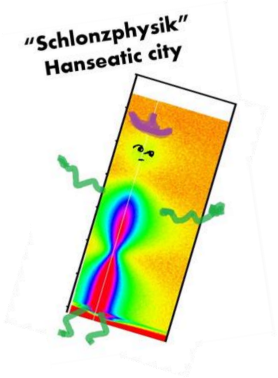
Langmuir trough GID setup at P08 of PETRA III and (pseudo) reflectivity acquisition through GISAXS



Chen Shen 沈辰
high resolution diffraction beamline P08 @ PETRA III
Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

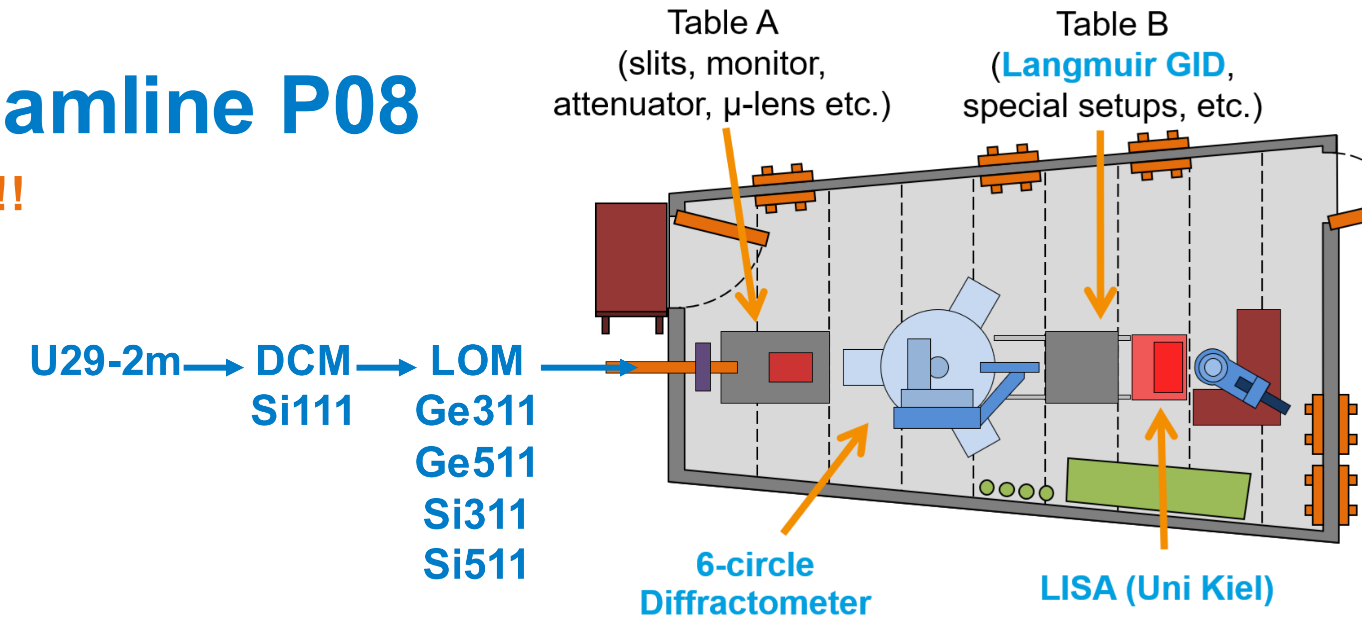
HELMHOLTZ

High resolution diffraction beamline P08

this is NOT a powder diffraction beamline!!!!!!!!!!!!

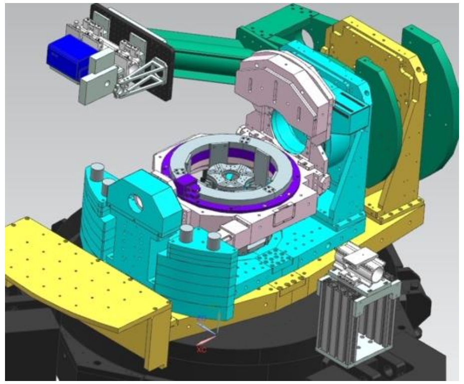
beamline parameters

energy range (keV)	:	5.4 – 29.4
beam size (HxV, µm, FWHM)	:	250 x 30 – 1000 x 500
beam divergence (HxV, µrad)	:	10 x 2
µ-mode	:	20x2 μm^2, 150x150 μrad^2
flux	:	10^{11} – 10^{12} ph/s
energy resolution (ΔE/E)	:	10^{-5}



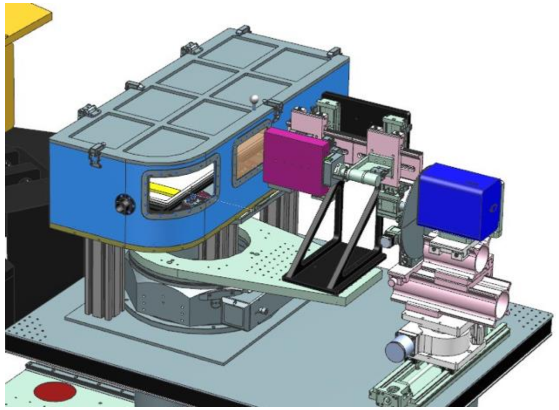
solid surface diffraction:

- high precision 6-circle diffractometer
  - XRR, high resolution XRD, CTRs, GIXS, (XRF)
  - lipid membrane chambers:
    solid-liquid chamber; hydration chambers
    *collaboration with SDU (DK) and CMU (USA)
  - stages for hard condensed matter surfaces

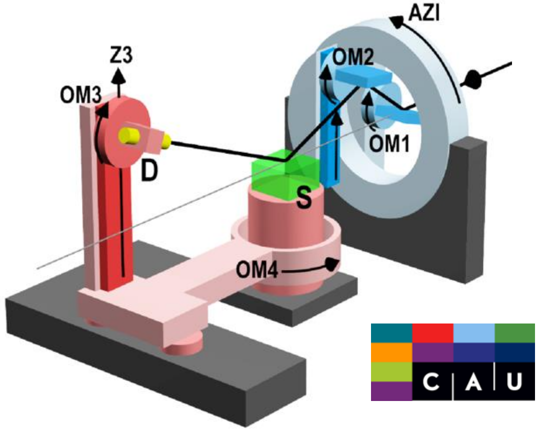


liquid surface diffraction:

- instrument 1: Langmuir trough GID setup
  - GIXS on Langmuir trough
- instrument 2: double crystal beam tilter LISA (Uni Kiel / ErUM-Pro)
  - XRR, XRNTR, (GIXS)



DOI: 10.1088/1742-6596/2380/1/012047



DOI: 10.1107/S1600577513026192

Langmuir trough GID setup at P08

ultra-low background grazing incidence instrument from Langmuir trough experiments

fixed incidence: 15keV, α_i = 0.07º (85% α_c)

GIXRD & GISAXS

lateral crystallographic / nanoscopic order

total reflection XRF

interfacial element accumulation

Langmuir trough: customized Kibron G4

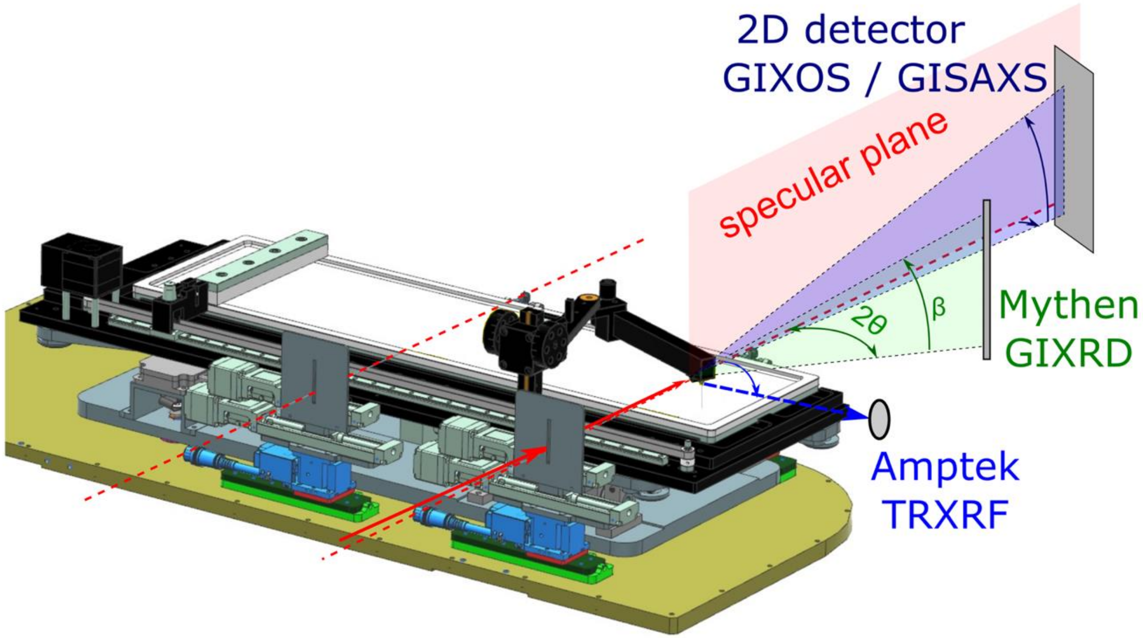
size (mm): 155x600; 155x350; 155x110; 155x60; 25x35

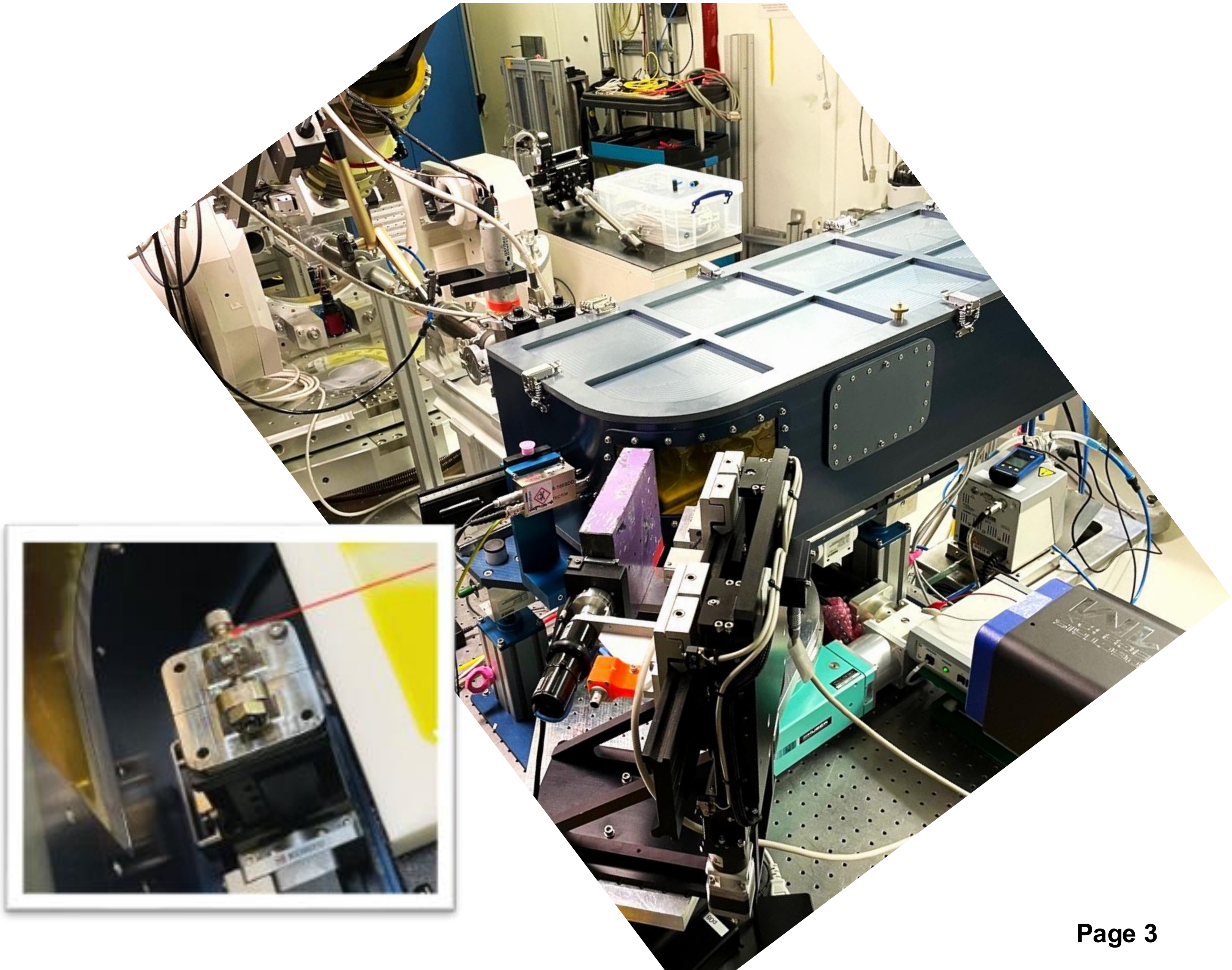
control: Tango + Sardana/SPOCK

prototype: automated subphase exchange and sample swap

advantage (vs beam tilting optics)

- better background shielding
- better signal-noise ratio from surface





Langmuir trough GID setup at P08

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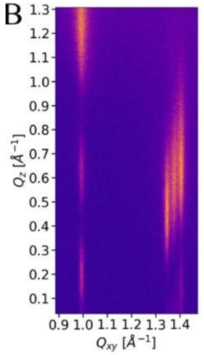
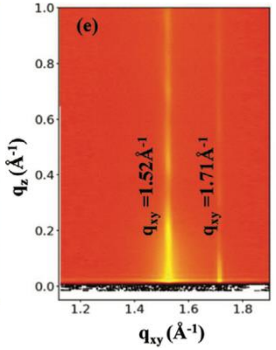
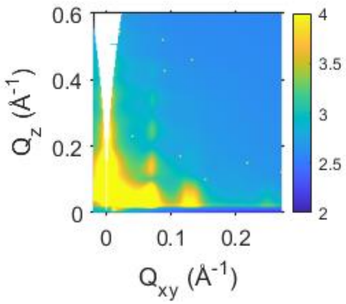


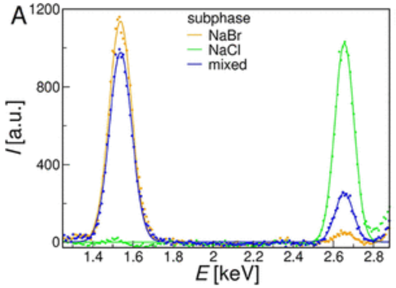
Mukhina, et. al. Langmuir 2022



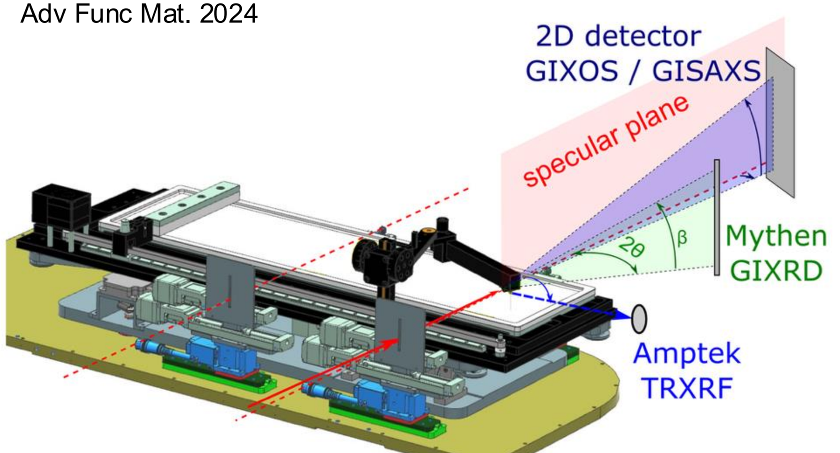
Chowdhury, et. al. Adv Func Mat. 2024



Courtesy: Jun Zhang



Mortara, et. al., Langmuir 2024

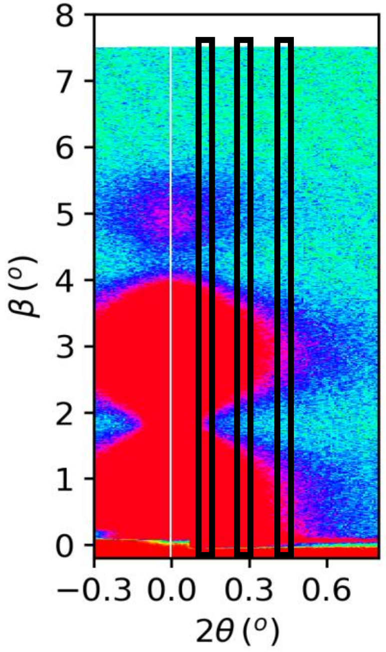


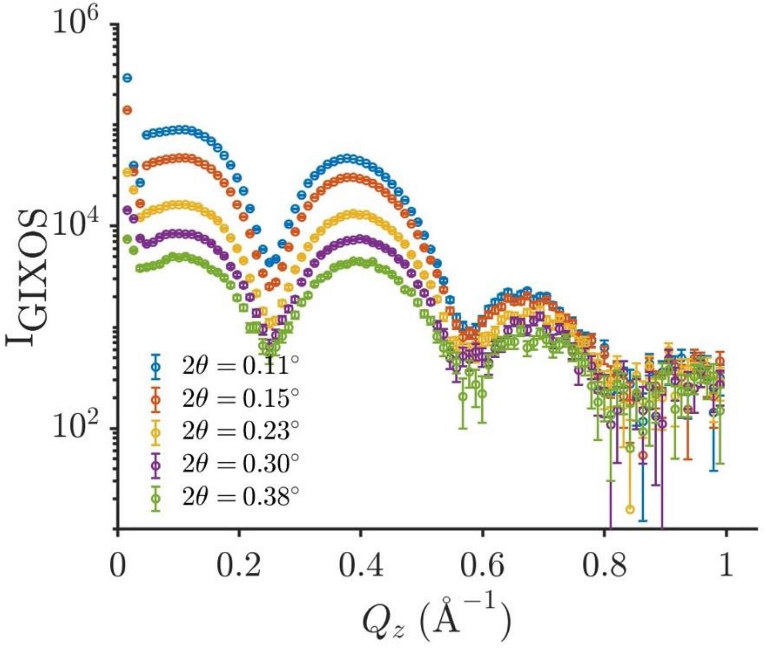
advantage (vs beam tilting optics)

- better background shielding
- better signal-noise ratio from surface

GIXOS [a-d] : (time resolved) layer structure

is GIXOS result consistent with XRR?





[a] Mora, et. al. Europhysics Letter (2004)
[b] Wiegart, et. al. Lanmguir (2005)
[c] Dai, et. al. Journal of Applied Physics (2011)
[d] Daillant, et. al. Soft Matter (2009)

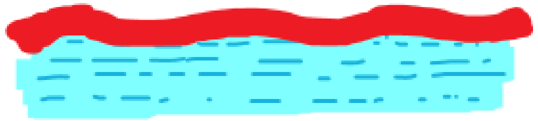
Extended Capillary Wave Model

liquid surface scattering theory development


Ben Ocko


Honghu Zhang

GISAXS measures the diffuse scattering R* around (total) specular reflection R



$$\langle h(\boldsymbol{Q}_{xy})h(-\boldsymbol{Q}_{xy})\rangle \propto \frac{k_B T}{\Delta\rho_m g+\gamma Q_{xy}^2+\kappa Q_{xy}^4}$$

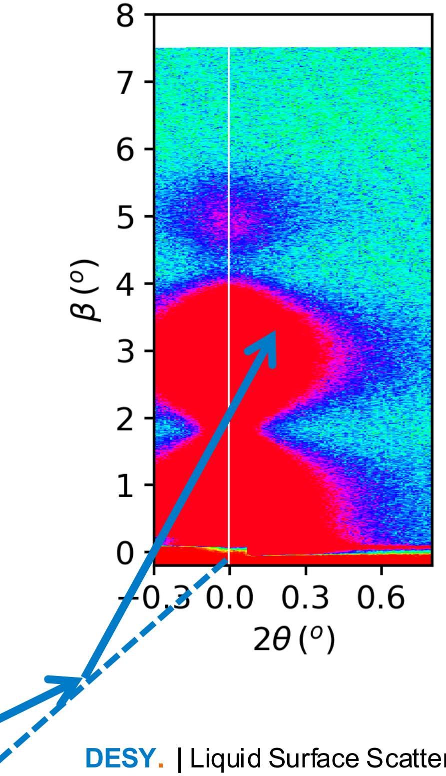
$$R, R^* = \left(\frac{Q_c}{2Q_z}\right)^4 |t_\alpha|^2 |t_\beta|^2 \cdot |\Phi(Q_z)|^2 \cdot \Psi(Q_{xy}, Q_z)$$

$$\eta = \frac{k_B T}{2\pi\gamma} Q_z^2 \qquad Q_{max} = \frac{\pi}{a_m} \qquad Q_\kappa = \sqrt{\frac{\gamma}{\kappa}}$$

$$\Psi(Q_{xy}, Q_z) = \frac{Q_z^4}{16\pi^2 \sin\alpha} \cdot \frac{\exp\left[\eta K_0\left(\frac{Q_\kappa}{Q_{max}}\right)\right]}{Q_{max}^\eta} \cdot \left[\underbrace{\frac{k_B T}{\gamma} \iint_{\Delta\beta,\Delta 2\theta} \frac{d\sin\beta\, d2\theta}{Q_{xy}^{2-\eta}}}_{\text{tension term}} + \underbrace{\frac{1}{Q_z^2} \iint_{\Delta\beta,\Delta 2\theta} C'(Q_{xy}, \eta, \kappa)\, d\sin\beta\, d2\theta}_{\text{stiffness term}}\right]$$

tension term
(standard capillary wave model)

stiffness term



Extended Capillary Wave Model

liquid surface scattering theory development

GISAXS measures the diffuse scattering R* around (total) specular reflection R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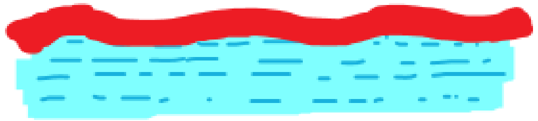

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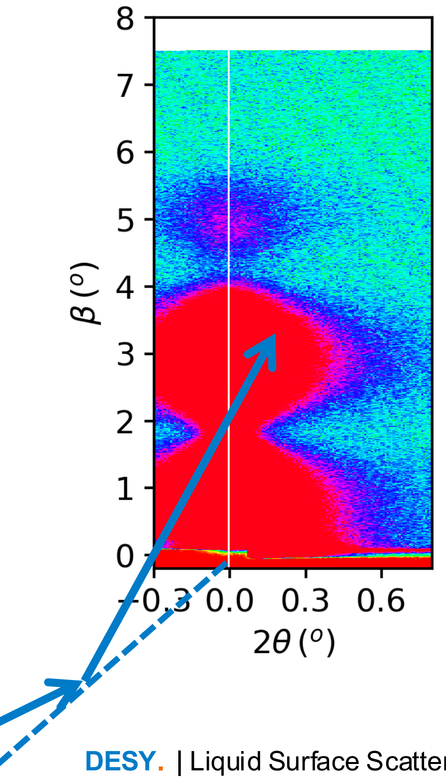
$$\eta = \frac{k_BT}{2\pi\gamma}Q_z^2 \qquad Q_{max} = \frac{\pi}{a_m} \qquad Q_\kappa = \sqrt{\frac{\gamma}{\kappa}}$$

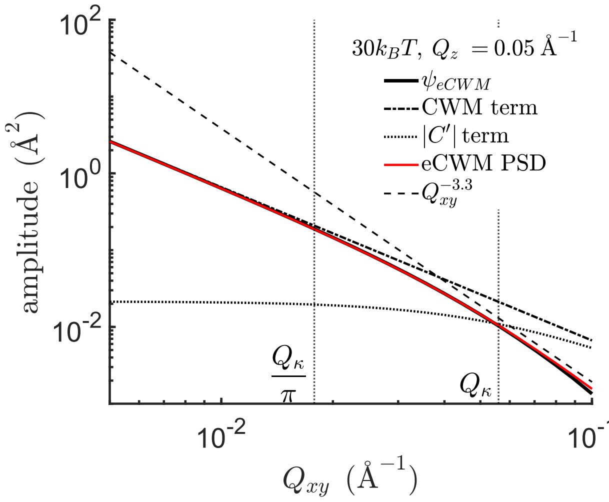
κ = 0
(standard capillary wave)

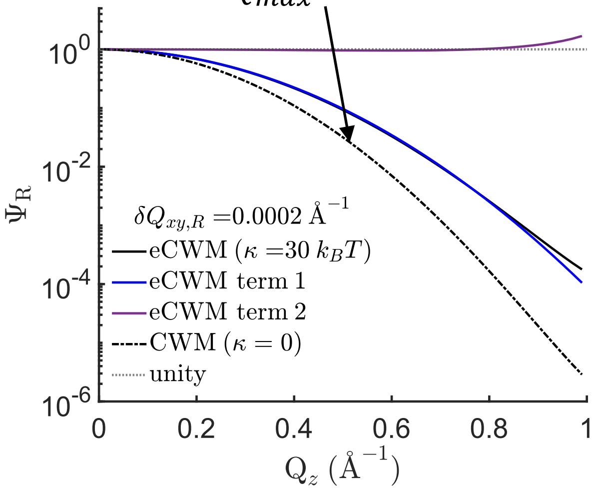
$$\Psi(Q_{xy}, Q_z) = \frac{Q_z^4}{16\pi^2 \sin\alpha} \cdot \frac{1}{Q_{max}^\eta} \cdot \frac{k_BT}{\gamma} \iint_{\Delta\beta,\Delta 2\theta} \frac{d\sin\beta\, d2\theta}{Q_{xy}^{2-\eta}}$$

$$R = R_F|\Phi(Q_z)|^2 \left(\frac{\delta Q_{xy,R}}{Q_{max}}\right)^\eta$$

$$\Psi_R = \left(\frac{\delta Q_{xy,R}}{Q_{max}}\right)^\eta \equiv \exp(-Q_z^2\sigma_R^2)$$







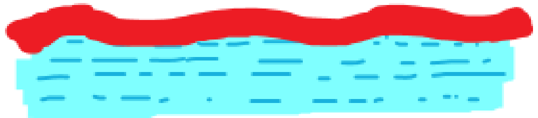
Extended Capillary Wave Model

liquid surface scattering theory development

GISAXS measures the diffuse scattering R* around (total) specular reflection R


Ben Ocko


Honghu Zhang



$$\langle h(\boldsymbol{Q}_{xy})h(-\boldsymbol{Q}_{xy})\rangle \propto \frac{k_BT}{\Delta\rho_m g+\gamma Q_{xy}^2+\kappa Q_{xy}^4}$$

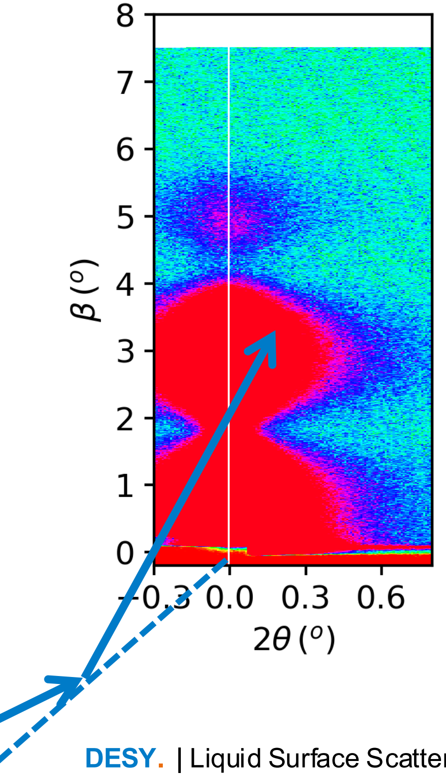
$$R, R^* = \left(\frac{Q_c}{2Q_z}\right)^4 |t_\alpha|^2|t_\beta|^2 \cdot |\Phi(Q_z)|^2 \cdot \Psi(Q_{xy}, Q_z)$$

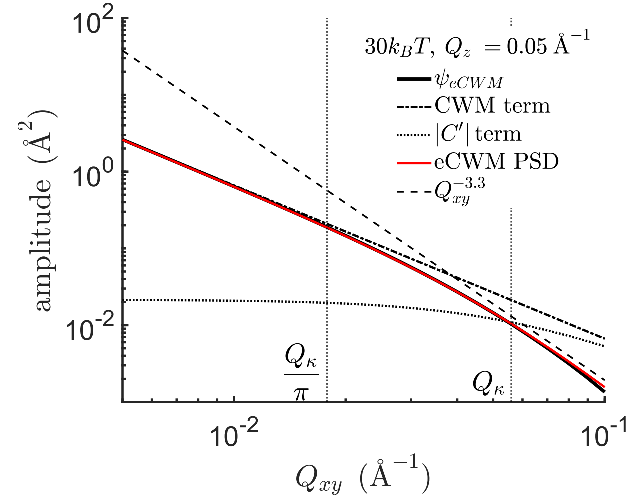
$$\eta = \frac{k_BT}{2\pi\gamma}Q_z^2 \quad Q_{max} = \frac{\pi}{a_m} \quad Q_\kappa = \sqrt{\frac{\gamma}{\kappa}}$$

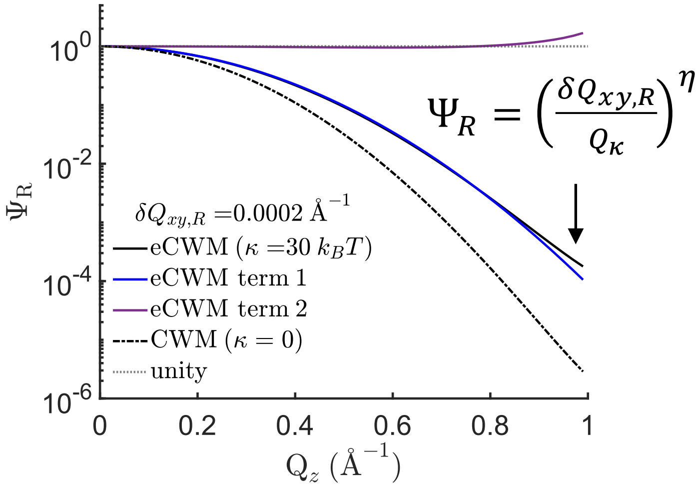
not too small κ
($Q_\kappa < 0.5Q_{max}$)

$$\Psi(Q_{xy}, Q_z) = \frac{Q_z^4}{16\pi^2 \sin\alpha} \cdot \frac{1}{Q_\kappa^\eta} \cdot \left[\frac{k_BT}{\gamma}\iint_{\Delta\beta,\Delta 2\theta}\frac{d\sin\beta\, d2\theta}{Q_{xy}^{2-\eta}} + \frac{1}{Q_z^2}\iint_{\Delta\beta,\Delta 2\theta} C'(Q_{xy},\eta,\kappa)\, d\sin\beta\, d2\theta\right]$$

$$R = R_F|\Phi(Q_z)|^2\left(\frac{\delta Q_{xy,R}}{Q_\kappa}\right)^\eta$$

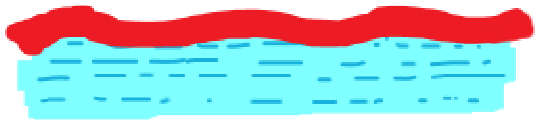






Extended Capillary Wave Model

experimental result



Ben Ocko

Honghu Zhang

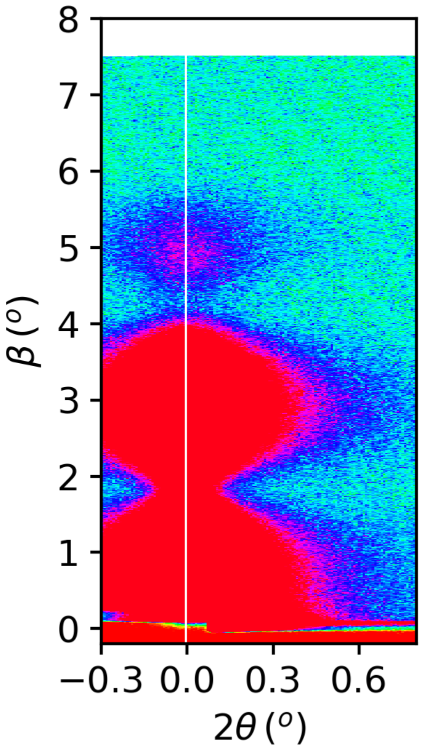
$$R, R^* = \left(\frac{Q_c}{2Q_z}\right)^4 |t_\alpha|^2 |t_\beta|^2 \cdot |\Phi(Q_z)|^2 \cdot \Psi(Q_{xy}, Q_z)$$

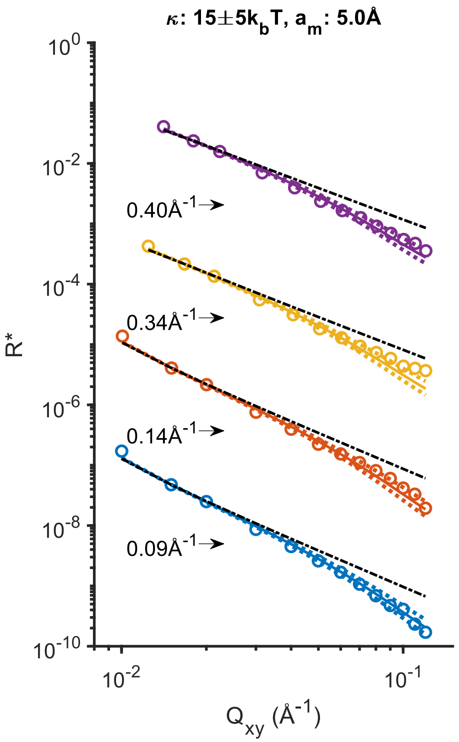
$$\Psi(Q_{xy}, Q_z) = \frac{Q_z^4}{16\pi^2 \sin\alpha} \cdot \frac{\exp\left[\eta K_0\left(\frac{Q_\kappa}{Q_{max}}\right)\right]}{Q_{max}^\eta} \cdot \left[\underbrace{\frac{k_B T}{\gamma} \iint_{\Delta\beta,\Delta 2\theta} \frac{d\sin\beta\, d2\theta}{Q_{xy}^{2-\eta}}}_{\text{tension term}} + \underbrace{\frac{1}{Q_z^2} \iint_{\Delta\beta,\Delta 2\theta} C'(Q_{xy}, \eta, \kappa)\, d\sin\beta\, d2\theta}_{\text{stiffness term}}\right]$$

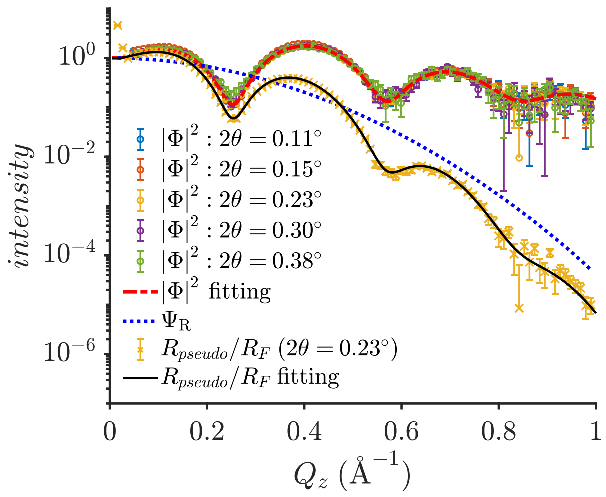
tension term (standard capillary wave model)

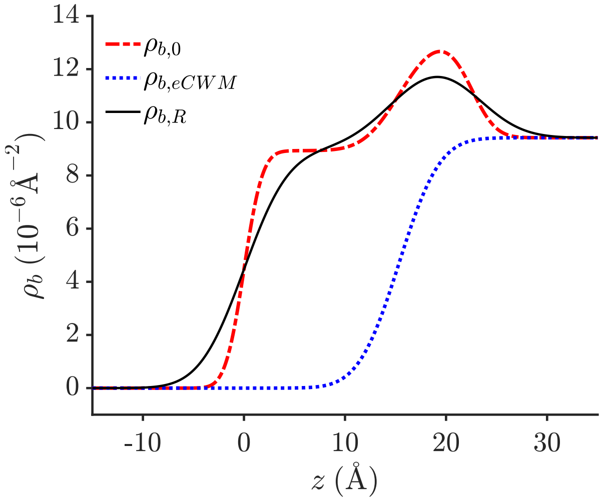
stiffness term

$$R^*(Q_{xy})\Big|_{Q_z} \rightarrow \kappa \qquad |\Phi(Q_z)|^2 = \frac{R^*(Q_{xy}, Q_z)}{\Psi(Q_{xy}, Q_z)} \cdot \frac{(2Q_z)^4}{Q_c^4 |t_\alpha|^2 |t_\beta|^2}$$









Pseudo reflectivity

obtaining reflectivity via GISAXS using eCWM

Ben Ocko

Honghu Zhang

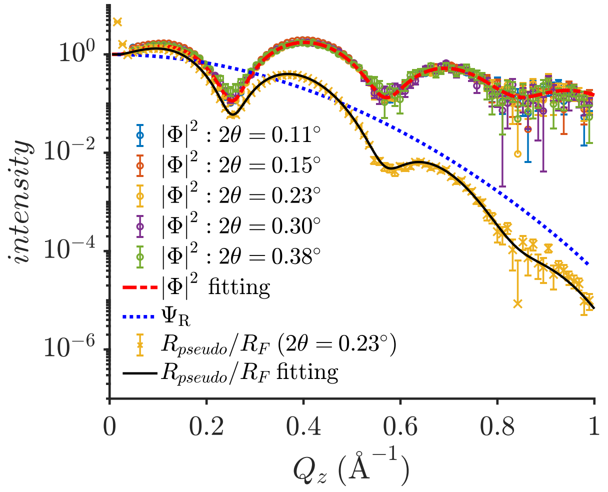
$$R, R^* = \left(\frac{Q_c}{2Q_z}\right)^4 |t_\alpha|^2 |t_\beta|^2 \cdot |\Phi(Q_z)|^2 \cdot \Psi(Q_{xy}, Q_z)$$

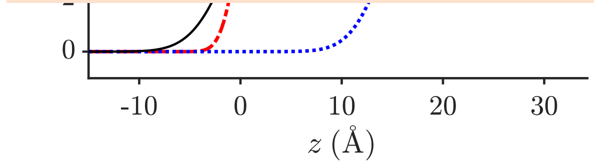
$$r(Q_z, Q_{xy}) = \frac{R^*(Q_z, Q_{xy})}{R(Q_z)} = \frac{\Psi}{\Psi(Q}$$

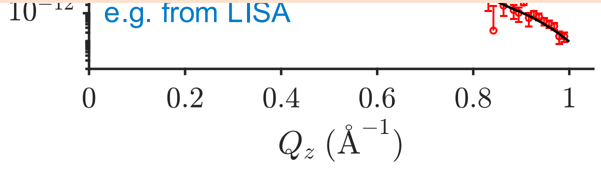
$$R_{pseudo}(Q_z) = \frac{R^*(Q_z, Q_{xy})}{r(Q_z, Q_{xy})}$$

GIXOS - pXRR vs specular XRR:
- **same curve !!!**
- higher real space resolution by acquiring high Q data
- quicker measurements and less radiation damage
- get the intrinsic structure
- get the bending modulus
- users can still use reflectivity analysis software

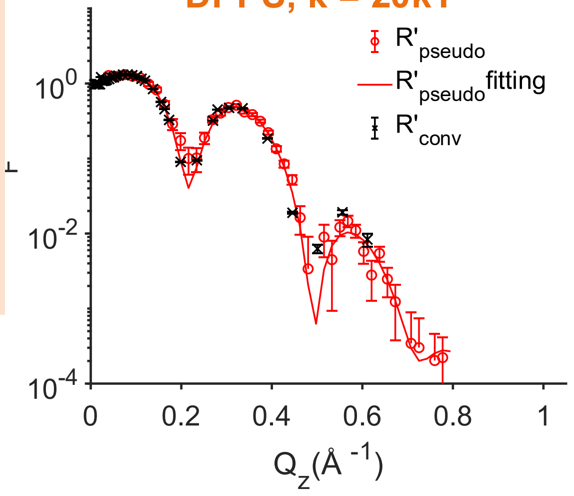
processed dataset from Langmuir trough GID setup
- $\kappa, \sigma_{R.eCWM}$
- $|\Phi(Q_z)|^2$
- $R_F|\Phi(Q_z)|^2$
- R_{pseudo}
- Ψ_R







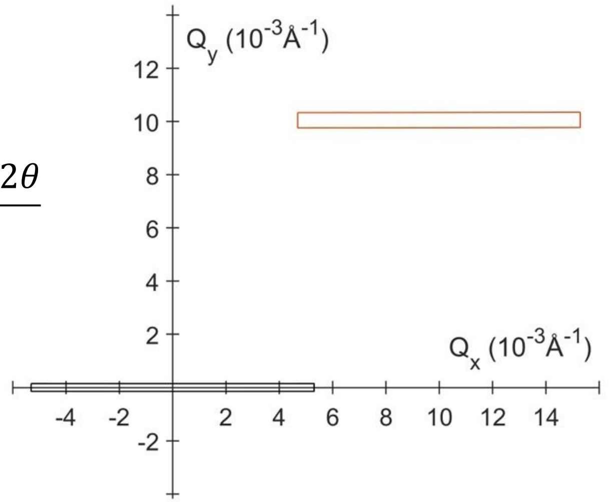
verification exp. at 12ID-OPLS, NSLS-II DPPS, κ = 20kT

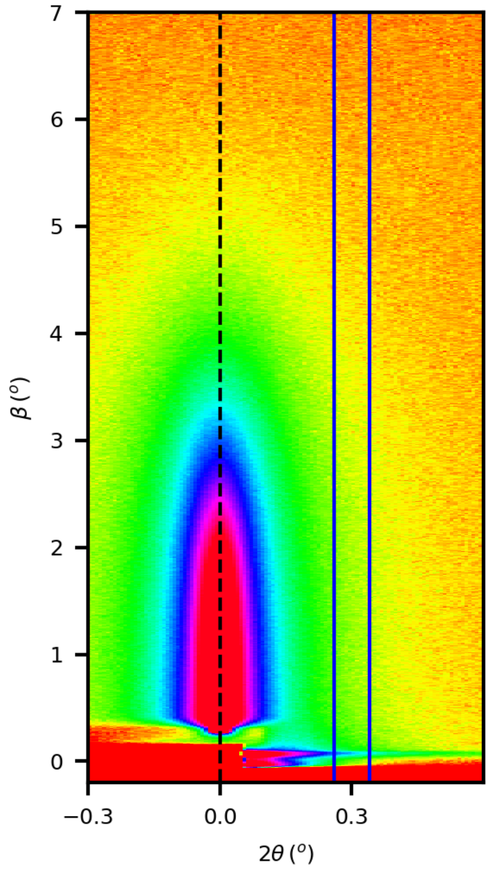


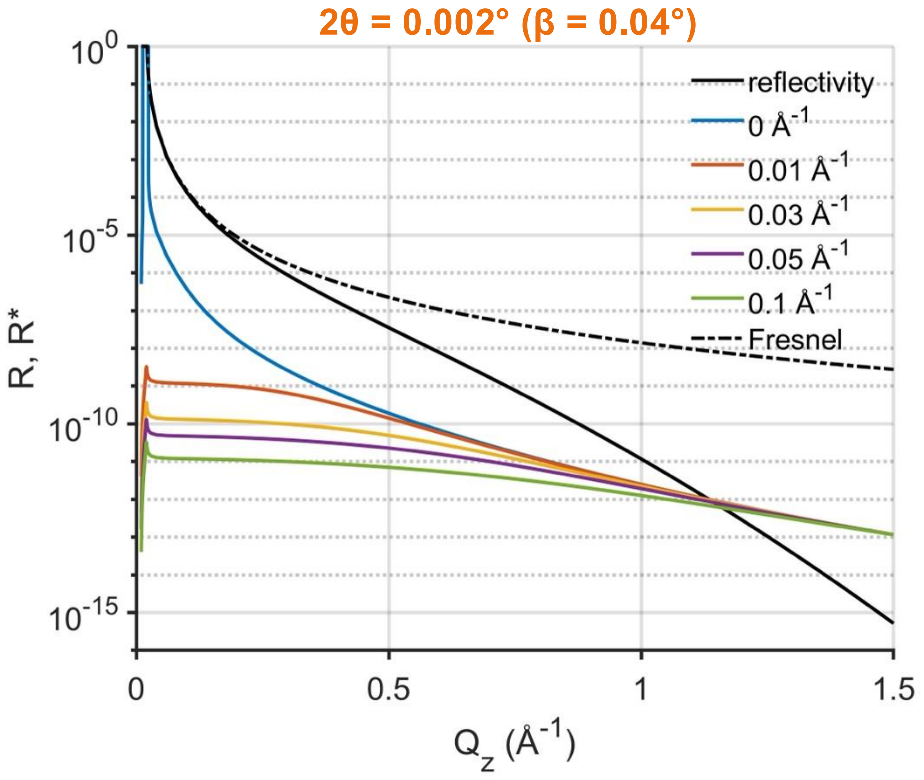
Pseudo reflectivity

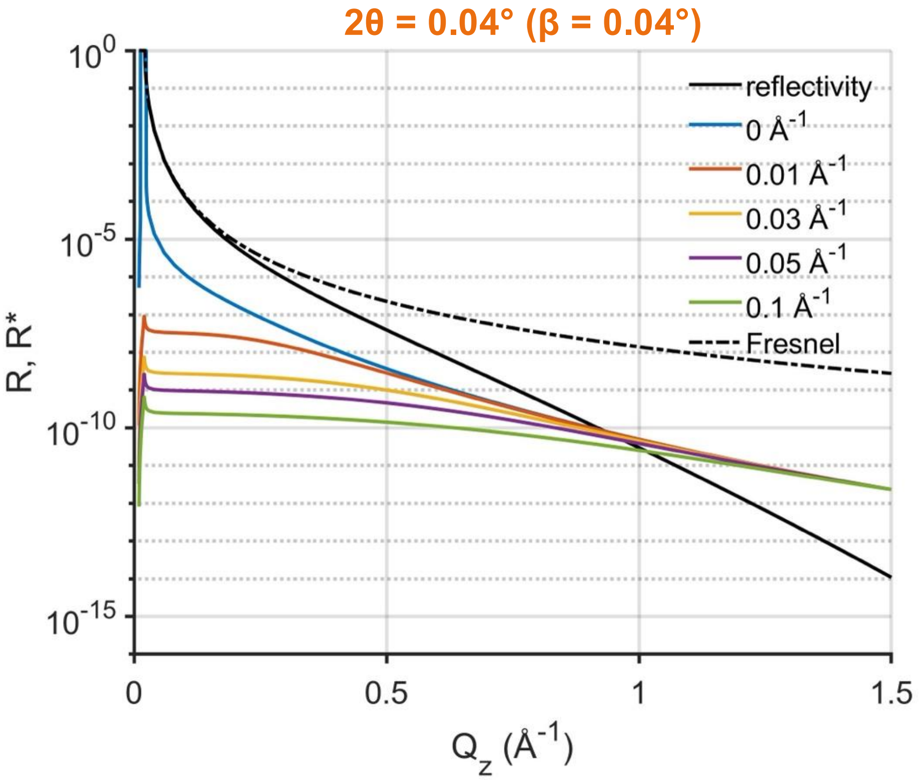
practical aspect: boost GIXOS resolution = boost SNR

$$R, R^* \propto \iint_{\Delta\beta, \Delta 2\theta} \frac{d \sin\beta \, d2\theta}{Q_{xy}^{2-\eta}}$$



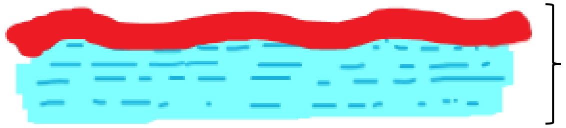






Pseudo reflectivity

practical aspect: background sources

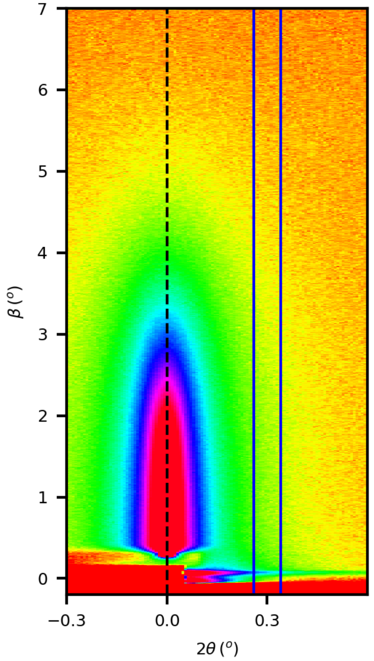


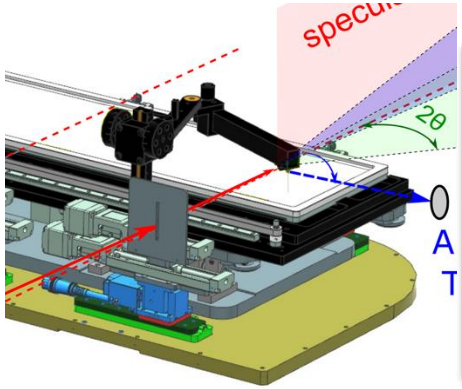
$\Lambda \sim 10nm$

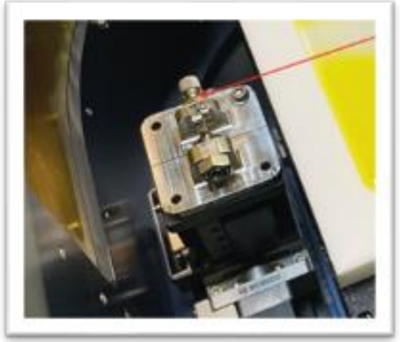
$$I_{total} = I_0 R^* + I_{instr.bkg} + I_{bulk}$$

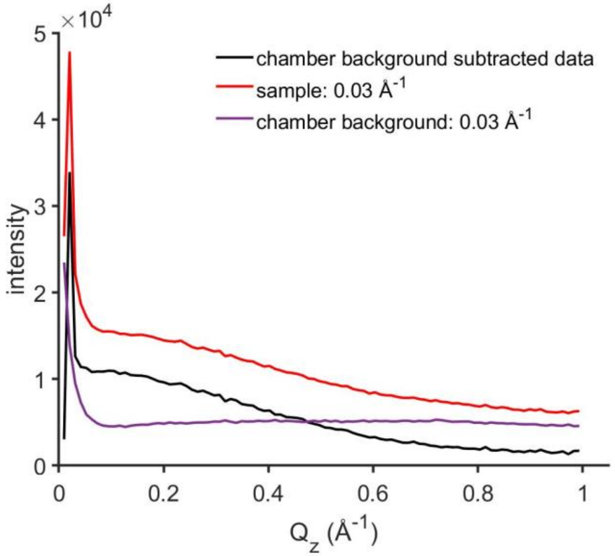
$I_{instr.bkg}$ front, two windows, atmosphere

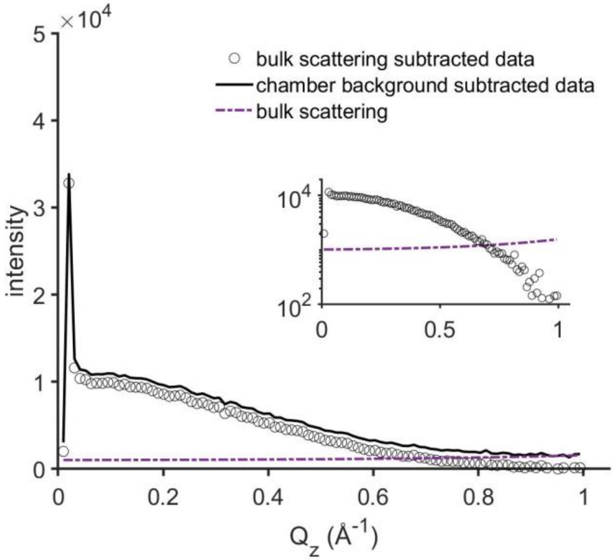
$$I_{bulk}(Q \to 0) \propto \frac{A_0 \rho_{b,\infty}^2 |t_\alpha|^2 |t_\beta|^2}{\sin\alpha} \cdot \frac{k_B T \kappa_T}{2Im(Q_{z,t})}$$

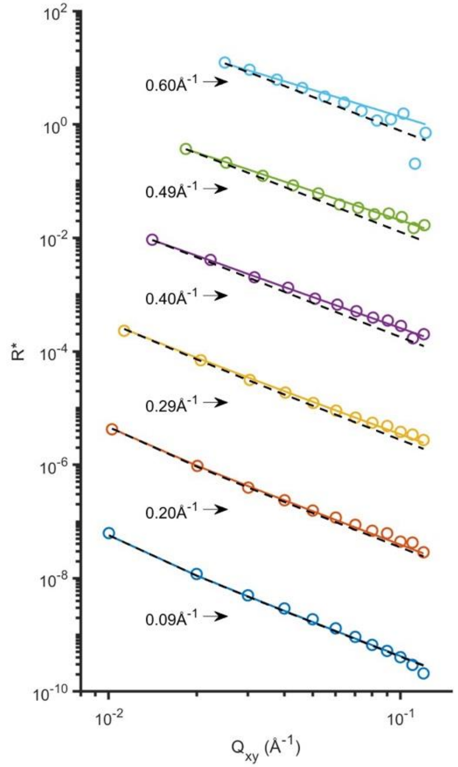








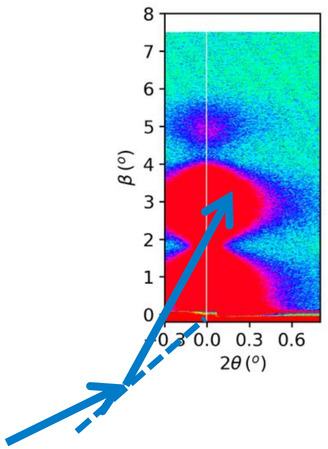


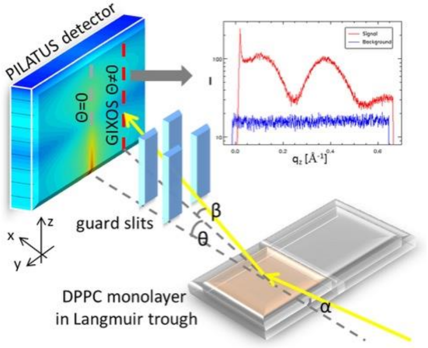


Pseudo reflectivity

practical aspect: two ways of GIXOS

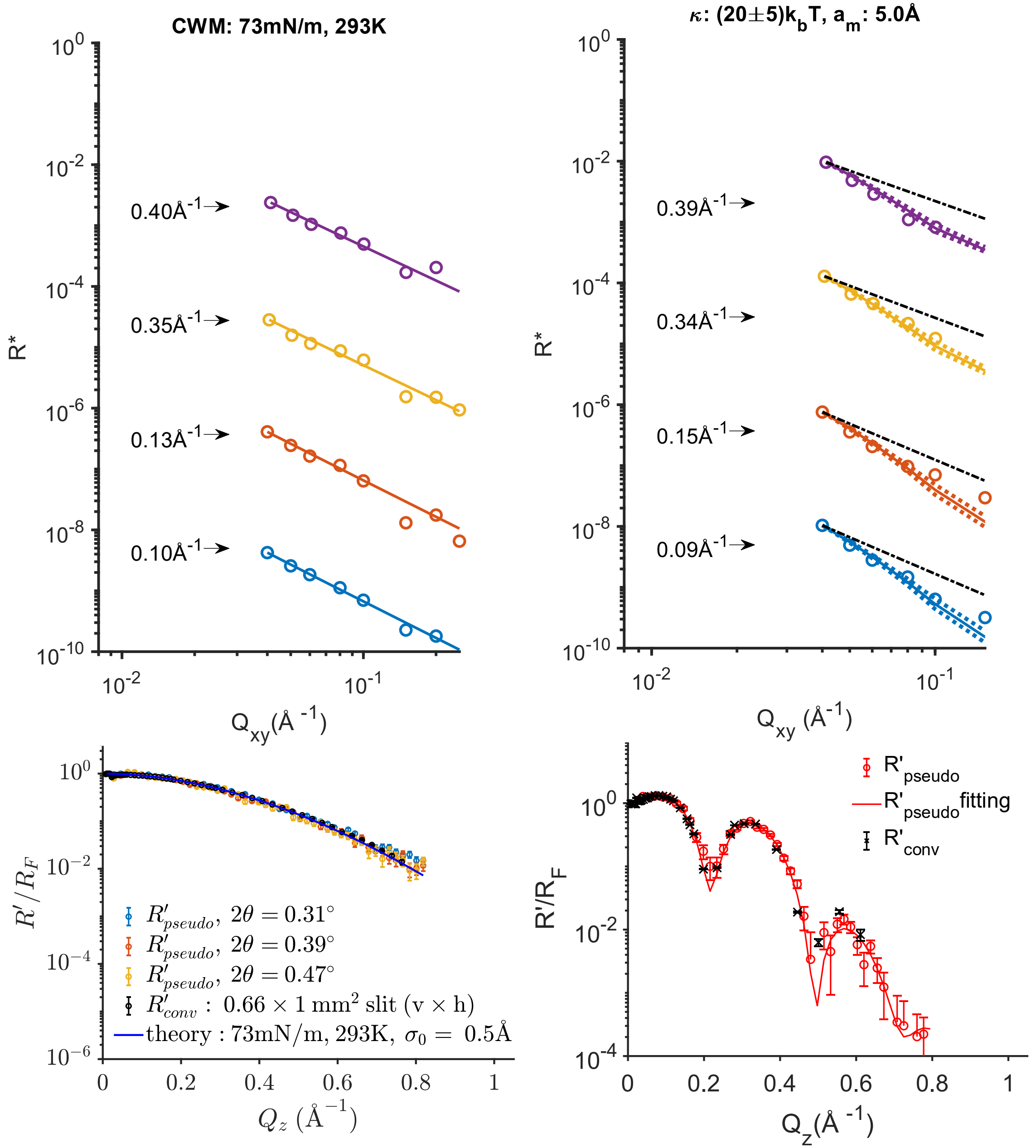
2D GISAXS	post-sample slit
one shot (quick)	Qxy scan (slow)
demanding background shielding: - in chamber beamstop / pinhole - feedthrough required	easy background shielding - slits act as collimation
very clean data	less clean data
low flexibility	high flexibility





Dai, et. al., J. Appl. Phys. (2011)

slit-GIXOS at 12ID-OPLS, NSLS-II



Cases when pseudo reflectivity is not suitable

- in-plane correlation signals overlapping with CW diffuse scattering, for example:
  - lipid layers with nanoscopic domains
  - conjugated polymers
- strong scattering signal from the bulk, for example:
  - multilayer crystals underneath the surface
  - thick polymer layer with strong SAXS signal

Future perspective of the Langmuir GID setup

- Eiger2 X 4M detector trough GID setup: covering Q-range to 2 /Å x 2 /Å in GIXS
- 1D µ-mode (HxV): (200 ~ 450) µm x 3 µm (with collaborations)
- versatile GIXOS environmental chambers (1st one from TU Dresden)

Acknowledgement

DESY
Florian Bertram (P08)
Rene Kirchhof (P08)
Bhaveshkumar Bharatiya (former P08)
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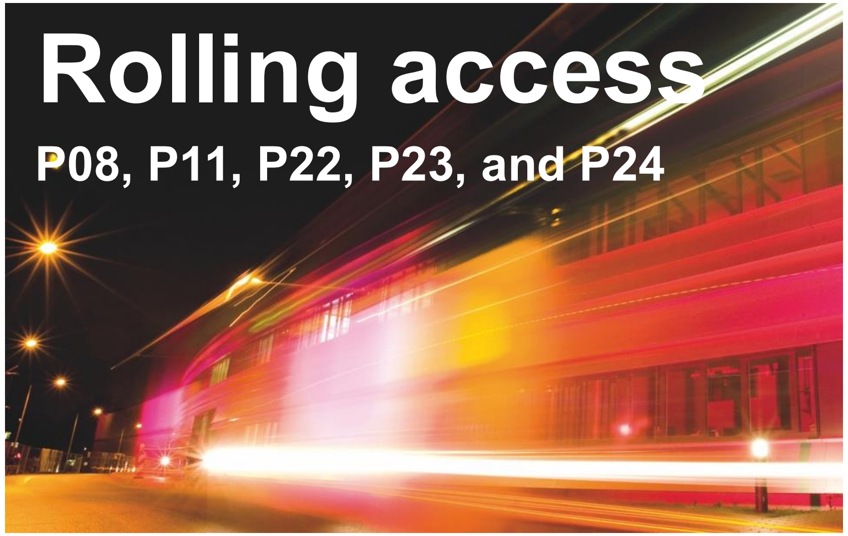
DanScatt
ErUM-Pro 05K19FK2 (Murphy) for Eiger2 1M detector

pseudo XRR on simple liquid surface
https://doi.org/10.1107/s1600576724002887
extended Capillary Wave Model
http://doi.org/10.1103/znt1-fmx6



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