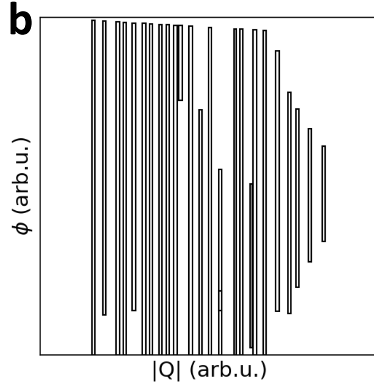
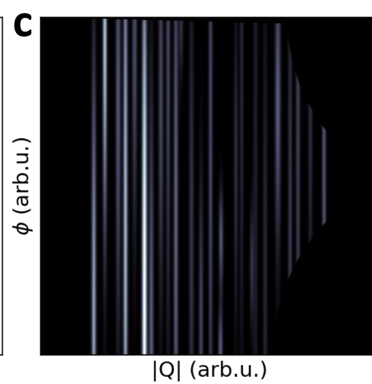
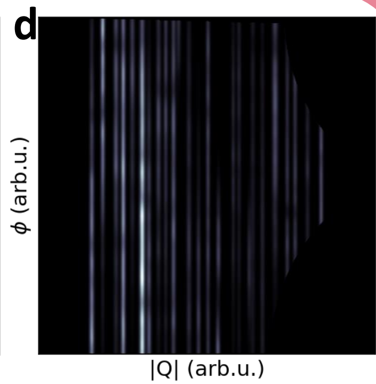
Trained on the simulated data

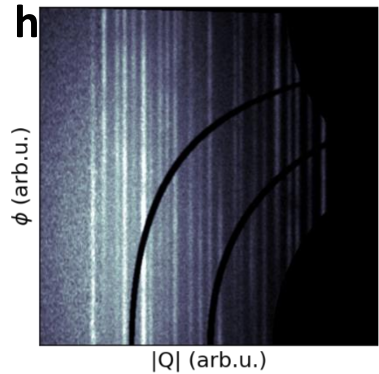
a. Simulate random peak positions & intensities | *b. Filter out strong overlaps* | *c. Generate 2D Gaussian peaks* | *d. Modulate intensities by Perlin noise*

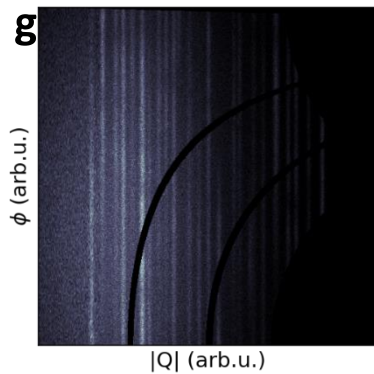


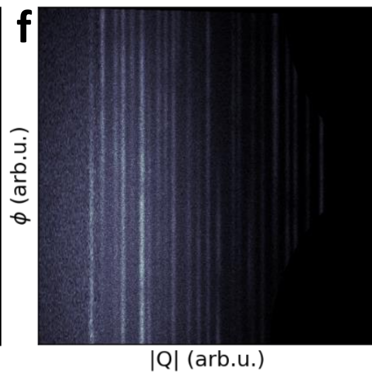


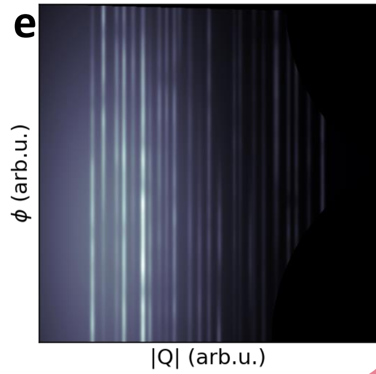












h. Smooth & correct contrast | *g. Add dark areas* | *f. Add noise* | *e. Add backgrounds*

Matched peaks	99.22 %
False negatives	0.78 %
False positives	0.25 %

- Material-agnostic image simulation with counting statistics, experimental artifacts, background scattering.
- Near-ideal performance on the simulated data

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Matching detected peaks with crystal structures

Start of the crystallization process

(a) (b) (c) (d)

Integrated intensity; 10^6; 10^3; MAPbBr$_3$; kapton

$||\mathbf{q}||$ (Å^{-1}); 0; 2; 4; 30; 40; t (s)

(322) (320) (300) (220) (211) (210) (200) (110) (100)

90; 45; 0; ϕ (°); 0; 2; 4; $||\mathbf{q}||$ (Å^{-1})

0; 1; 2; 3; q_{xy} (Å^{-1})

Simulated diffraction profile for the expected crystal structure

The detected peaks **(d)** are **matched against a set of expected crystal structures** and the corresponding simulated diffraction peaks.

The matching is performed for each time-frame of in situ measurements **(b)**.

Based on the matching results, we can **extract integrated intensities** corresponding to the matched structures **(a)**

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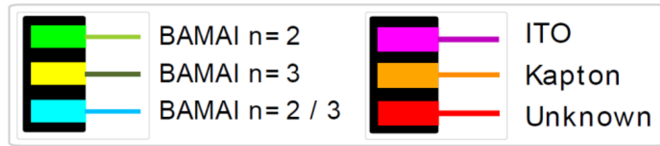
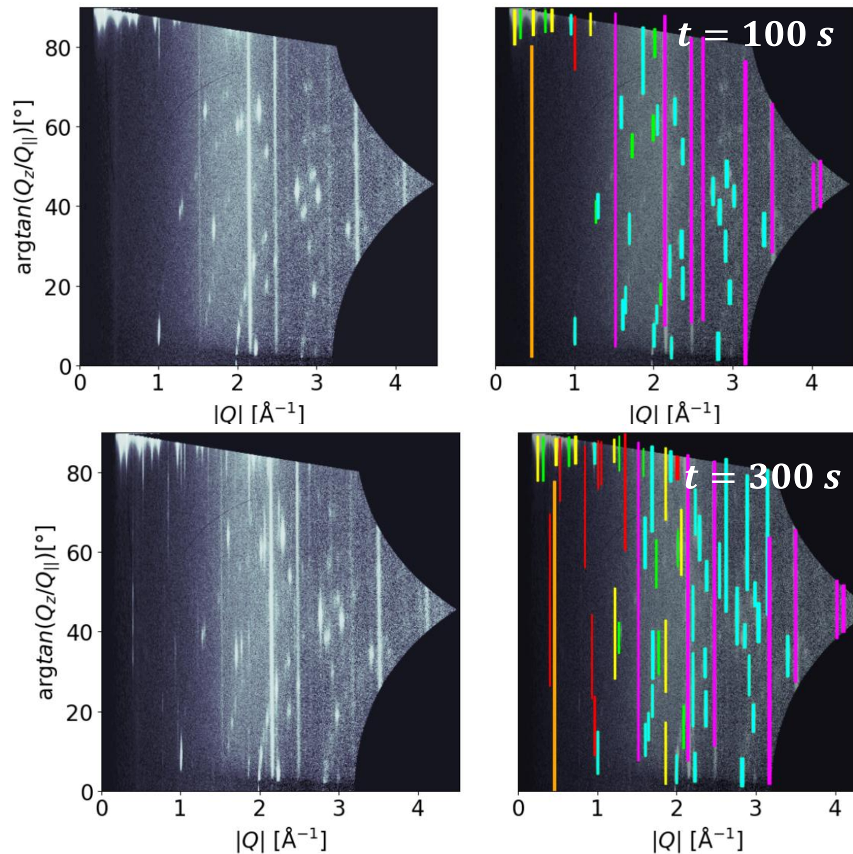
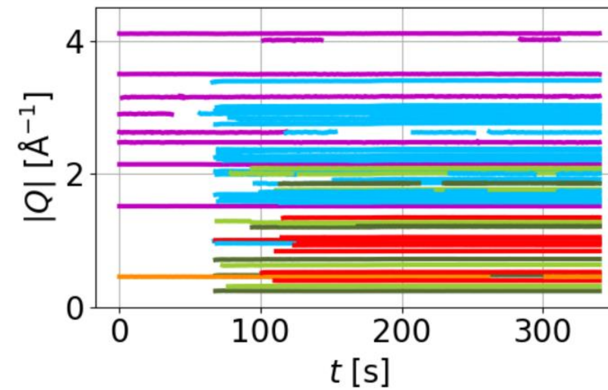
Automated phase & lattice parameters determination

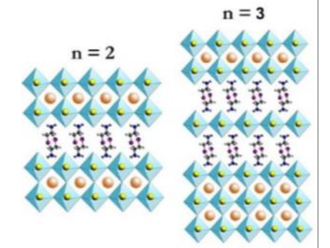


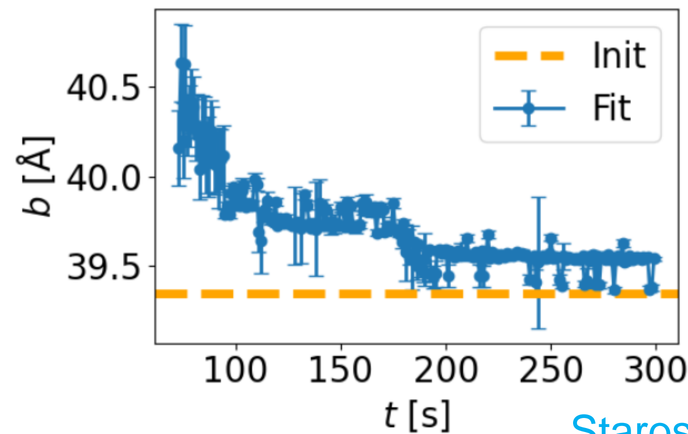
Matching algorithm identifies structures based on detected peaks





Identified radial peak positions vs time pinpoint the start of the crystallization processes





Lattice parameter refinement reveals a shift in spacer molecule length. Such subtle processes are frequently overlooked by the manual analysis

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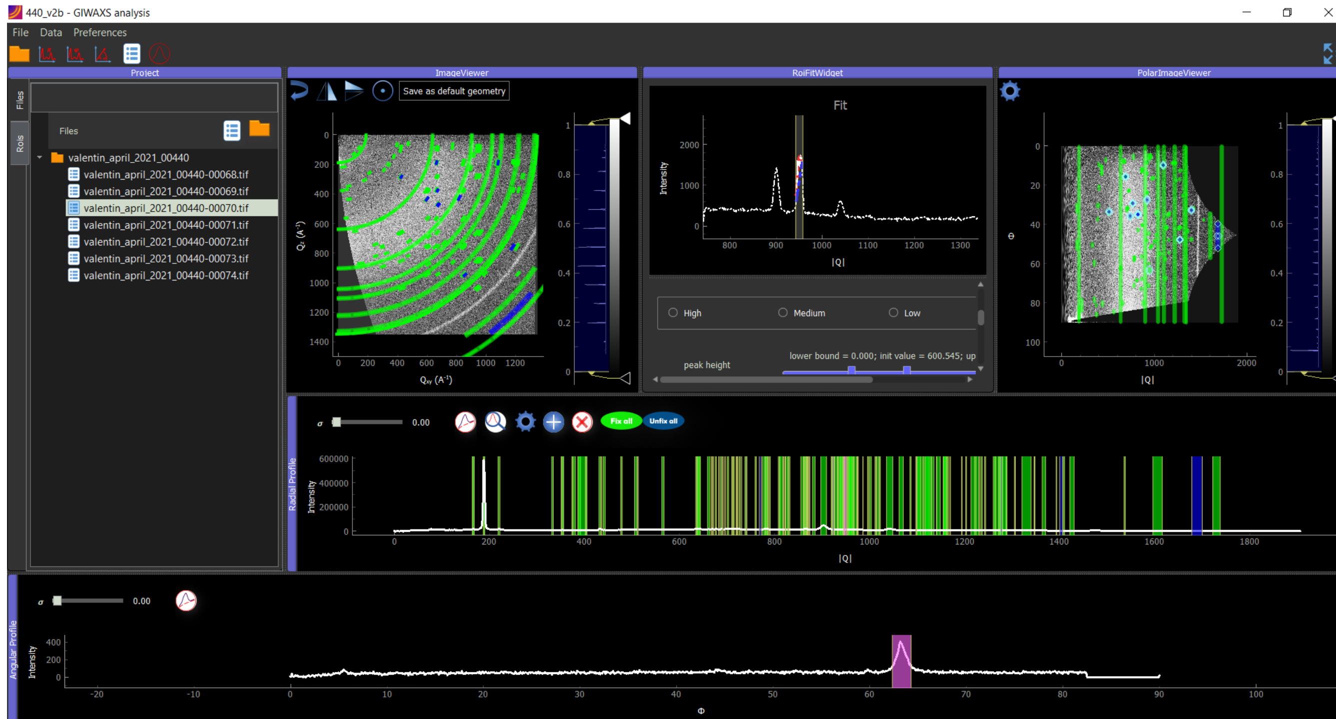
Tested on the experimental data

Designed GUI for GIWAXS data annotation



- 3 confidence levels (high, med, low)
- ML predicts lower number of false positives compared to students
- Low intensity peaks are still hard to detect

Test results on the experimental data

Metric	Master / Bachelor students	Deep learning model
High confidence peaks detected	96 %	95 %
Medium confidence peaks detected	86 %	76 %
Low confidence peaks detected	74 %	42 %
Share of false positives	18 %	**7 %**
Time per image	2 hours	**8 ms**

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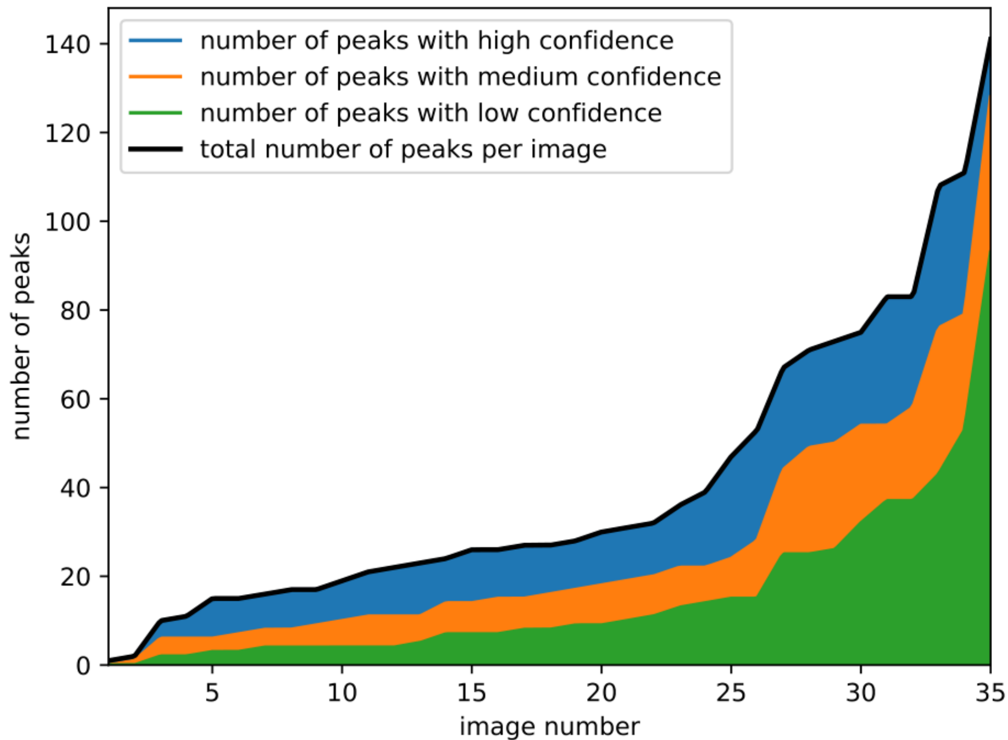
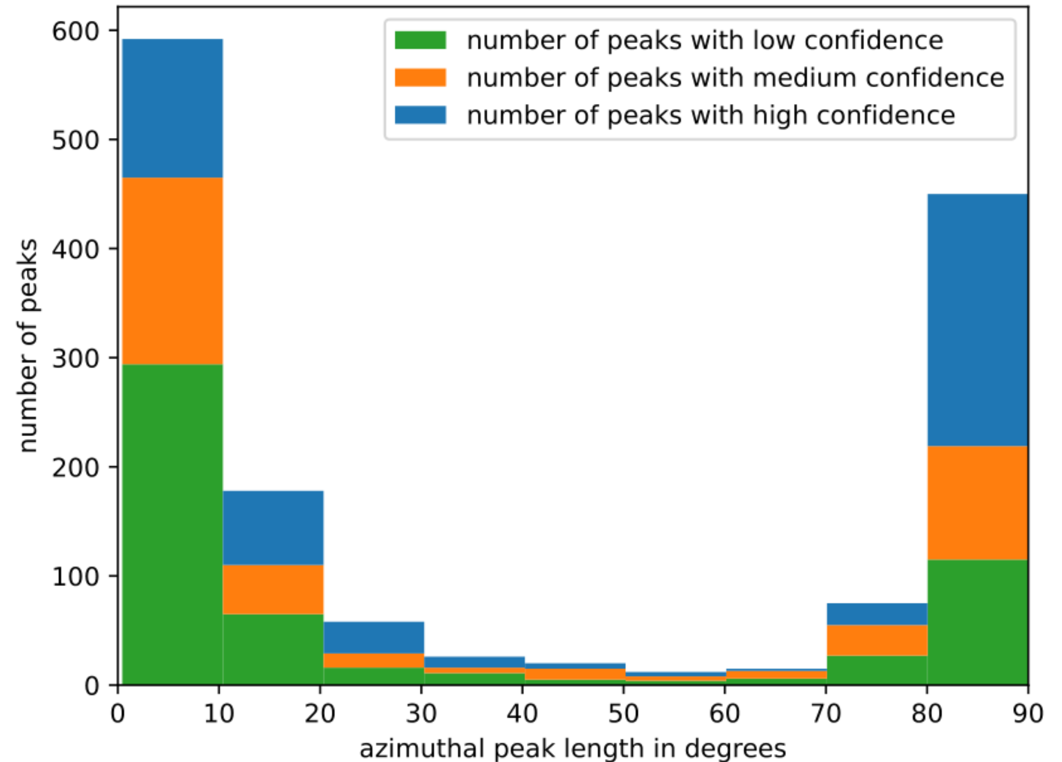
Labelled experimental data

35 GIWAXS images with ~1600 fitted peaks





C. Voelter, *et al.*, in preparation

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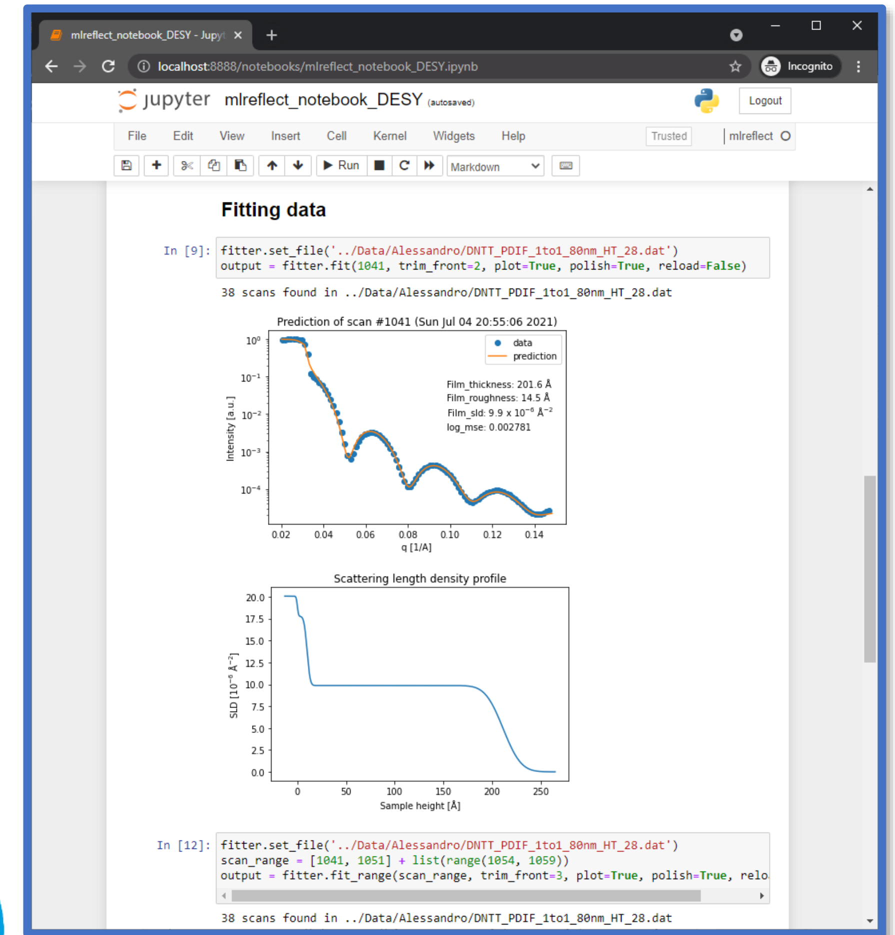
Neural network is part of the *mlreflect* package

Python package *mlreflect* was developed for a BMBF project in collaboration with DESY

- Available on GitHub
- Installable via PyPI
- Online documentation available on Read the Docs
- Can be used with Jupyter notebooks as GUI
- Is already installed on our home XRR machine
- Also installed at DESY, PETRA III
- New “prior aware” model allows definition of more complex models without retraining



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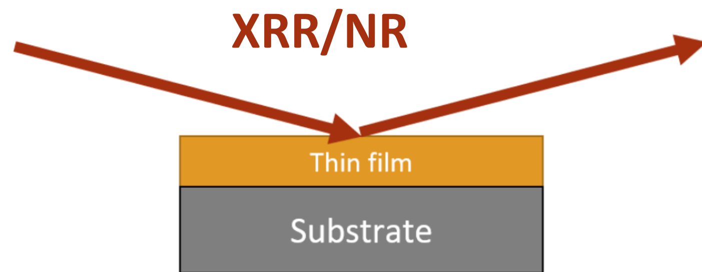
für Bildung
und Forschung

Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)
Munteanu et al., submitted

Conclusion

- Large labeled datasets for surface scattering (reflectometry + GIWAXS) are very rare.
- Issue 1: Collecting *diverse* data
- Issue 2: Preparing high quality labels for collected data
- Develop ML tools to enhance labeling process



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https://doi.org/10.5281/zenodo.6497438 (Open XRR dataset)

nfdi Nationale Forschungsdaten Infrastruktur

DFG Deutsche Forschungsgemeinschaft

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