Deutsches Elektronen-Synchrotron DESY
Ein Forschungszentrum
der Helmholtz-Gemeinschaft

CLOSED-LOOP EXPERIMENTS USING ML-BASED ONLINE ANALYSIS AT SYNCHROTRON BEAMLINES: A CASE STUDY IN X-RAY REFLECTOMETRY

Linus Pithan, Vladimir Starostin, Valentin Munteanu,

Alexander Hinderhofer, Stefan Kowarik, Frank Schreiber

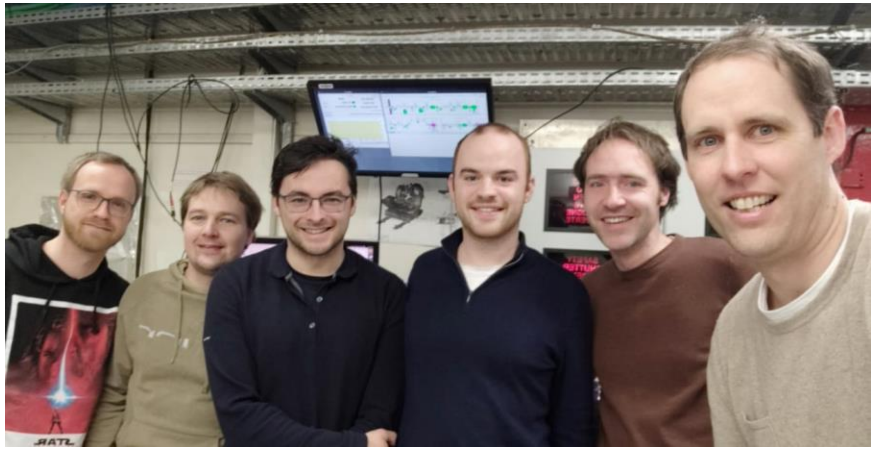
Agenda – It's about ML and Reflectometry

A story of an excellent, long-standing collaboration

PETRA III.

ESRF The European Synchrotron

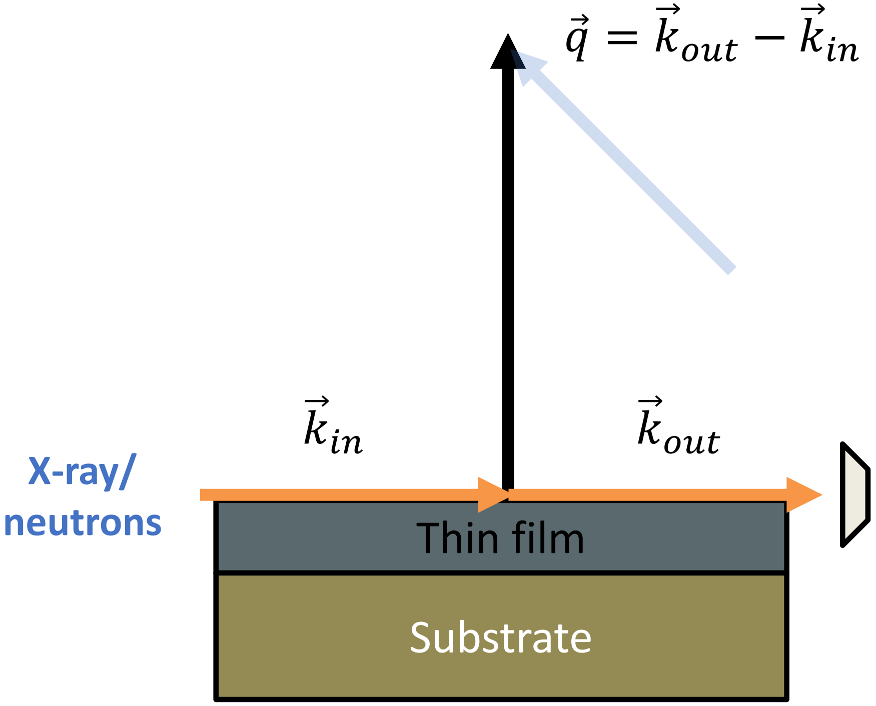
Frank Schreiber's group (Uni Tübingen)
Stefan Kowarik's group (Uni Graz)
Vladimir Starostin (ML Cluster Tübingen)



X-ray and neutron reflectivity (XRR/NR)

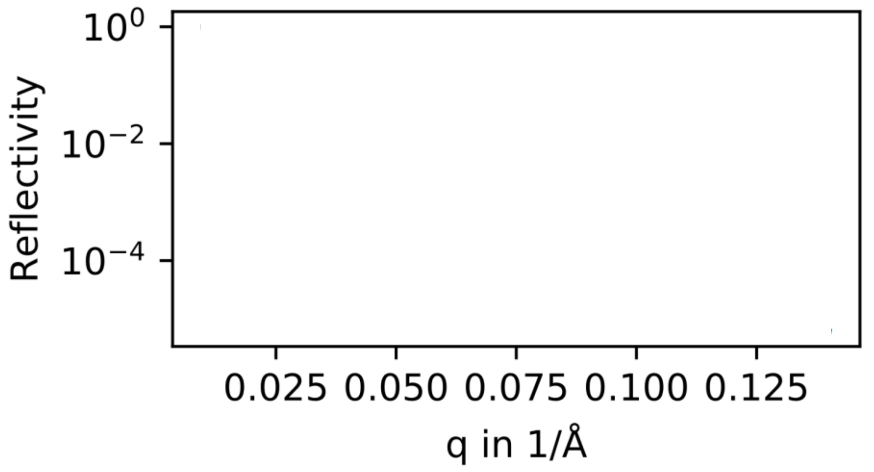
Experiment

X-ray beam at certain discrete angles



Data

Reflected beam intensity for each angle



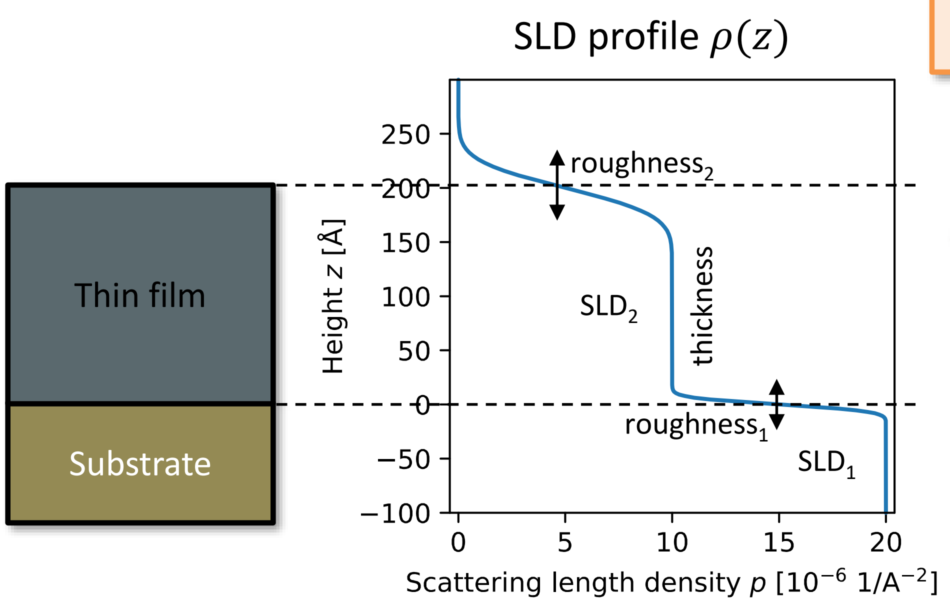
Shape of reflectivity curve provides information about thin film properties

Greco et al. J. Appl. Crystallogr., 2019, 52, 1342

ML: Modeling of the "back-transformation"

$$R(q_z) \propto q_z^{-4} \left| \int \frac{d\rho(z)}{dz} e^{iq_z z} dz \right|^2$$

Fourier transform with phase loss!

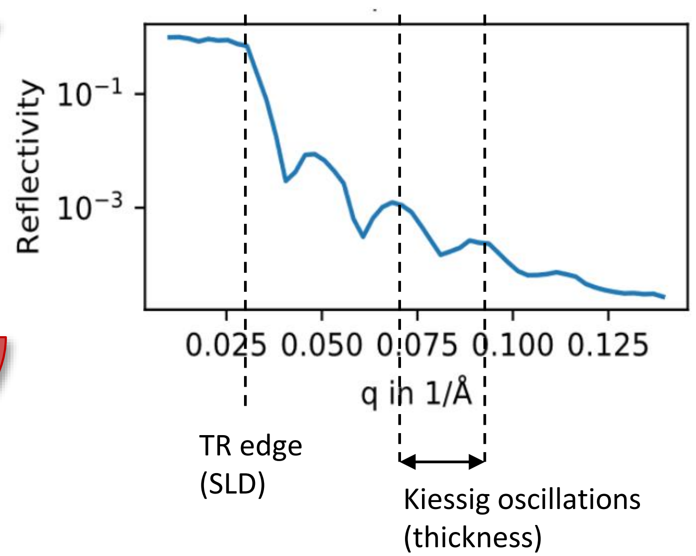


Theoretical models
(Parratt, matrix method, kinematic approximation)

Iterative fitting
(LMS, χ^2, posterior probability estimation)

No analytical "back-transformation"!

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



SLD profile usually parameterized, *i.e.*
$\rho(z) = \rho(z;\boldsymbol{p})$, with $\boldsymbol{p}$ = (thickness, roughness, SLD)

Greco et al. J. Appl. Crystallogr., 2019, 52, 1342

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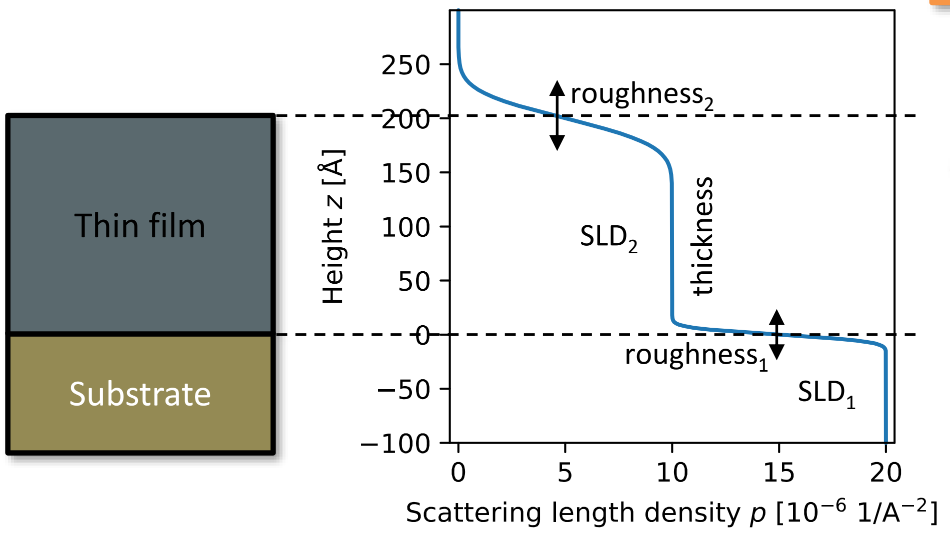
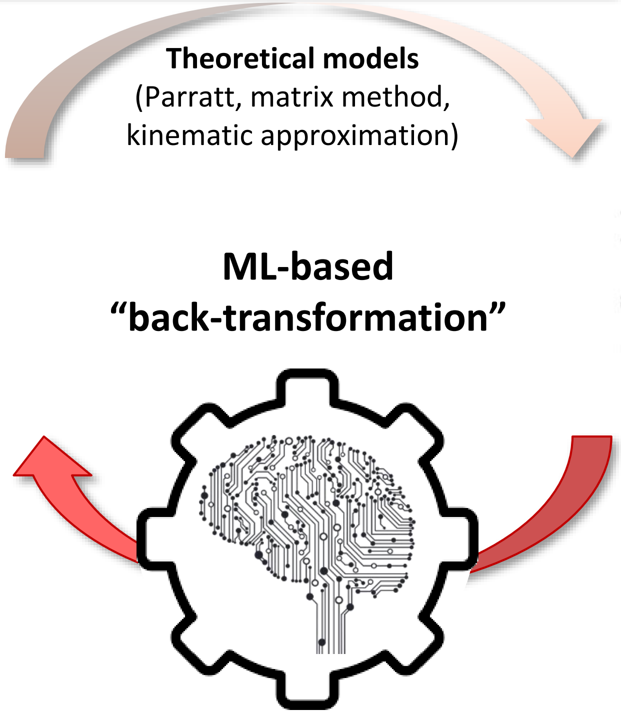
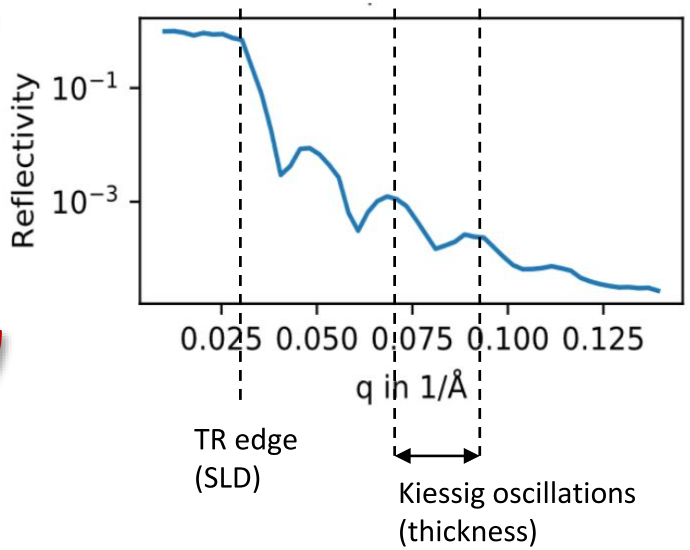
SLD profile $\rho(z)$





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



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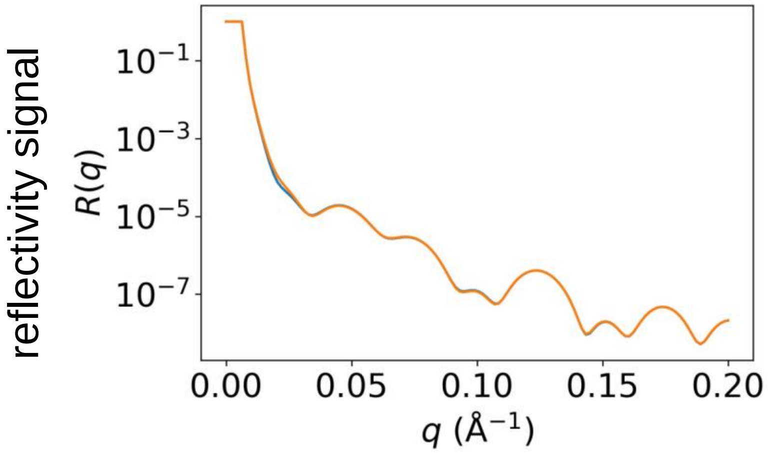
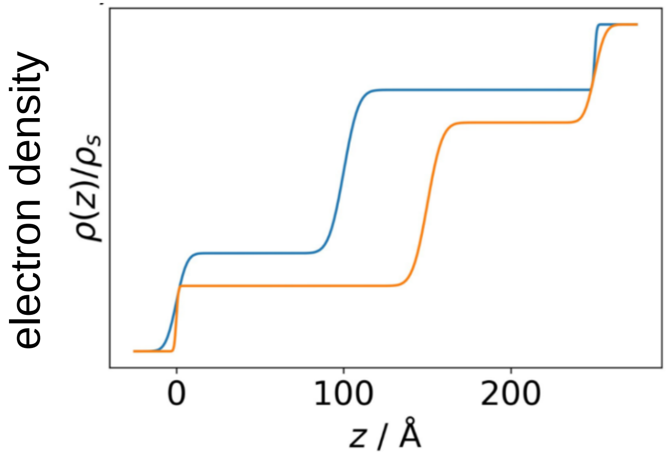
Greco et al. J. Appl. Crystallogr., 2019, 52, 1342

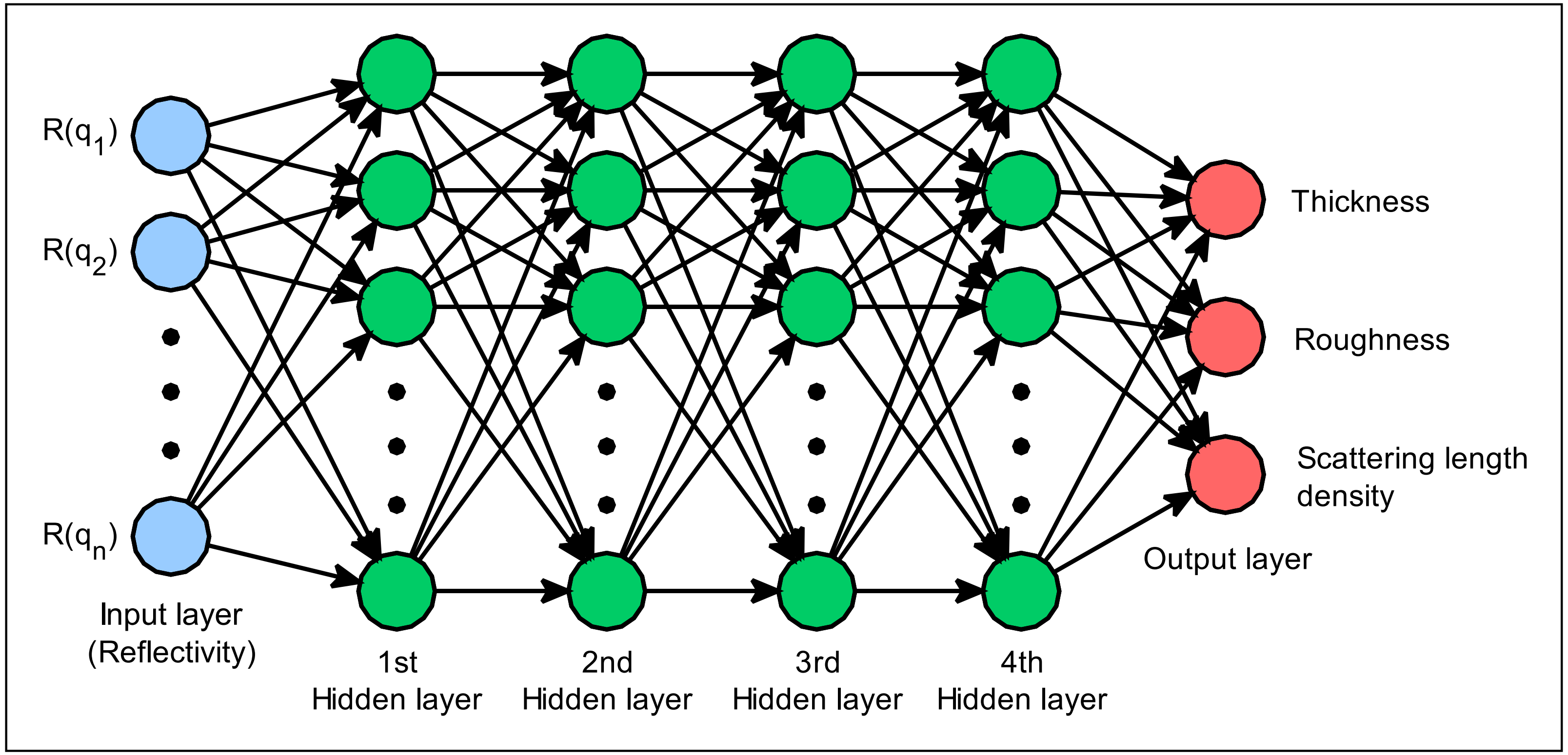
ill-posed inverse problems



scattering vector



sample depth profile



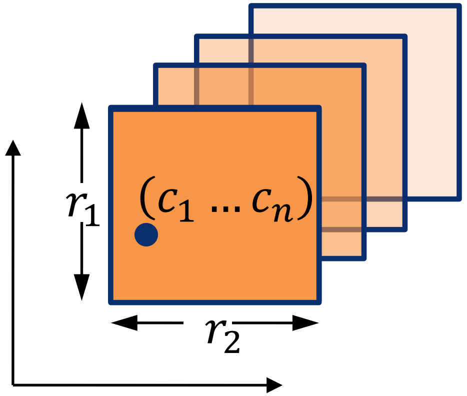
- Attempting to train a neural network as a point estimator over the domain containing the non-uniqueness leads to incorrect predictions corresponding

V. Munteanu et al., J. Appl. Cryst. 2024, 57,
DOI: 10.1107/S1600576724002115

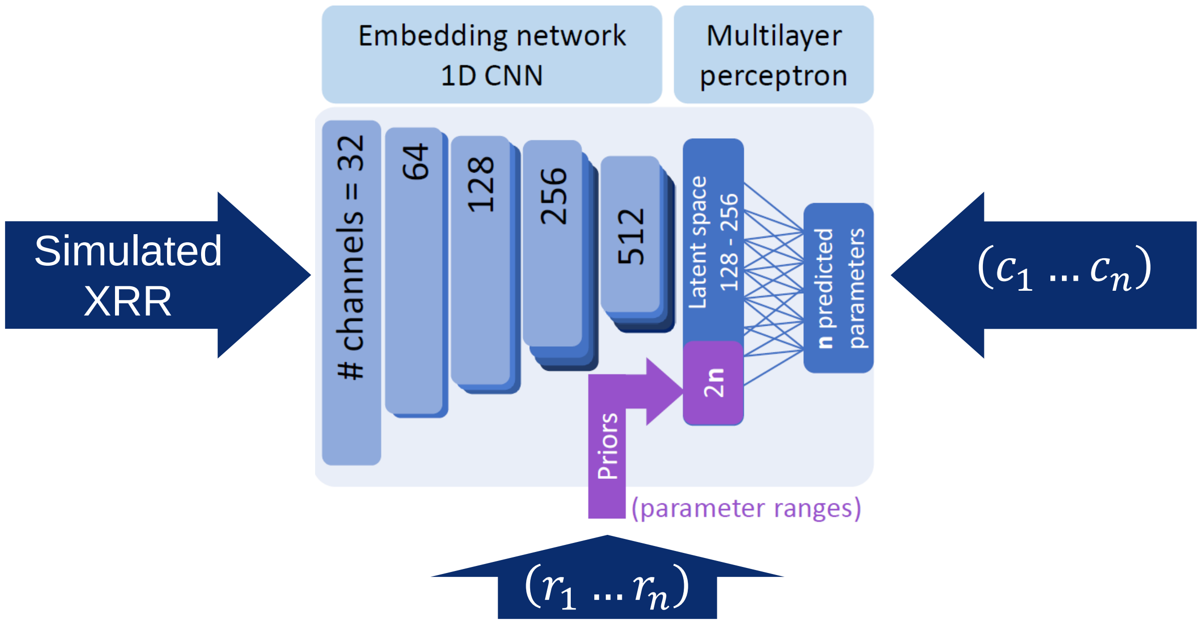
Reflectorch: Including prior knowledge

- How to tackle ill-posed inverse problems by taking advantage of the *a priori* knowledge of experimentalists?

During Training:



parameter space



Key aspects:

- Think of the training process as simultaneously learning on all possible subdomains (or a subset of subdomains) of the full parameter space

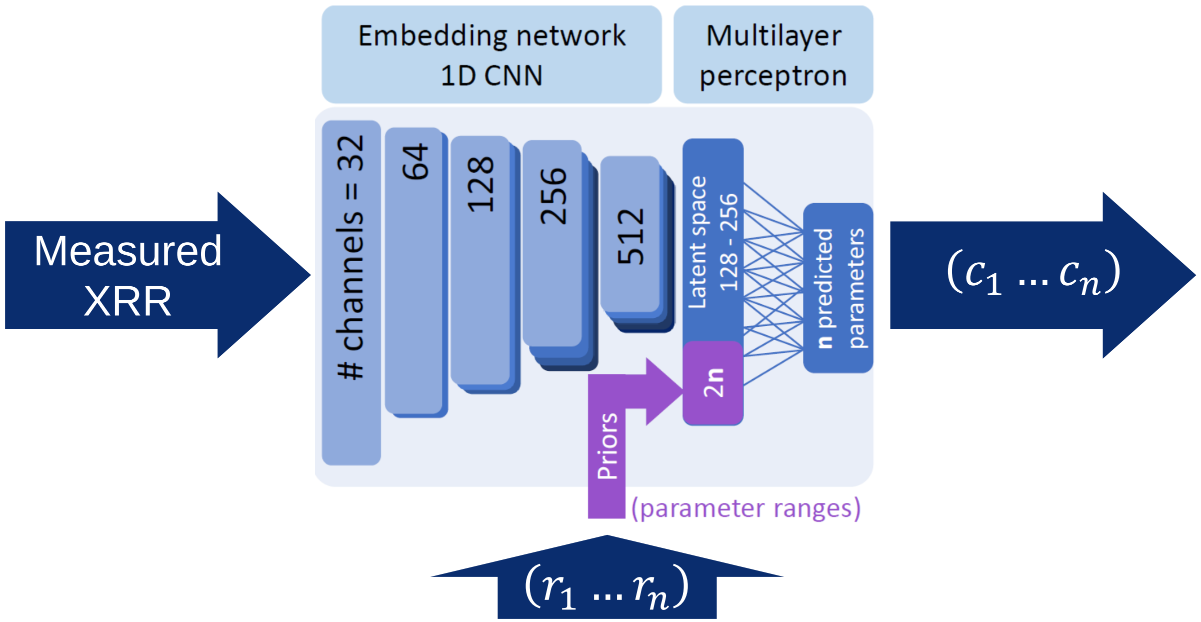
- **github.com/schreiber-lab/reflectorch**

V. Munteanu, , V. Starostin, A. Greco, L. Pithan, A. Gerlach, A. Hinderhofer, S. Kowarik and F. Schreiber. J. Appl. Cryst. **57, 2024, DOI** 10.1107/S1600576724002115

Reflectorch: Including prior knowledge

- How to tackle ill-posed inverse problems by taking advantage of the *a priori* knowledge of experimentalists?

At inference:



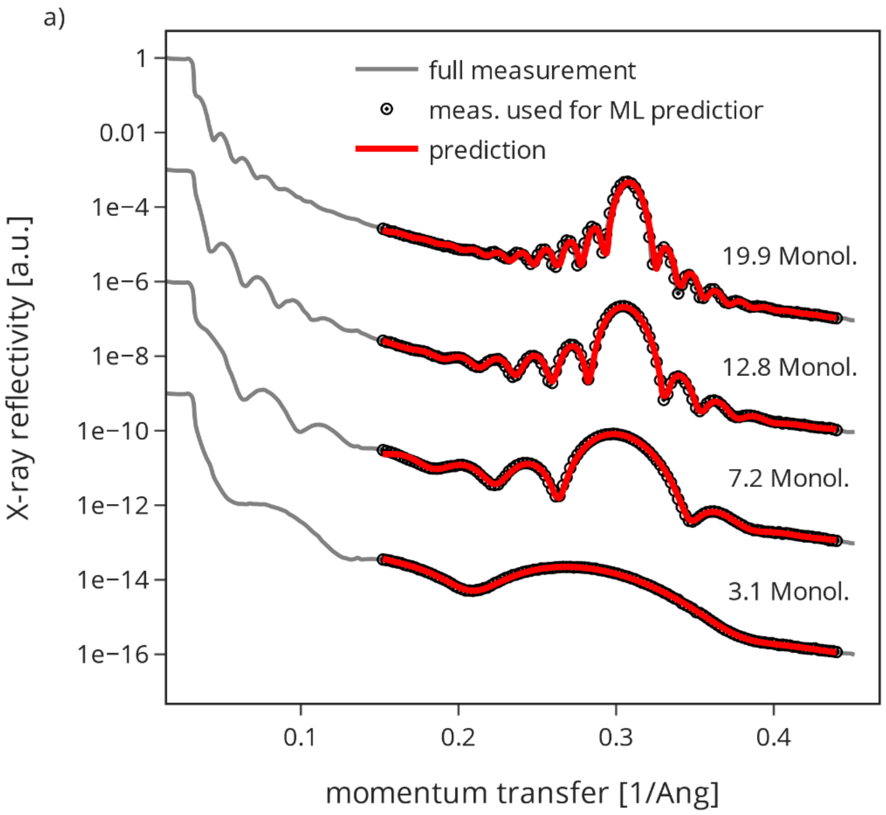
Key aspects:

- Think of the training process as simultaneously learning on all possible subdomains (or a subset of subdomains) of the full parameter space
- Combination of high speed of neural networks with the flexibility of conventional fitting procedures
- Embedding network produces latent embedding of XRR/NR data
- **github.com/schreiber-lab/reflectorch**

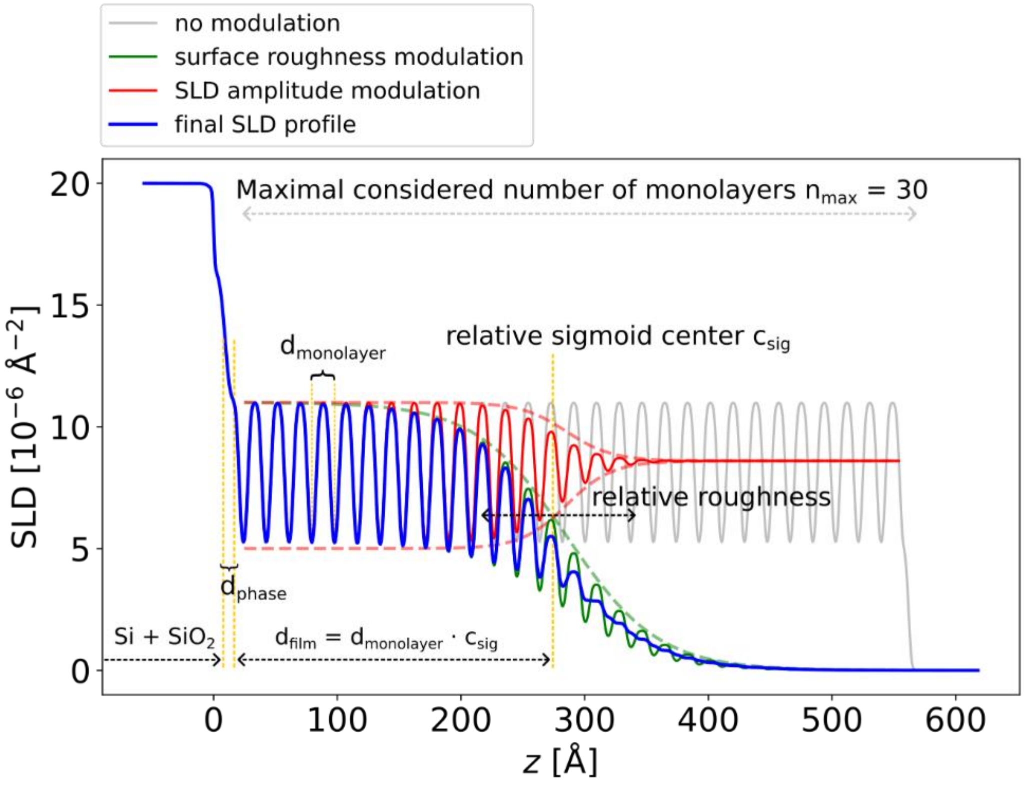
V. Munteanu, , V. Starostin, A. Greco, L. Pithan, A. Gerlach, A. Hinderhofer, S. Kowarik and F. Schreiber. J. Appl. Cryst. **57, 2024,** **DOI** 10.1107/S1600576724002115

High-dimensional parameter spaces

XRR data during growth (PTCDI-C8)



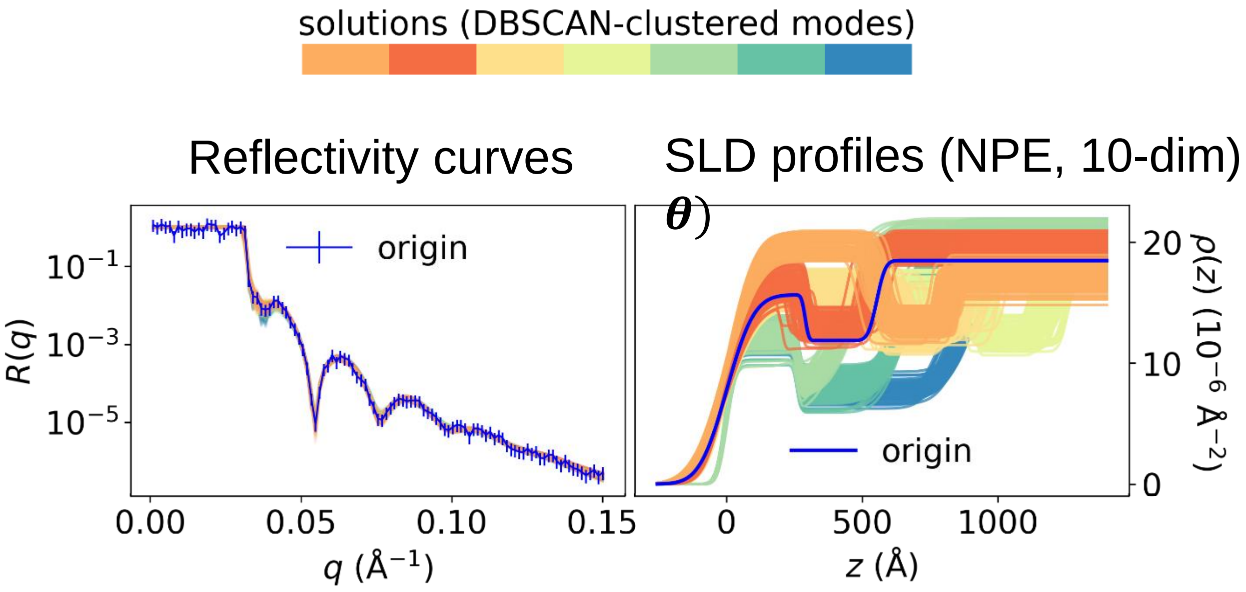
Multilayer model for (PTCDI-C8)



- Enables physics informed parametrization
- Demonstrated to work for live feedback with more than 15 open parameters during film growth

L. Pithan et al, *Closing the loop: Autonomous experiments enabled by machine-learning-based online data analysis in synchrotron beamline environments*, J. Synchrotron Rad. **30** (2023) 1064

Simulation-based inference (SBI) – general ML framework for Bayesian inference



Main Idea:
- Swap NN with e.g. Normalizing Flows
- (take symmetries in the data into account)

→ Get to probabilistic model

Reliable

No missed solutions

Dax *et al. Phys. Rev. Lett.*, 2023, **130**, 171403

Accurate

Result easily validated and **corrected** (Neural Importance Sampling)

Müller *et al., ACM Trans. Graph.*, 2019, **38**, 1

Fast

Analysis performed in seconds

Fast PyTorch simulator developed

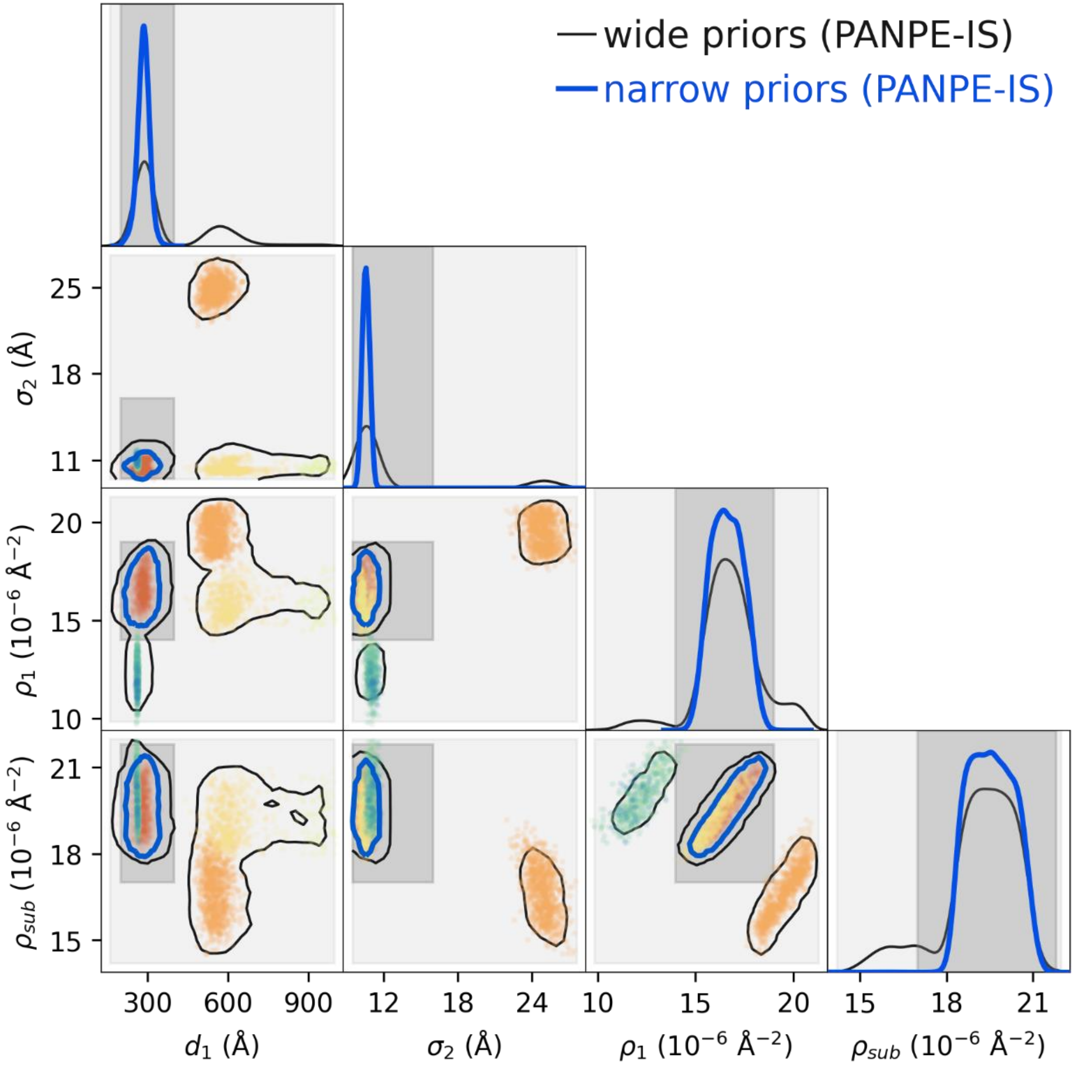
Flexible

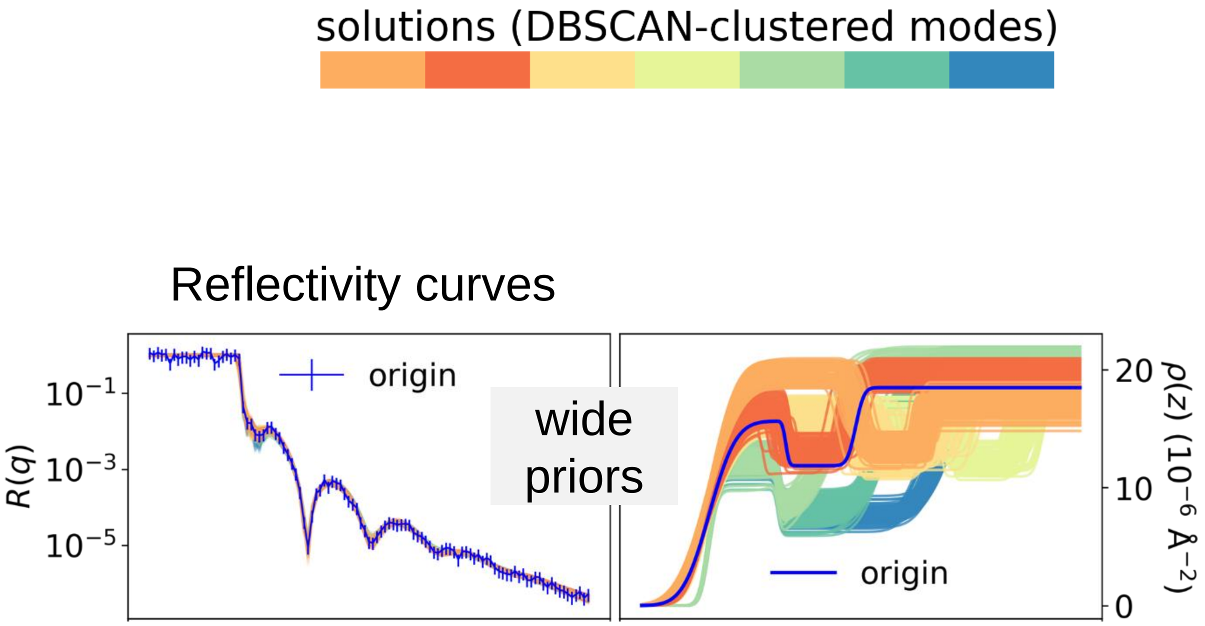
Prior-Amortized Neural Posterior Estimation (PANPE) approach enables incorporation of prior information

Starostin et al., 2024

V. Starostin et al., 2024, 10.48550/arXiv.2407.18648

Incorporating dynamic prior information into SBI





V. Starostin et al., 2024, 10.48550/arXiv.2407.18648 ml science colaboratory

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arXiv > physics > arXiv:2407.18648

Physics > Applied Physics

[Submitted on 26 Jul 2024]

Fast and Reliable Probabilistic Reflectometry Inversion with Prior-Amortized Neural Posterior Estimation

Vladimir Starostin, Maximilian Dax, Alexander Gerlach, Alexander Hinderhofer, Álvaro Tejero-Cantero, Frank Schreiber

Subjects: **Applied Physics (physics.app-ph)**; Soft Condensed Matter (cond-mat.soft); Machine Learning (cs.LG); Machine Learning (stat.ML)

Cite as: arXiv:2407.18648 **[physics.app-ph]**

(or arXiv:2407.18648v1 **[physics.app-ph]** for this version)

https://doi.org/10.48550/arXiv.2407.18648

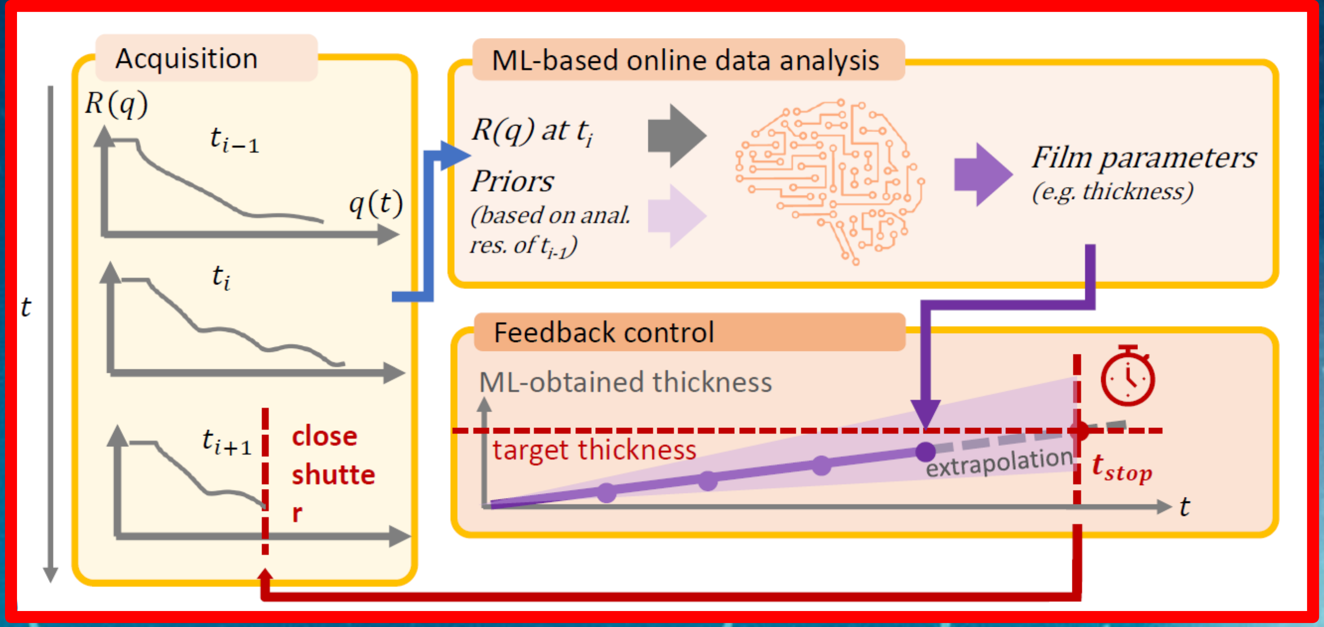
submitted to ScienceAdcances

Machine learning

closed loop

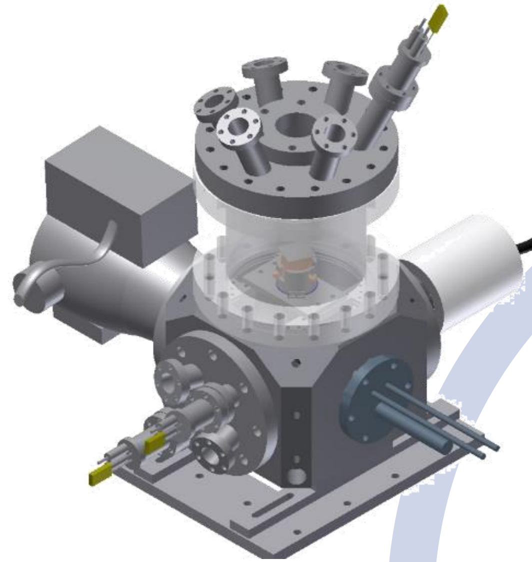
Autonomous Experiments

Experiment control



L. Pithan et al, J. Synchrotron Rad. **30** (2023) 1064

A simple autonomous experiment

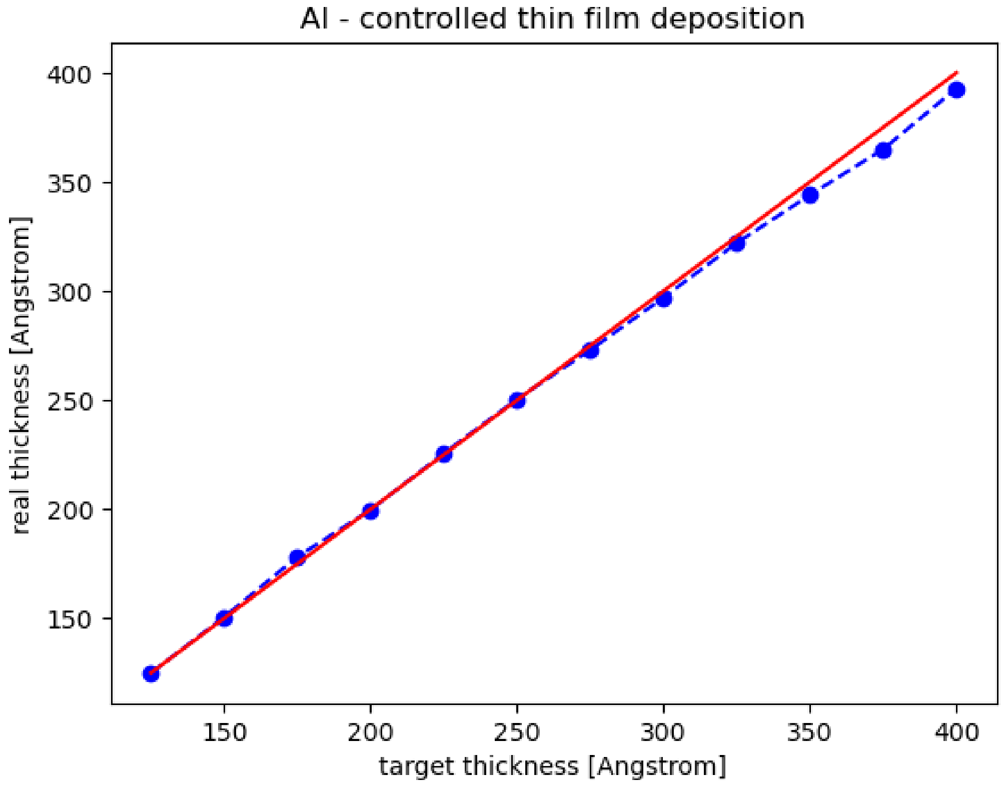


measure reflectivity

process data on the fly

Predict physical parameters with machine learning

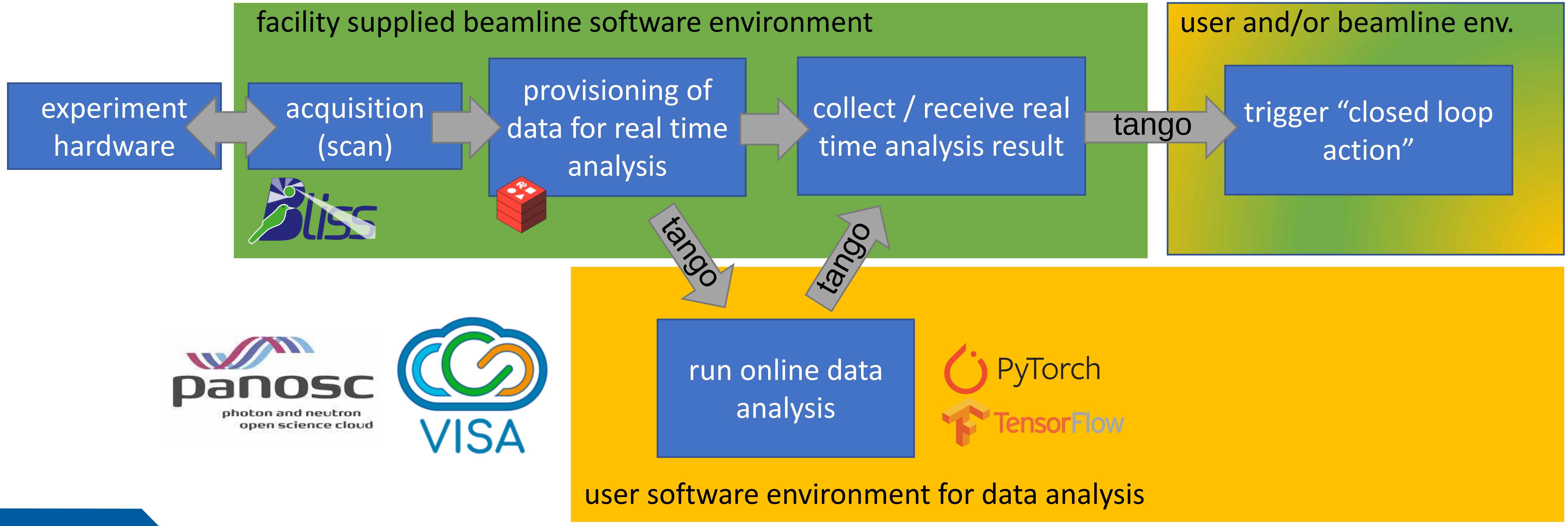
act on sample environment



L. Pithan et al, J. Synchrotron Rad. **30** (2023) 1064
J.-F. Perrin, ESRF Highlights 2022, 158-159 (2022).
https://www.tango-controls.org,
https://bliss.gitlab-pages.esrf.fr/bliss

Software environments

- No need for user software to be installed in the beamline environment



L. Pithan et al, J. Synchrotron Rad. **30** (2023) 1064

Outlook: ML integration into loop beamline operation

- What infrastructure should facilities provide to enable visiting users to systematically perform autonomous experiments ?
- How transferable can these solutions between facilities in order to be transparent for the user?

PETRA IV.
Which control system to choose?

Bliss
sardana
bluesky

ROCK-IT

https://www.rock-it-project.de/

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

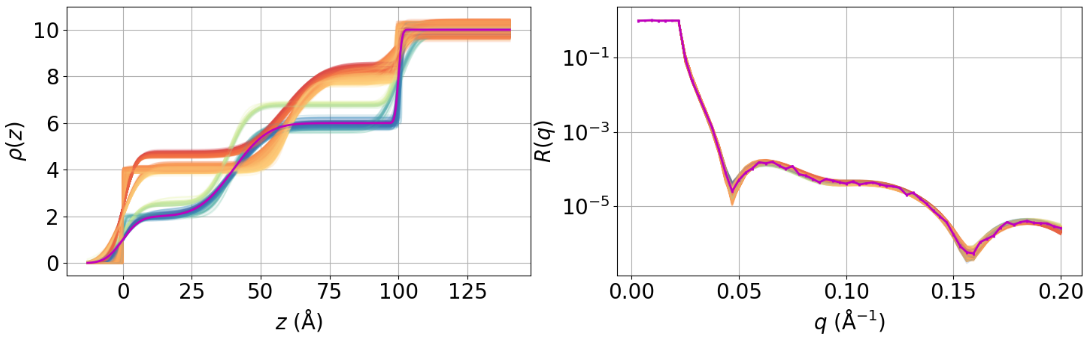
DIGITAL LEAPS

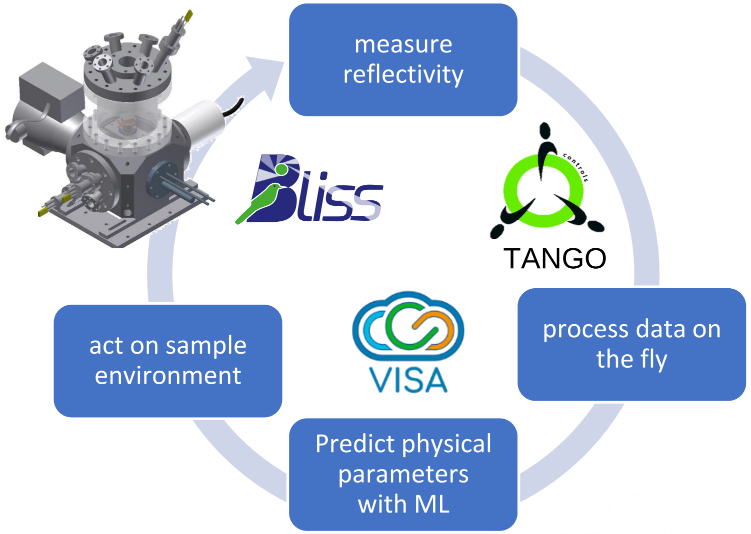
Fully Automated Beamline

Please get in touch with me in case you'd like to be involved!

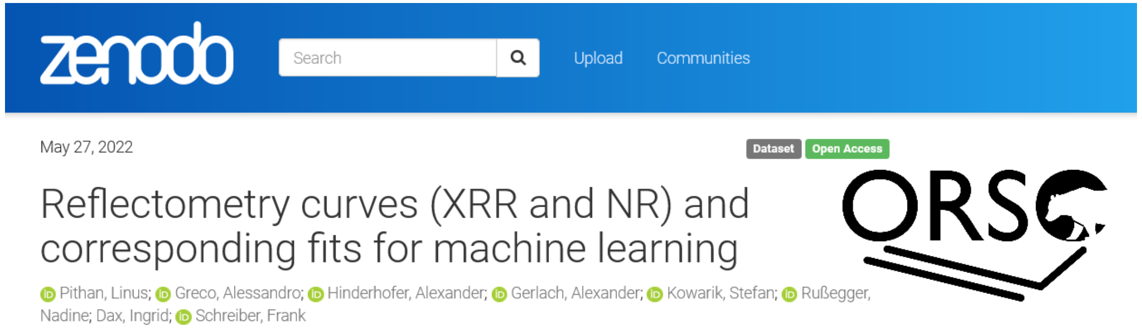
Summary

- Neural networks for “fitting” XRR data
  - Multilayer Percepton (ML-Reflect package)
  - Incorporation of prior knowledge (Relectorch package)
  - Prior-Amortized Neural Posterior Estimation (PANPE)
- Closed loop feedback & beamline integration





Main references:
github.com/schreiber-lab
L. Pithan et al, J. Synchrotron Rad. 30 (2023) 1064
A. Greco et al., *J. Appl. Cryst.* 55, 362 (2022)
A. Hinderhofer et al., J. Appl. Cryst. 56 (2023)
V. Munteanu et al., J. Appl. Cryst. 57 (2024)
V. Starostin et al., 2024, 10.48550/arXiv.2407.18648



Labelled reflectometry data

L.Pithan et al. 2022 https://doi.org/10.5281/zenodo.6497438

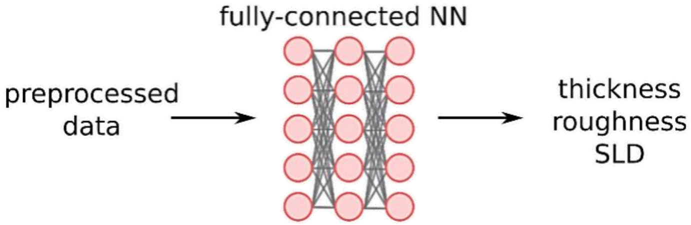
Different approaches to inverse problem with ML

In general, inverse problem in reflectometry is ill-posed due to *the phase problem* → *possible multimodal solutions*

***Point estimators**. To avoid ambiguity, **the task should be narrowed down to specific cases** (e.g., silicon + silicon oxide + organic layer)*

***Probability density estimators.** Resolves the ambiguity issue*

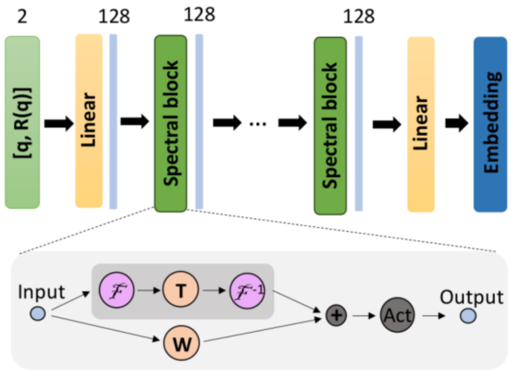
Regression via NNs (MLP, CNN, …)



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

- Simple implementation
- Fast inference

Fourier Neural Operator (FNO)



V. Munteanu et al., J. Appl. Cryst. 2024, 57, DOI: 10.1107/S1600576724002115

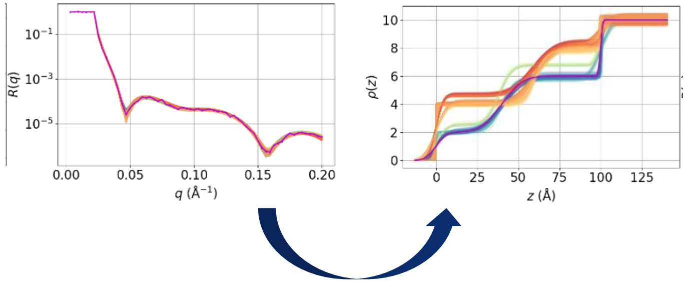
- Discretization-invariant learning

- Fails on multimodal cases
- Does not account for parameter distribution
- Does not provide error bars / uncertainty estimation
- Likely that retraining the ML-model is needed for different use cases

Normalizing Flows

neural posterior estimation

Experimental curve   Sampled SLD profiles



Sample profiles via conditional inverse Normalizing Flows transformation

Starostin et al., 2024

- Accelerated Bayesian analysis
- Resolves ambiguity problem
- Provides error bars
- No retraining required
- More difficult to implement

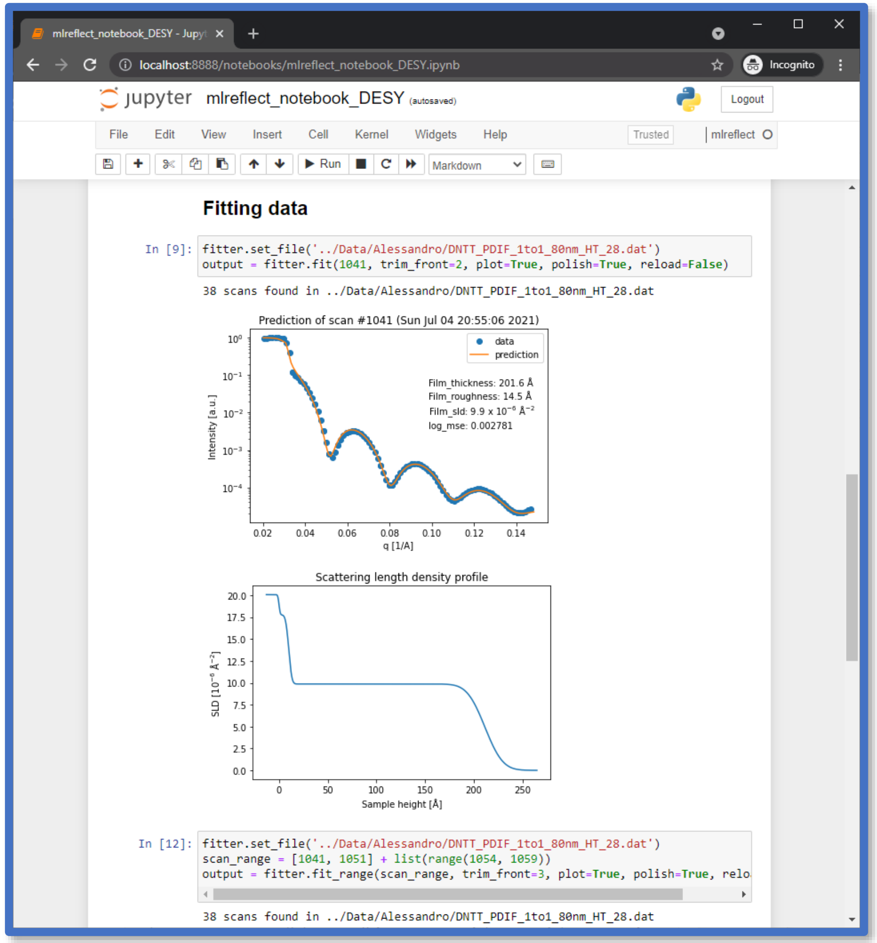
Generation 1 (2019-2022): MLReflect

Python package *MLReflect*
BMBF project in collaboration with DESY

- Available on GitHub / PyPI
- Online documentation available on Read the Docs
- Can be used with Jupyter notebooks as GUI
- Installed at DESY, PETRA III, P08

Remaining challenges

- Parameter ranges are defined during training stage.
- Discretization to be defined during training stage.
- Does not work reliable for complex multilayer structures
- No estimation of error bars



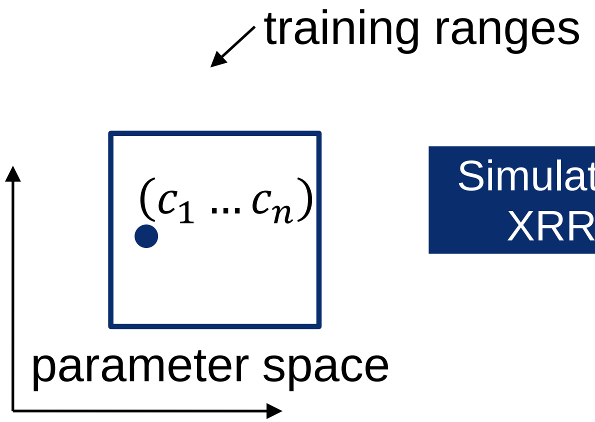
github.com/schreiber-lab/mlreflect
Greco et al., *J. Appl. Cryst.* **55**, 362 (2022)

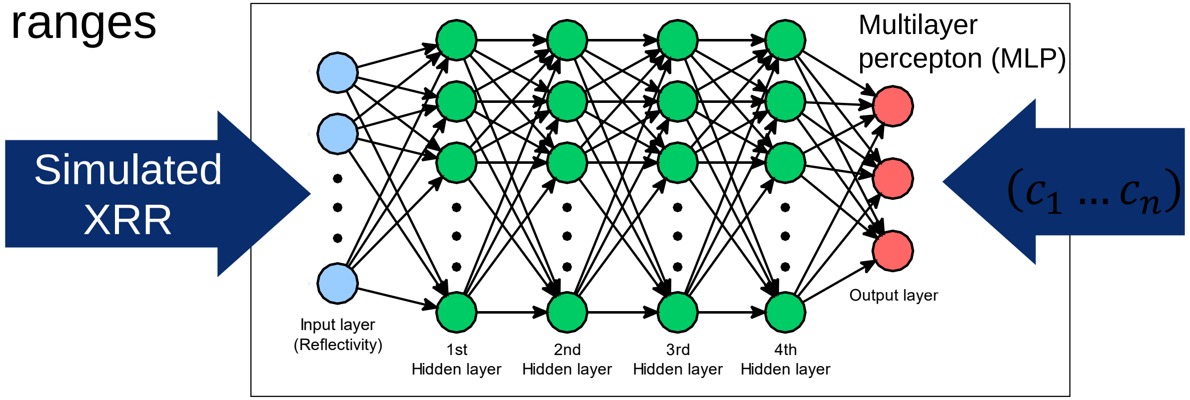
Bundesministerium für Bildung und Forschung

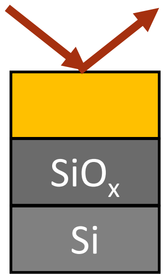
Generation 2: Including prior knowledge

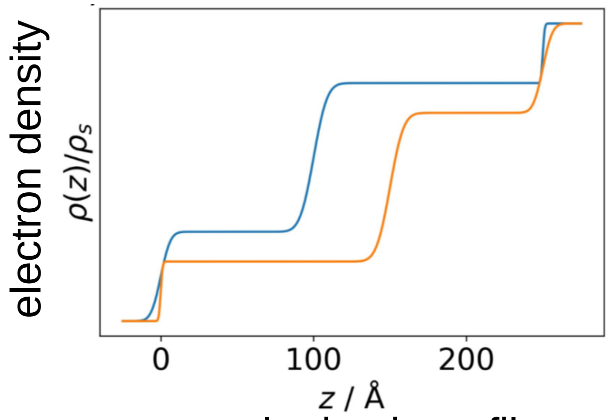
- How to tackle ill-posed inverse problems by taking advantage of the *a priori* knowledge of experimentalists?

“Standard” / classical approach to train the ML model:









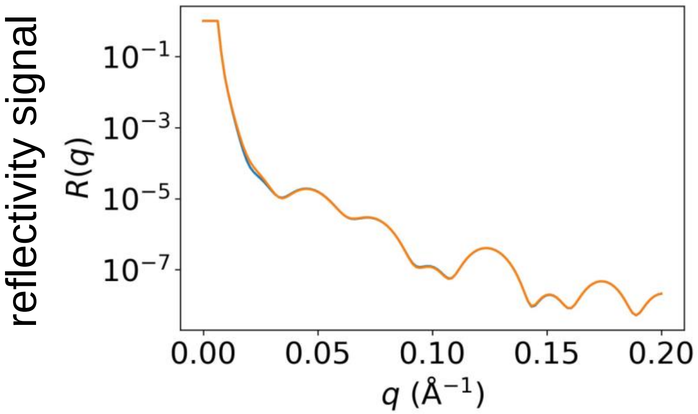
sample depth profile

Classical approach for model training:

- sample from param. space
- simulate XRR + noise
- gradient descent in MLP

Limitations:

- Model can only be trained for “small” parameter space, where a one-to-one mapping can be assumed!
- Consequence: make sure that e.g. only the orange curve falls in training ranges
- Training of “general” model not possible
- No option for the experimentalist to “guide” the model

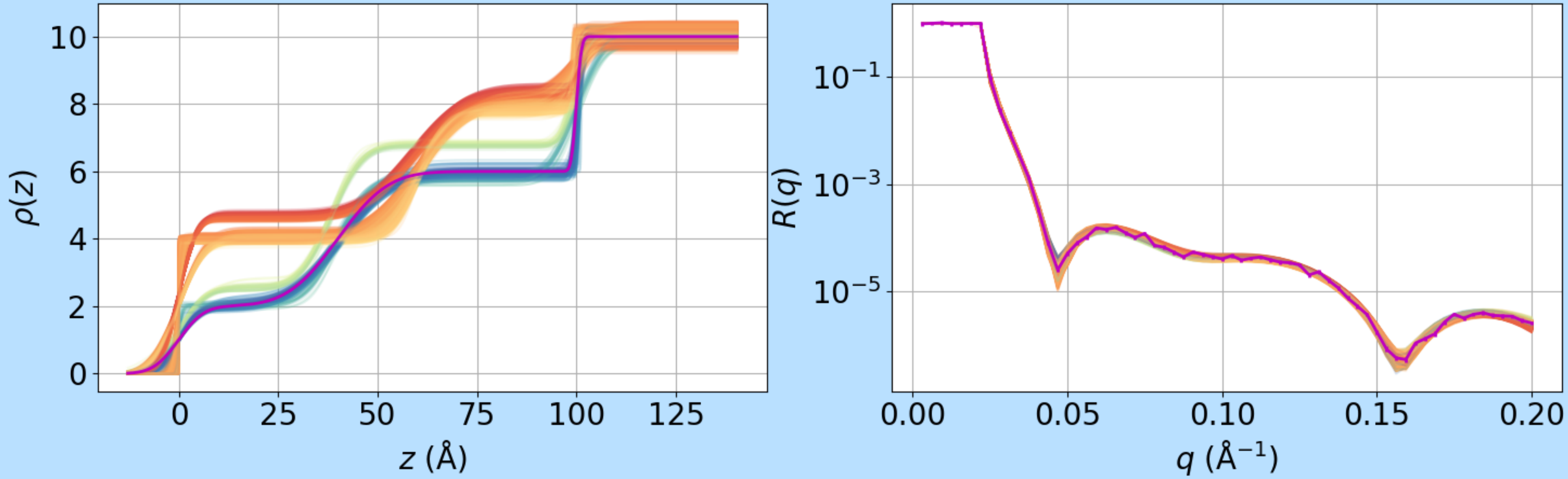


scattering vector

V. Munteanu et al., J. Appl. Cryst. (2024, accepted), 57,
DOI: 10.1107/S1600576724002115

Underdetermined inverse problems

- forward process maps parameters of physical system to an observable signal
- inverse problem describes the retrieval of physical parameters from
  a (possibly noise-corrupted) measurement of the signal
    - **one-to-one mapping**: training of neural network straightforward to approximate inverse function
    - **many-to-one** mapping (forward process): leads to an underdetermined, ill-posed inverse problem with corresponding **one-to-many** inverse mapping that cannot be approximated via regression.

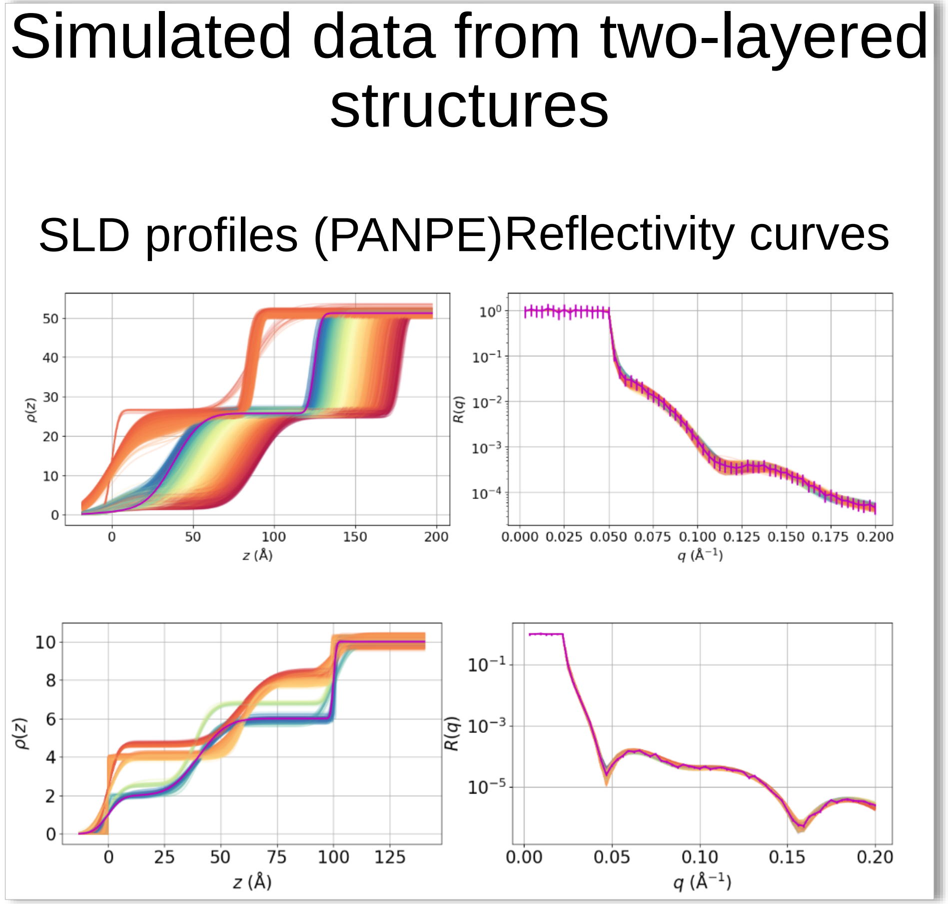


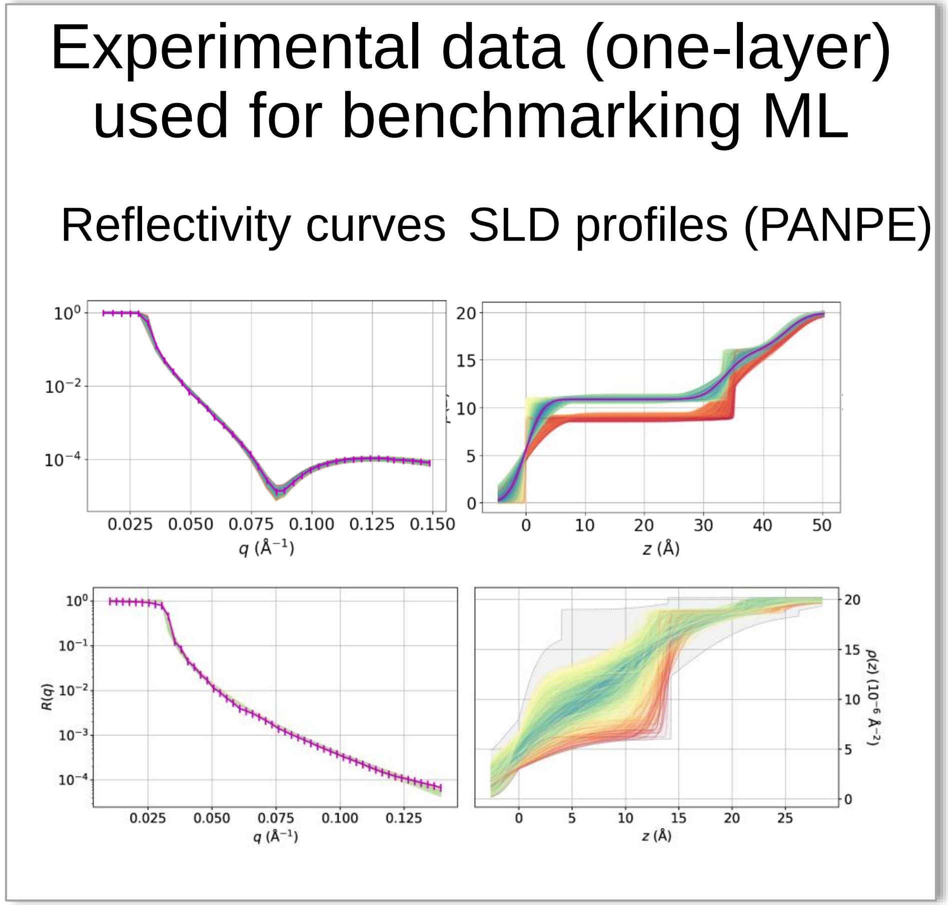
depth in sample

V. Munteanu et al, J. Appl. Cryst. **accepted, (2024)**
V. Starostin et. al **, in prep.**

scattering vector

Confirmed hypothesis about high levels of ambiguity



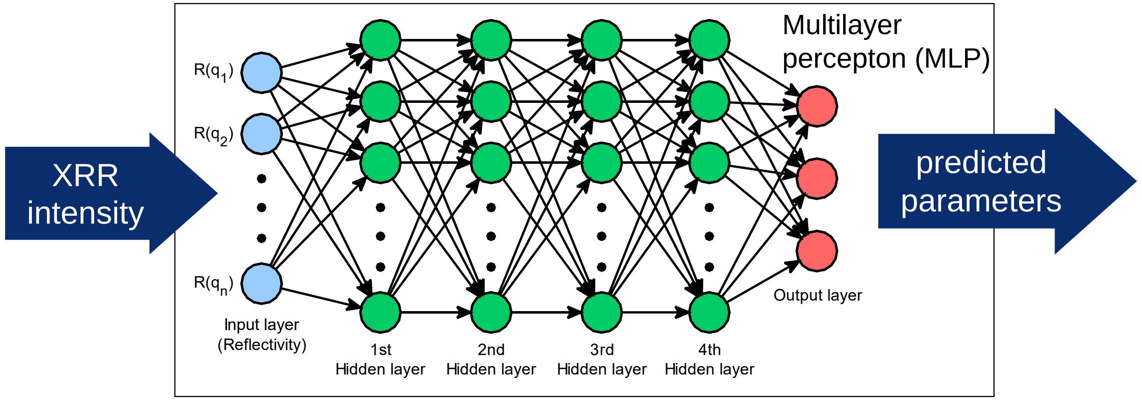


V. Starostin et al., 2024

Gen 2.2: Discretization-invariant learning

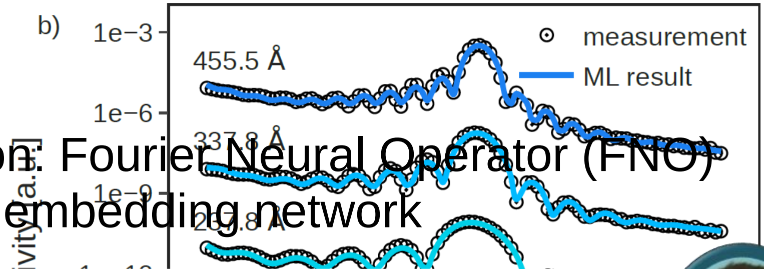
EBERHARD KARLS UNIVERSITÄT TÜBINGEN

- in traditional approach: Only XRR curves as input, q-base is fixed

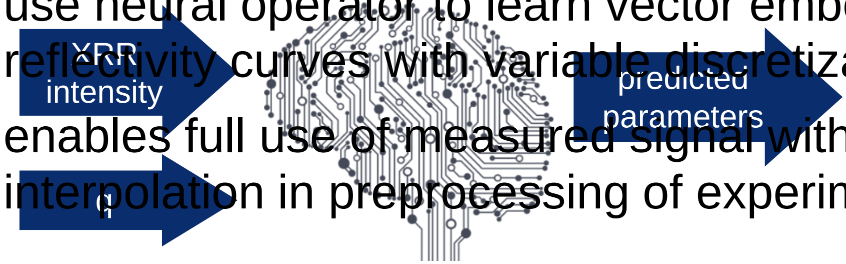


Problem:

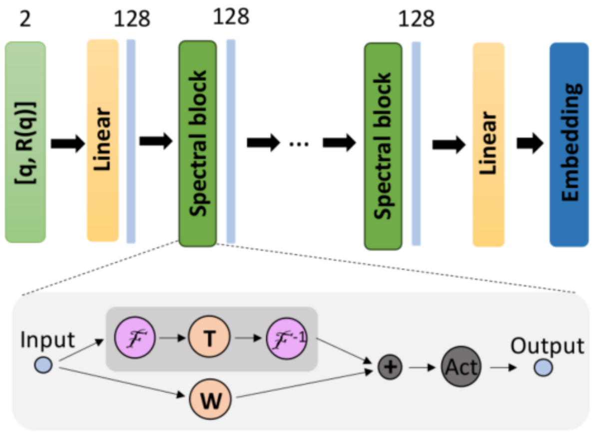
- Neural networks can only learn mappings between finite-dimensional vector spaces.
- Multilayer percepton requires a fixed discretization (range and resolution) of the input



- How to become more flexible in q-discretization?
- use neural operator to learn vector embedding for reflectivity curves with variable discretizations
- enables full use of measured signal without relying on interpolation in preprocessing of experimental data



- Solution: Fourier Neural Operator (FNO) as the embedding network

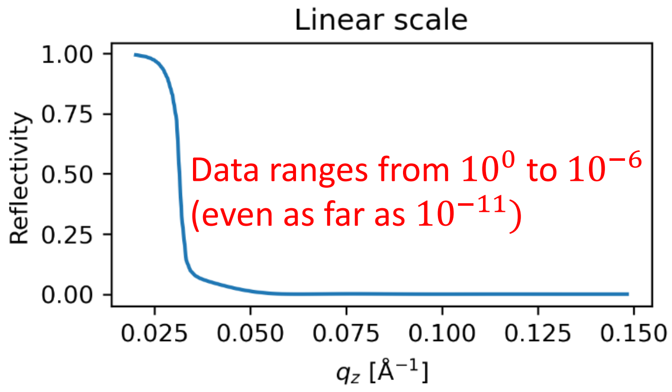


Concept developed by V. Munteanu, PhD student in Frank Schreiber's lab

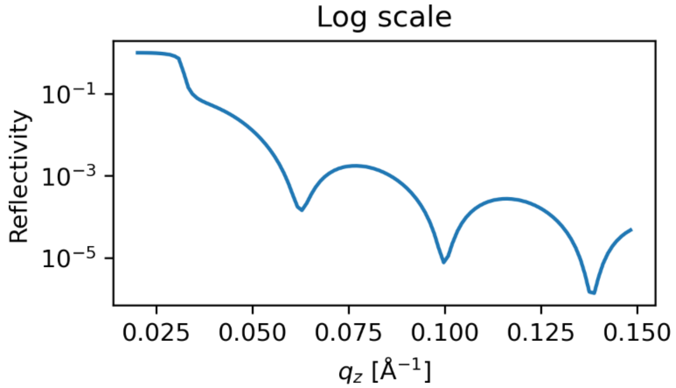
V. Munteanu et al., J. Appl. Cryst. (2024, accepted), 57, DOI: 10.1107/S1600576724002115

Challenges for ML specific to reflectometry

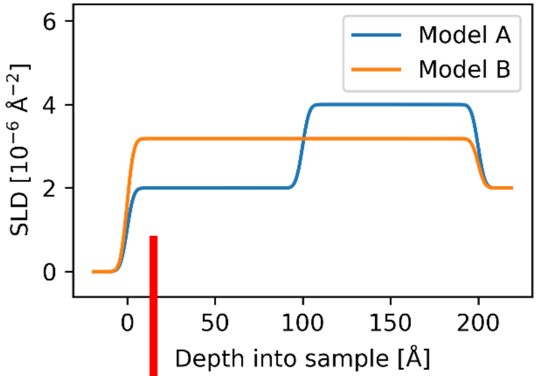
1. High dynamic range



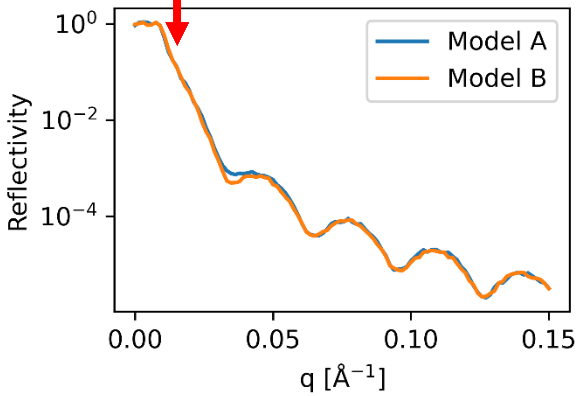
Log scale can help, but inputs are still not equally distributed



2. Phase problem/ambiguity

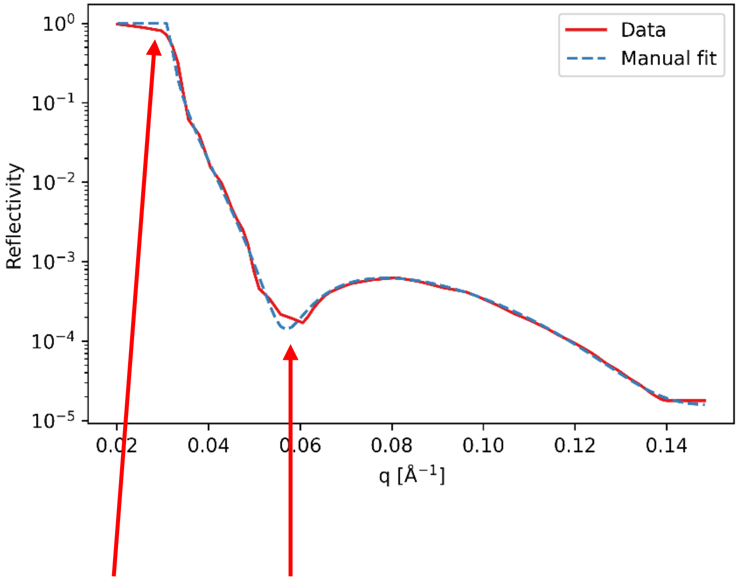


Different samples create the same reflectivity signal!



3. Experimental artifacts

The neural network is trained with simulated data, but meant to be used with experimental data!



Experiment and theory do not follow the same distribution!

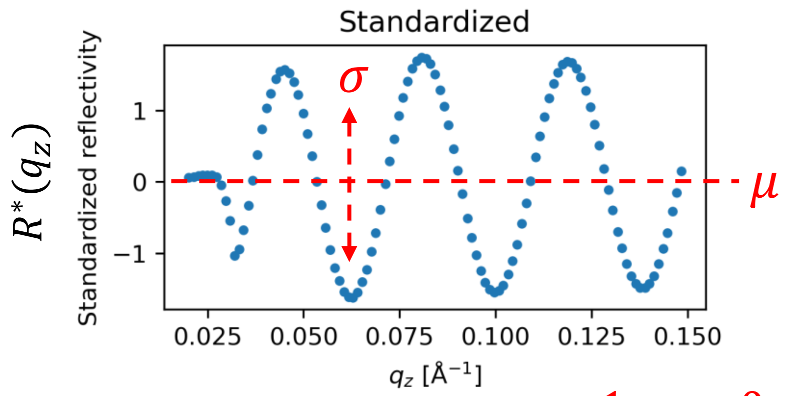
Solutions for reflectometry-related challenges

1. High dynamic range

Standardize input

$$R^*(q_z) = \frac{R(q_z) - \bar{R}(q_z)}{\hat{R}(q_z)}$$

- $\bar{R}(q_z)$: mean
- $\hat{R}(q_z)$: standard deviation
- derived from training set with artificial noise



$\sigma = 1, \mu = 0$

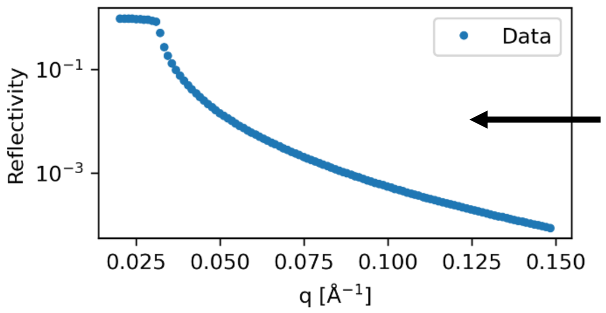
2. Phase problem/ambiguity

Reduce solution space (add "priors")

E.g., 3 thin film parameters with a certain range

Thin film	varied	Thickness 20 – 1000 Å
Oxide	fixed	Roughness 0 – 40 Å
Silicon	fixed	SLD 1 – 14 10^{-6}Å^{-2}

Remove "featureless" curves



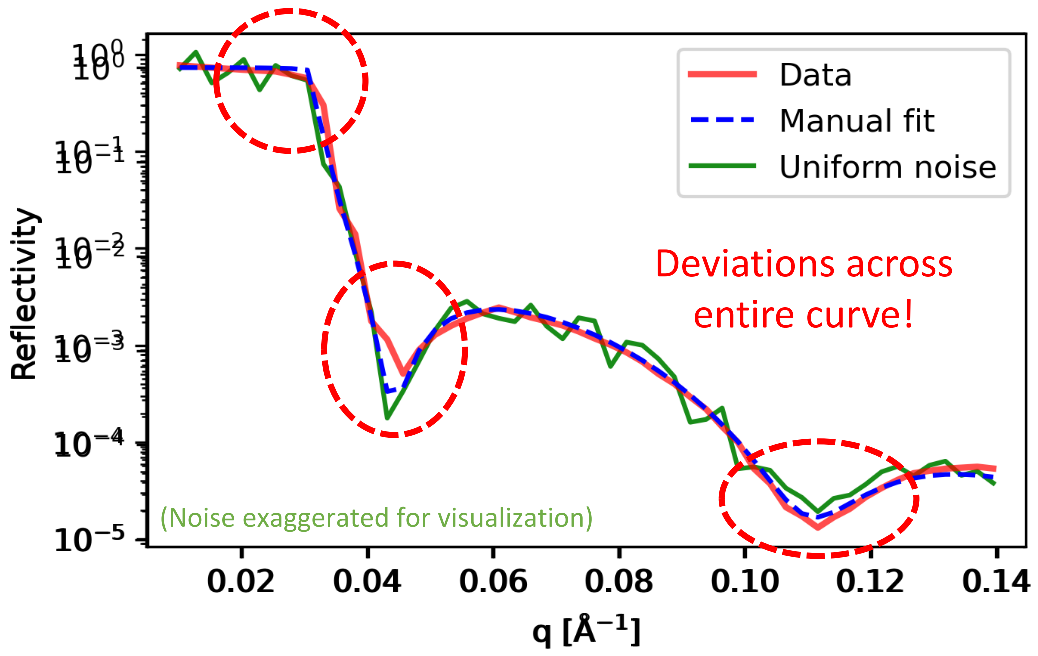
Exclude from training:

- Low thickness: < 20 Å
- Low contrast: < 10^{-6} Å^{-2}
- High roughness: > 40 Å

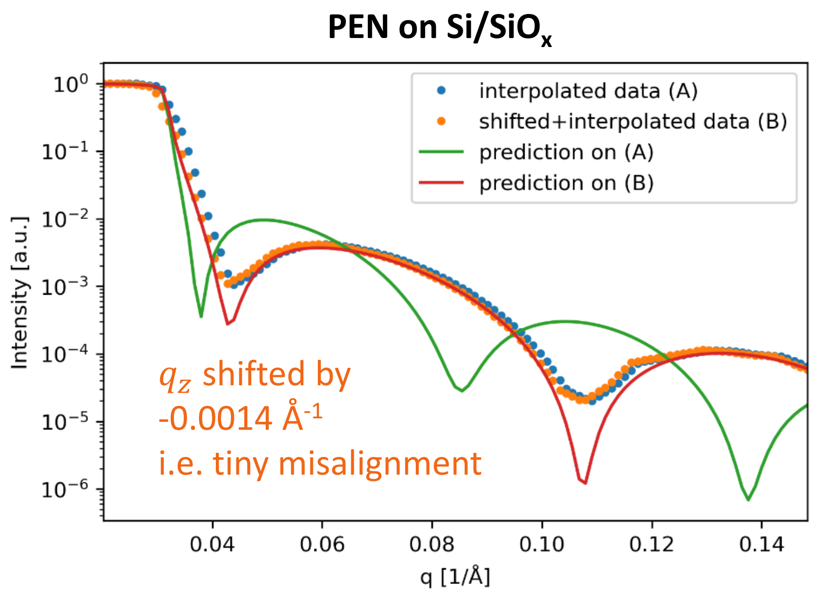
Greco *et al. Mach. Learn.: Sci. Technol.*, 2021, **2**, 045003

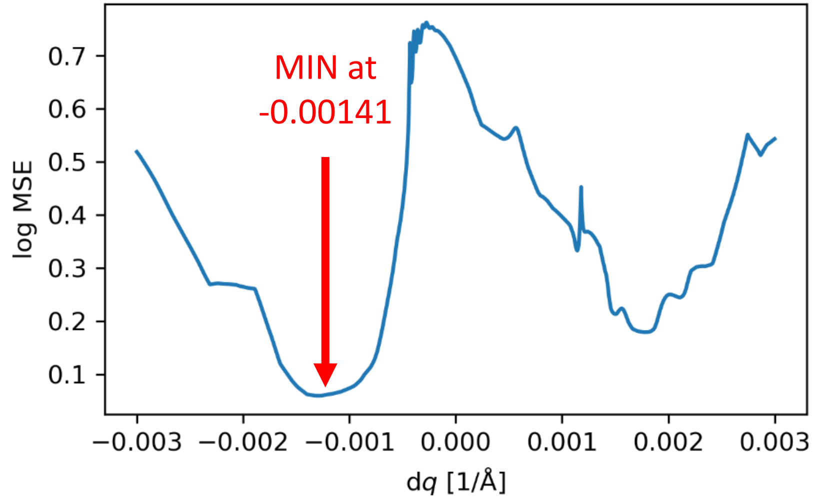
Solutions for reflectometry-related challenges

3. Experimental artifacts (noise, misalignement, …)

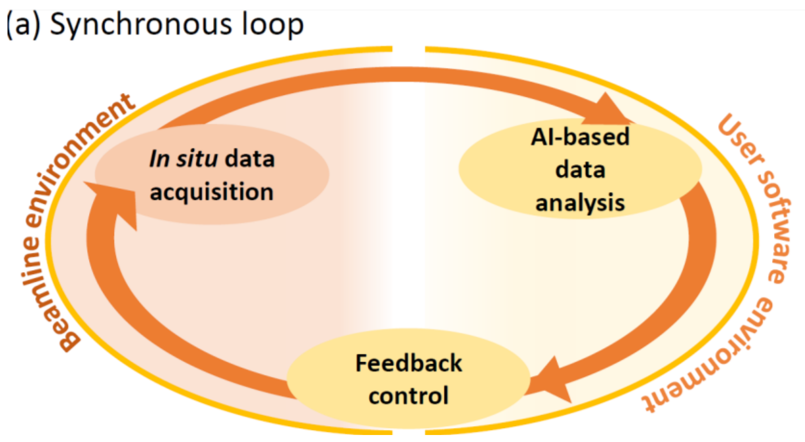


E.g. uniform noise:
each input multiplied with a value between $1 - n$ and $1 + n$





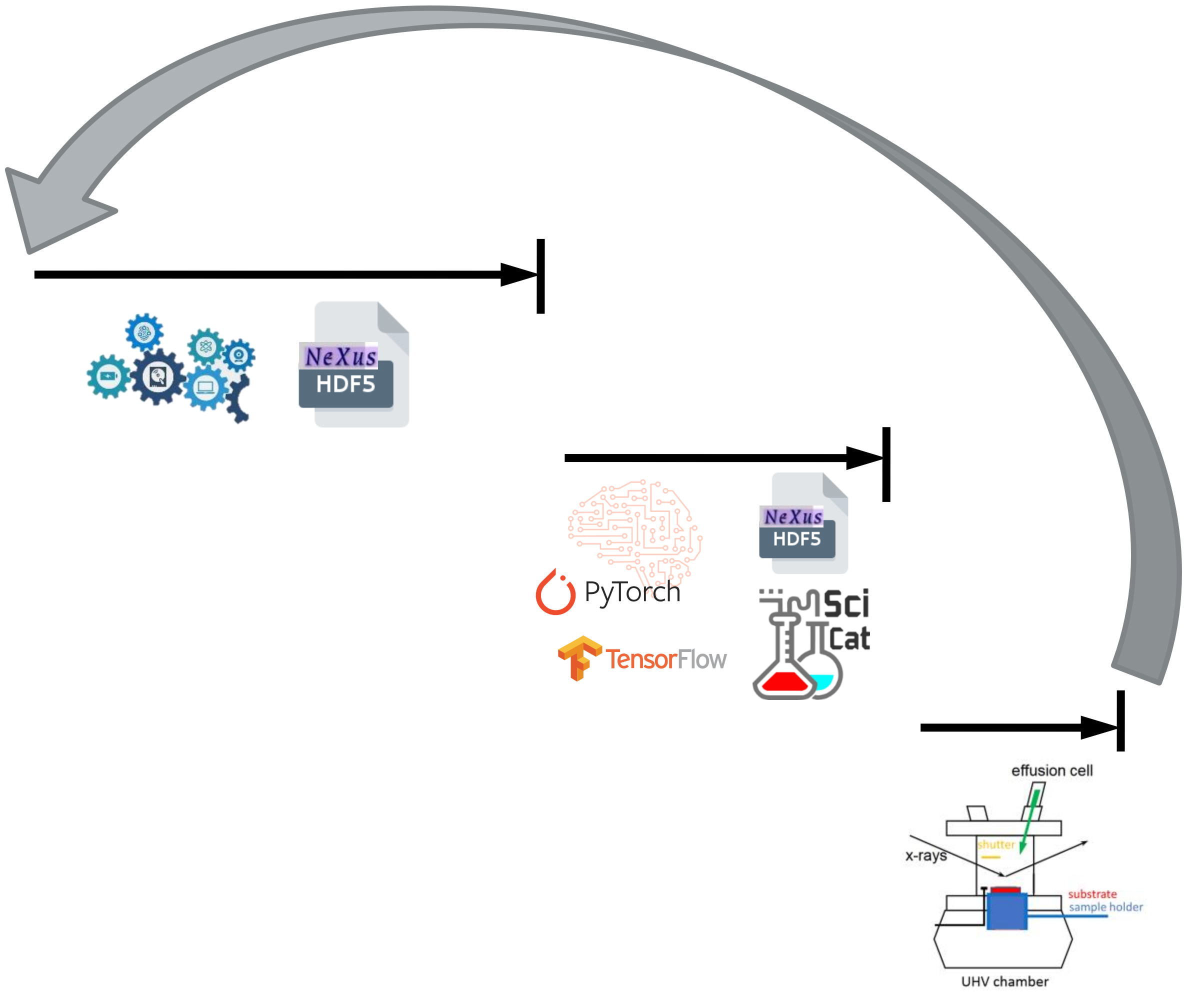
Feedback scheme




```
> while(True):
    s = a2scan(…)

    d = predict(s)

    if d > 50:
     stop_growth()
     break
```



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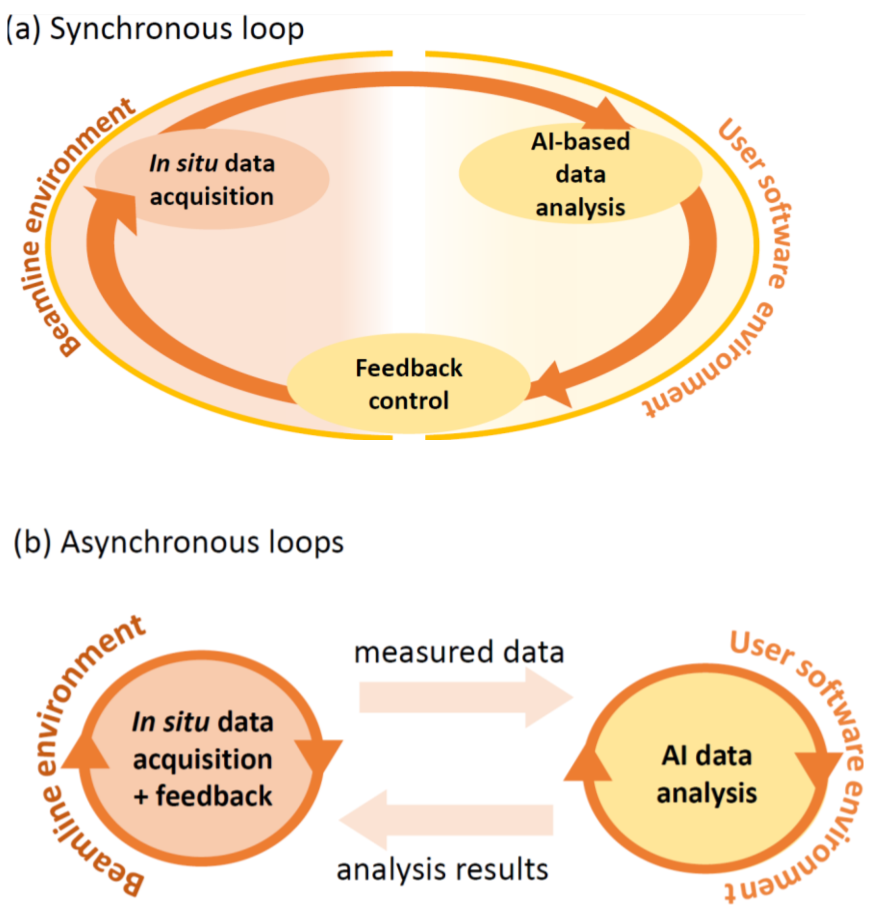
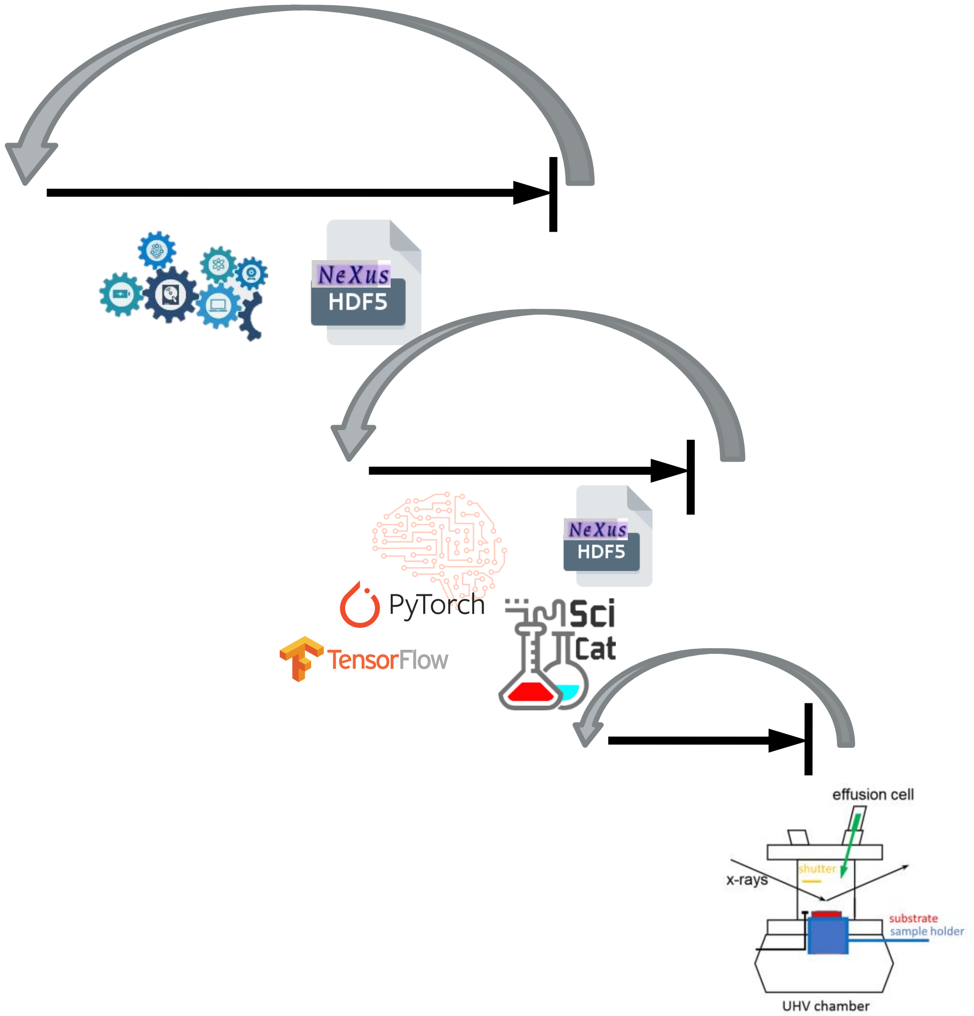
Feedback scheme





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Public XRR dataset

zenodo

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May 27, 2022 | Dataset | Open Access

Reflectometry curves (XRR and NR) and corresponding fits for machine learning

Pithan, Linus; Greco, Alessandro; Hinderhofer, Alexander; Gerlach, Alexander; Kowarik, Stefan; Rußegger, Nadine; Dax, Ingrid; Schreiber, Frank

Public reflectometry data collection

- about 250 experimental XRR profiles
- labelled data:
  sample parameters & classical fits are published together with measurements
- indented to grow
  - call for contributing datasets
  - plans for dedicated infrastructure

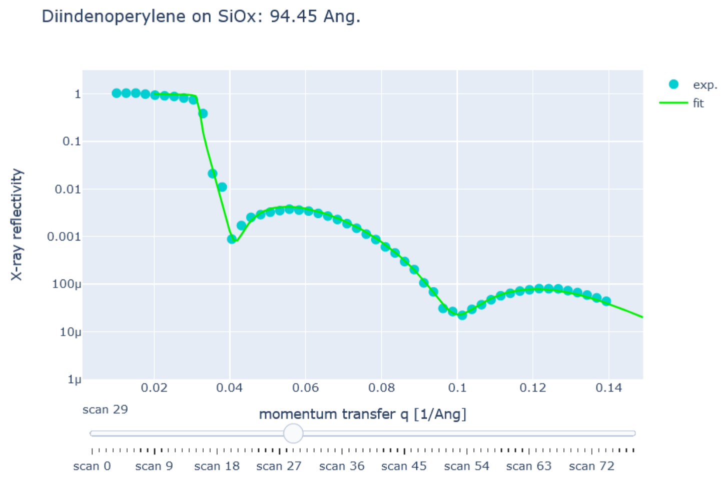
In [1]:

```
#prepare jupyter notebook for plots shown below
%run prepare_plot.py

#imports
from silx.io.dictdump import nxtodict     #read NeXus hdf5 to python dict
import numpy as np
import pandas as pd
from IPython.display import display

#use same q_range as during fitting
q_fit = np.linspace(0.02,0.15,130,endpoint=False)

#produce plots
for key, ds in nxtodict("xrr_dataset.h5").items():
    if "@" not in key:   #skip nexus attributes

        print("Dataset: ",key)
        print("Experimentalists: ",ds["metadata"].pop("Experimentalists","?"))
        ds["metadata"].pop("@NX_class", "")
        display(pd.DataFrame.from_dict({"Dataset":[key],**ds["metadata"]}))

        fig = prepare_figure(ds,q_fit,str(ds["metadata"]["Layer_material"])+" on SiOx")
        fig.show()
```

NeXus

```
Dataset:  DIP_1
Experimentalists:  ['Kowark, Stefan']
```

	Dataset	Layer_CAS	Layer_formula	Layer_material	Substrate_temperature	Substrate_temperature@unit	instrument	q_max_
0	DIP_1	188-94-3	C32H16	Diindenoperylene	303	K	ESRF, ID10b	0



Foreseen collaboration:

DAPHNE 4NFDI

SciCat

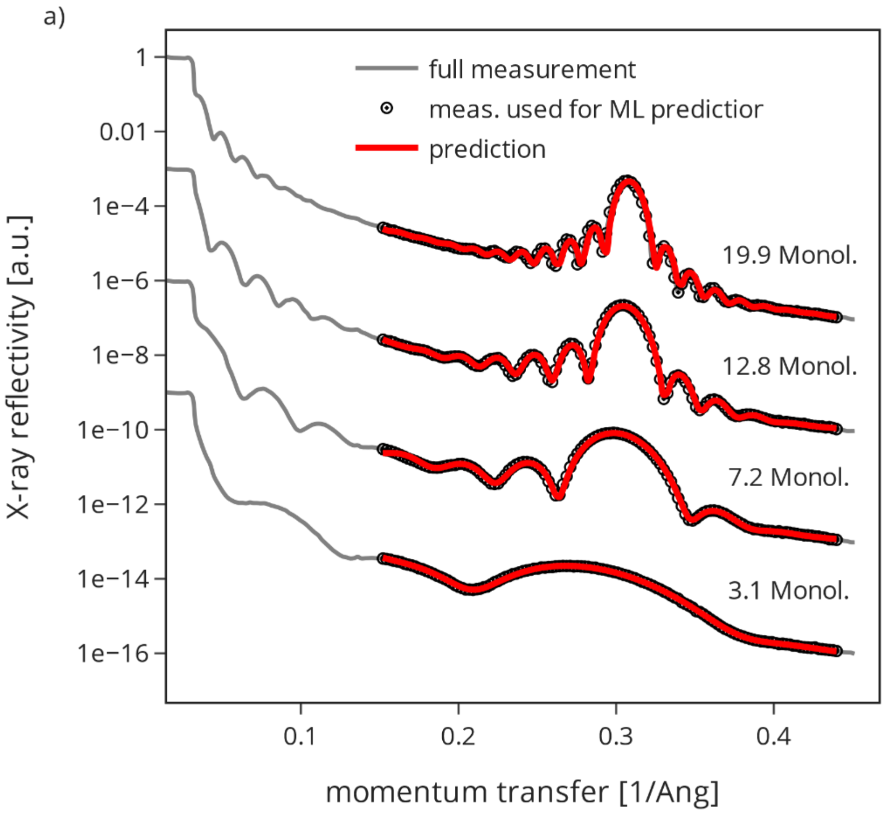
ORSO

CAU Kiel University Christian-Albrechts-Universität zu Kiel

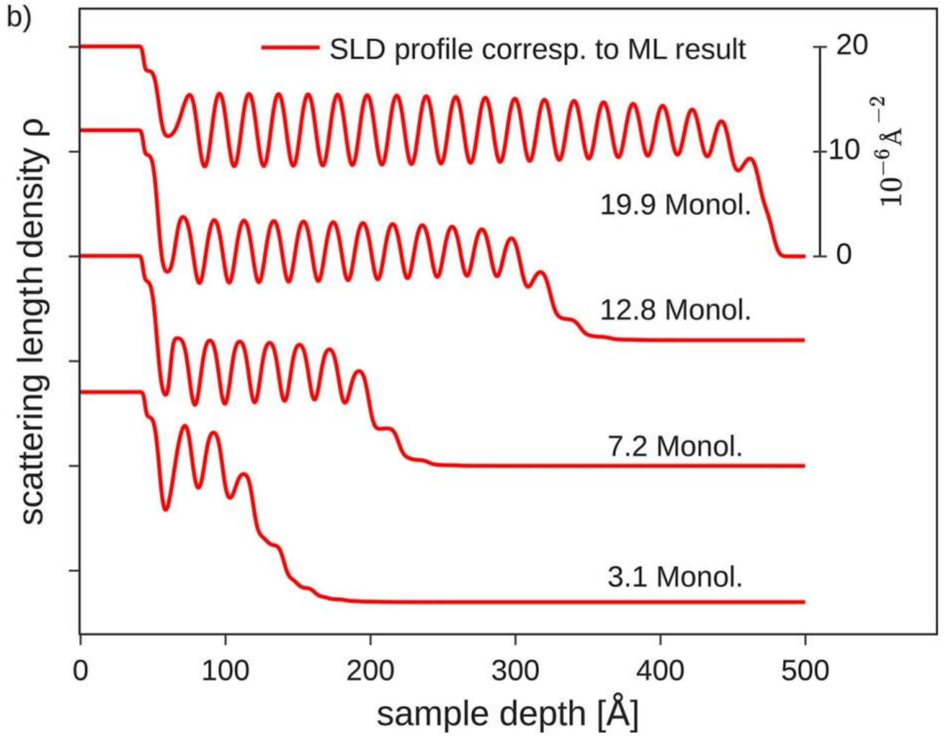
L.Pithan et al. 2022 https://doi.org/10.5281/zenodo.6497438

Multilayers and 15+ parameters

XRR data during growth (PTCDI-C8)



SLD profiles for (PTCDI-C8)



L. Pithan et al, *Closing the loop: Autonomous experiments enabled by machine-learning-based online data analysis in synchrotron beamline environments*, J. Synchrotron Rad. **30** (2023) 1064