Grazing Incidence Total Scattering at PETRA III: Extending Local Structure Analysis to Ultrathin Films on Single-Crystal Substrates

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HELMHOLTZ

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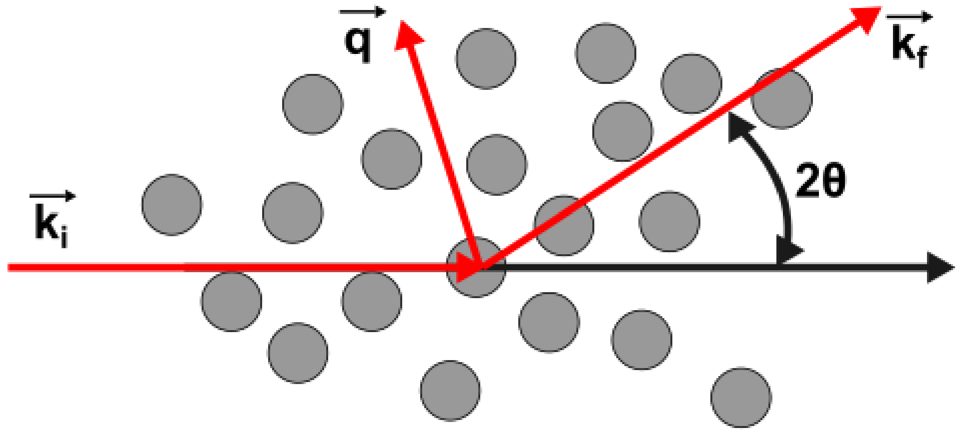
High energy scattering

High energy X-ray scattering

X-ray interaction with matter

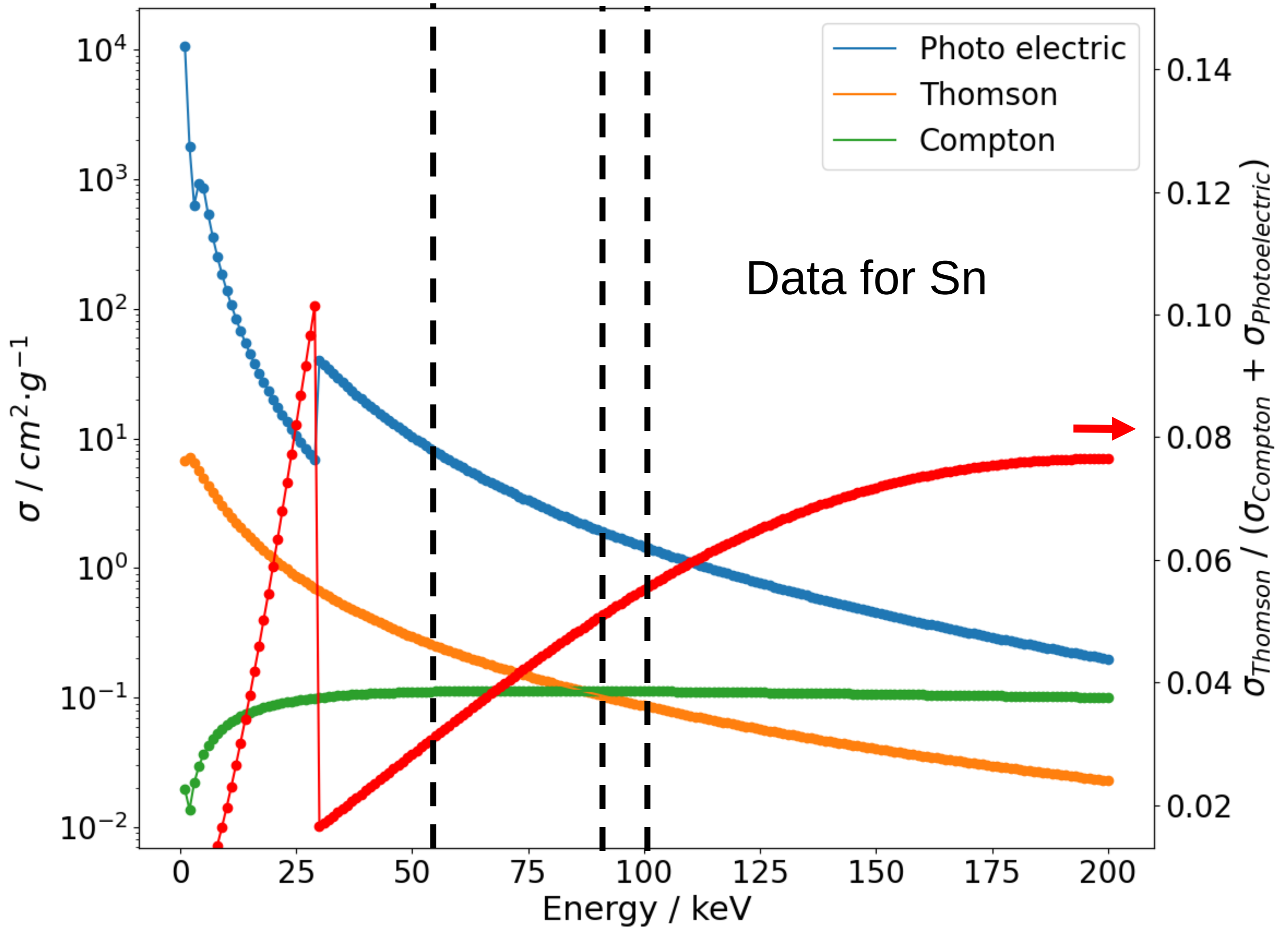
- X-ray absorption (photo electric effect)
- Compton (inelastic) scattering
- **Thomson (elastic) scattering**



$$\lambda = 2d_{hkl}\sin(\theta)$$

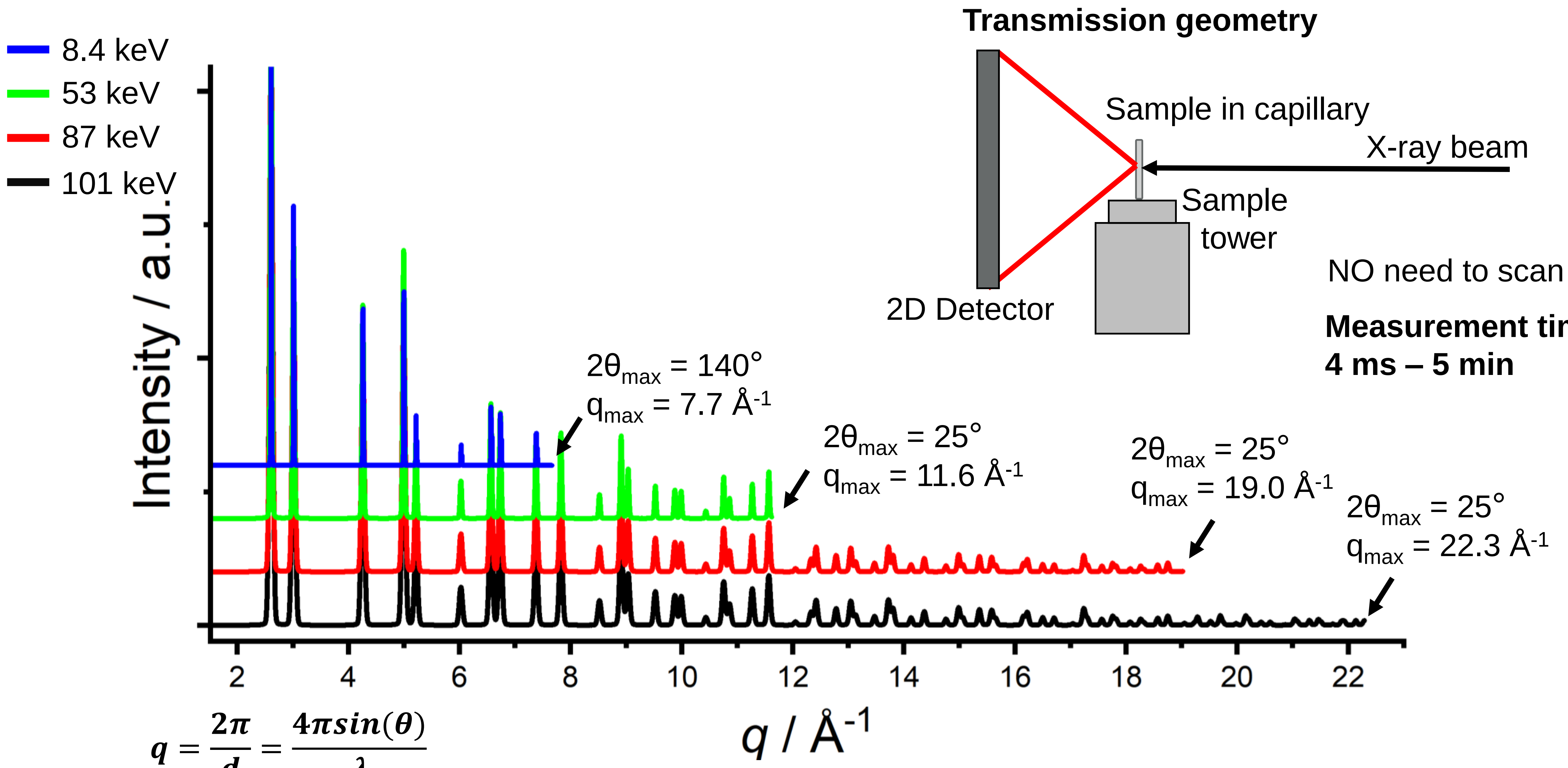
$$q = \frac{2\pi}{d} = \frac{4\pi\sin(\theta)}{\lambda}$$

$$E = \frac{hc}{\lambda}$$



- Low absorption
  - Probing bulk materials
  - Low beam damage

High energy X-ray scattering



$$q = \frac{2\pi}{d} = \frac{4\pi sin(\theta)}{\lambda}$$

NO need to scan

Measurement time
4 ms – 5 min

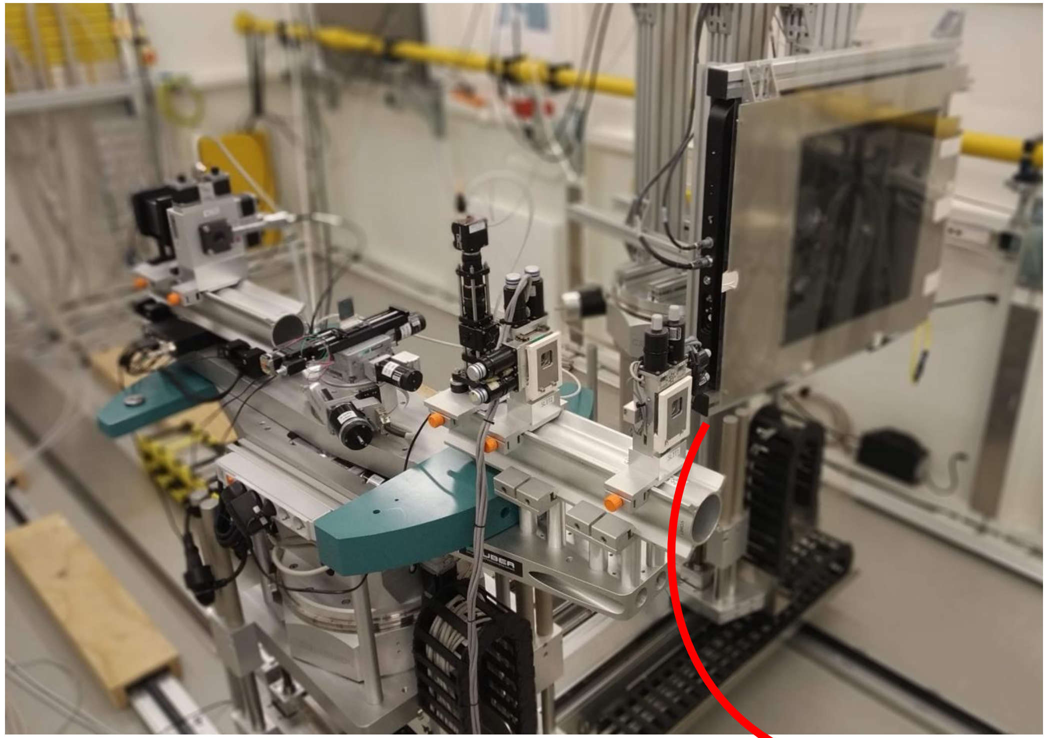
Detector stage

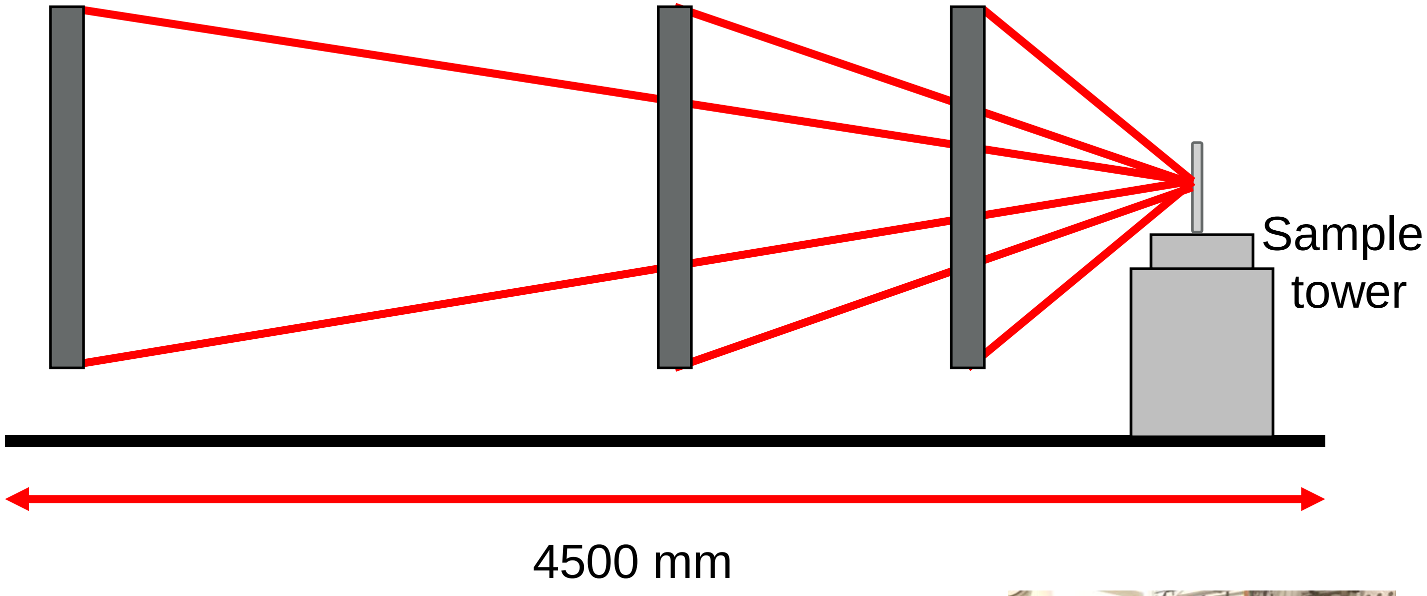
*values @ 101 keV

SAXS
sdd = 4000 mm
q_{max} = 2.3 Å^{-1}

XRD
sdd = 1000 mm
$\Delta q/q = (2.5 – 0.38)\times10^{-2}$
q_{max} = 10.4 Å^{-1}

TS
sdd = 400 mm
$\Delta q/q = (3.6 – 0.86)\times10^{-2}$
q_{max} = 24 Å^{-1}





Perkin Elmer XRD1621

area: 410 x 410 mm^2, speed: 15 Hz
pixel size: 200 x 200 µm^2
no gaps

Pilatus3 X 2M CdTe

(PETRA III shared device)
area: 253.7 x 288.8 mm^2, speed: 250 Hz
pixel size: 172 x 172 µm^2
Single-photon counting
Threshold energy

Grazing incidence total scattering

Grazing incidence scattering

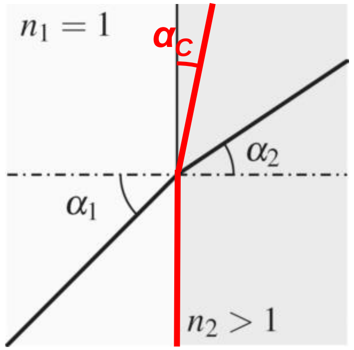
$$\eta = 1 - \delta + i\beta$$

Absorptive term ($i\beta$)

Dispersive term (δ)

Complex refraction index (η)

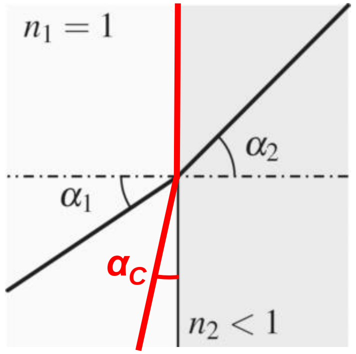
Visible light: $\mathrm{Re}(\eta) > 1$



Total internal reflection

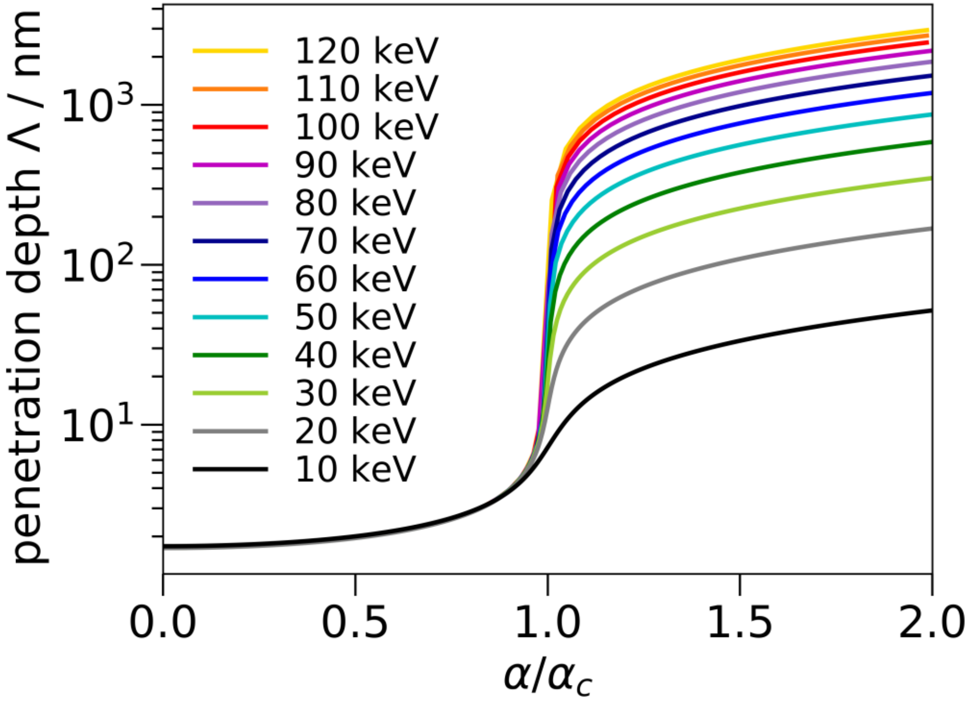
$$\alpha_C = 90 - asin\left(\frac{n_1}{n_2}\right)$$

X-rays: $\mathrm{Re}(\eta) < 1$

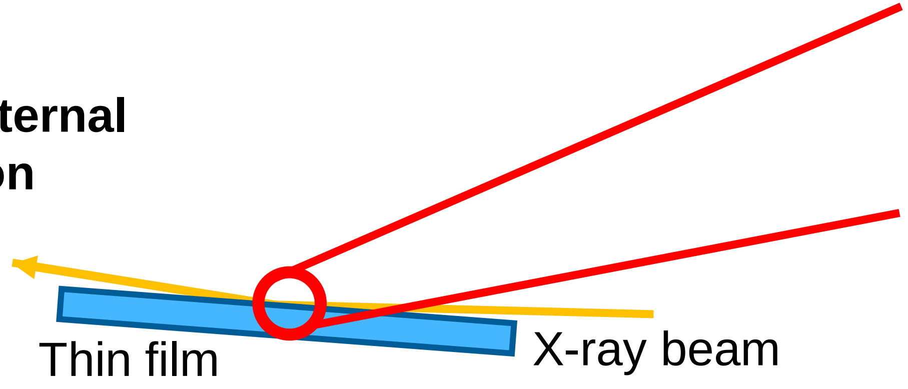


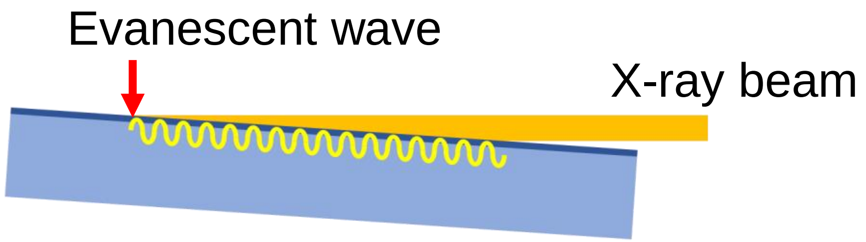
Total external reflection

α_C(@ 101keV) ≈ (10 – 50)×10^{-3} °



R. Feidenhans'l. Surf. Sci. Rep. 10 (1989), 105.



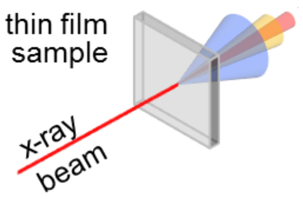


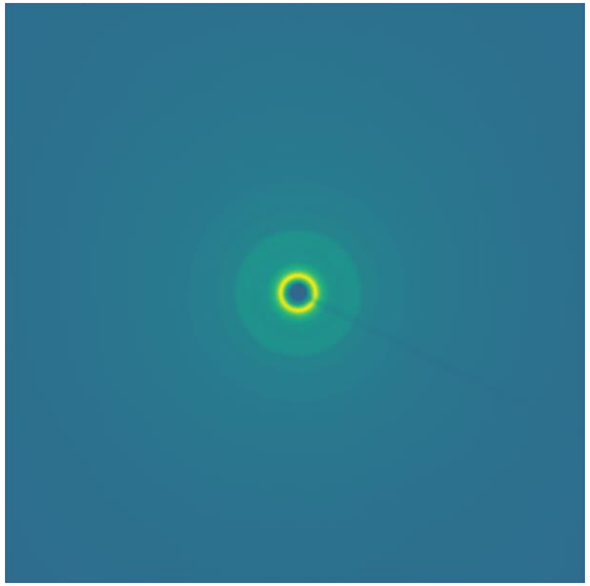
- The X-ray beam is constrained to propagate through the surface of the film
- Surface sensitive amplified scattering

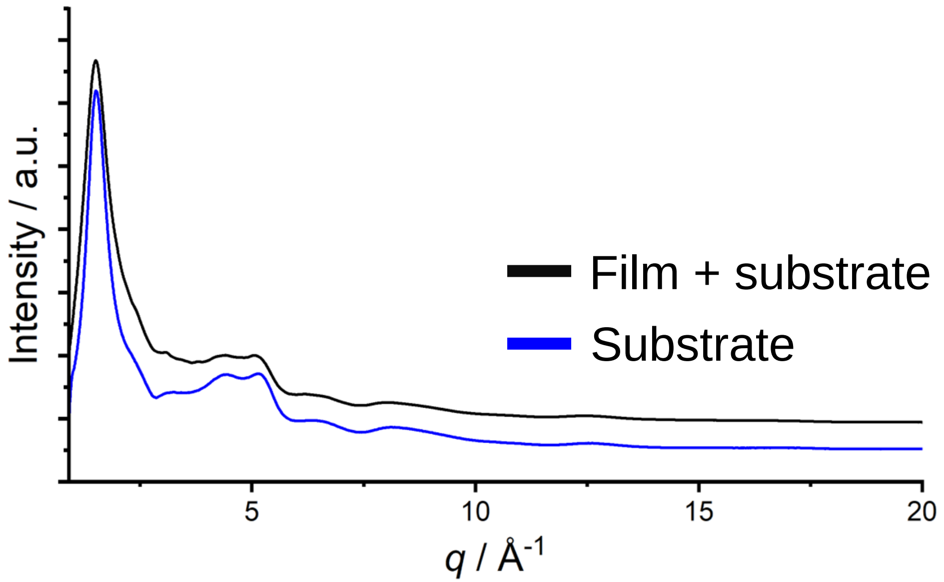
Grazing incidence scattering

Transmission

50 nm HfO_2 film on 100 µm fused silica substrate

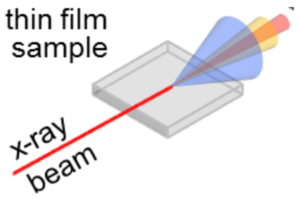


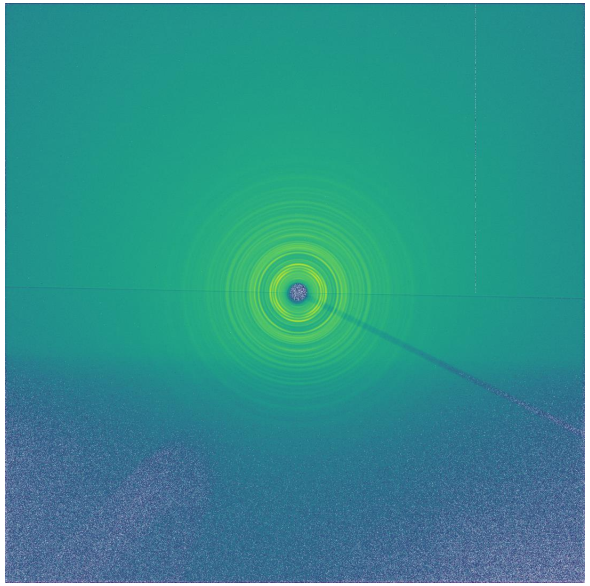


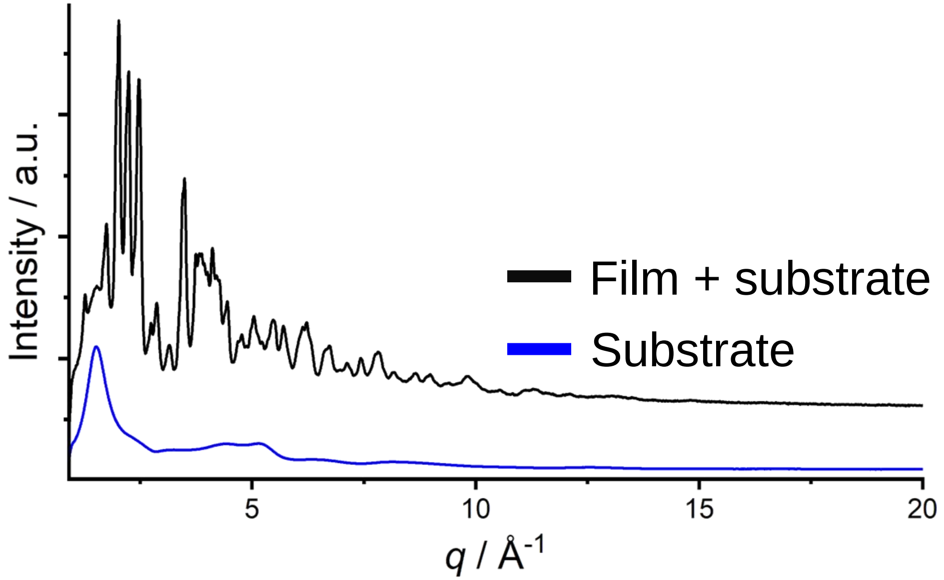


Grazing incidence

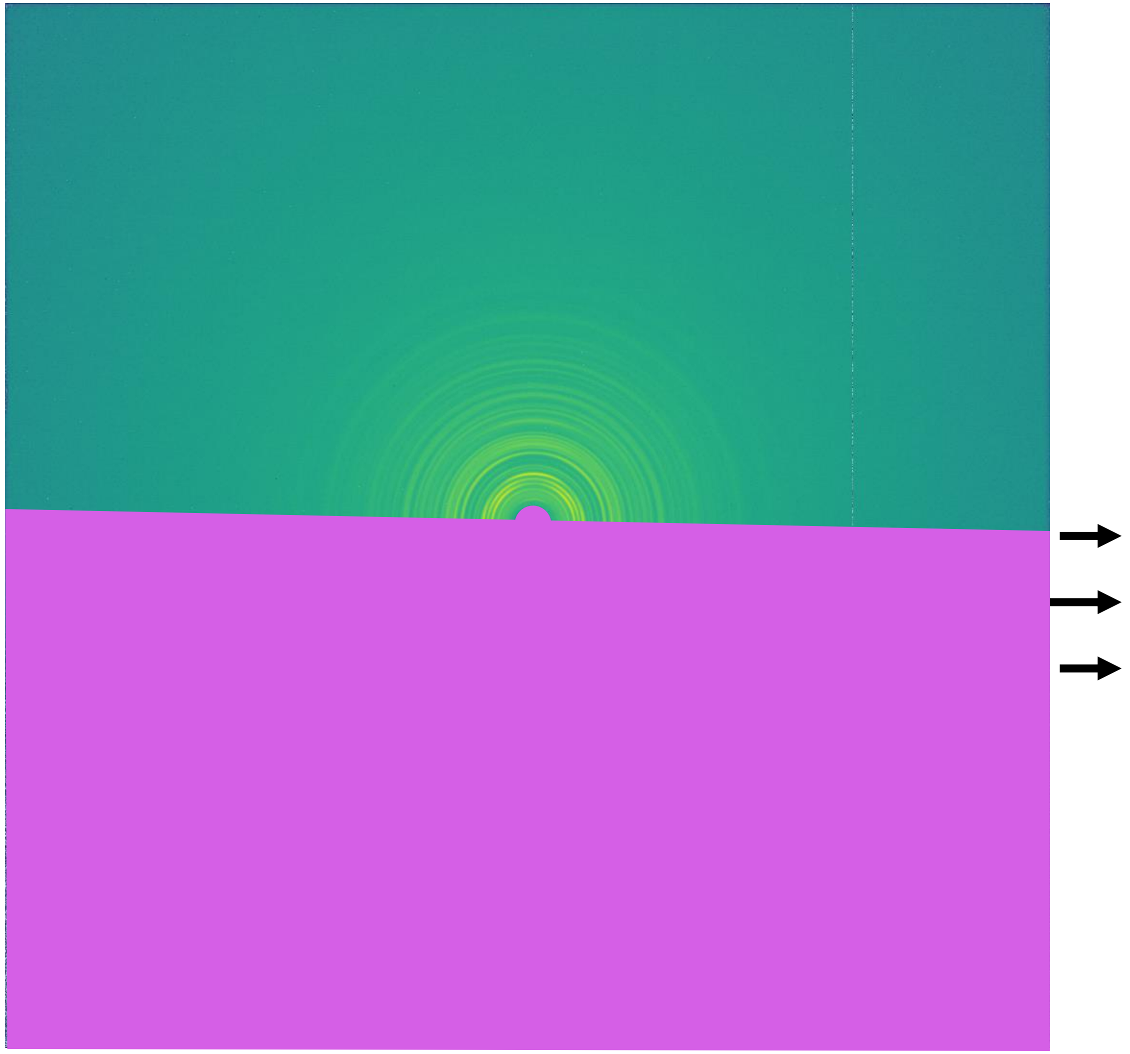
50 nm HfO_2 film on 1 mm fused silica substrate



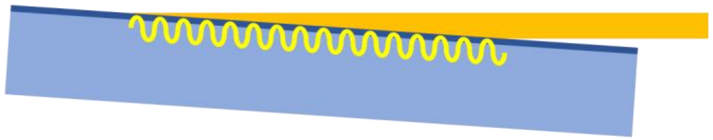




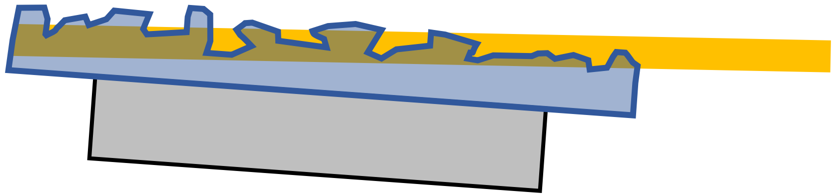
Grazing incidence scattering



Ideal surface (flat):

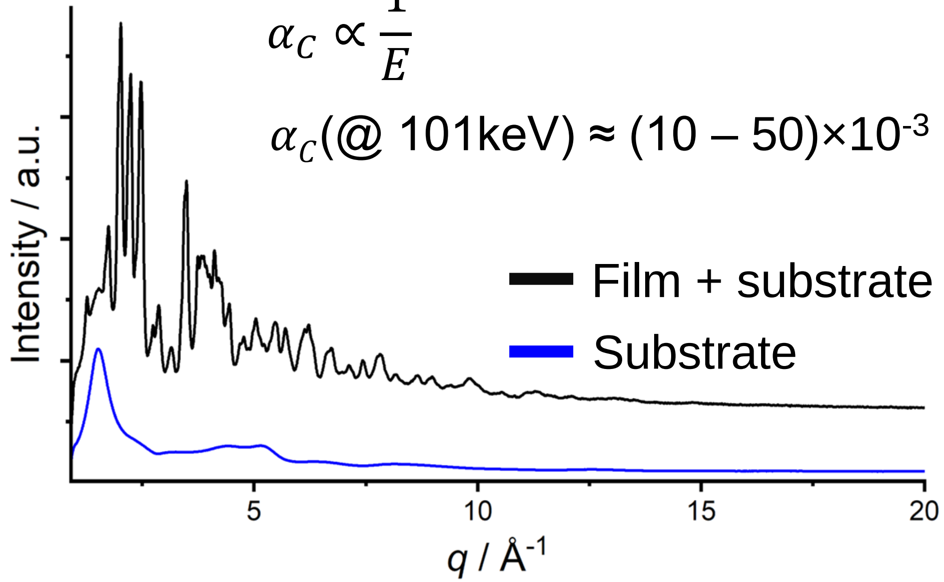


Real surface (rough):



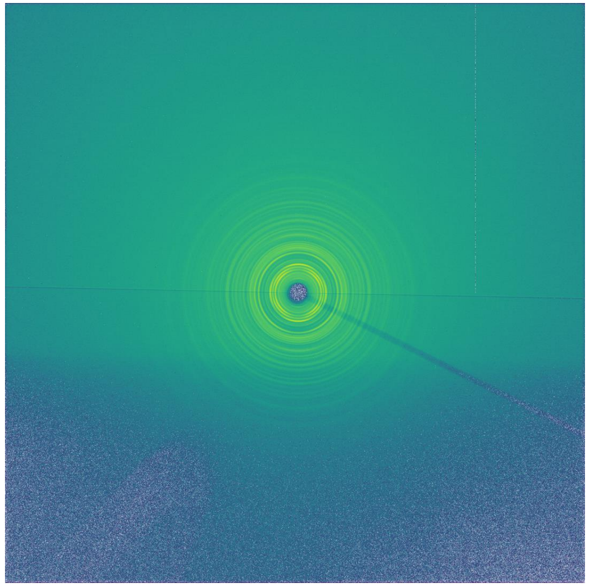
$\alpha_C \propto \frac{1}{E}$

α_C(@ 101keV) ≈ (10 – 50)×10^{-3} °



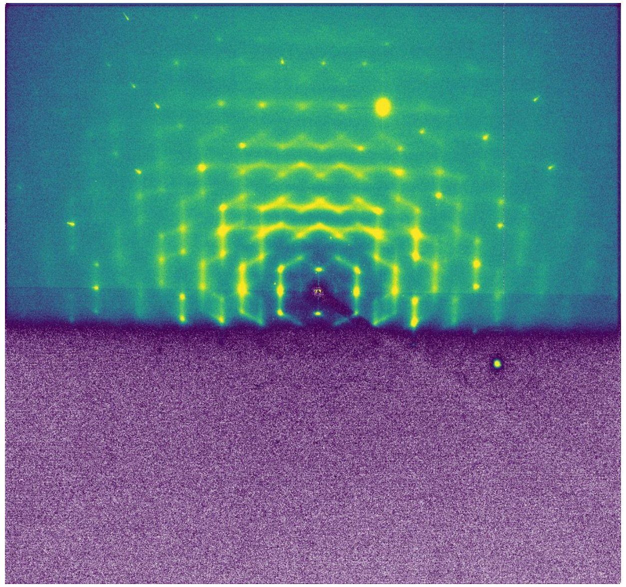
Amorphous vs. single crystalline substrates

Fused silica (amorphous)



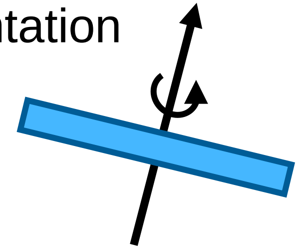
- ✓ Isotropic
- ✓ Straightforward
- ✓ Universal
- ⊗ Large background
- ⊗ Mostly not representative of application cases

Silicon (single crystalline)



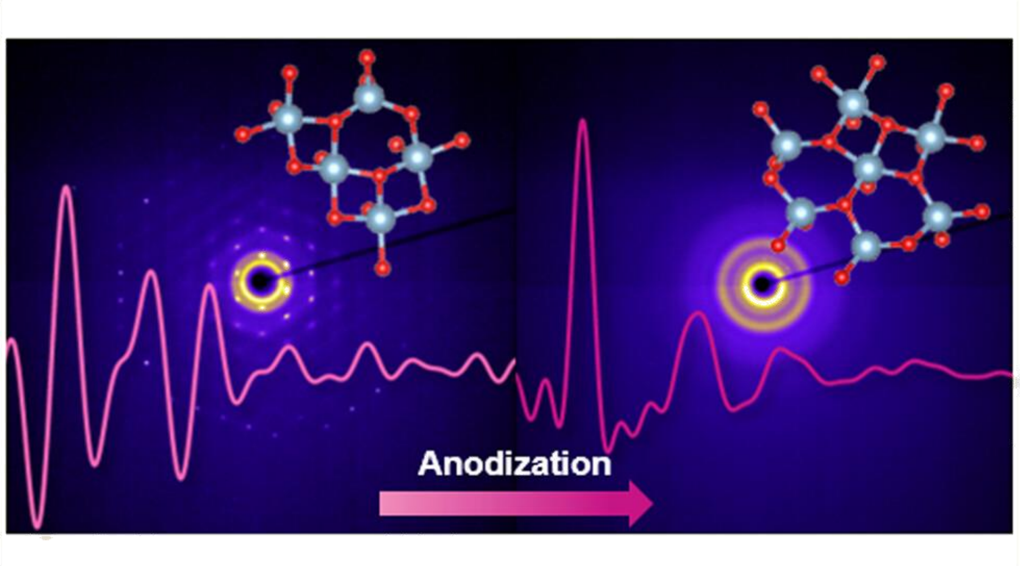
- ✓ Low background signal
- ✓ In line with application
- ✓ High quality
- ⊗ Anisotropic (diffuse)
- ⊗ External background does not reproduce background signal

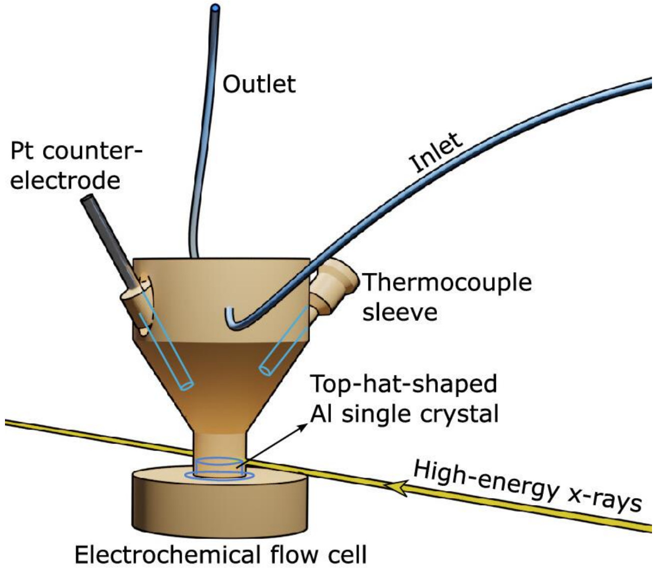
Random azimuthal orientation

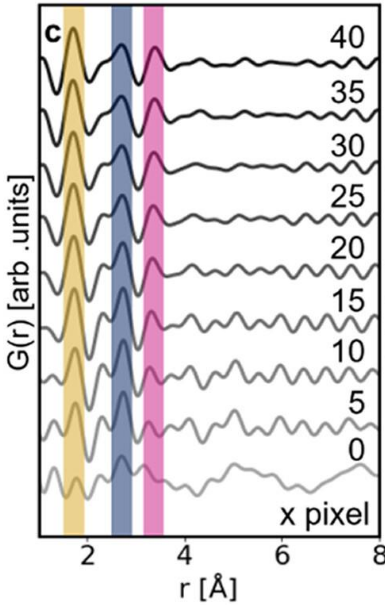


Grazing incidence total scattering in single-crystalline substrates

Al-native oxide pdf from Al single crystalline background



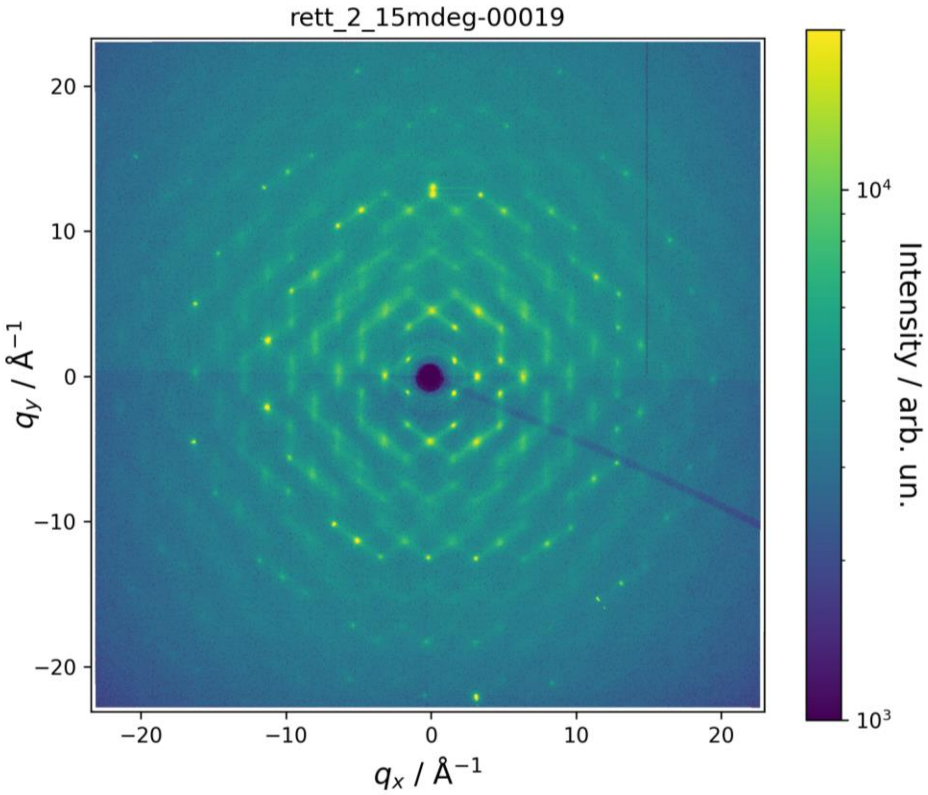


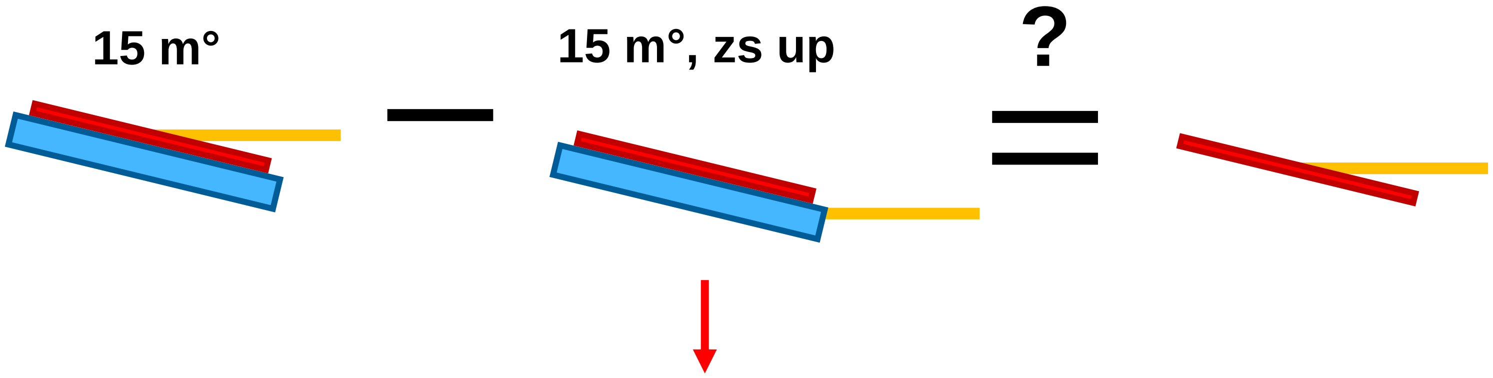


- At t = 0, there is significant diffraction from the single crystalline Al substrate
- Masking the Bragg spots enables to process the film scattering into pdf
- The retrieved pdf in consistent with the Al native oxide structure

Magnard, N. P., Igoa Saldaña, F., Dippel, A.-C., et al. (2025). *ACS Applied Materials & Interfaces*.

Measurement conditions



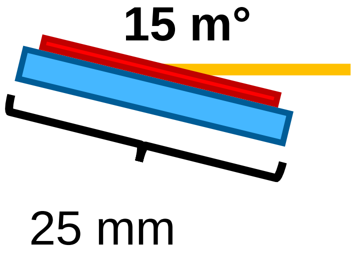


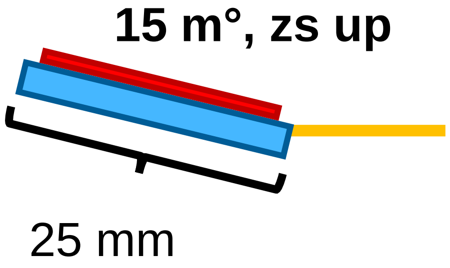
Can minimal sample movement to probe bulk substrate reproduce the background scattering?

Sample courtesy of Alexander Rettie

2D background subtraction

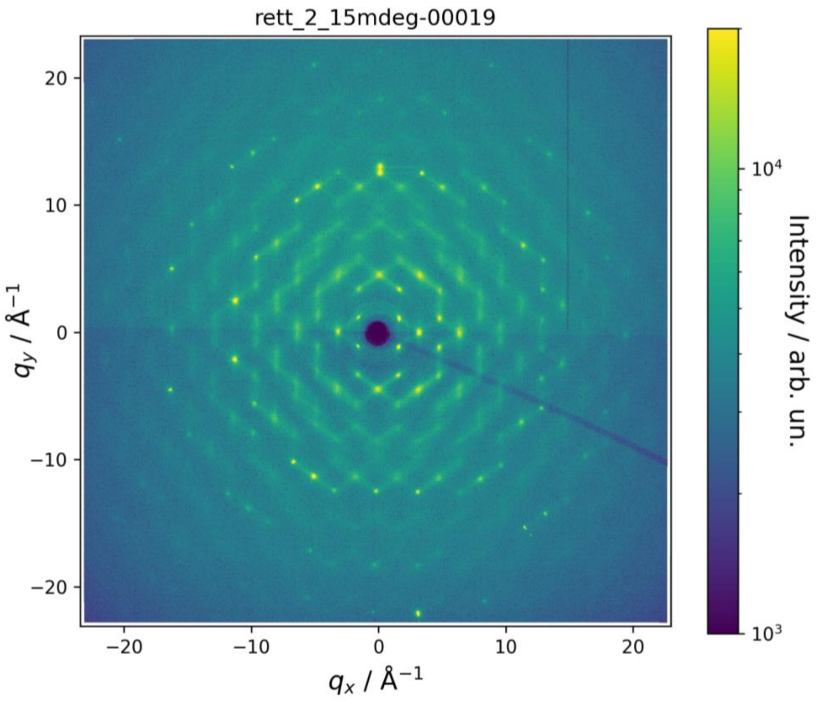
$Li_{2.75}Al_1P_{1.25}O_{5.1}$

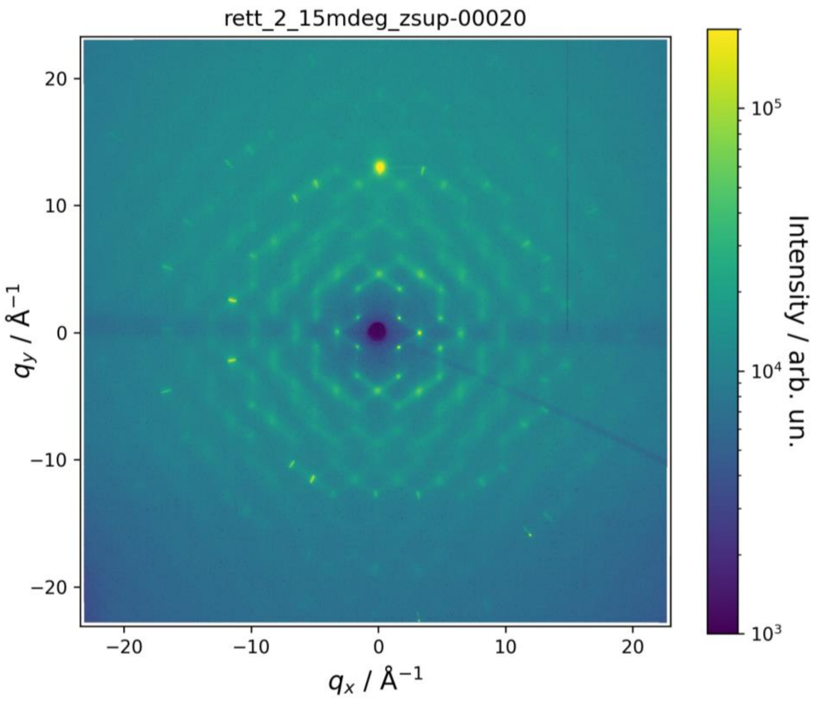


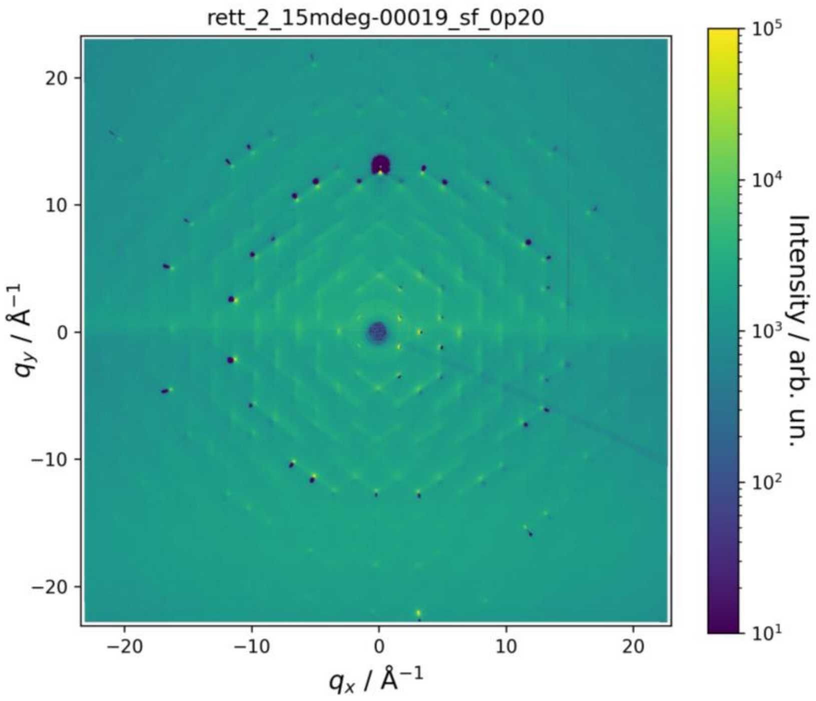


Subtraction

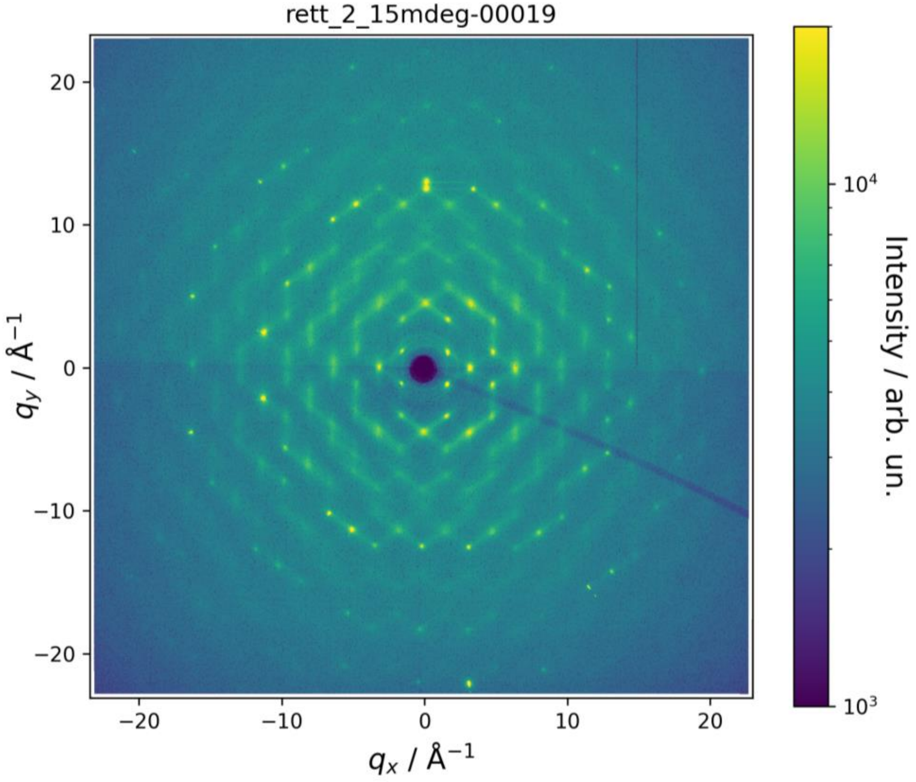
Unsuccessful subtraction, pure substrate and film background not matching

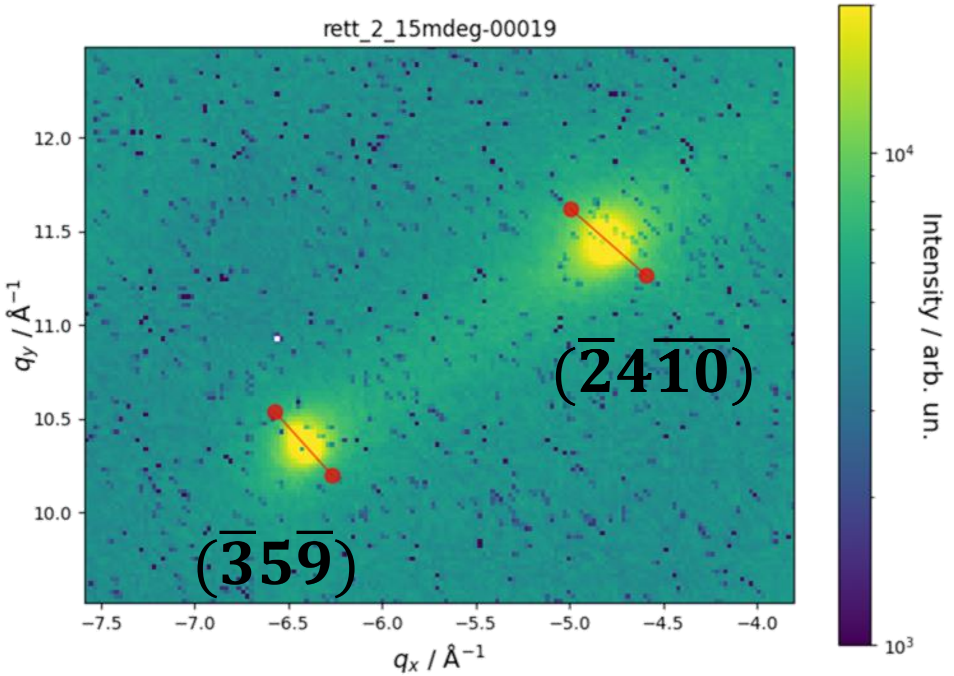


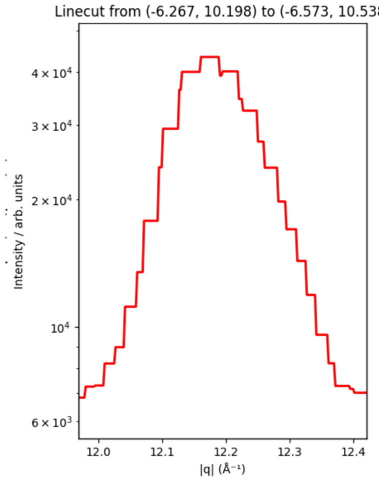


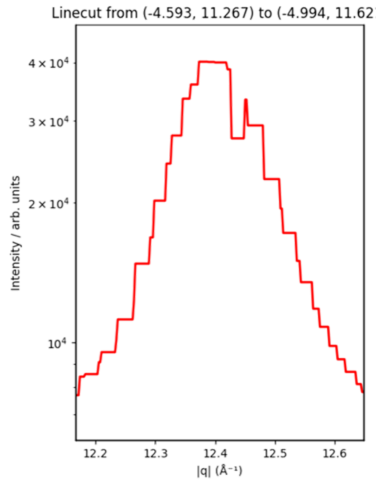


A closer look to the subtraction problem



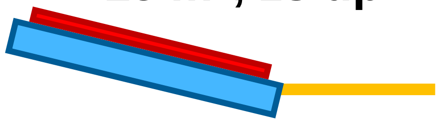


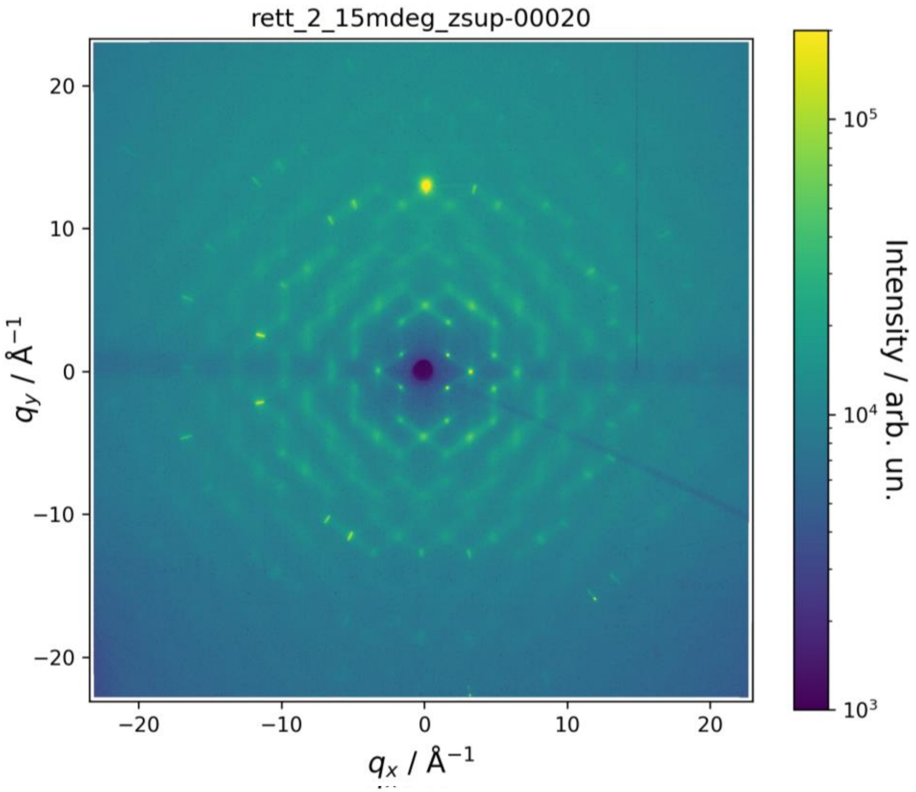


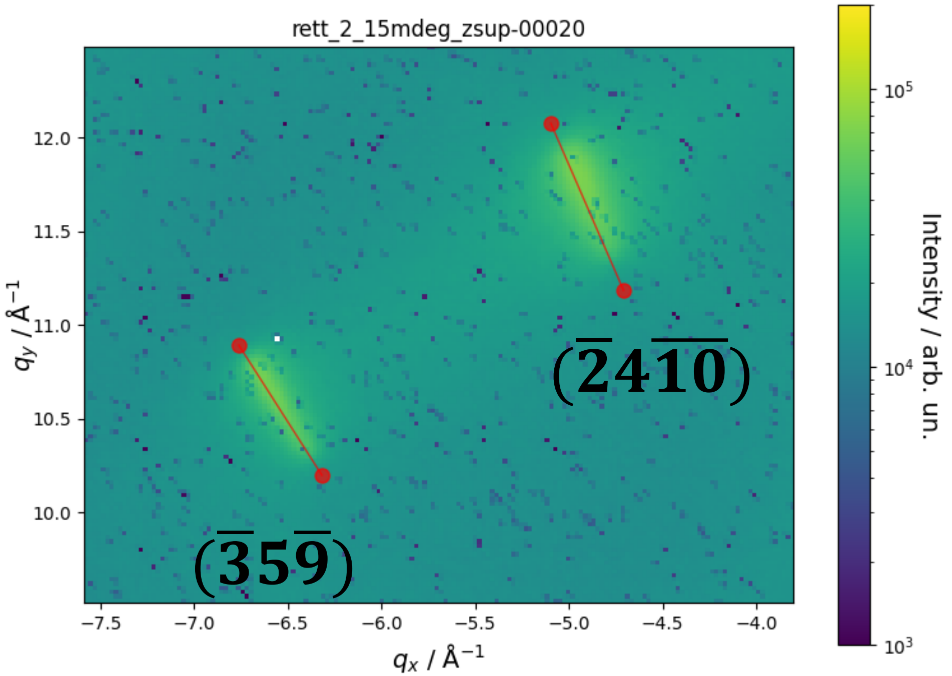


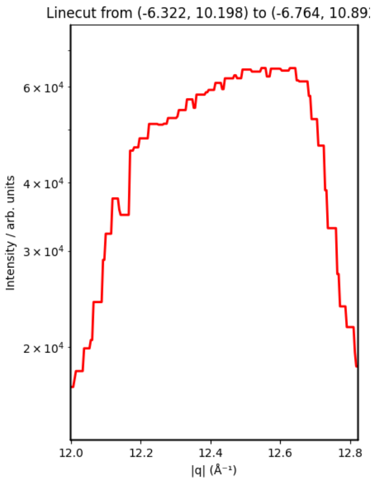
A closer look to the subtraction problem

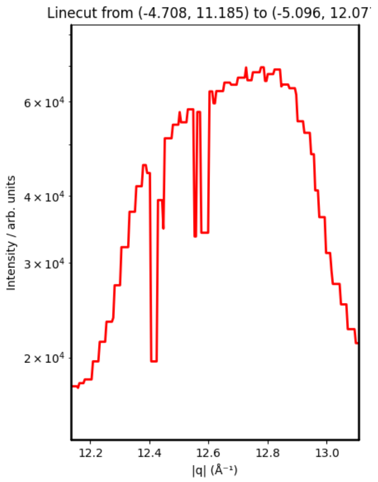
15 m°, zs up





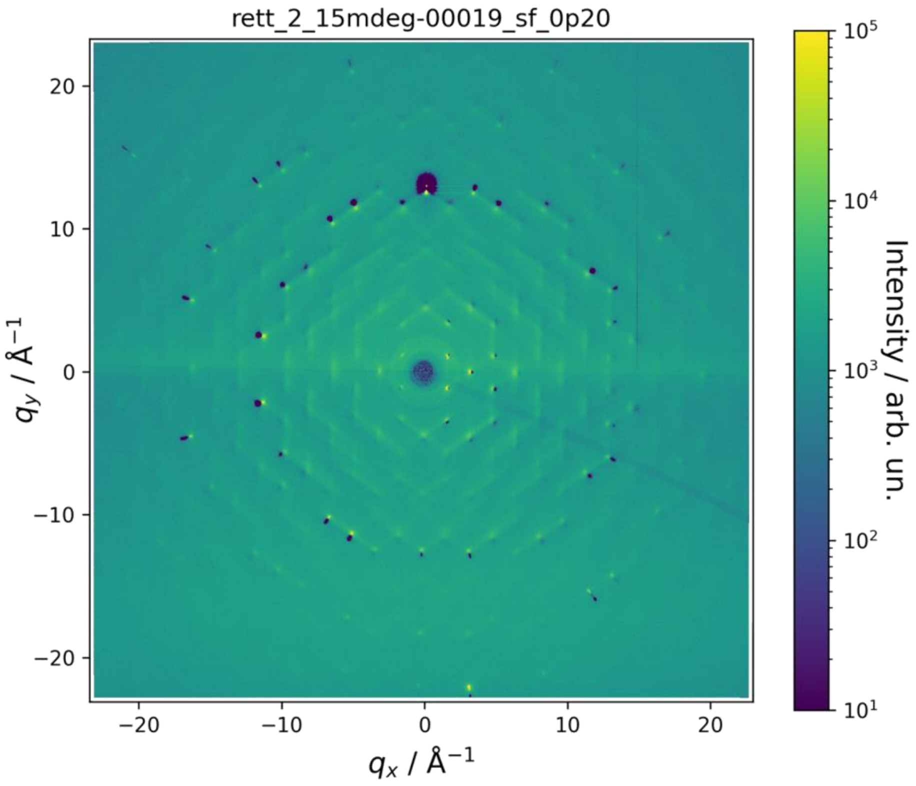


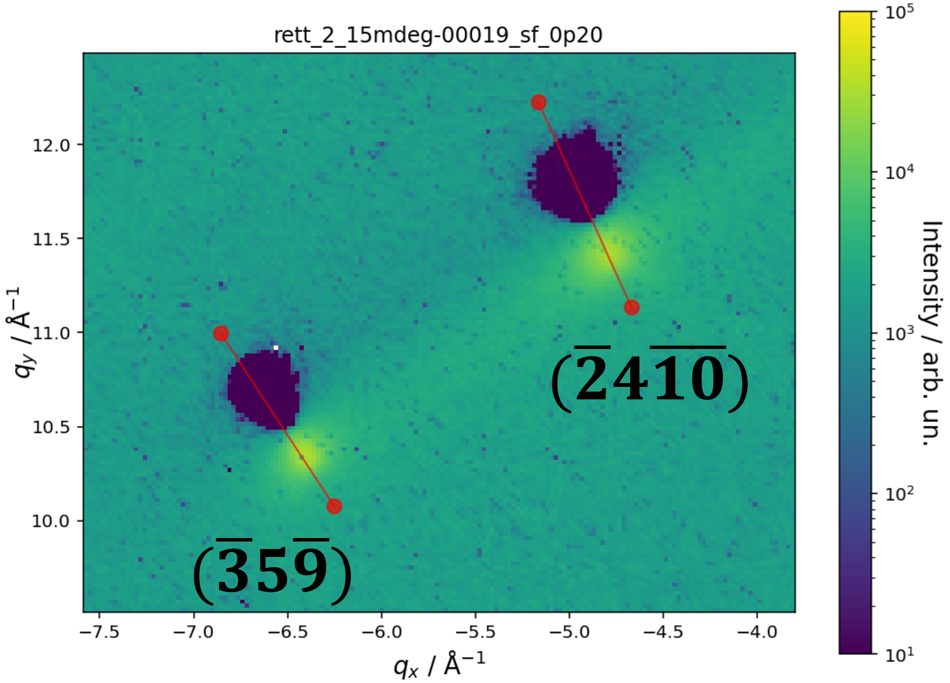


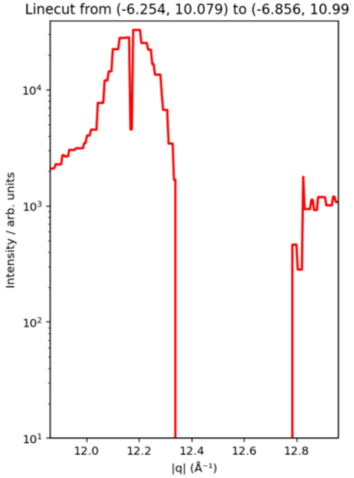


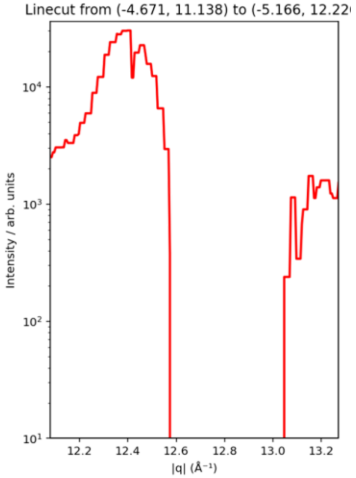
A closer look to the subtraction problem

Subtraction



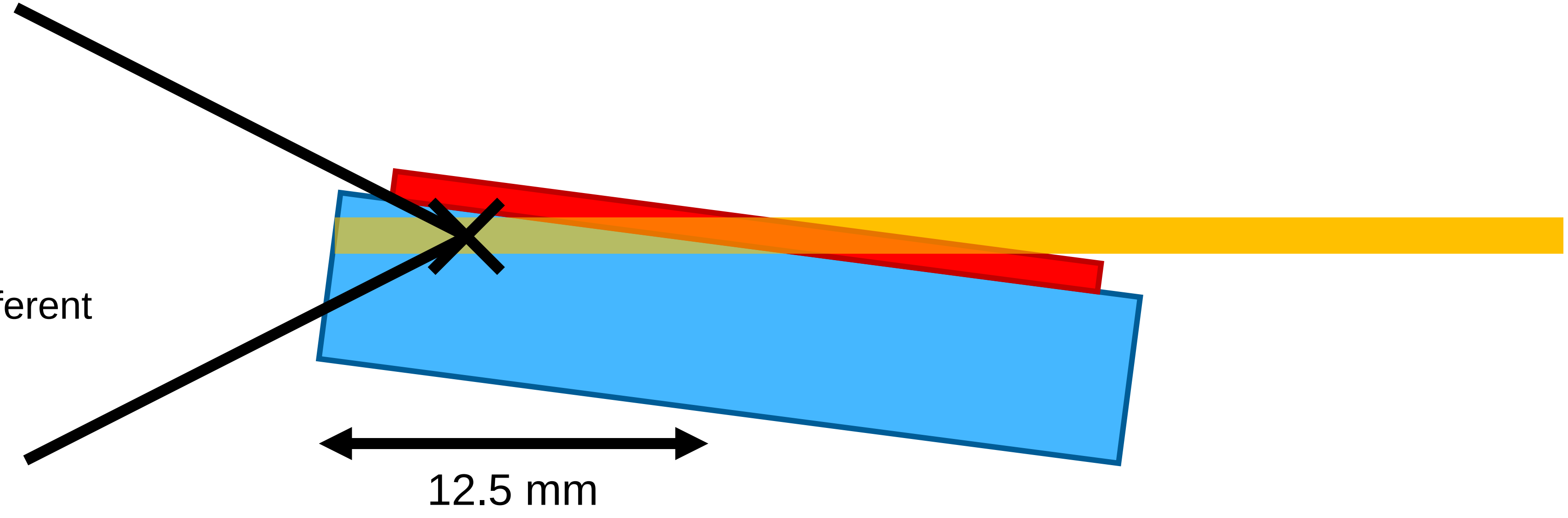




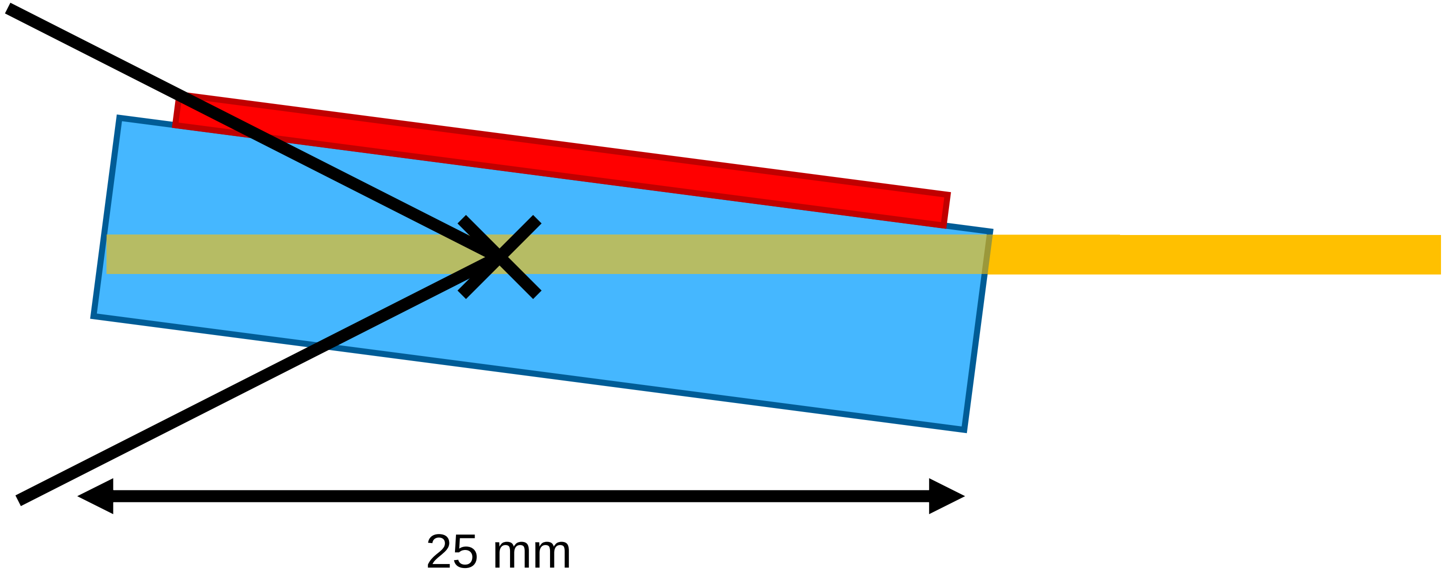


A closer look to the subtraction problem

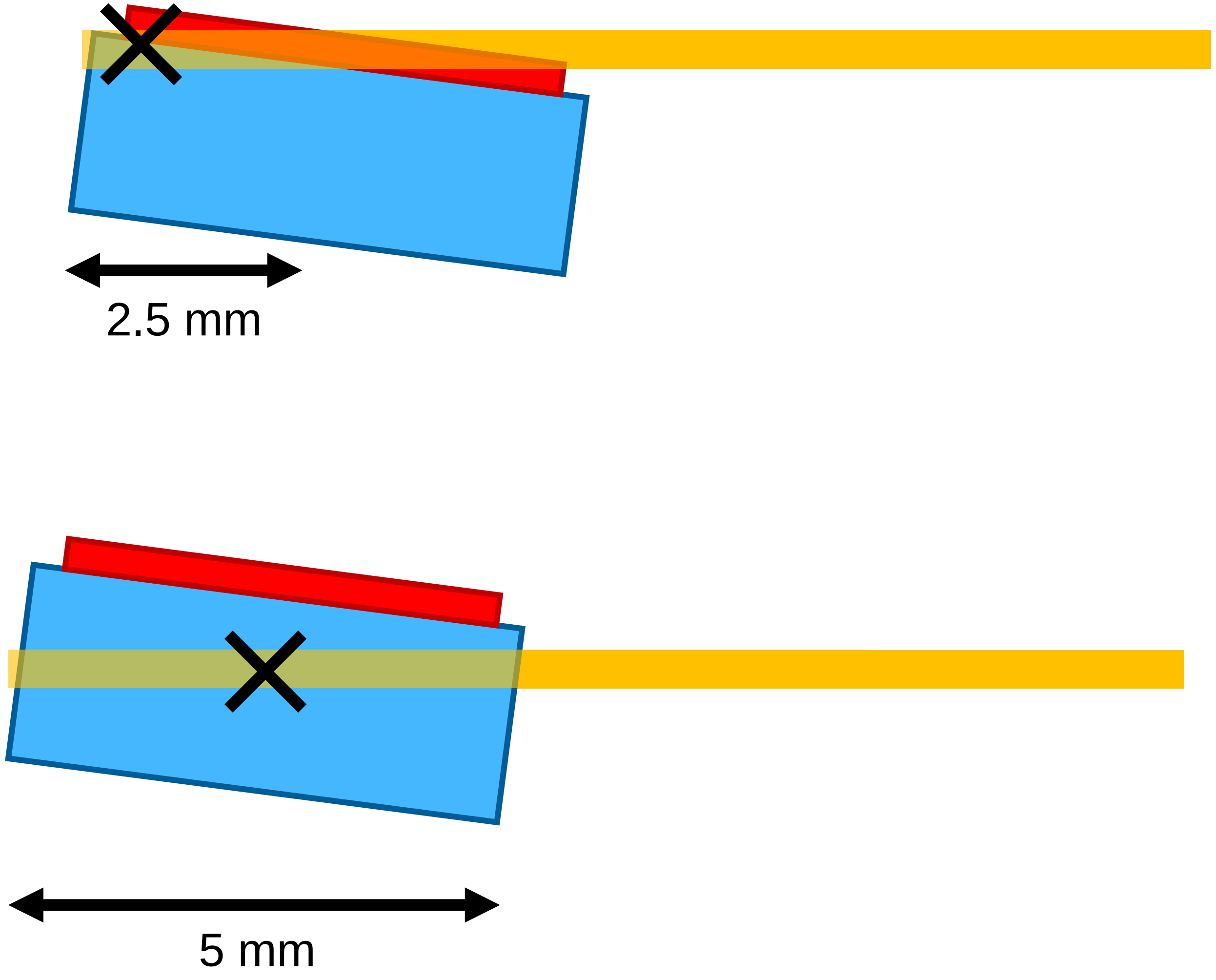
Different mean origins give different sample-to-detector distances



A broad distribution of sample-to-detector distances causes broad scattering profiles, irreproducible in grazing incidence



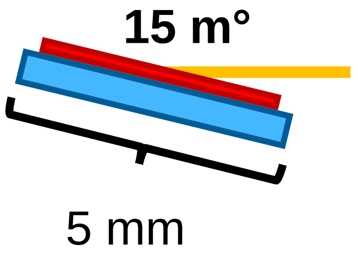
A closer look to the subtraction problem

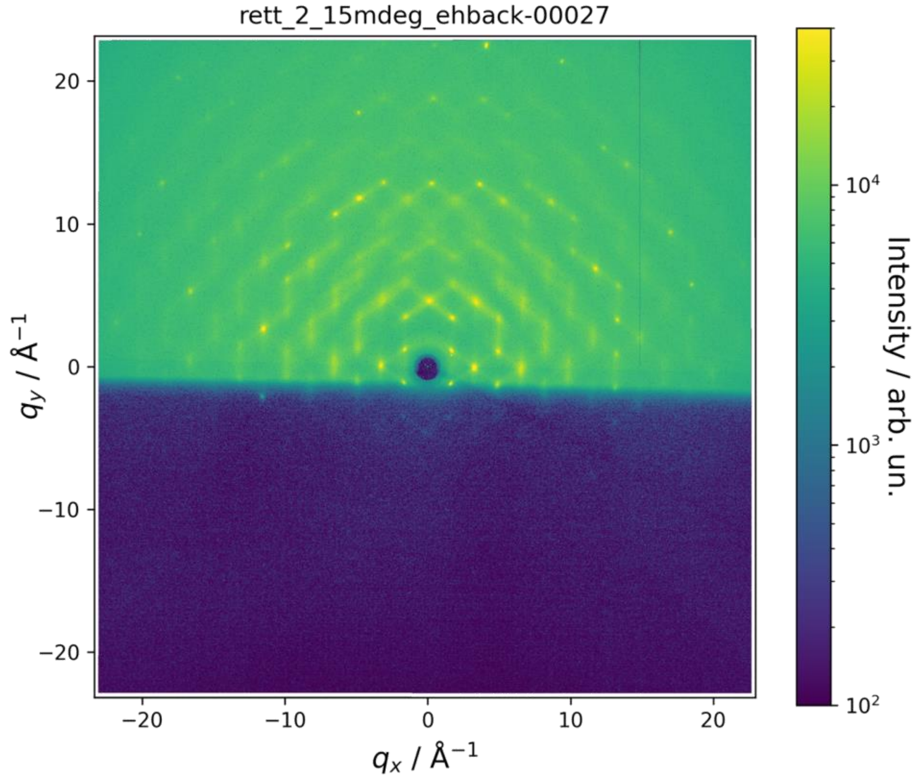


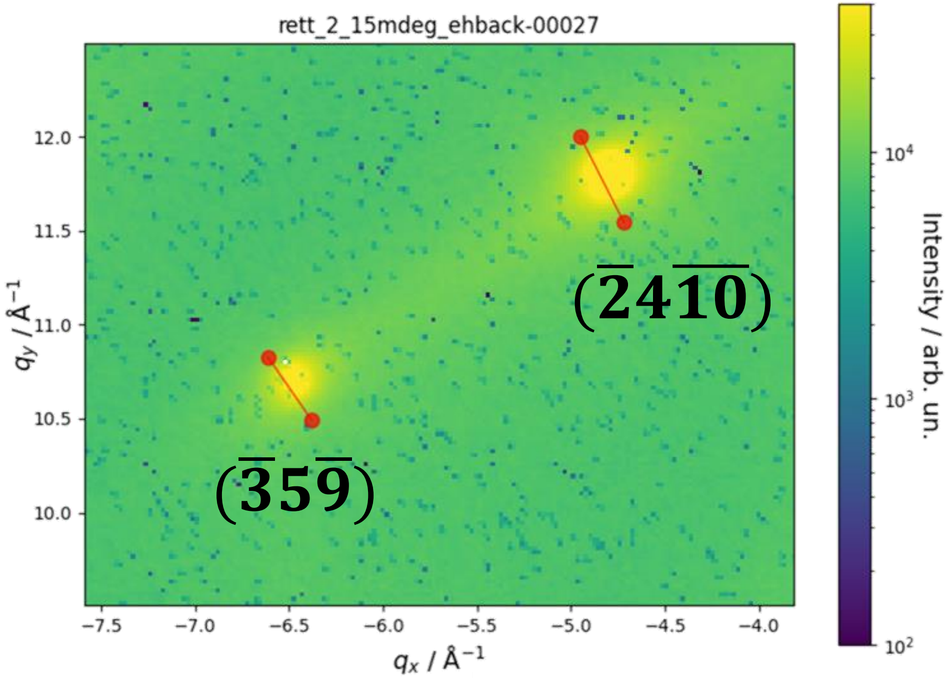
Reducing the sample dimensions could minimize the sample-to-detector differences?

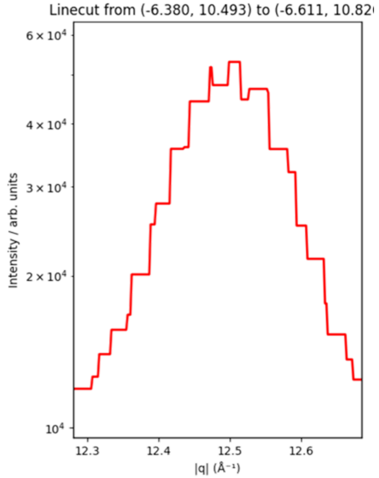
Cut sample to 5 mm

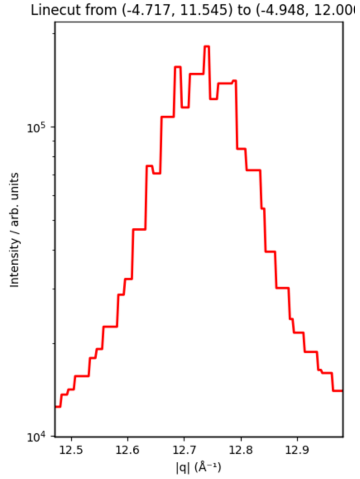
$Li_{2.75}Al_1P_{1.25}O_{5.1}$





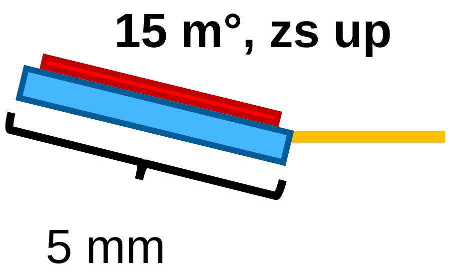


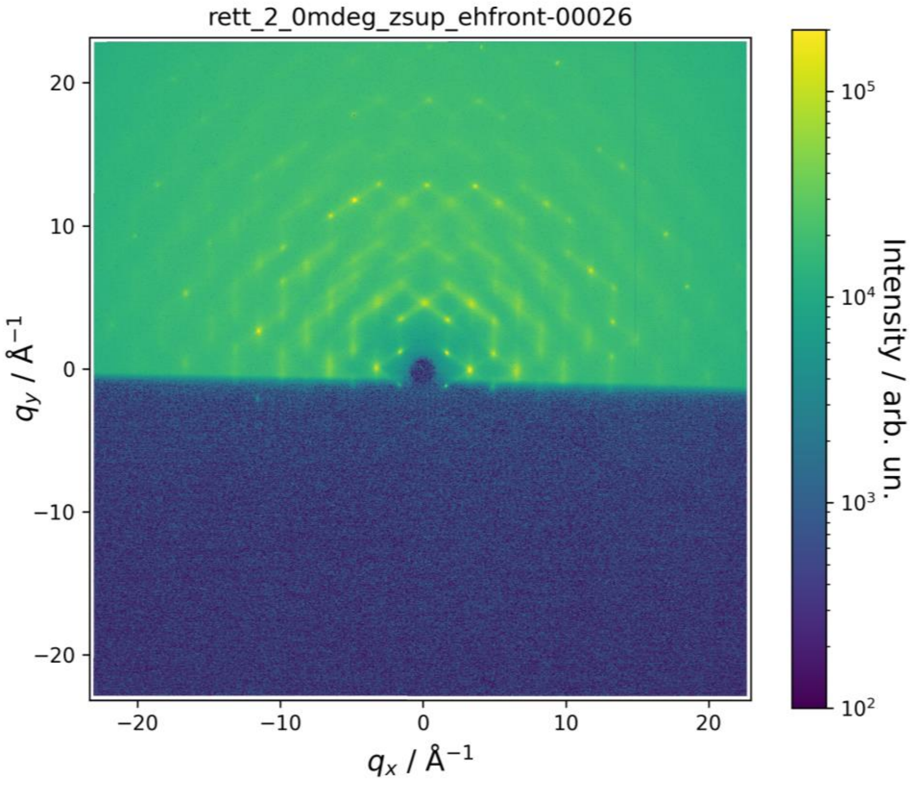


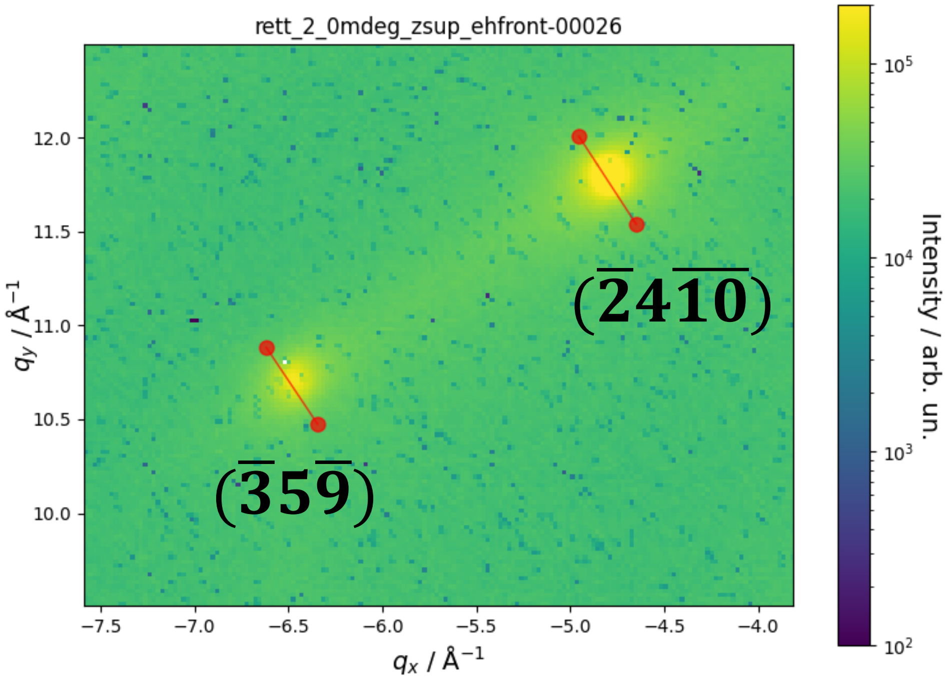


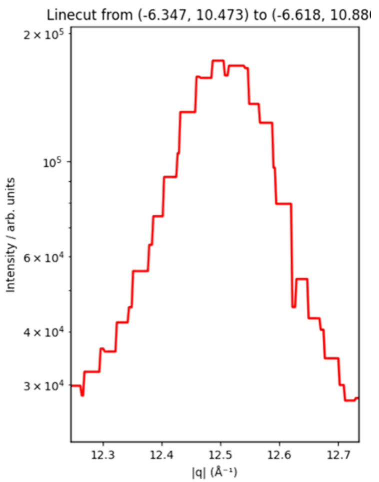
Cut sample to 5 mm

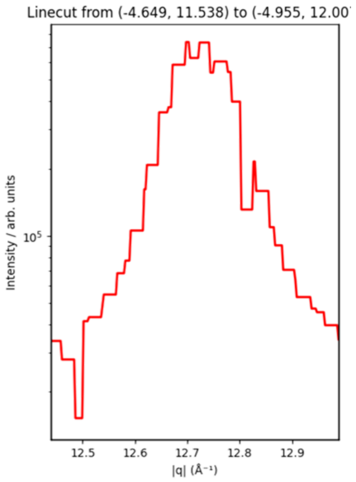
$Li_{2.75}Al_1P_{1.25}O_{5.1}$







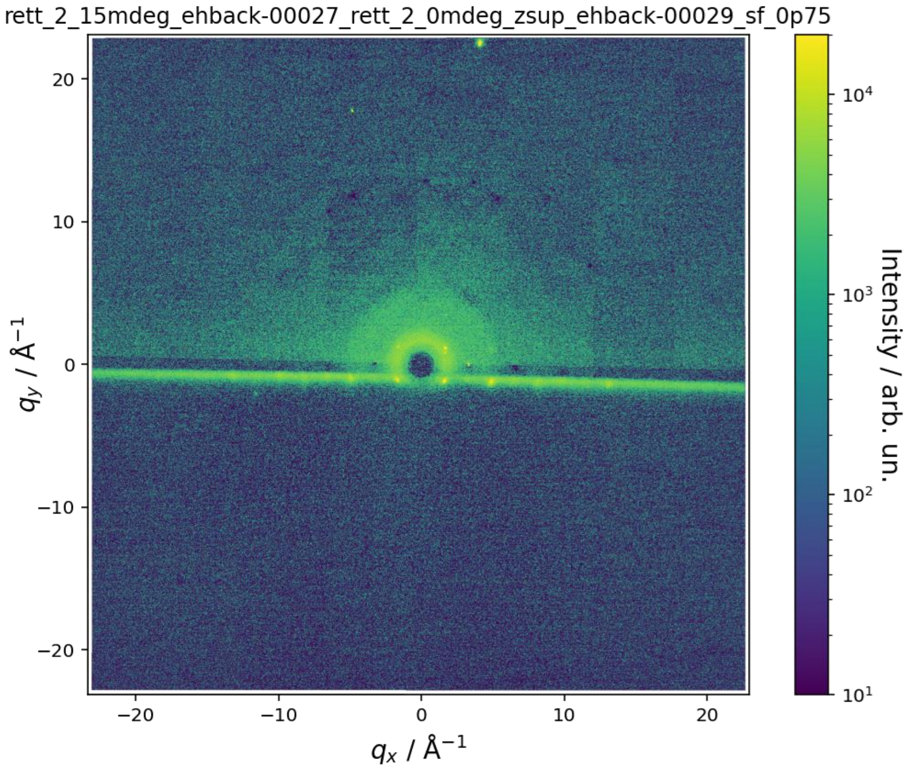


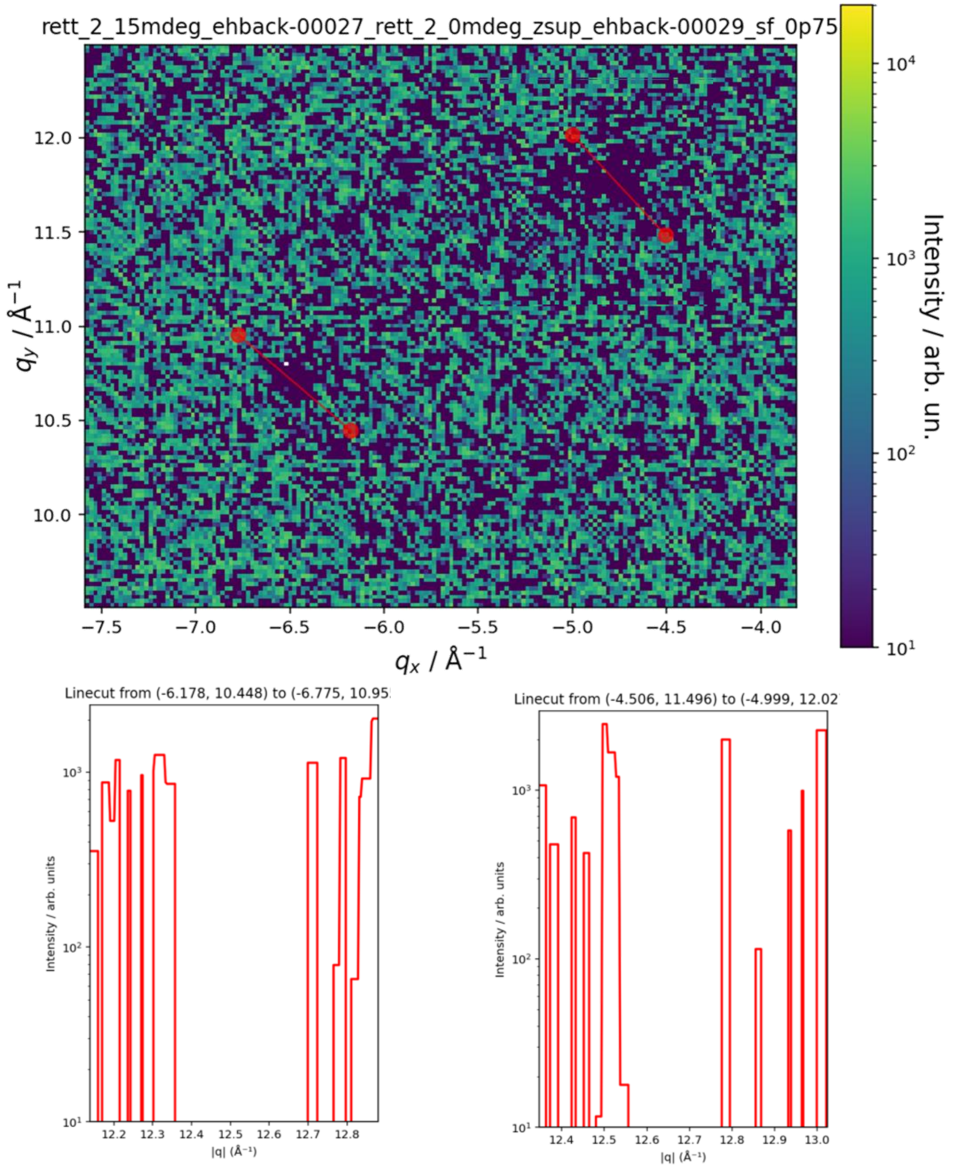


Cut sample to 5 mm

$Li_{2.75}Al_1P_{1.25}O_{5.1}$

Subtraction

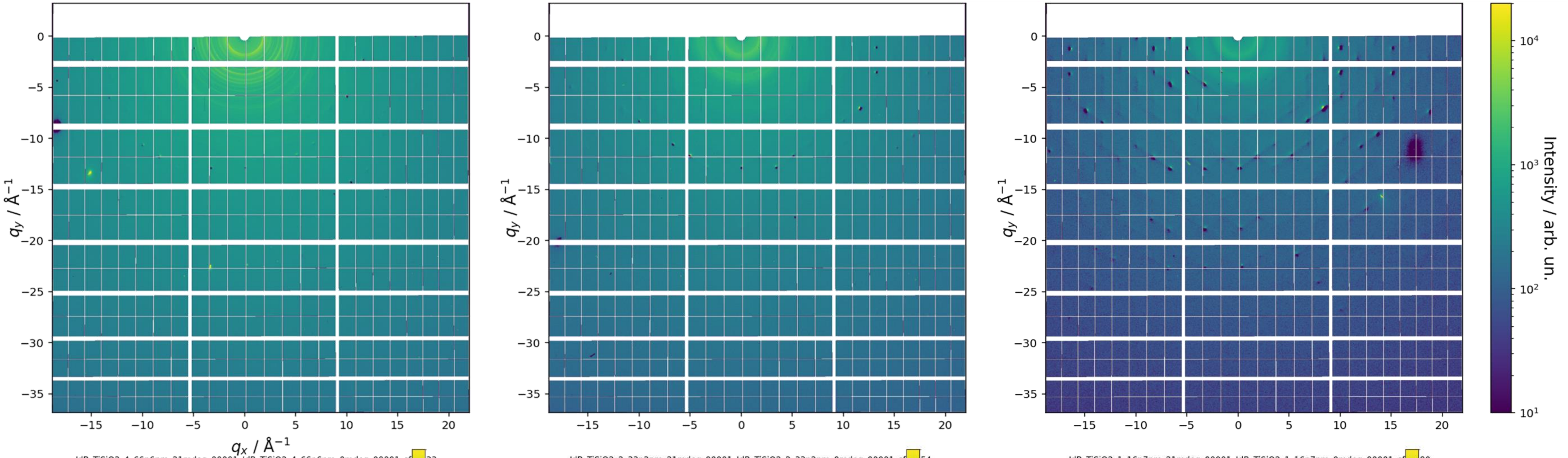


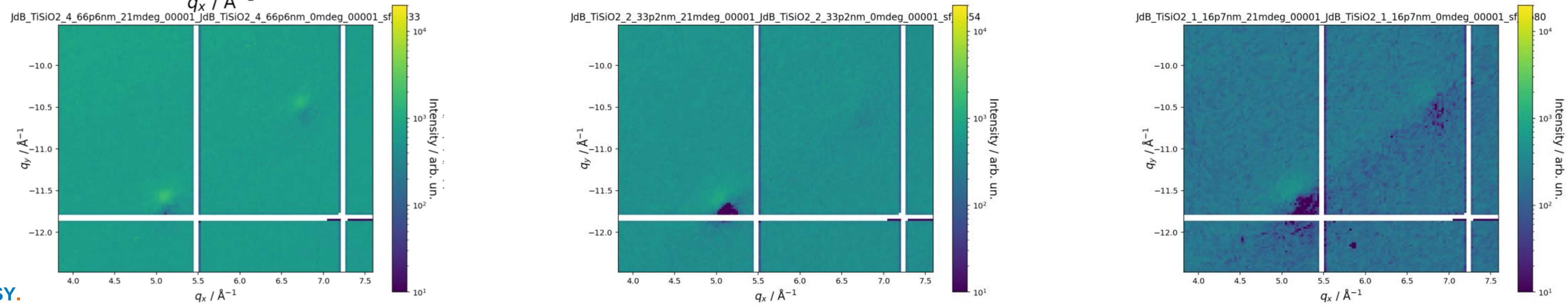


Sample courtesy of Jorden de Bolle

Is the problem really gone?

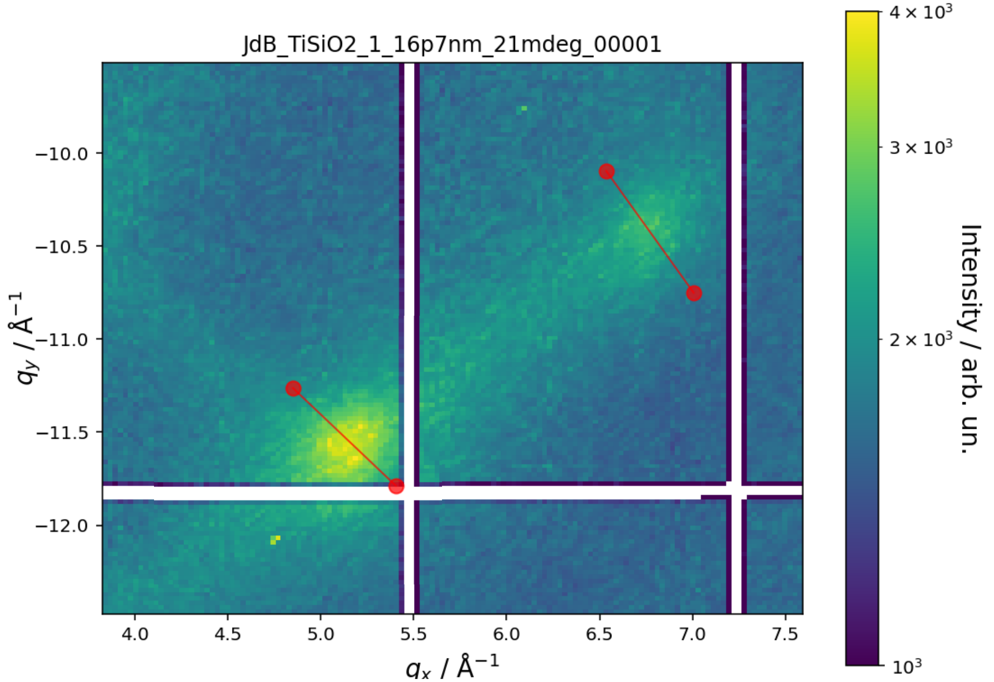
$Ti_xSi_{1-x}O_2$ 66.6 nm on Si | **$Ti_xSi_{1-x}O_2$ 33.2 nm on Si** | **$Ti_xSi_{1-x}O_2$ 16.7 nm on Si**

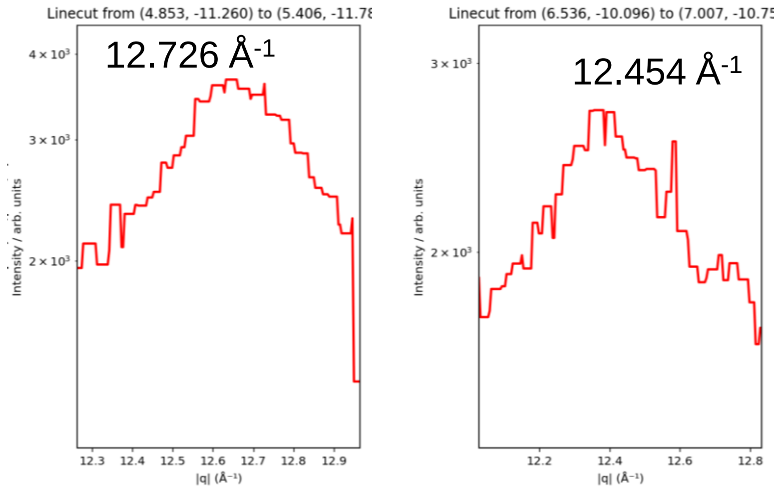




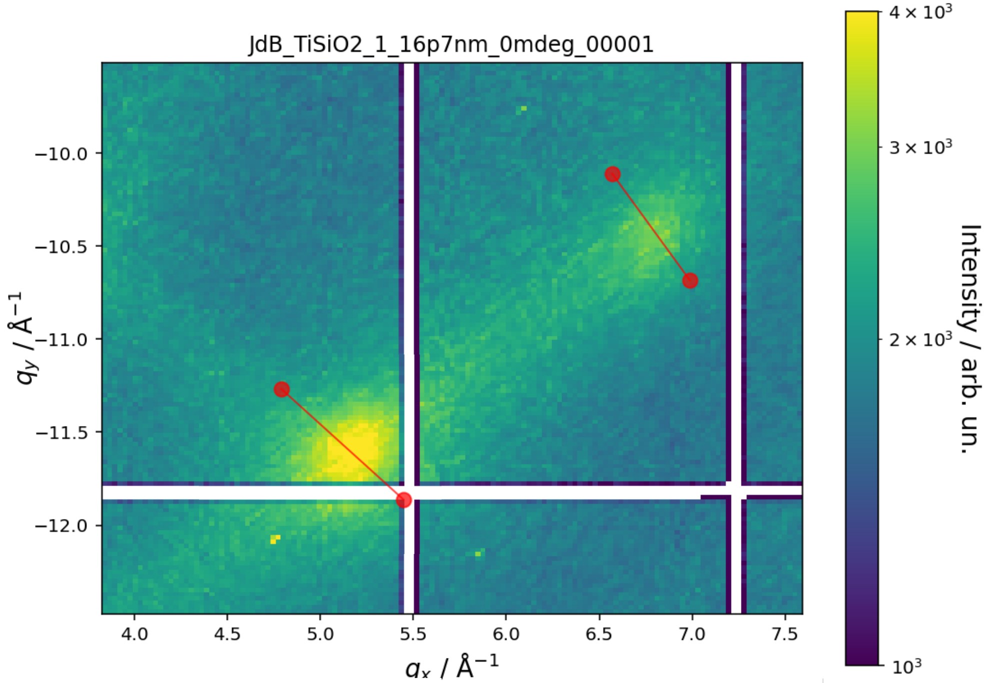
$TiSiO_2$ 16.7 nm on Si

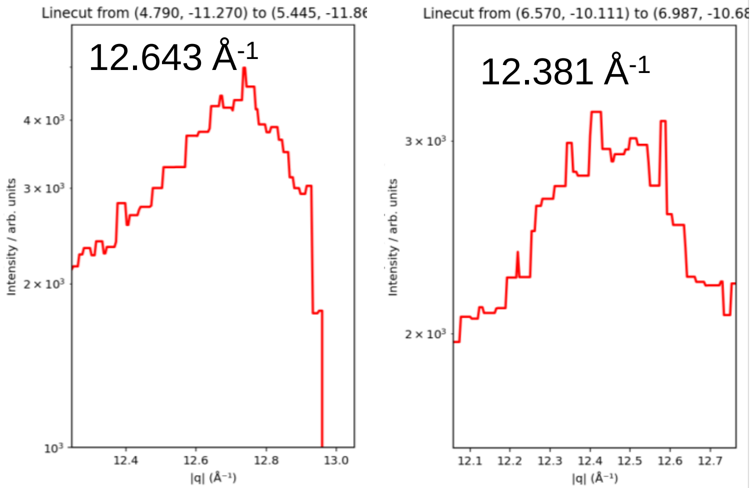
Thin film



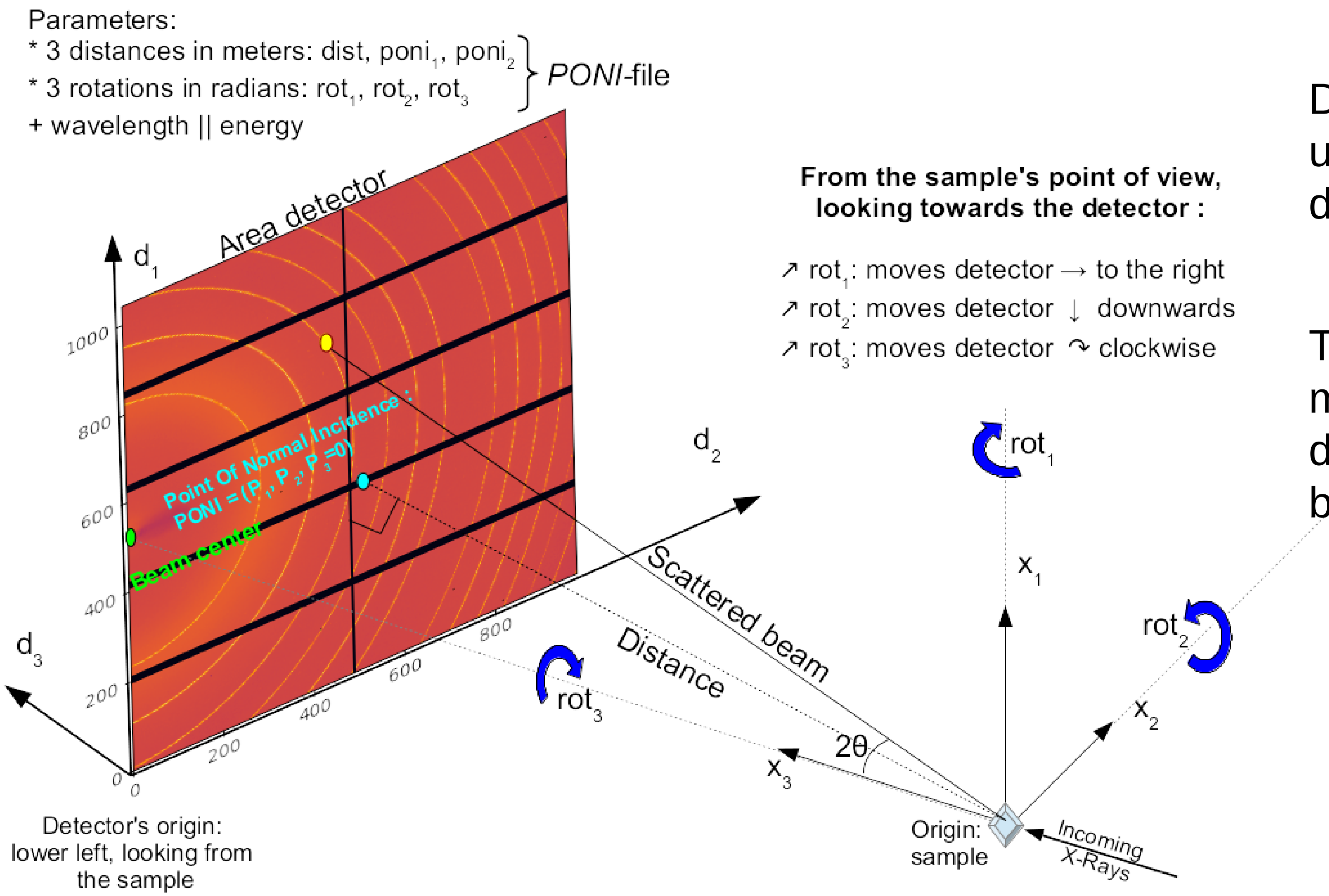


Substrate





Background sample-to-detector distance correction

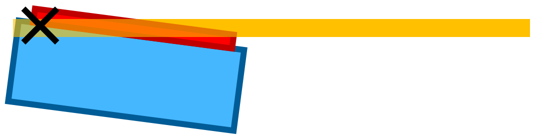


Detector calibration parameters are used standardly to reconstruct/integrate detector images

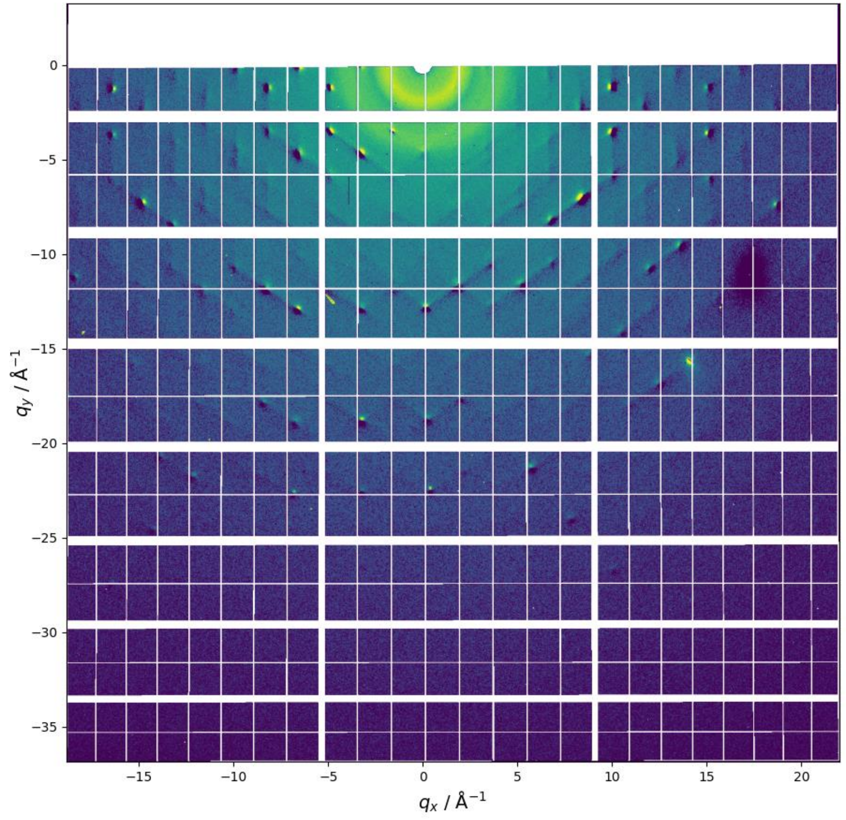
The distance parameter can be manipulated to account for the intrinsic distance change when measuring the background

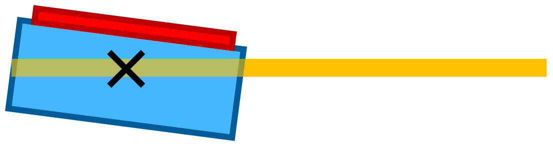
Kieffer, J., Valls, V., Blanc, N., & Hennig, C. (2020). New tools for calibrating diffraction setups. *Synchrotron Radiation*, *27*(2), 558-566.

Background sample-to-detector distance correction

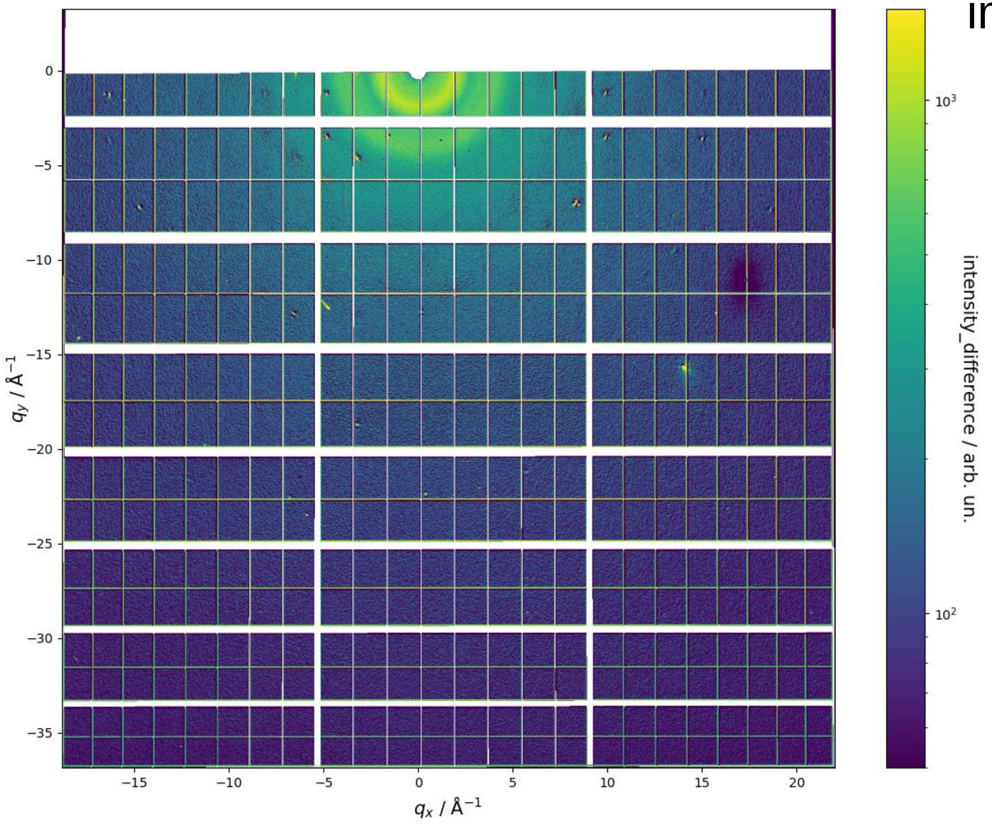


$SDD_{Background}$ = 300.82 mm





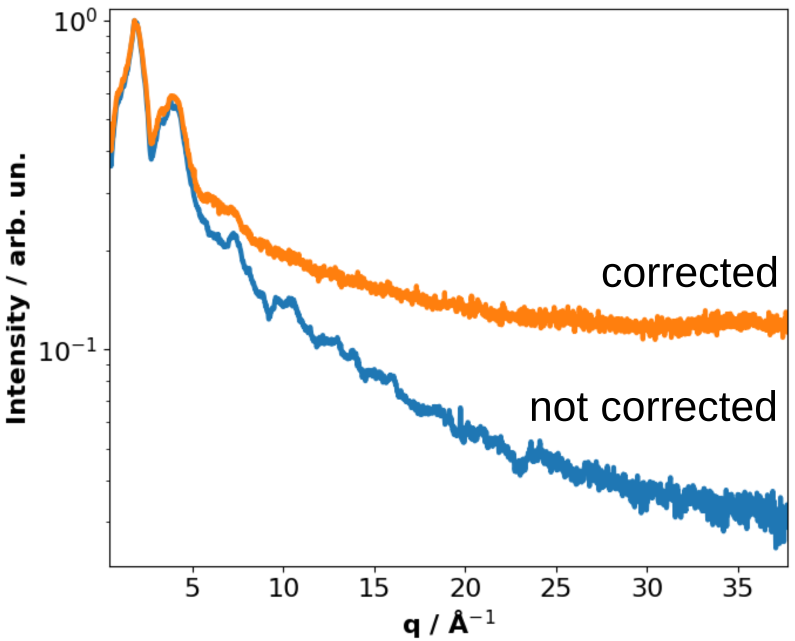
$SDD_{Background}$ = 301.82 mm



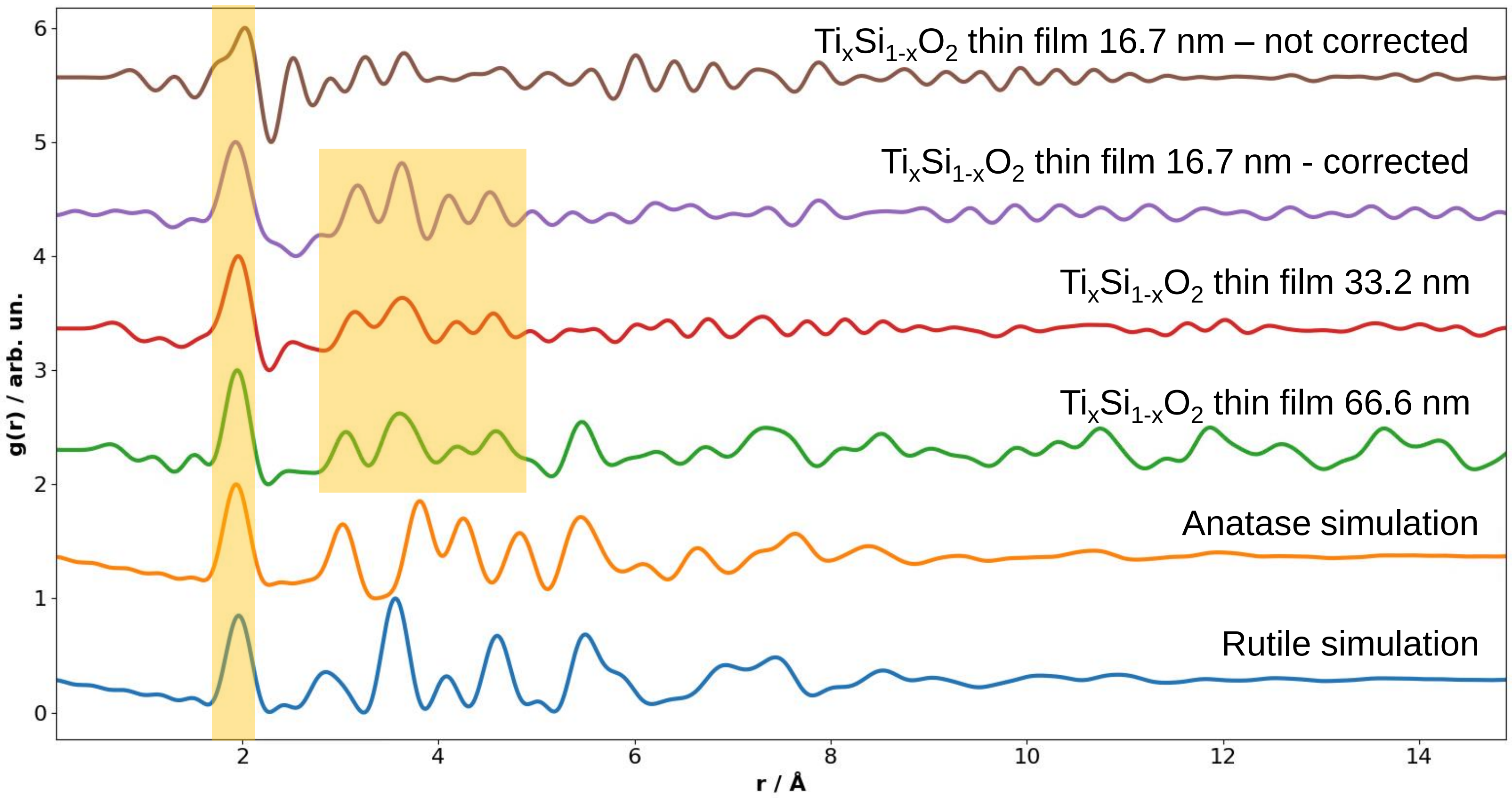
- Sample-to-detector distance discrepancies can be accounted for in the 2D images

Background sample-to-detector distance correction

Reciprocal space

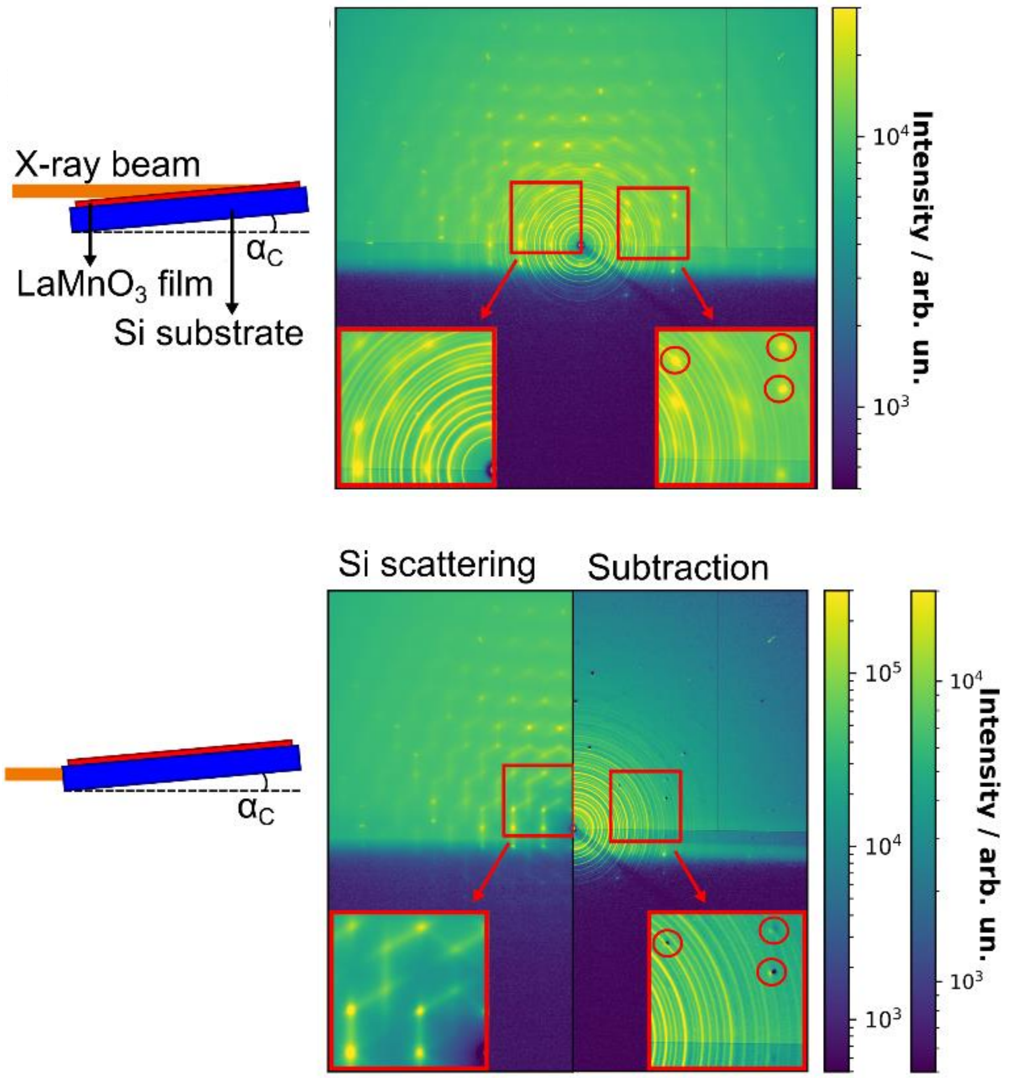


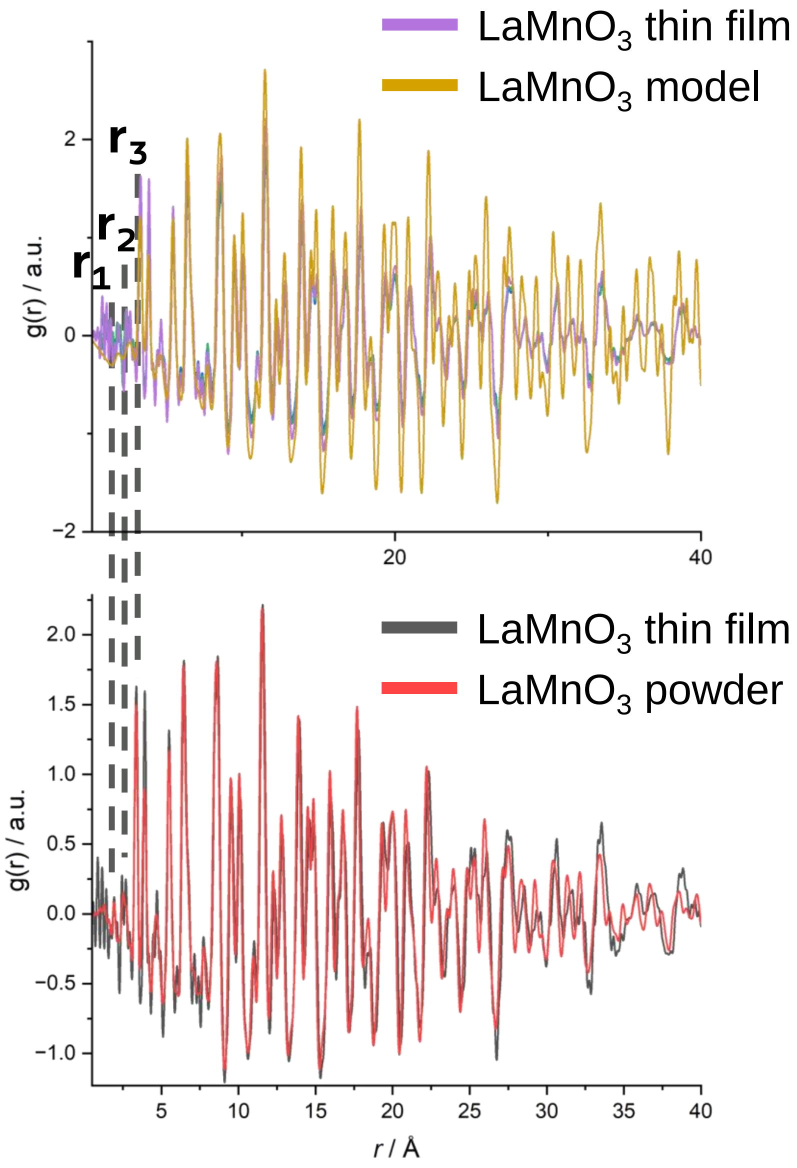
Real space

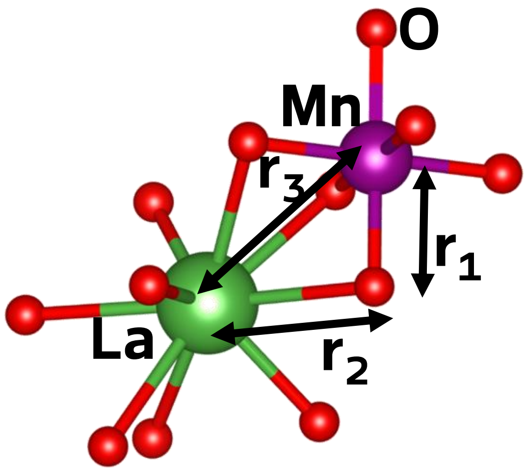


- ➢ The corrected data shows a smooth scattering decay in *q*-space
- ➢ The corrected data presents similar features in real-space to the thicker and more crystalline counterparts. Reliable correlation peaks can be observed up to 5 Å

Validation in crystalline material







- Crystalline $LaMnO_3$ showed well agreement between thin film – model and thin film – powder pdfs

Summary

- Scattering in grazing incidence conditions highly enhances the signal
- Substrate background is always present: isotropic for amorphous and anisotropic for single crystalline
- To subtract the background in a single crystalline substrate, a small (5 mm) length has to be kept
- Discrepancies between substrate and film background become more notorious the less the sample scatters
- The geometrical discrepancies can be corrected for by 2D *q*-space reconstruction prior subtraction
- The recovered pattern fits with powder and modelled data for a crystalline sample

Thank you

fernando.igoa@desy.de

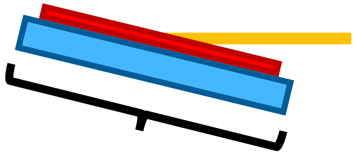
Beamlines P07(DESY) & P21.1 @ PETRA

2D subtraction method validation

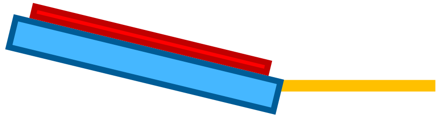
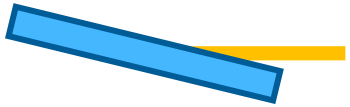
Measurement conditions

Sample courtesy of Xiao Sun

Sample-detector = 400 mm
Energy = 101.45 keV
Grazing incidence angle = 20 mdeg
Detector = Perkin Elmer
Sample = $LaMnO_3$ thin film on Si (111) substrate
Focused beam dimensions = 2.6 x 200 μm



5 Background subtraction strategies were considered:

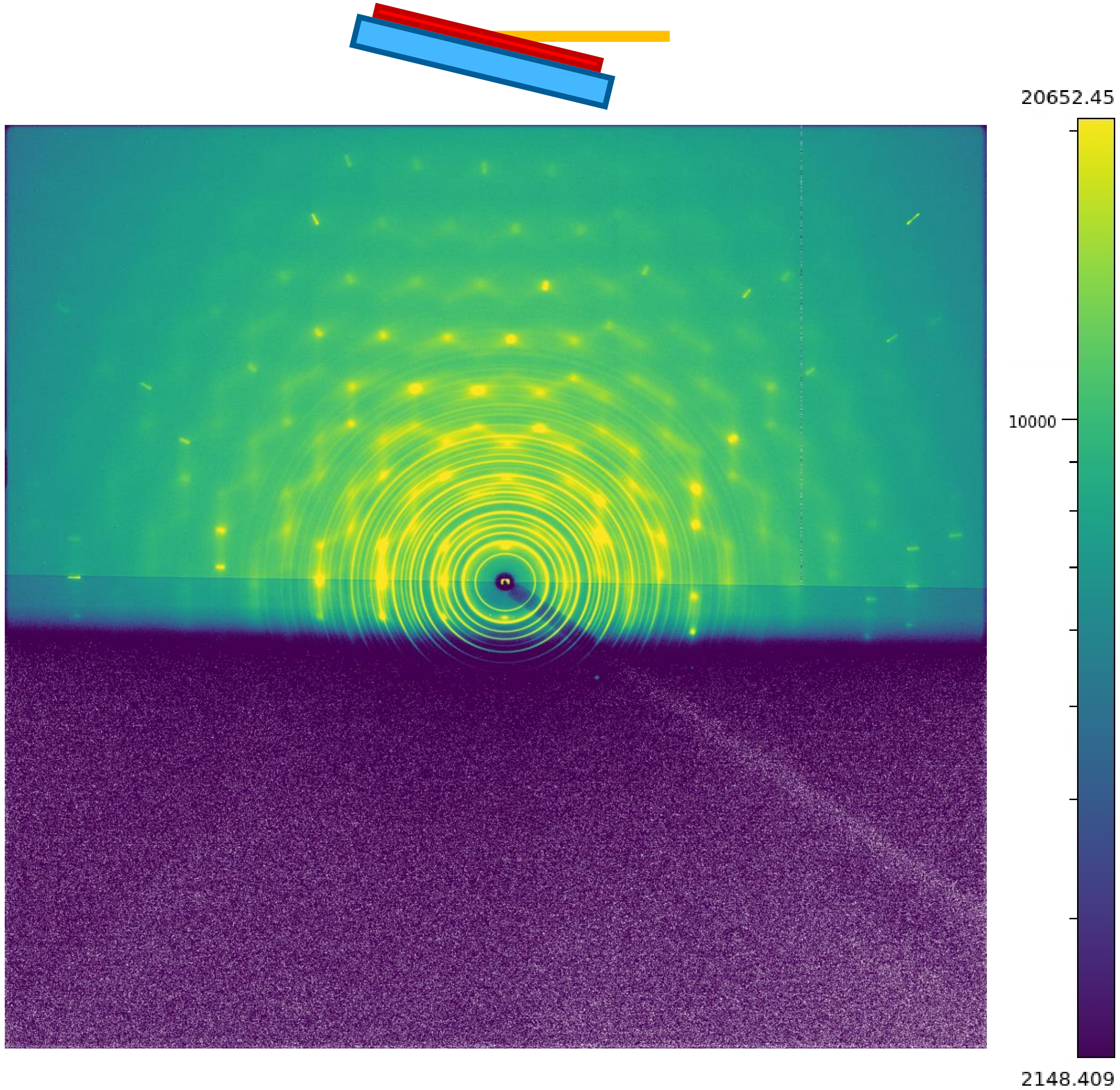




	0 m°	**20 m°, zs up**	**20 m°, Si substrate**
2D subtraction	✓	✓	✗
1D subtraction	✓	✓	✓

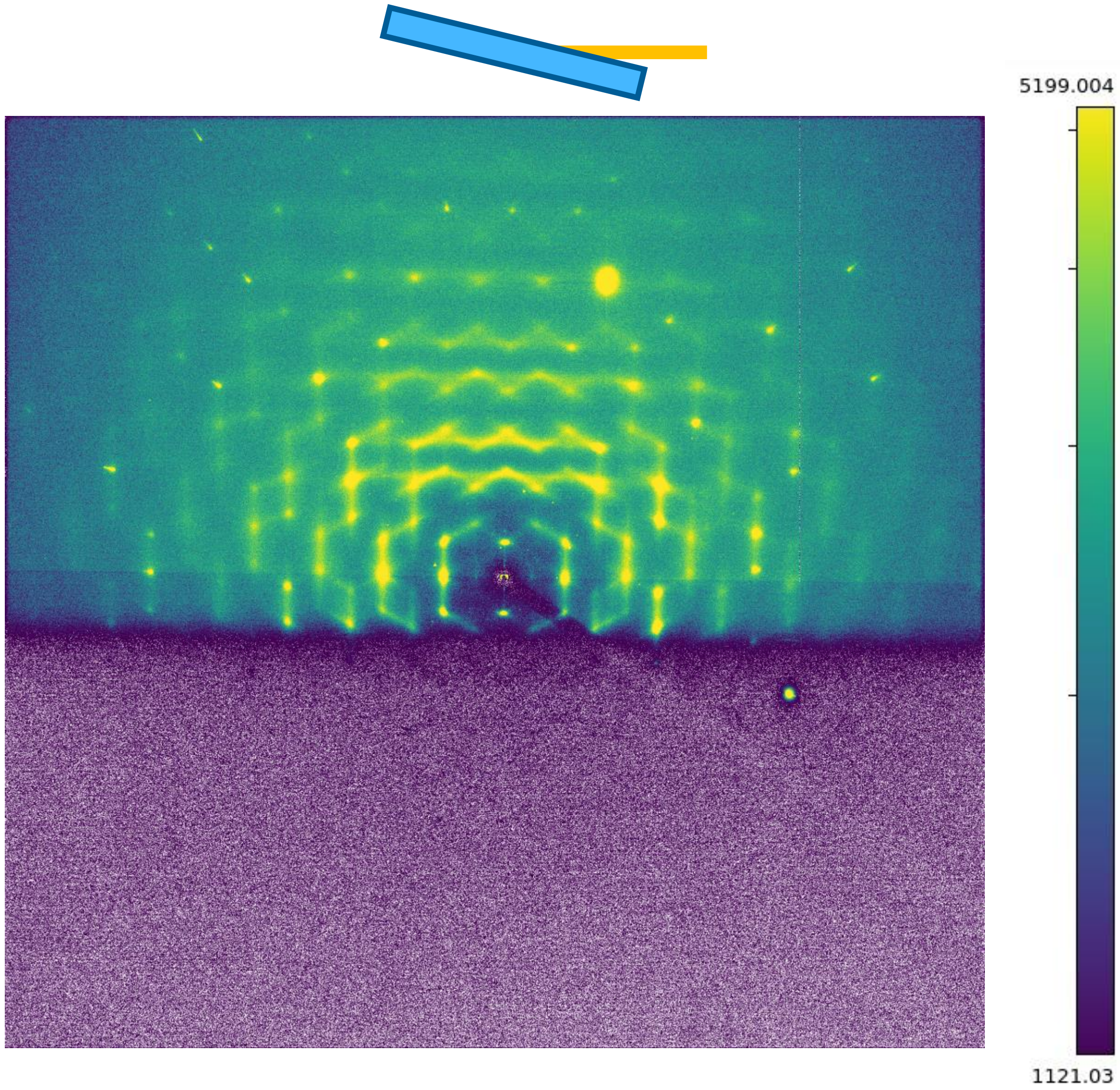
2D backgrounds: Si substrate (alone)

In the Si substrate background, Bragg and diffuse scattering differ significantly from the thin film measurement

20 m°

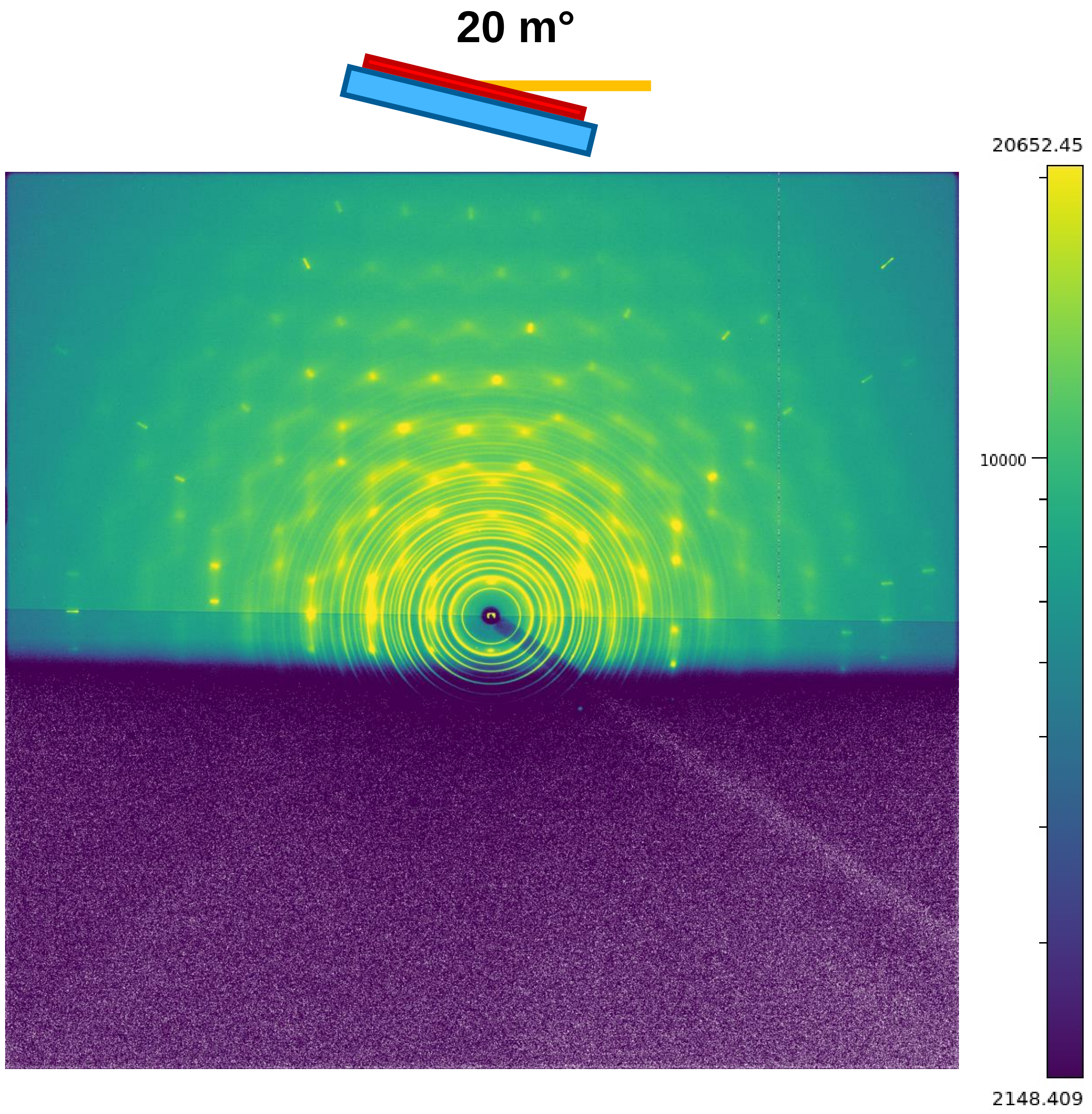


20 m°, Si substrate

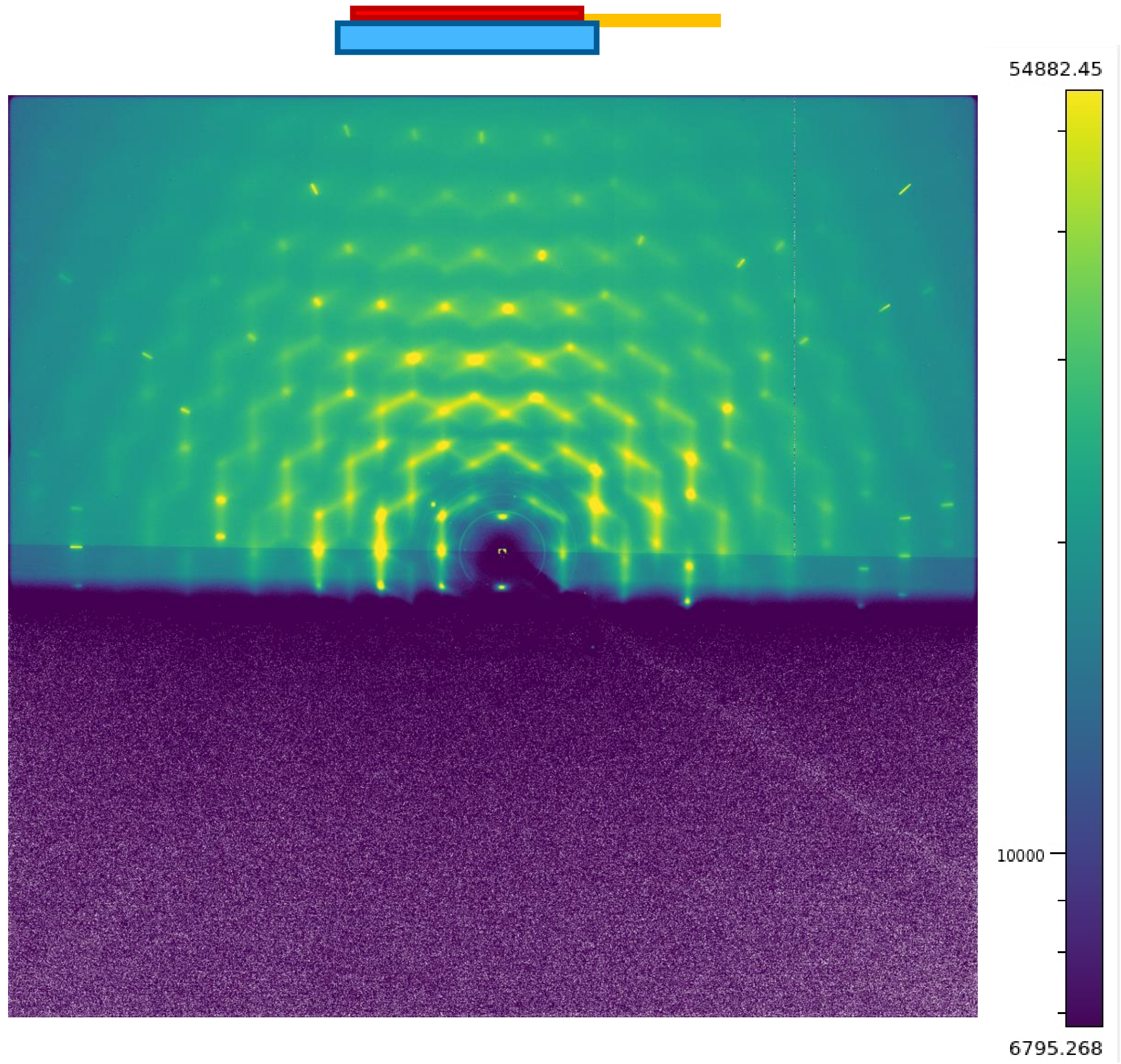


2D backgrounds: 0 m° tilt

The 0 m° background strategy leaves a background resembling much more the thin film, but with remaining thin film scattering data (ring at low q for instance)



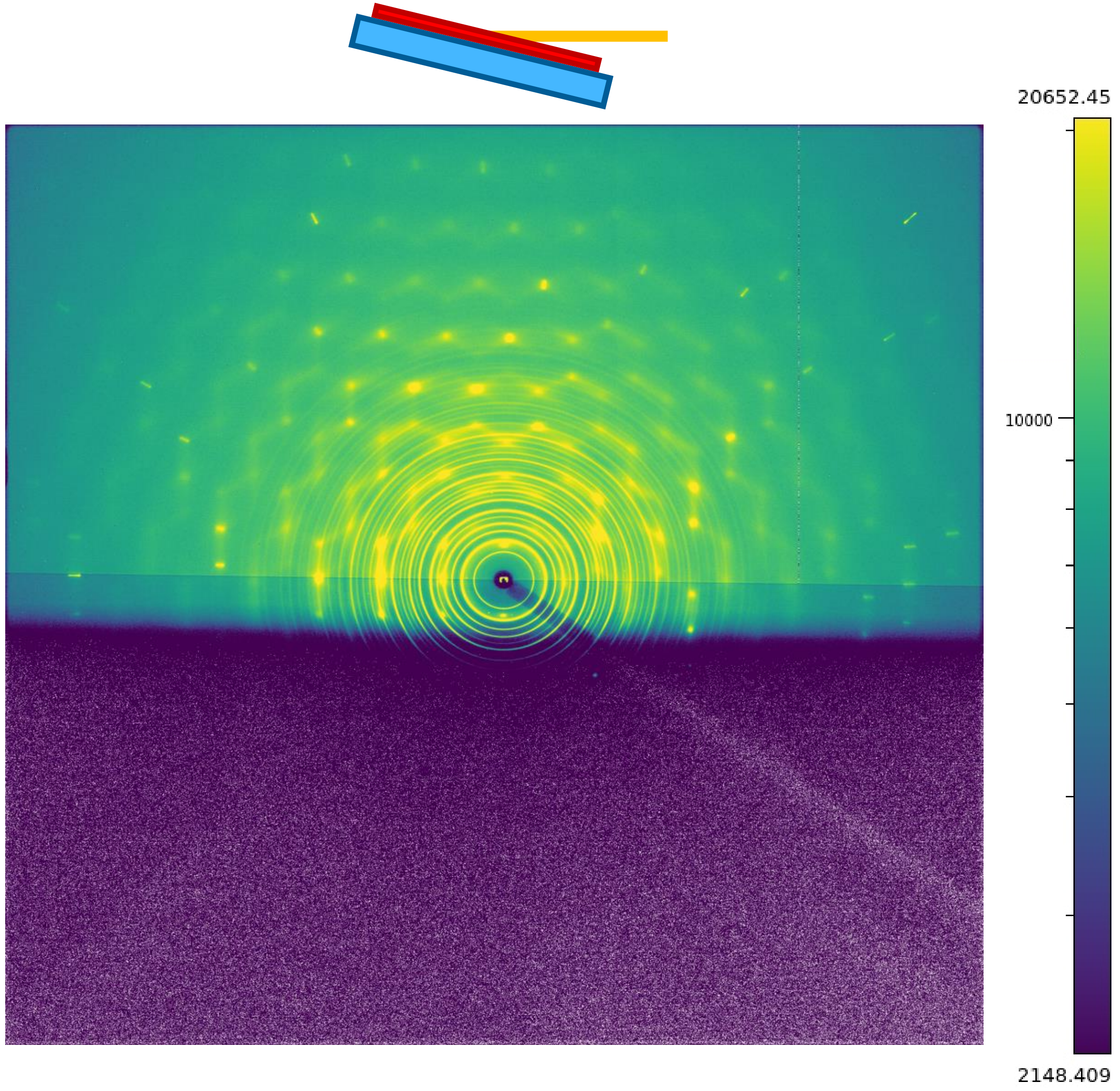
0 m°



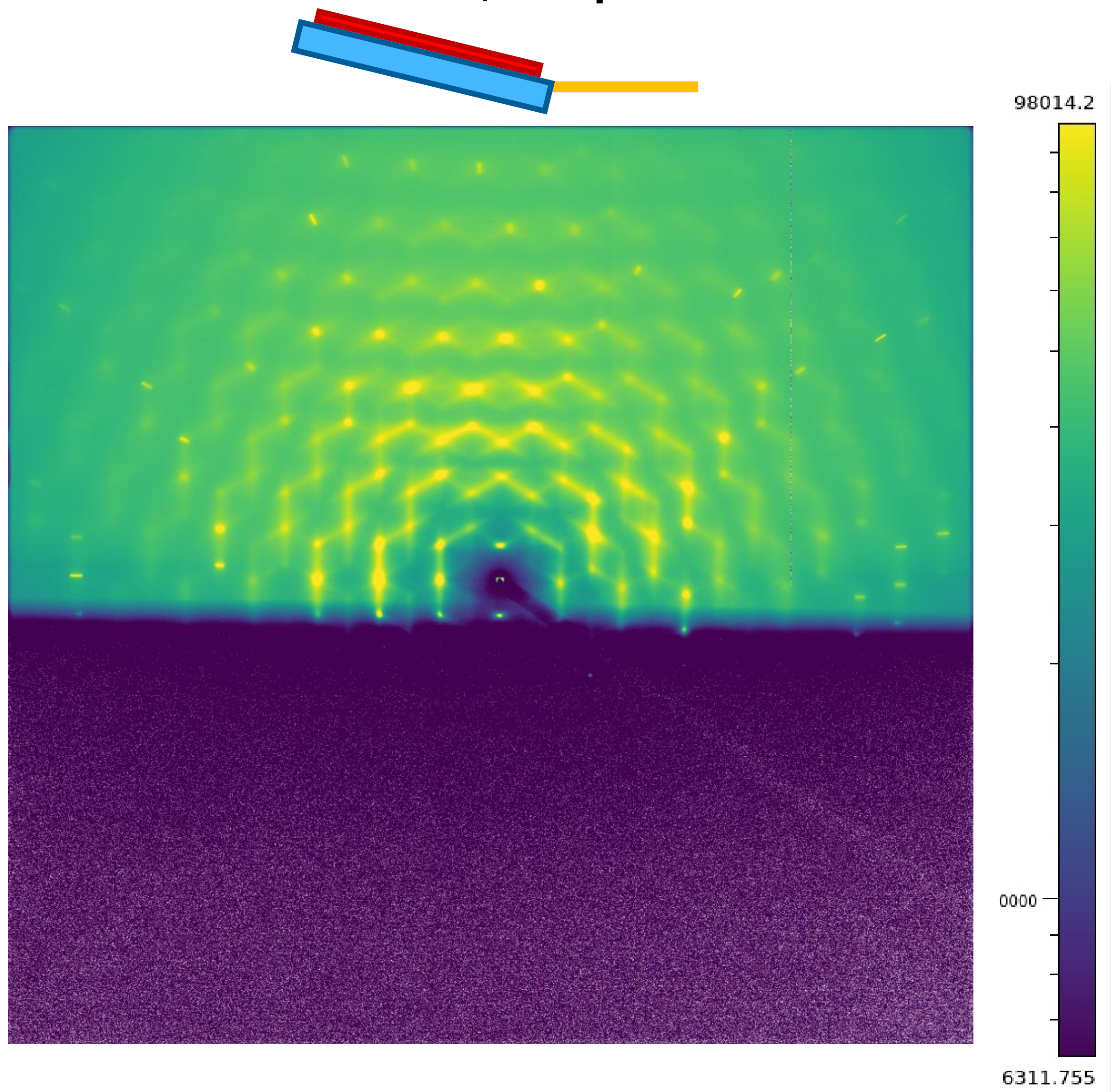
2D backgrounds: 20 m° tilt and zs lifted up

The zs up strategy gives a similar background than the previous but without thin film scattering signal

20 m°



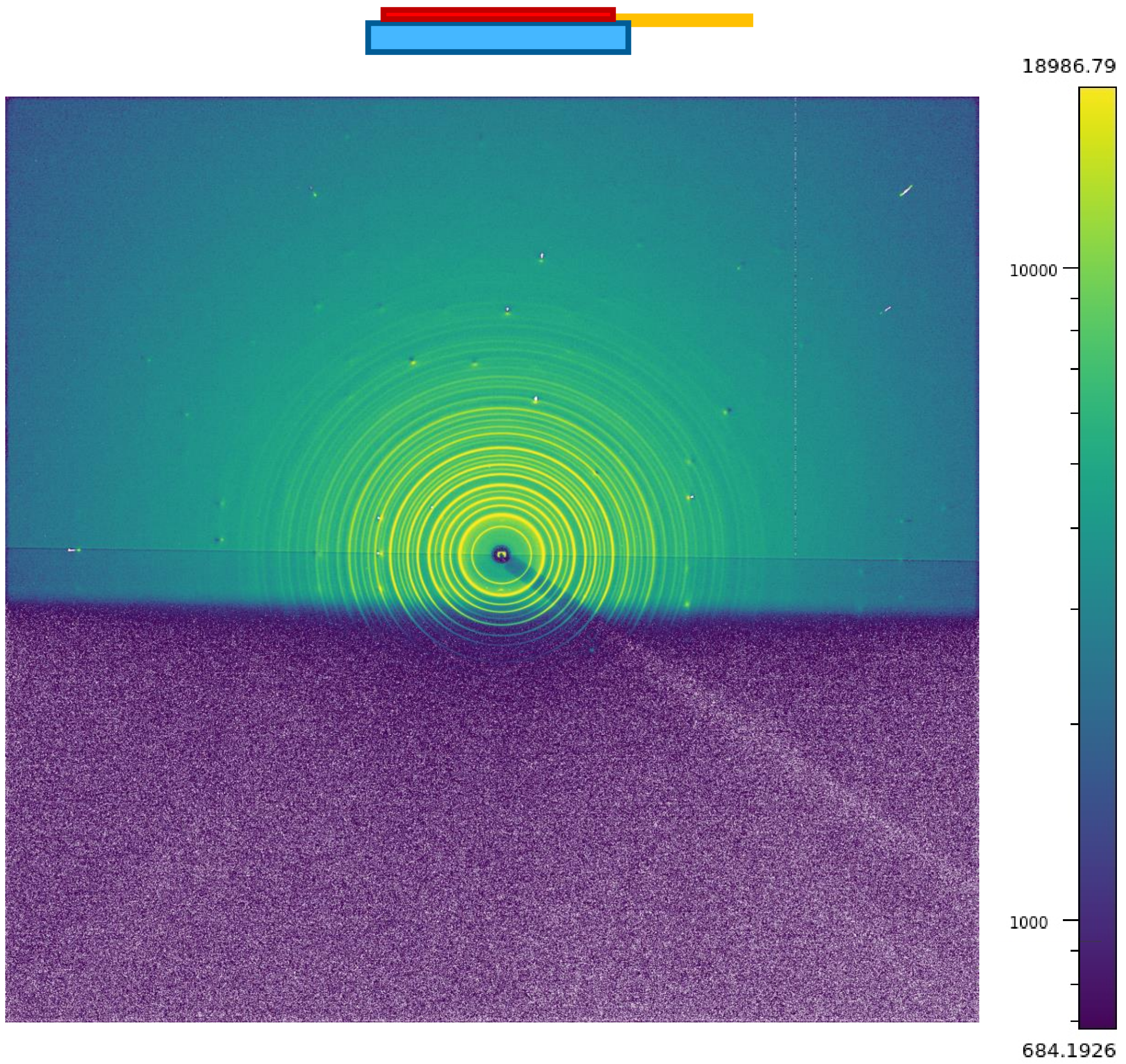
20 m°, zs up



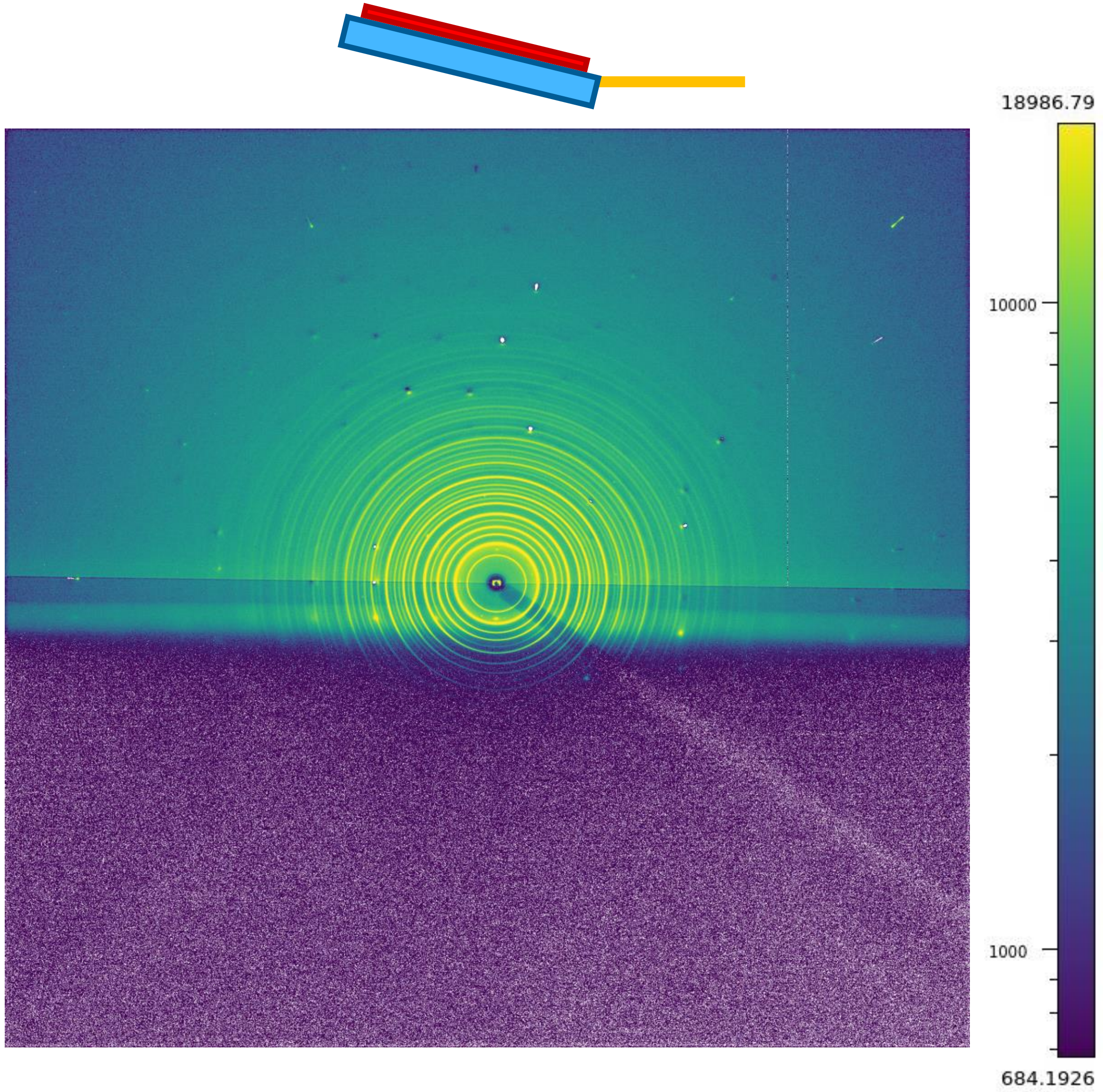
2D background subtractions

In both former cases, 2D subtractioncompletely eliminates the diffuse scattering and leaves only concentrated intensities around some Bragg positions. In both cases the subtraction required scaling

0 m°

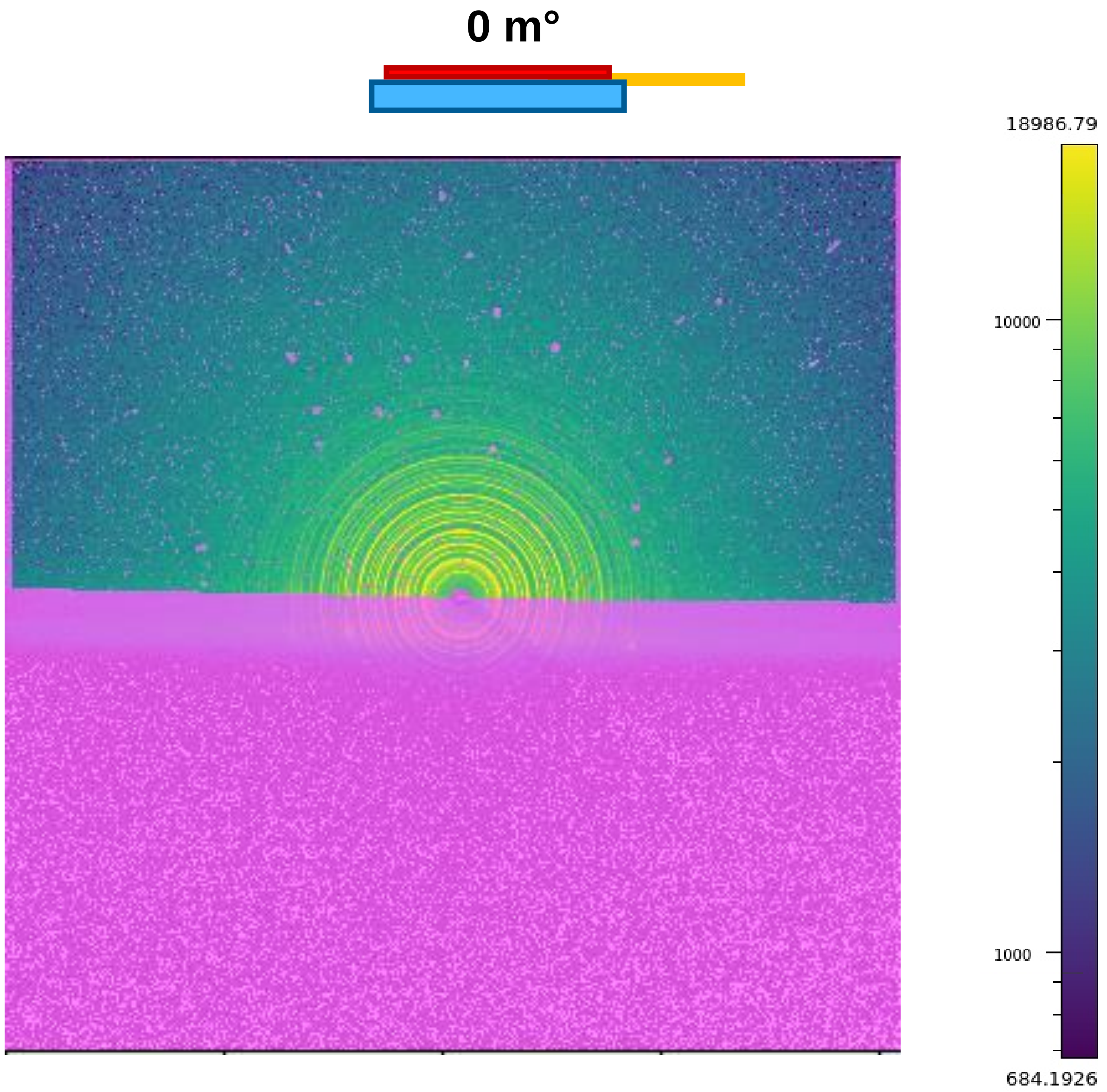


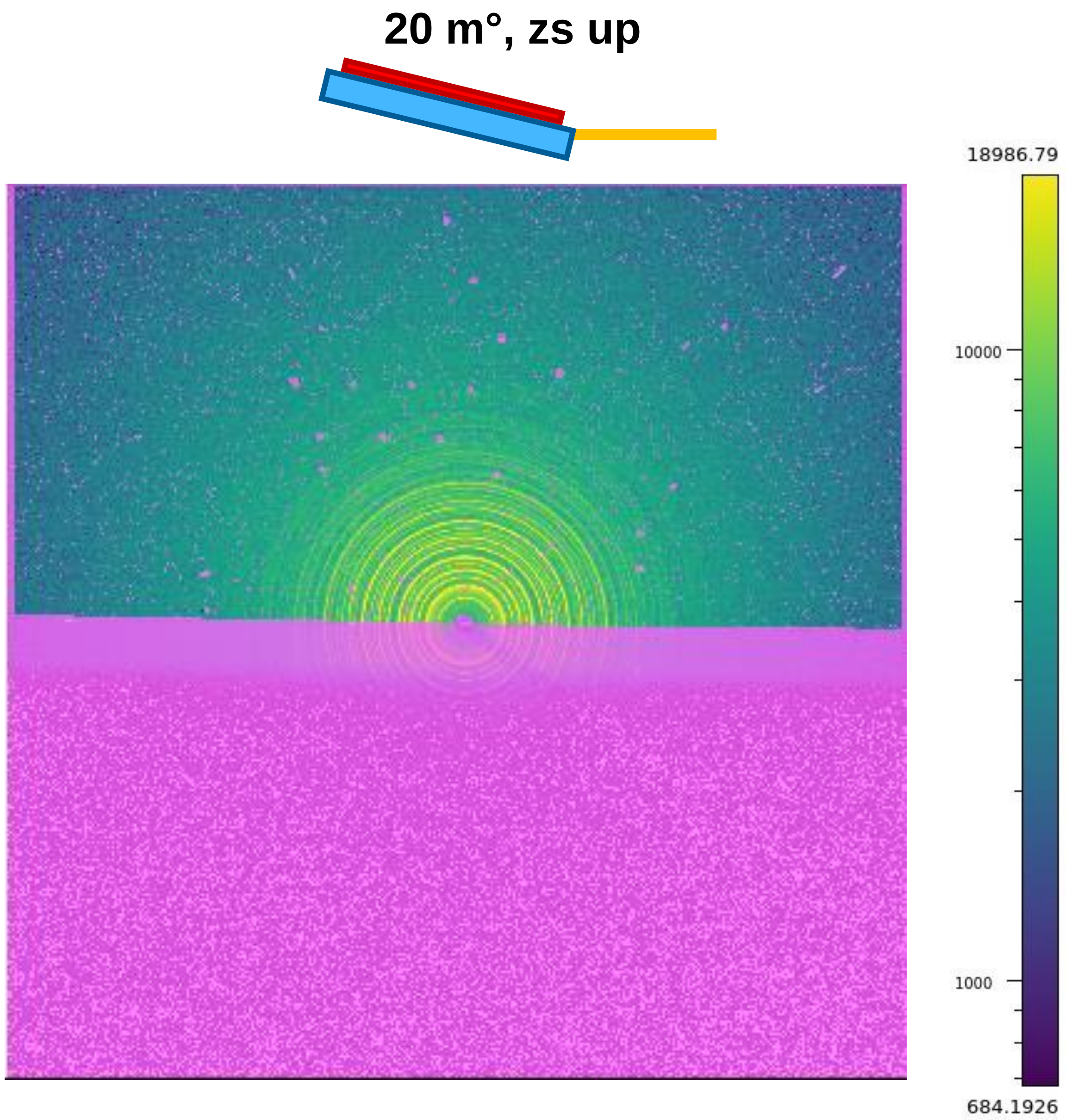
20 m°, zs up



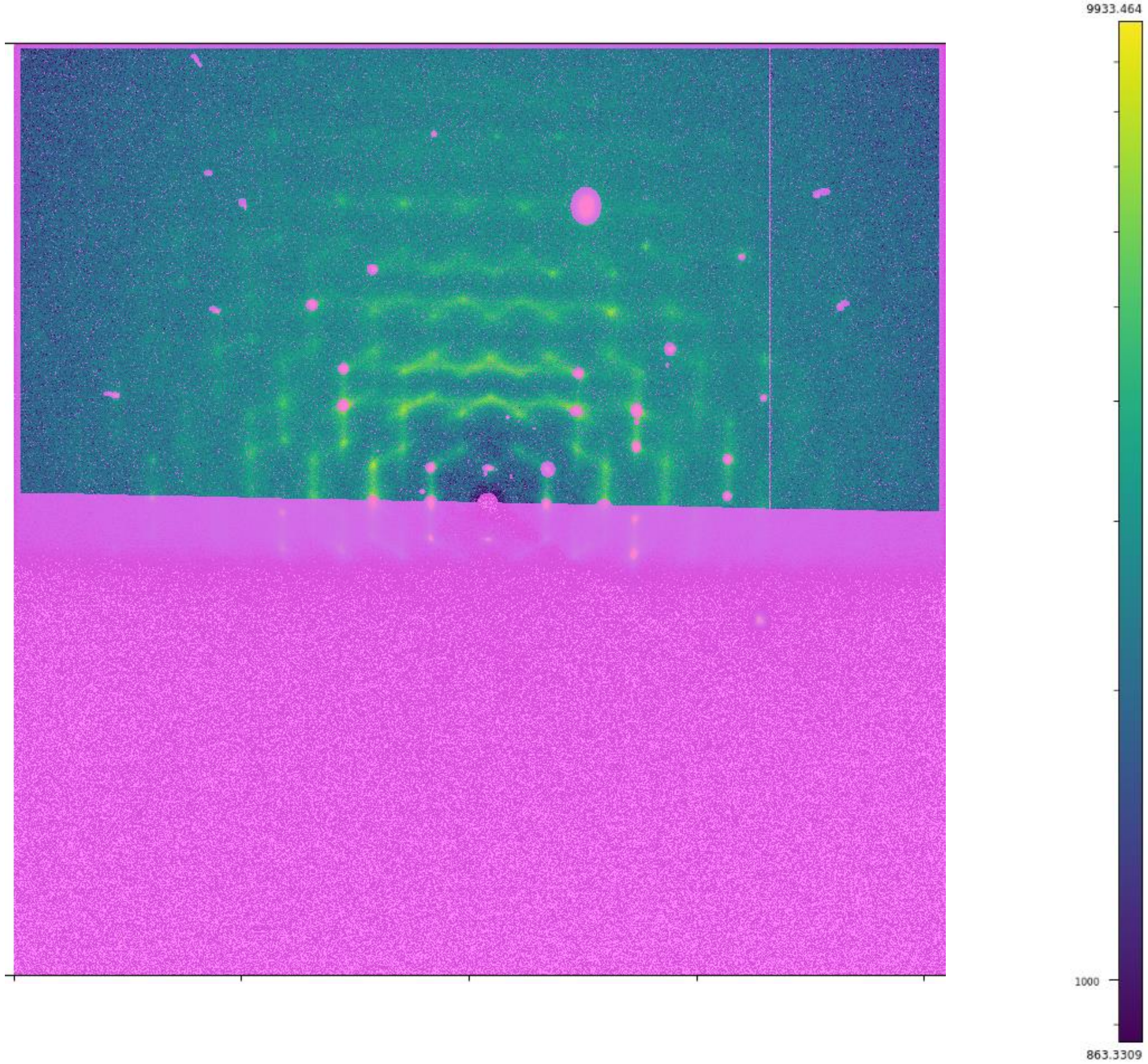
2D background subtractions and masking

These remaining background intensities can be masked

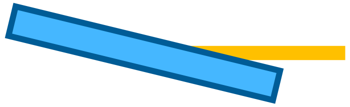




Si substrate background masking



20 m°, Si substrate

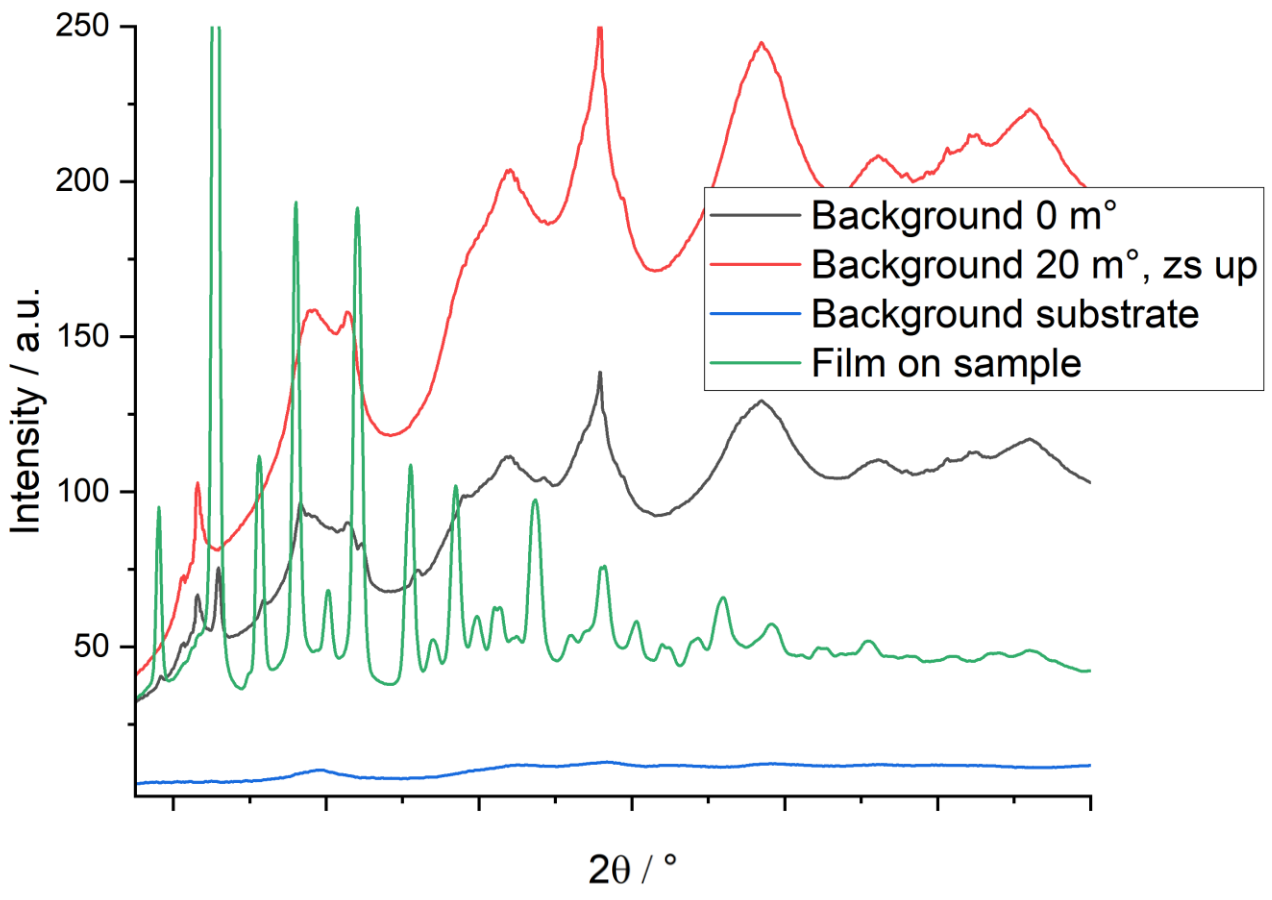


Masking of the Si substrate alone background for integration and posterior 1D subtraction. Only the regions in Bragg condition (or approximately) were masked

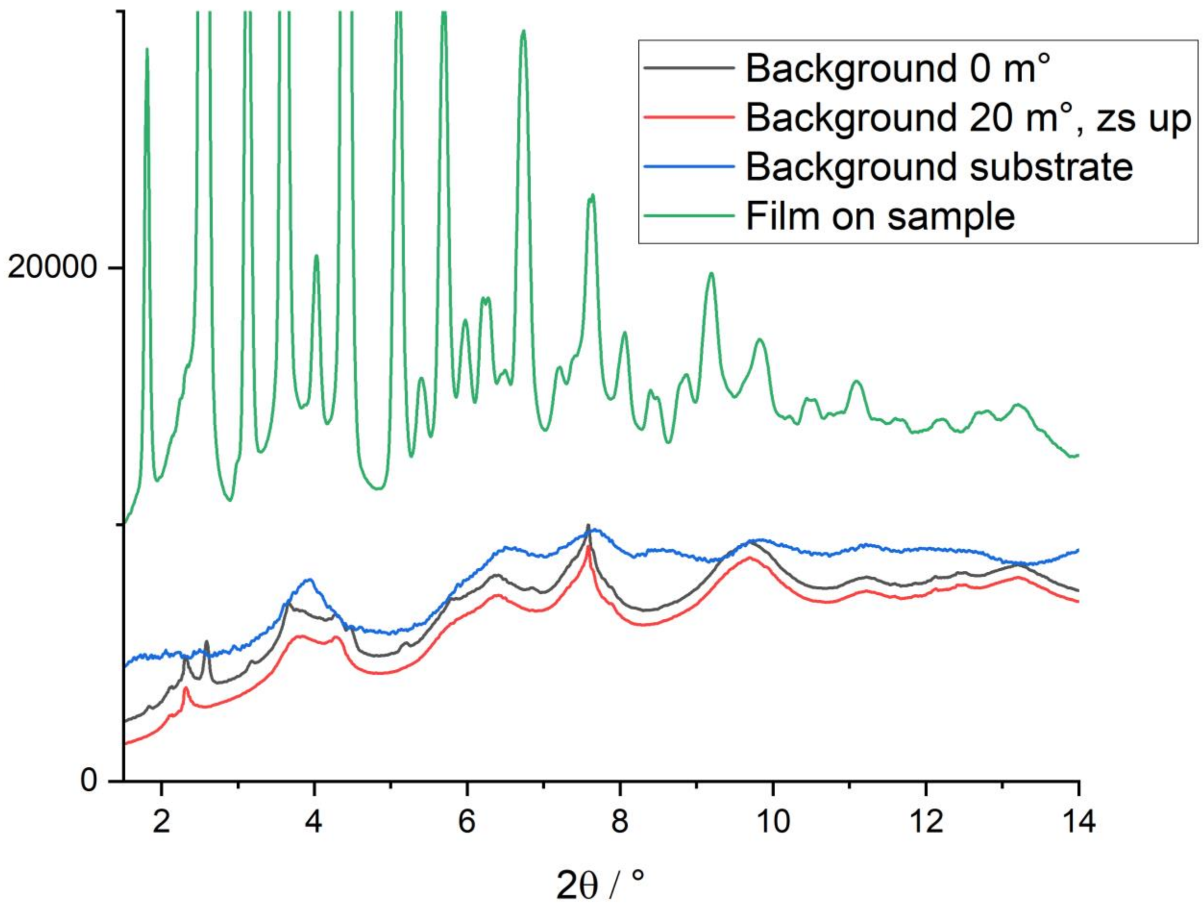
1D backgrounds

The 1D backgrounds are obtained from integrating the 2D backgrounds shown before with their corresponding masks.

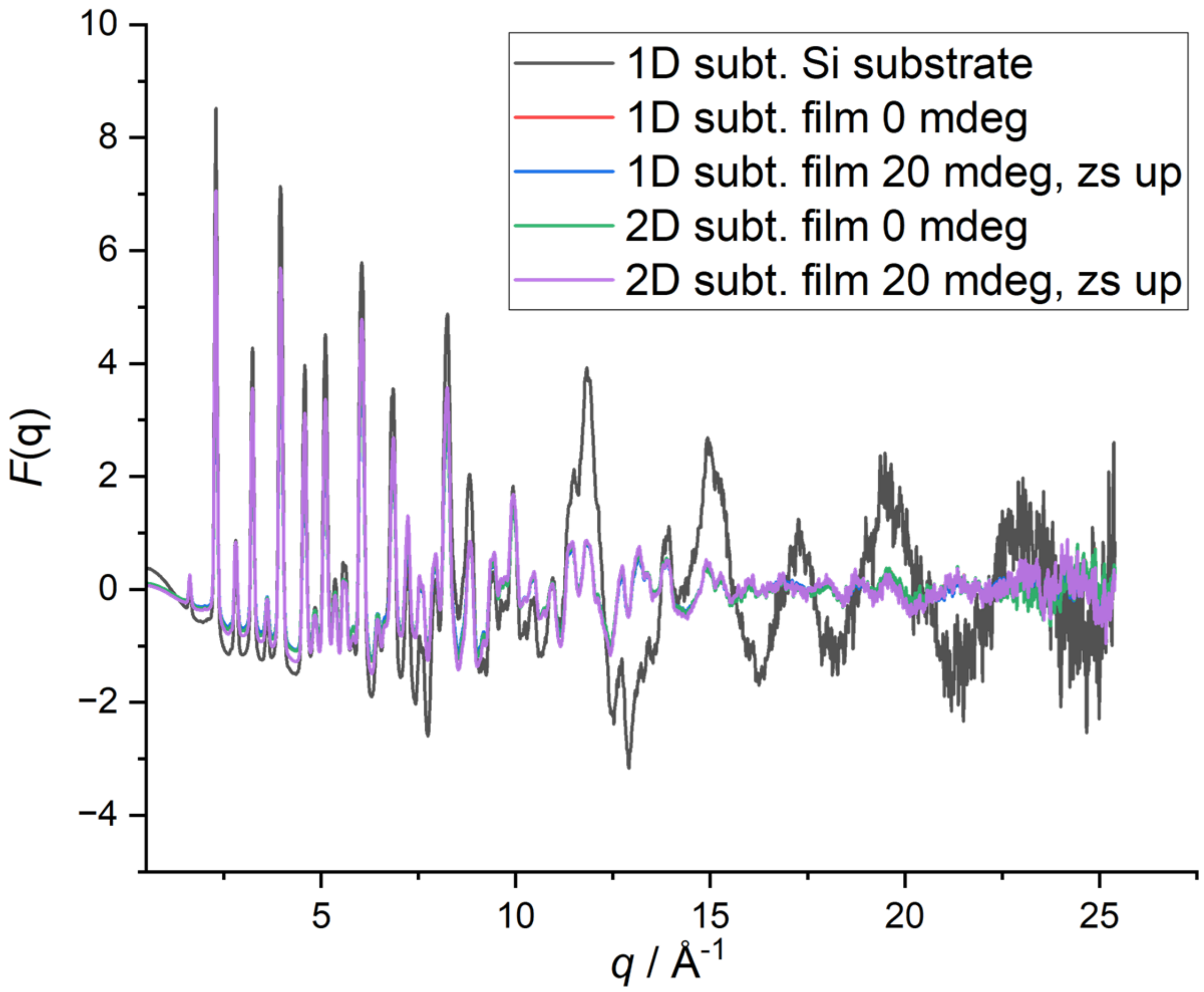
Raw integrated data normalized for exposure time



Scaled integrated backgrounds



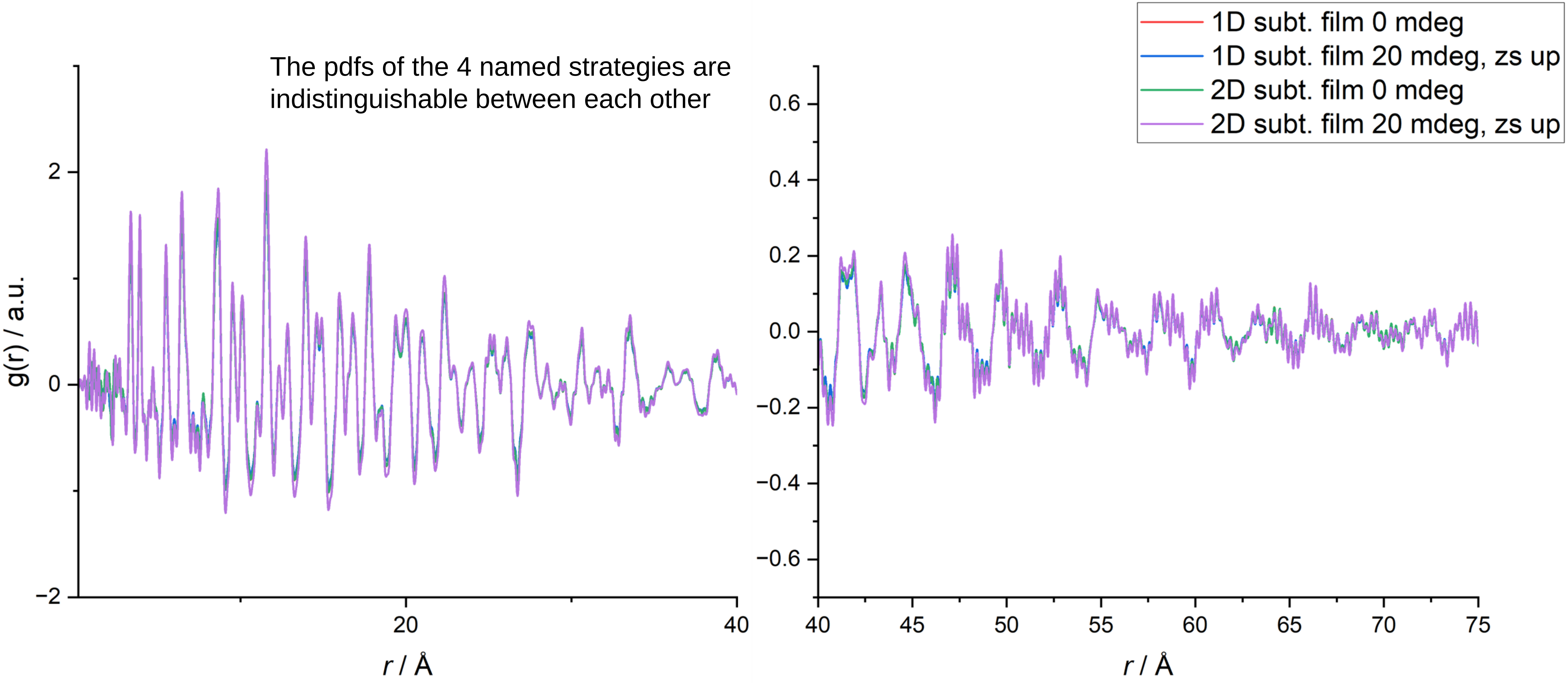
F(q) for each background subtraction



When the 5 background strategies are compared in F(q), they all seem similar except for the Si substrate strategy

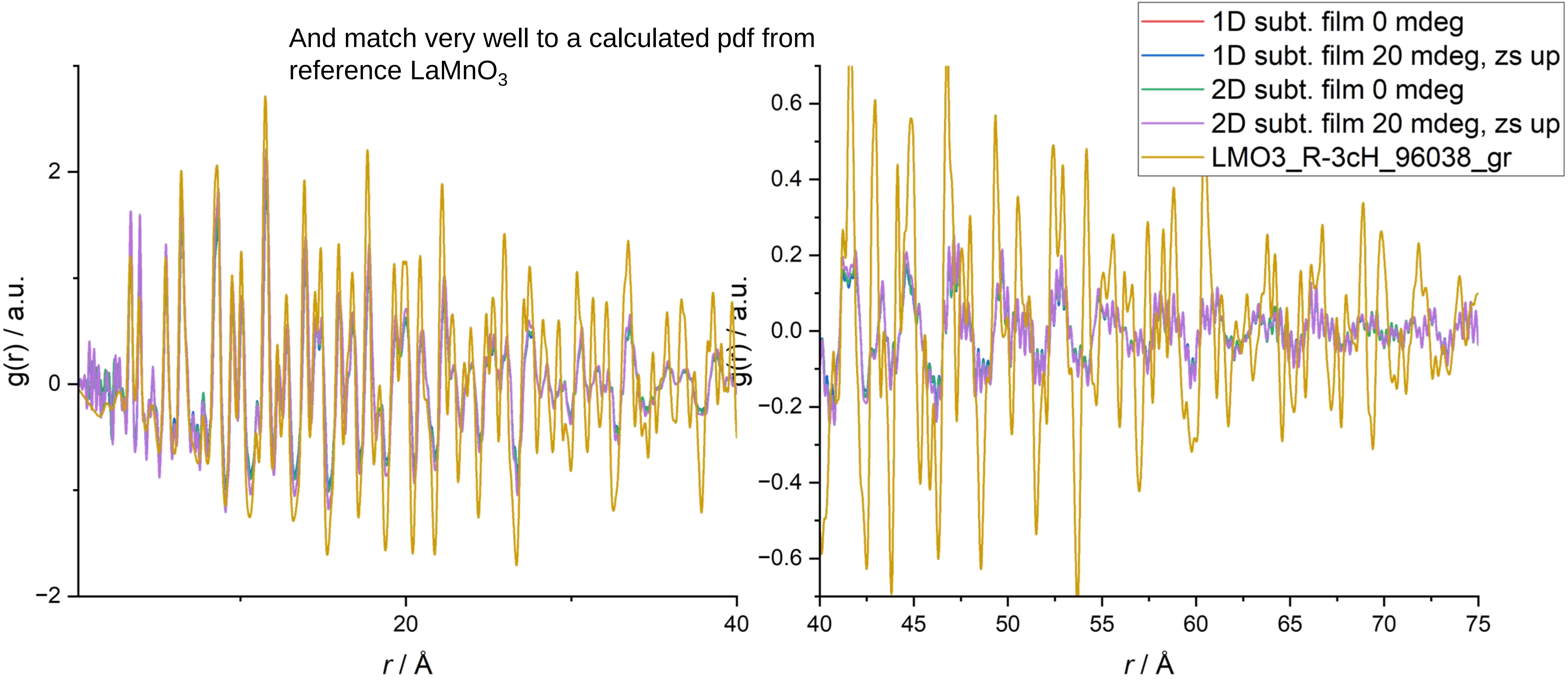
g(r) for each background subtraction

The pdfs of the 4 named strategies are indistinguishable between each other



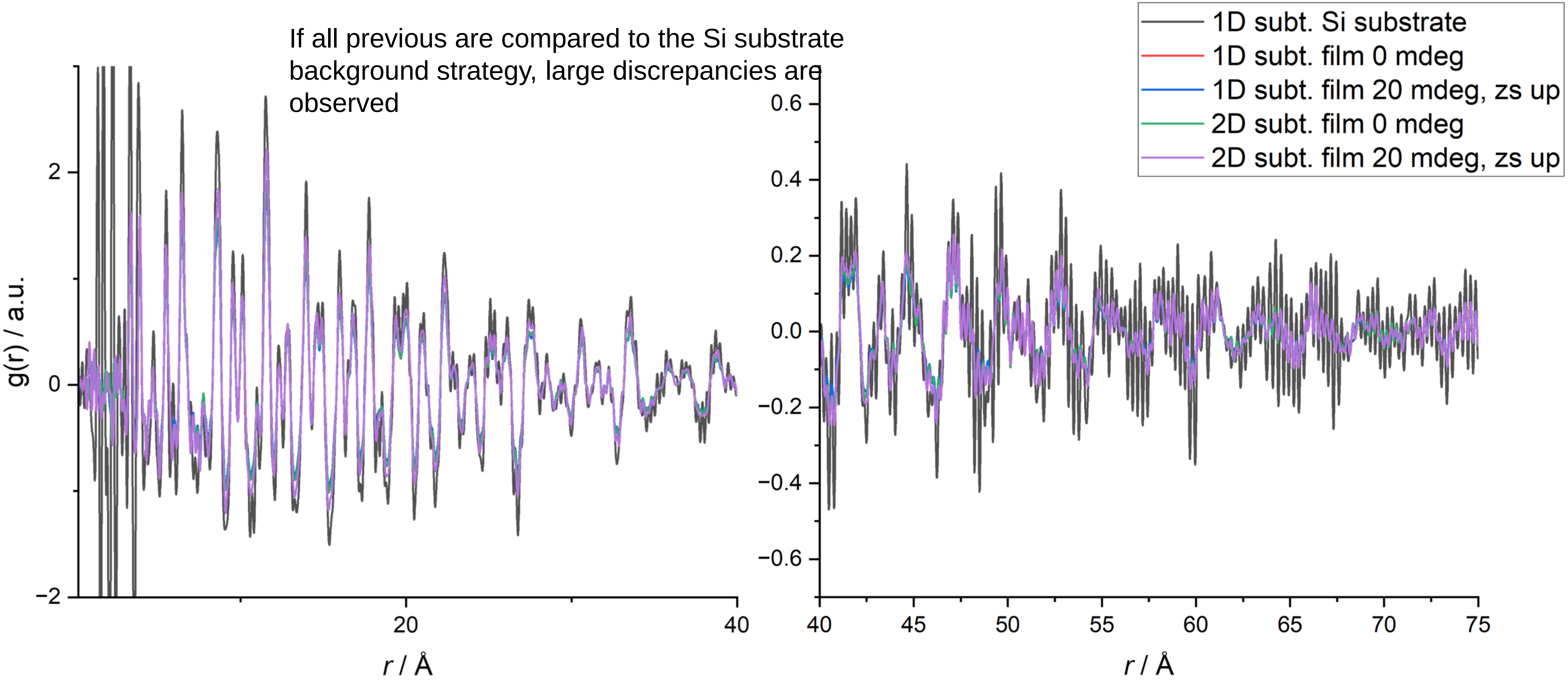
g(r) vs $LaMnO_3$ reference

And match very well to a calculated pdf from reference $LaMnO_3$



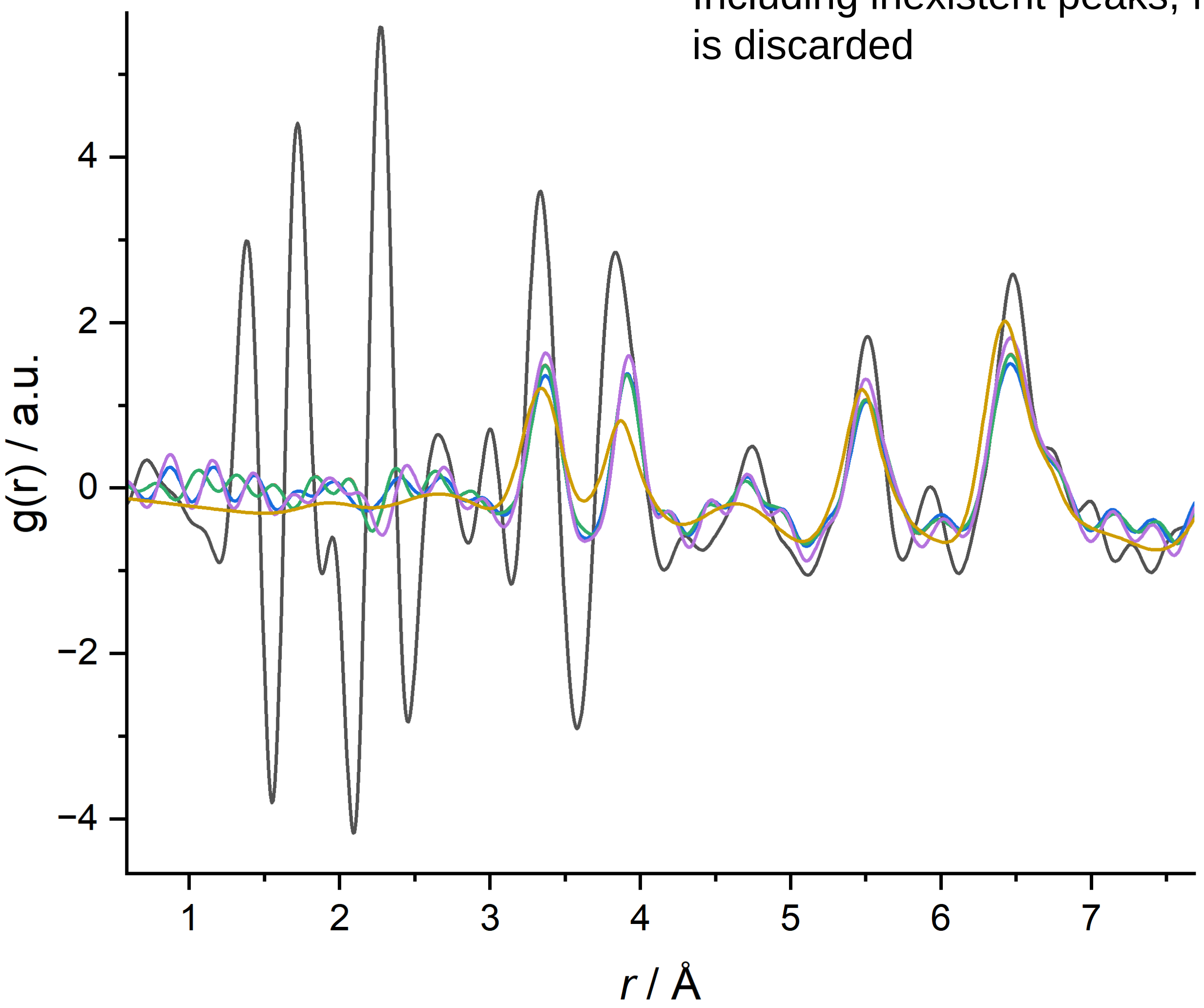
g(r) vs Si substrate (alone) background

If all previous are compared to the Si substrate background strategy, large discrepancies are observed



g(r) vs Si substrate (alone) background

Including inexistent peaks, for which this strategy is discarded



g(r) vs powder measurement

Finally, the thin film pdfs also match very well to experimental $LaMnO_3$ powder pdf measurement