ESRF	**Experiment title:** Controlled thin film growth through machine-learning based closed-loop feedback with online X-ray scattering analysis.	**Experiment number:** MI-1462
Beamline: ID10	**Date of experiment:** from: 01 March 2023 to: 06 March 2023	**Date of report:** 28/06/2023
Shifts: 15	**Local contact(s):** Maciej Jankowski, Oleg Konovalov	*Received at ESRF:*

Names and affiliations(* indicates experimentalists, † indicates proposers):

Linus Pithan *†(1), Vladimir Starostin *†(1), David Mareček *(2), Lukas Petersdorf *(3), Constantin Völter *(1), Alexander Gerlach †(1), Alexander Hinderhofer (1), Bridget Murphy (3), Stefan Kowarik* (2), Frank Schreiber †(1)

(1) Universität Tübingen Germany, (2) Universität Graz Austria, (3) Universität Kiel Germany

Report:

In recent years, X-ray scattering has received a significant boost due to the increased use of machine learning strategies in the analysis of data acquired at synchrotron sources. This experiment contributed to the use of online data analysis in XRR at the ESRF. However, the tested approach is well suited for a broader range *of in situ* and *in operando* experiments for processes in dynamic equilibrium where the information extracted from the scattering data itself can be used to stabilize the equilibrium.

The full analysis of the results produced in this experiment is available in [1-3], including a public dataset released in the ESRF data catalog before the end of the embargo period. The project was realized in close collaboration with DAPHNE4NFDI [4].

Description of the experiment:

We leveraged ML based real-time data analysis for XRR integrated into the ID10 beamline environment. As a proof-of-concept, we grew organic molecular thin films of Alq3 (C27H18AlN3O3) and PTCDI-C8 (N,N′-dioctyl-3,4,9,10-perylene tetracarboxylic diimide). The growth was monitored with X-ray reflectivity and we established a closed loop between real-time data acquisition, ML-based online data analysis and the sample environment to tailor the deposition process of organic thin films on the molecular monolayer level. Through a strong integration via TANGO and BLISS all ML-based fit results are saved together with raw data in the dataset published in the ESRF data catalogue.

Results:

There are main results from this experiment. First there are technical (software) designs how user developed mashine learning (ML) code [1,3,5] can be integrated into existing beamline infrastructure (Fig. 1). In this context we focused on the perspective of visiting facility users and strategies to provide an elementary data analysis in real time during the experiment without introducing the additional software dependencies in the beamline control software environment. A closed loop feedback system based on real time data analysis was put in place.

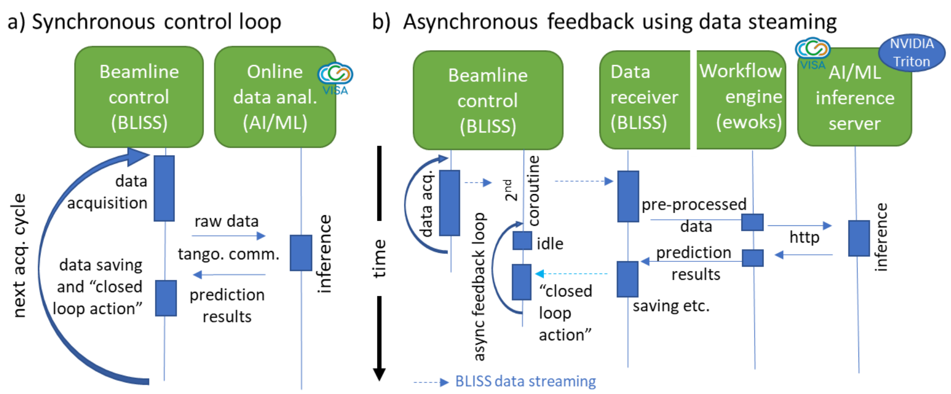

Fig 1: Architecture illustration of the implemented synchronous and asynchronous feedback loop based on BLISS. In a) data acquisition, ML inference and feedback action follow each other strictly in time (synchronous over distributed system) while in b) acquisition and data analysis + feedback are separated in independent coroutines to decouple independent processes. Figure taken from [1]

Second, beyond the engeneering level, there are the actual results produced using our ML code and the corresponding integration into a closed loop feedback configuration. In Fig. 2 expemplary fits using the real time (online) data anaysis are shown. Here we successfully grew molecular thin films of predefined thickness, where the ML-based closed loop took control over the termination of the growth process by closing the relevant deposition shutter of the growh chamber.

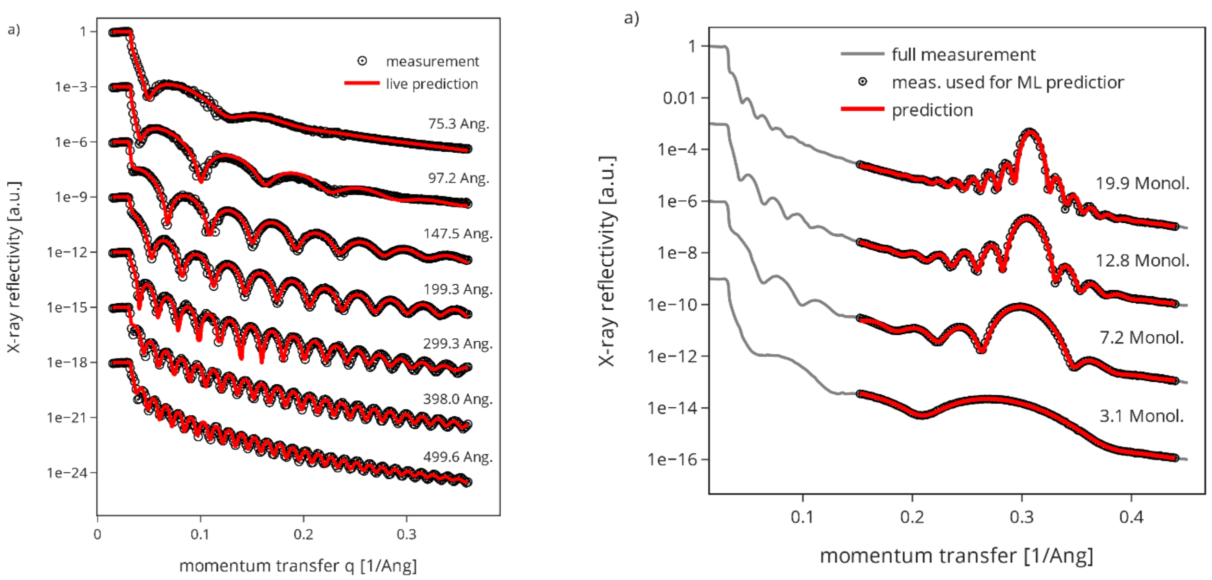

Fig2: ML based online data analysis of XRR measurements for a single molecular layer on top of Si (left) and crystalline multilayer structure with Bragg peak (right). Figures taken from [1]

References:

[1] **Pithan et al. 2023**, https://doi.org/10.48550/arXiv.2306.11899 preprint based on the results of this experiment, submitted to J. Sync Rad., Closing the loop: Autonomous experiments enabled by machine-learning-based online data analysis in synchrotron beamline environments

[2] **Pithan et al. 2023**, https://doi.esrf.fr/10.15151/ESRF-DC-1249105707, Public dataset based on the results of this experiment, (released before the end of embargo period)

[3] **Munteanu et al. 2023,** (in preparation), Neural network analysis of neutron and X-ray reflectivity data: incorporating prior knowledge for tackling the phase problem

[4] **Barty et al. 2023**, https://zenodo.org/record/8040606, DAPHNE4NFDI consortium: DAta from PHoton and Neutron Experiments for NFDI, https://www.daphne4nfdi.de

[5] **Greco et al. 2022**, J. Appl. Cryst. 55, 362, mlreflect