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Polarised Neutron Reflectometry

—

a complementary method to RIXS and ARPES

spectroscopy workshop on novel materials
PSI - SYN
Beatenberg, 3. − 7. May 2011

. . . did so far:

- chemistry studies
- γ-Compton spectroscopy on GaAs

Konstanz

- x-ray diffraction (resonant! HASYLAB, ESRF) on GaAs and ZnSe in electric fields

Potsdam

- neutron optics development
- instrument scientist, reflectometry
- PNR on layerd magnetic films
  - YBCO/LCMO (C. Bernhard, B. Keimer)
  - div. (F. Miletto)
  - LSMO/YBCO (M. Radovic)

PSI

outline

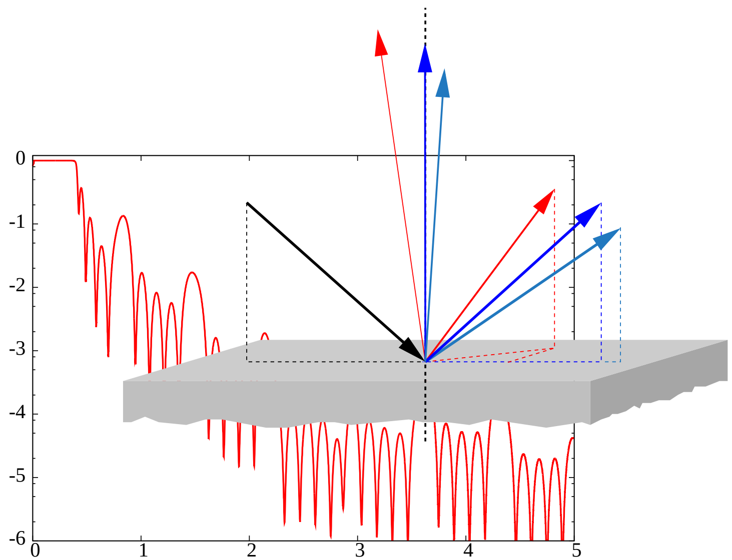
- CV

  → done.

- intro to PNR

  → reflectometry in general
  → . . . with neutrons
  → . . . on magnetic samples

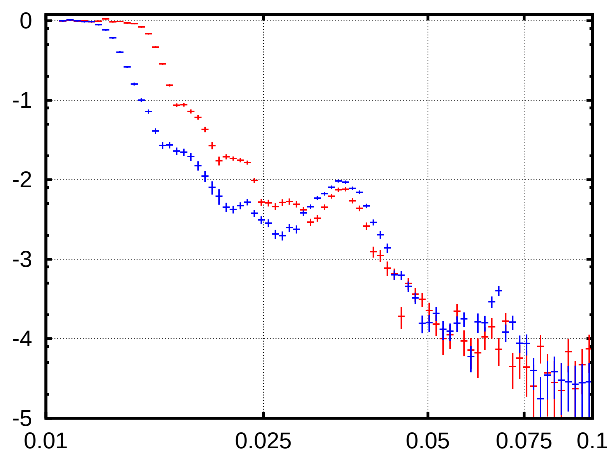
  → experimental set-up



- experiments: LSMO / YBCO interfaces

  → bi-layers (Y. Sassa, M. Radovic)

  → multilayers (M. Radovic)

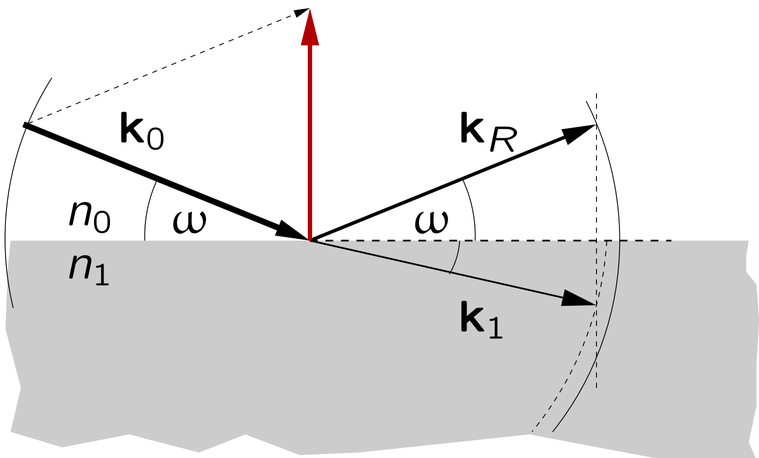


analogy to visible light

flat surfaces partly reflect light
→ picture of the boot

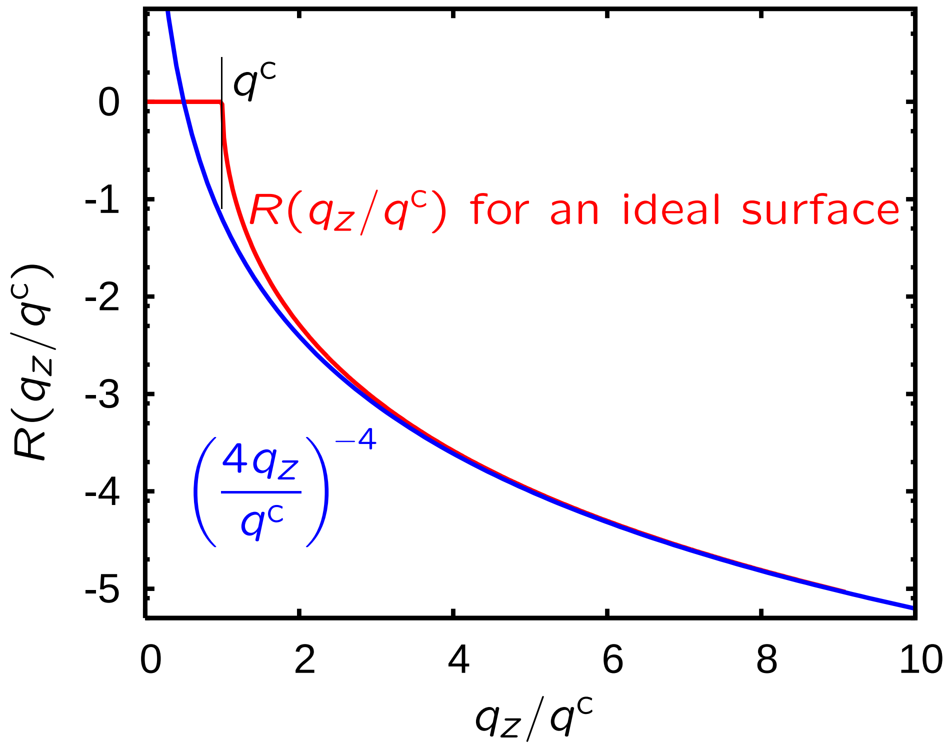
some media also transmit light
→ ground below the water

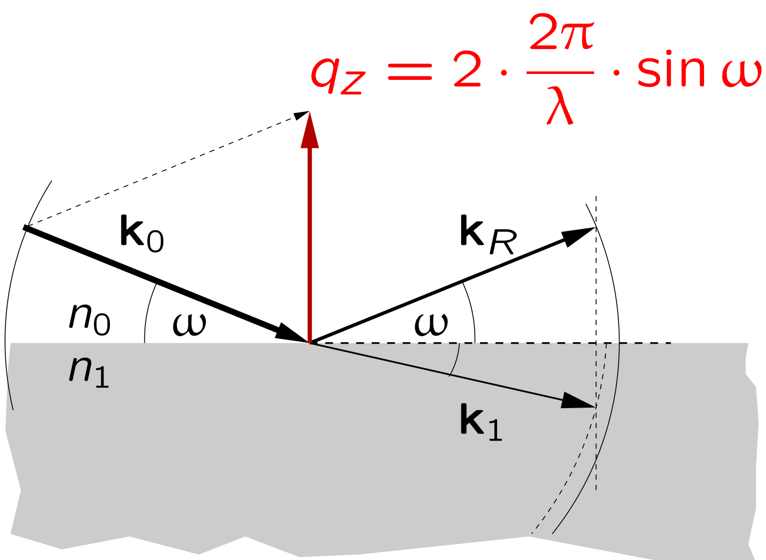
parallel interfaces
→ colorful soap bubbles



Fresnel reflectivity

- reflectivity of a sharp flat surface
- total external reflection for $q_z < q^c$
- exponential decay of $R(q_z)$ for $q_z > q^c$





neutrons / x-rays:

$\lambda \in \{1 \ldots 20\,\text{Å}\}$

$\omega^c < 1^\circ$

refraction of transmitted beam

$\Rightarrow$ dynamical scattering theory

reflected intensity of a multilayer

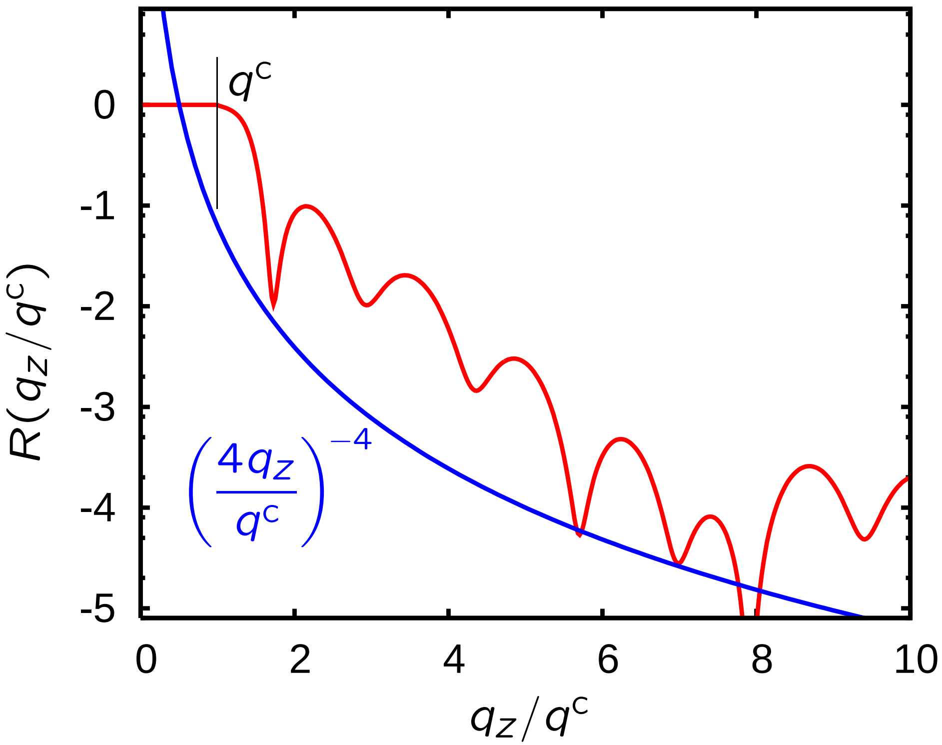
several parallel interfaces:

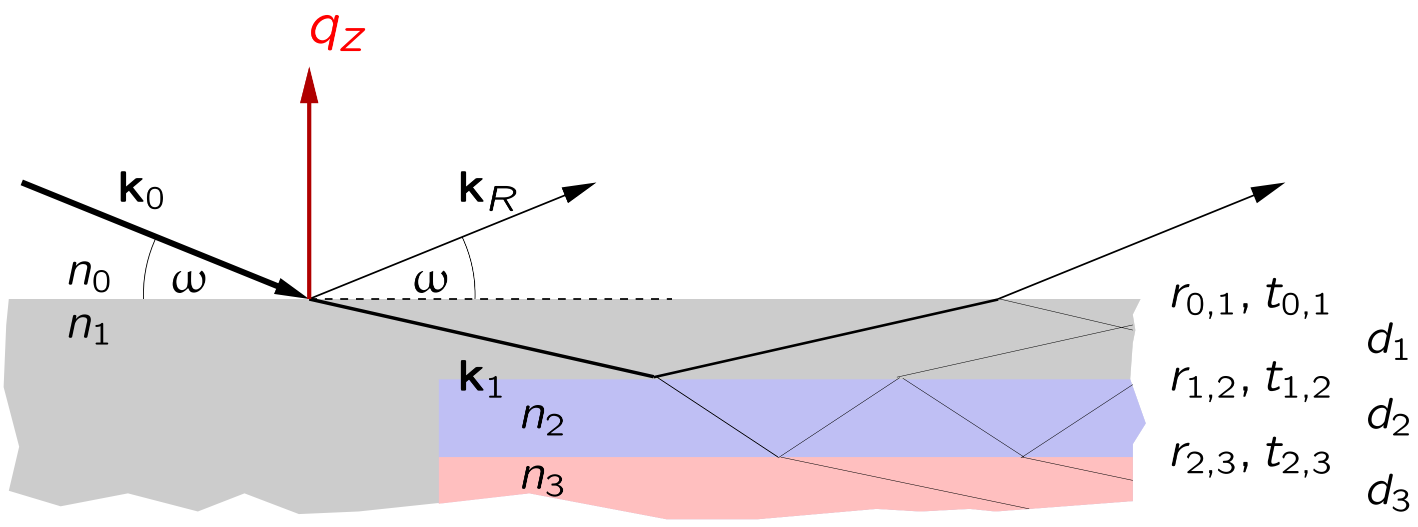
interference of all waves

$\Rightarrow$ complex reflectance

$$r = r(q_z, n_0, n_1, n_2, \ldots, d_1, d_2, \ldots)$$

$$R(q_z) = |r(q_z)|^2$$





d_1 thickness of layer 1

$r_{2,3}$ reflectance of interface 2/3

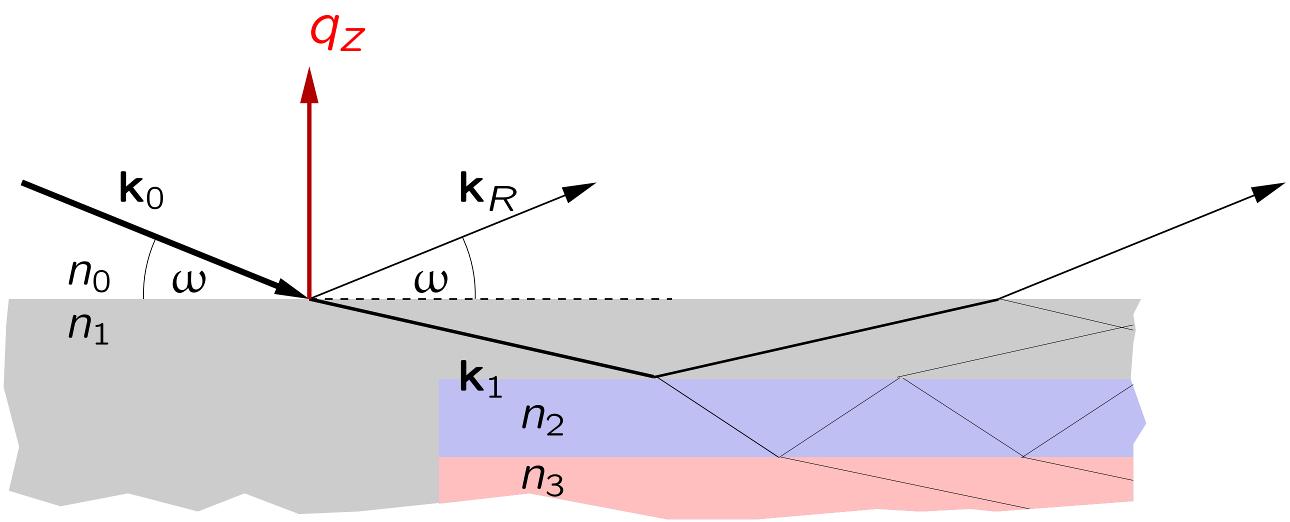
reflected intensity of a multilayer

$$R(q_z) = |r(q_z)|^2$$

$\Rightarrow$ all phase information is lost

$\Rightarrow$ one way road:

$\Rightarrow$ calculation of $R(q_z)$ using a model
and
comparison to measured curve(s)

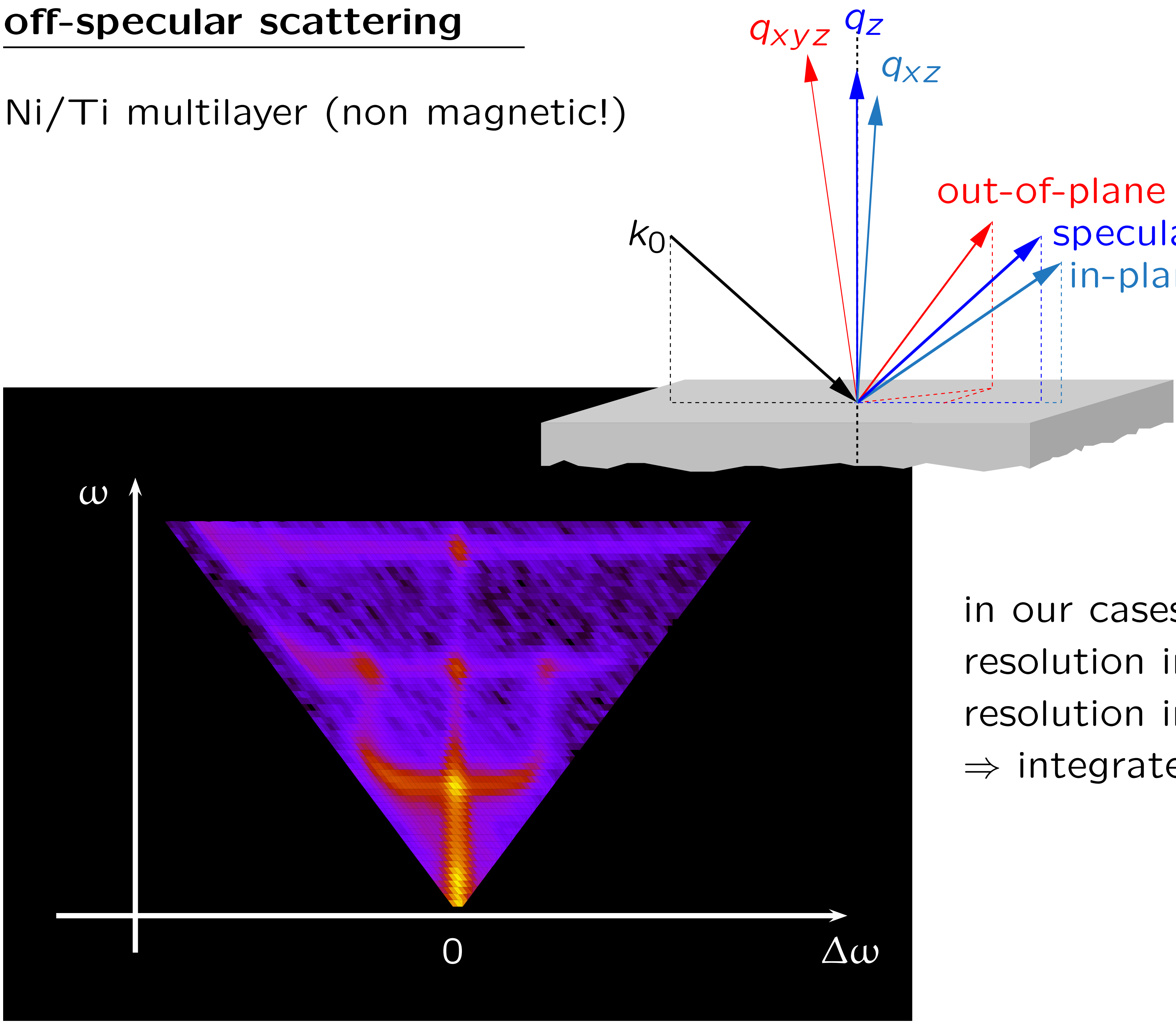


real effects
to be taken into account:

– illumination of the sample
– resolution of the set-up $\Delta\omega$, $\Delta\lambda$
– non-sharp interfaces
– inhomogeneous layers

off-specular scattering

Ni/Ti multilayer (non magnetic!)



in our cases:

resolution in x: $\approx 0.01^\circ$

resolution in y: $> 1^\circ$

$\Rightarrow$ integrated over y

reflectometry, in general :

J. Daillant, A. Gibaud:
X-ray and Neutron Reflectivity
Lect. Notes Phys. 770 (Springer 2009)

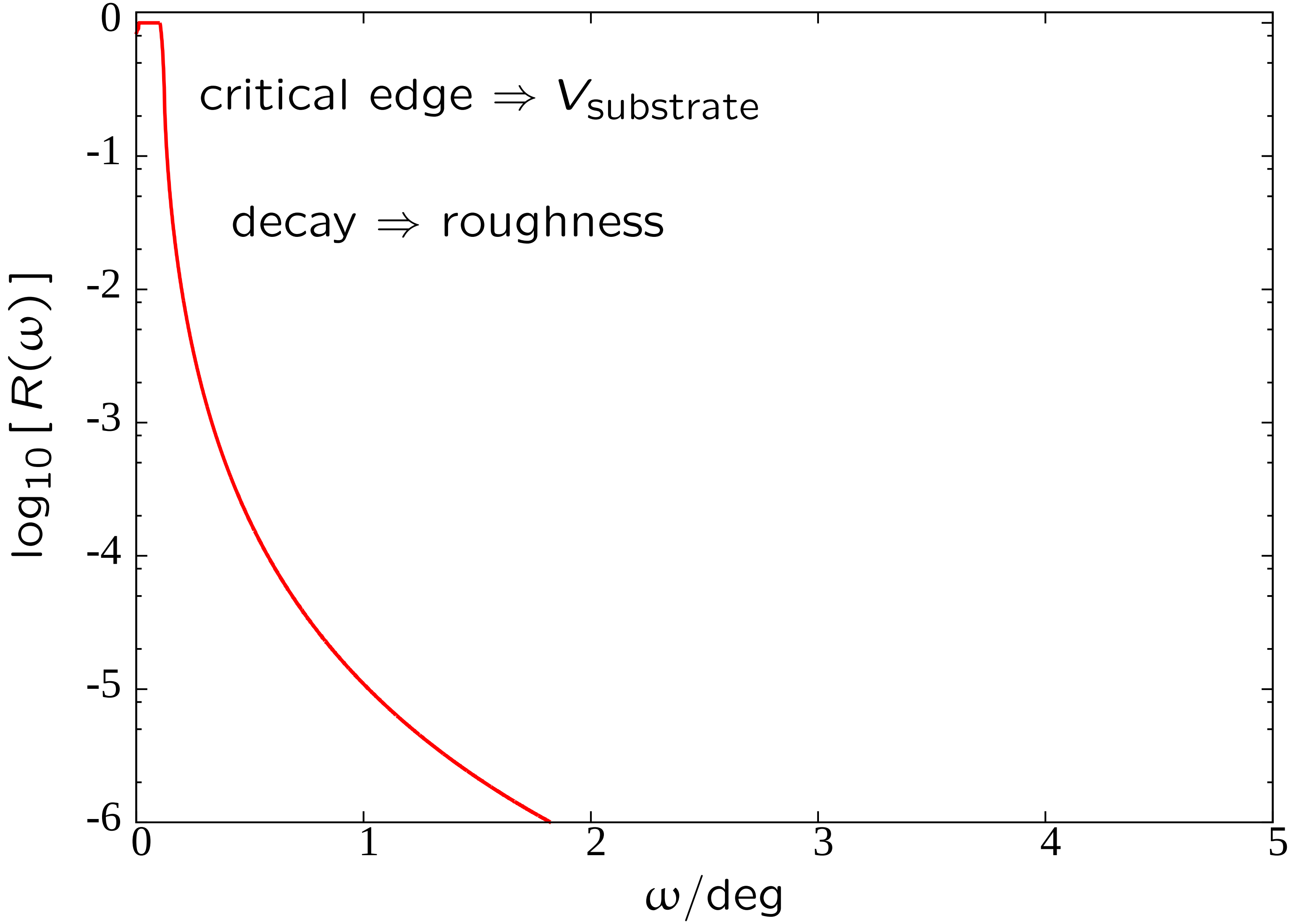
U. Pietsch, V. Holý, T. Baumbach:
High-Resolution X-Ray Scattering
(Springer 2004)

J. Stahn:
Introduction to polarised neutron and resonant x-ray reflectometry
`http://people.web.psi.ch/stahn/publications`

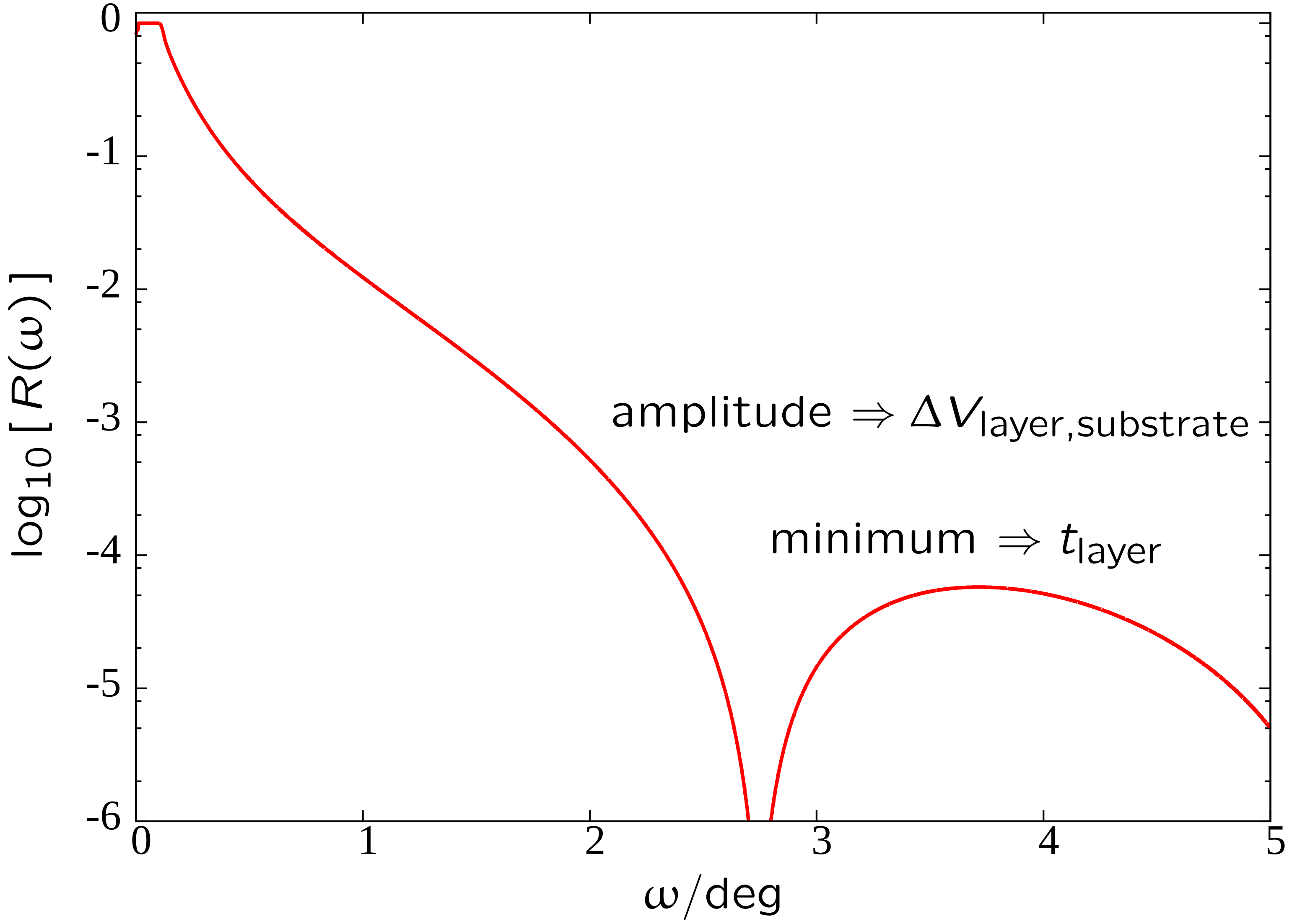
. . . on magnetic systems

F. Ott:
Neutron scattering on magnetic surfaces
C. R. Physique **8**, 763-776 (2007)

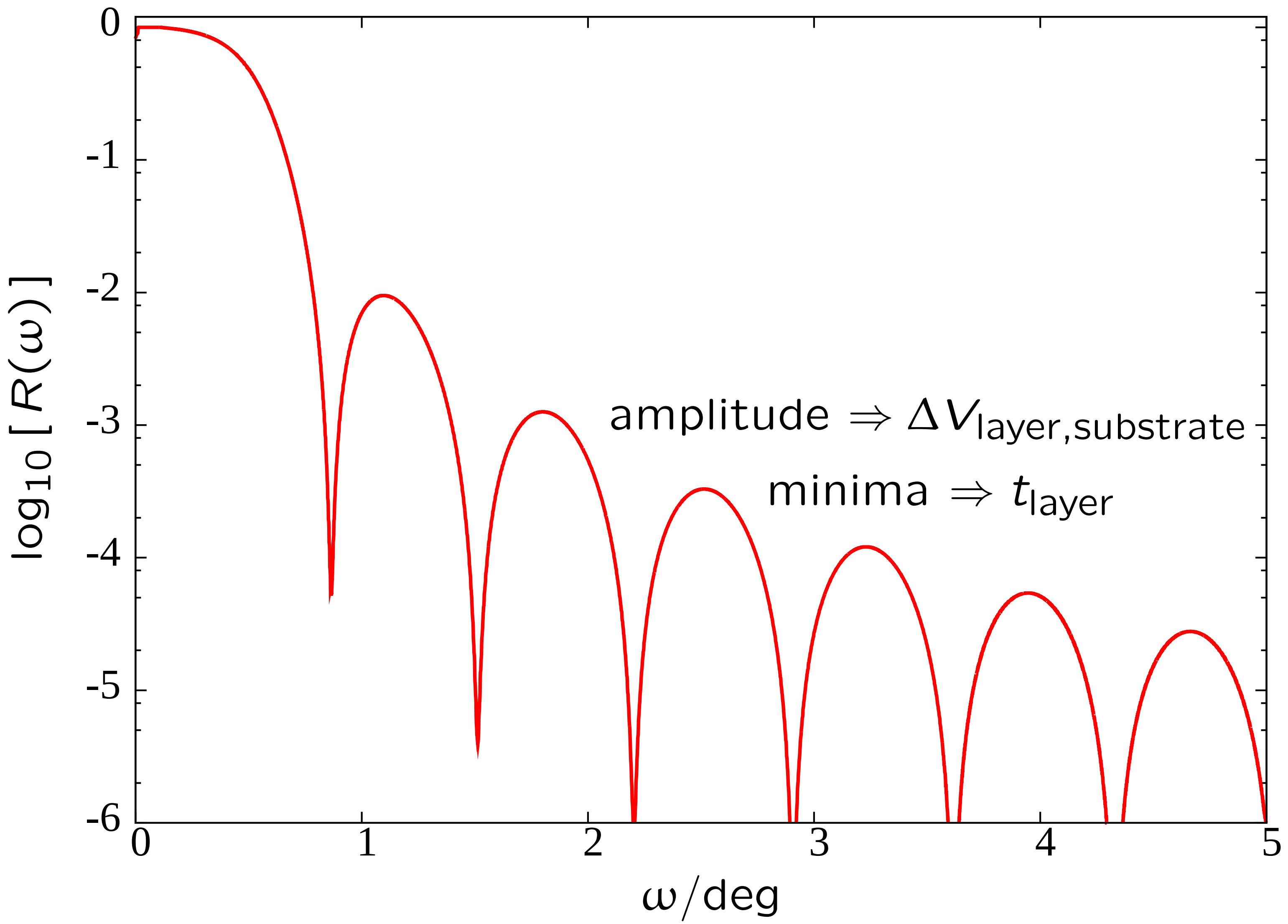
. . . of a surface

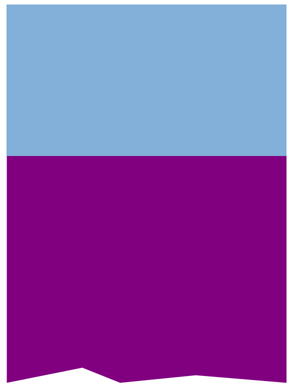

0
-1
-2
-3
-4
-5
-6
0
1
2
3
4
5
critical edge ⇒ $V_{substrate}$
decay ⇒ roughness
$\log_{10}[\,R(\omega)\,]$
ω/deg

. . . of a thin layer

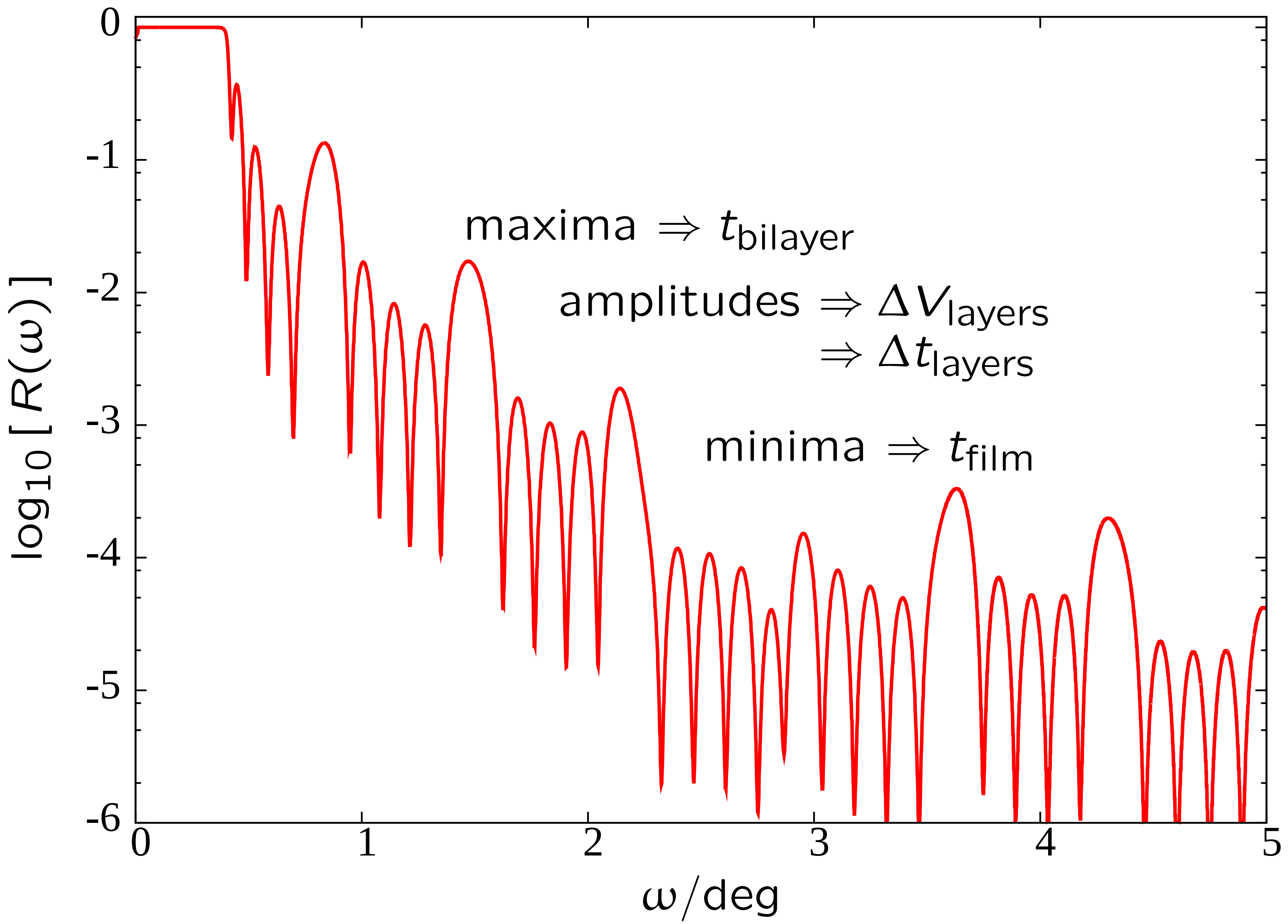


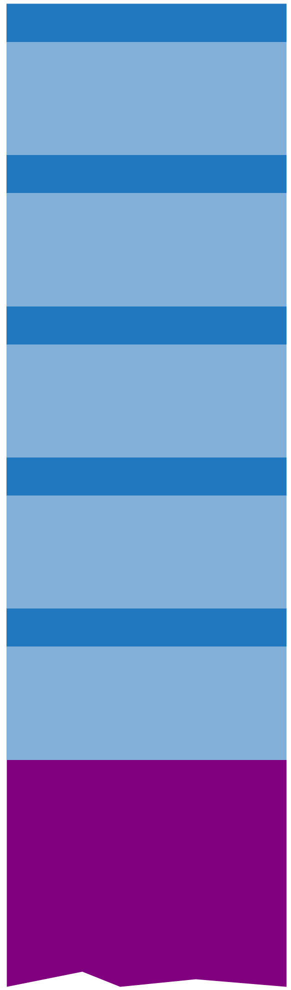
. . . of a thick layer





. . . of a periodic multilayer





what is V_i for neutrons?

interaction neutron / nucleus j with $\lambda \gg r_{\text{nucleus}\,j}$

$$V_j^{\text{Fermi}} = b_j \frac{2\pi\hbar^2}{m} \delta(\mathbf{r})$$

$$V_i^{\text{n}} = \frac{1}{Vol} \int_j V_j^{\text{Fermi}} \, \mathrm{d}\mathbf{r}$$

$$= \frac{2\pi\hbar^2}{m} \frac{1}{Vol} \sum_j b_j$$

$$:= \frac{2\pi\hbar^2}{m} \rho^b$$

b: nuclear scattering length

$\Rightarrow$ isotope specific

complement to XR

interaction neutron magnetic moment $\boldsymbol{\mu}$ / magnetic induction $\mathbf{B}$

$$V^{\text{m}} = \boldsymbol{\mu}\,\mathbf{B}$$

$$:= \frac{2\pi\hbar^2}{m} \rho^{\text{m}}$$

$\boldsymbol{\mu} \uparrow\uparrow \mathbf{B} \Rightarrow V^{\text{m}} = +\mu B$

$\boldsymbol{\mu} \uparrow\downarrow \mathbf{B} \Rightarrow V^{\text{m}} = -\mu B$

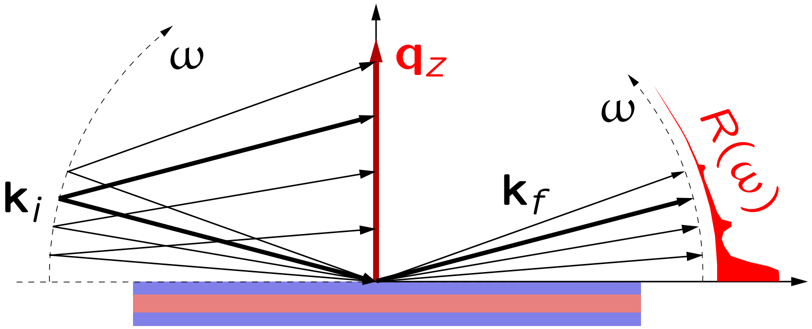
$\boldsymbol{\mu} \perp \mathbf{B} \Rightarrow V^{\text{m}} = 0$

$$R = R(q_z) = R(\lambda, \omega) \qquad q_z = 4\pi \frac{\sin\omega}{\lambda}$$

angle-dispersive set-up

variation of ω with fixed λ

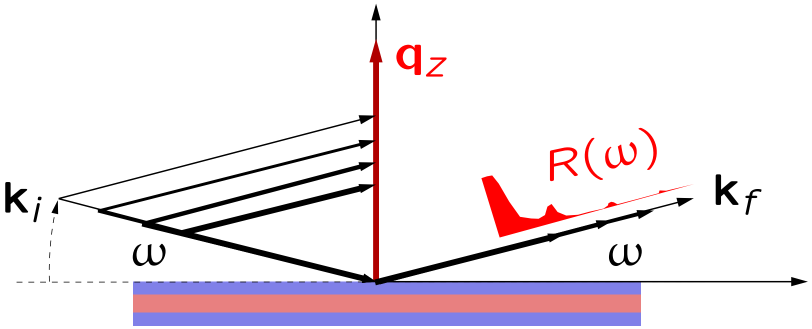
detection under 2ω



energy-dispersive set-up

variation of λ with fixed ω

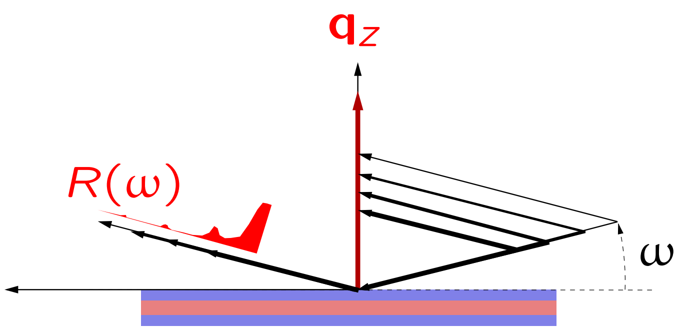
detection via time-of-flight

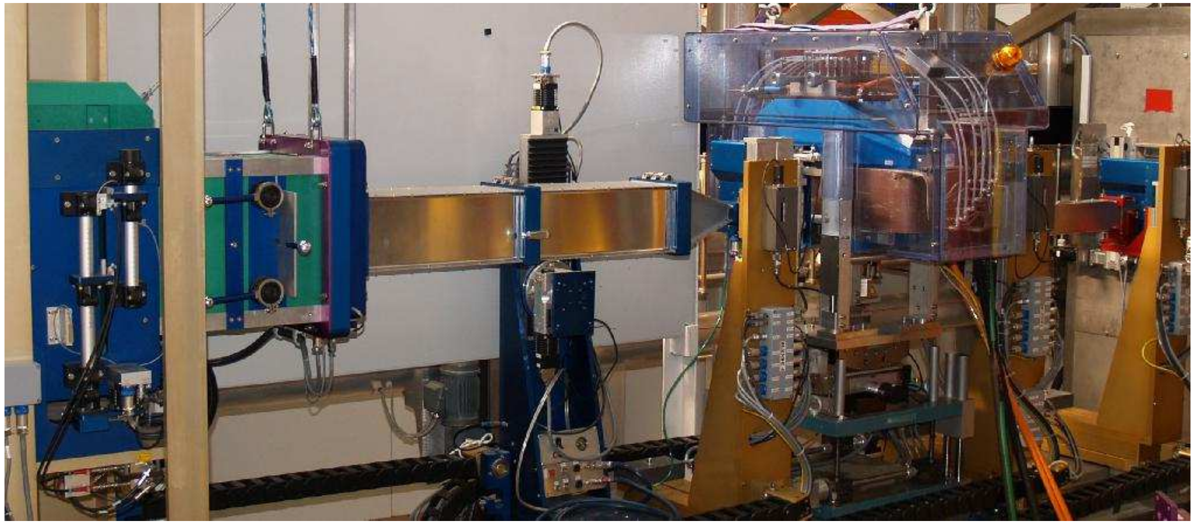


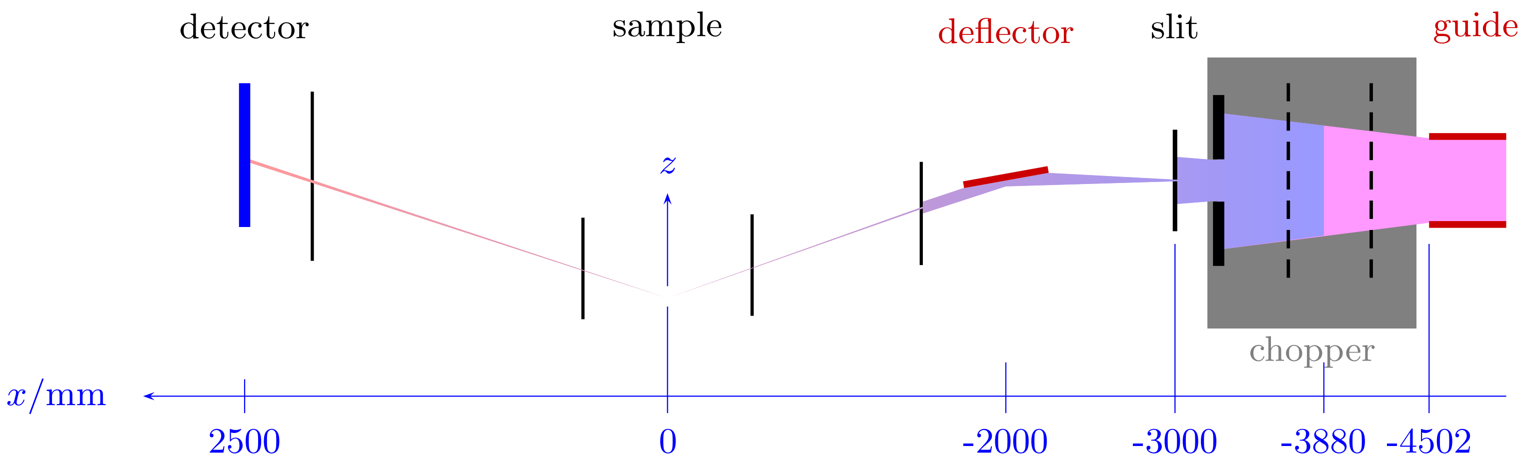
energy-dispersive set-up

neutron reflectometer

Amor at SINQ

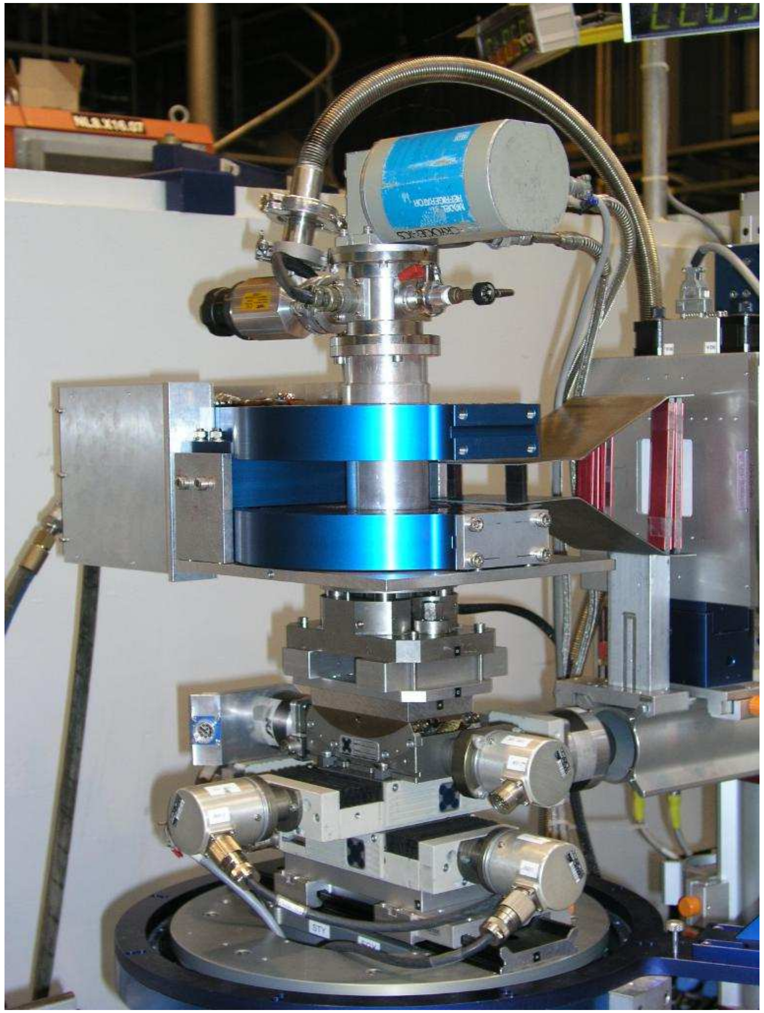






time-of-flight / energy encoding

cooling with a
closed cycle refrigerator $8\,\mathrm{K} < T < 450\,\mathrm{K}$



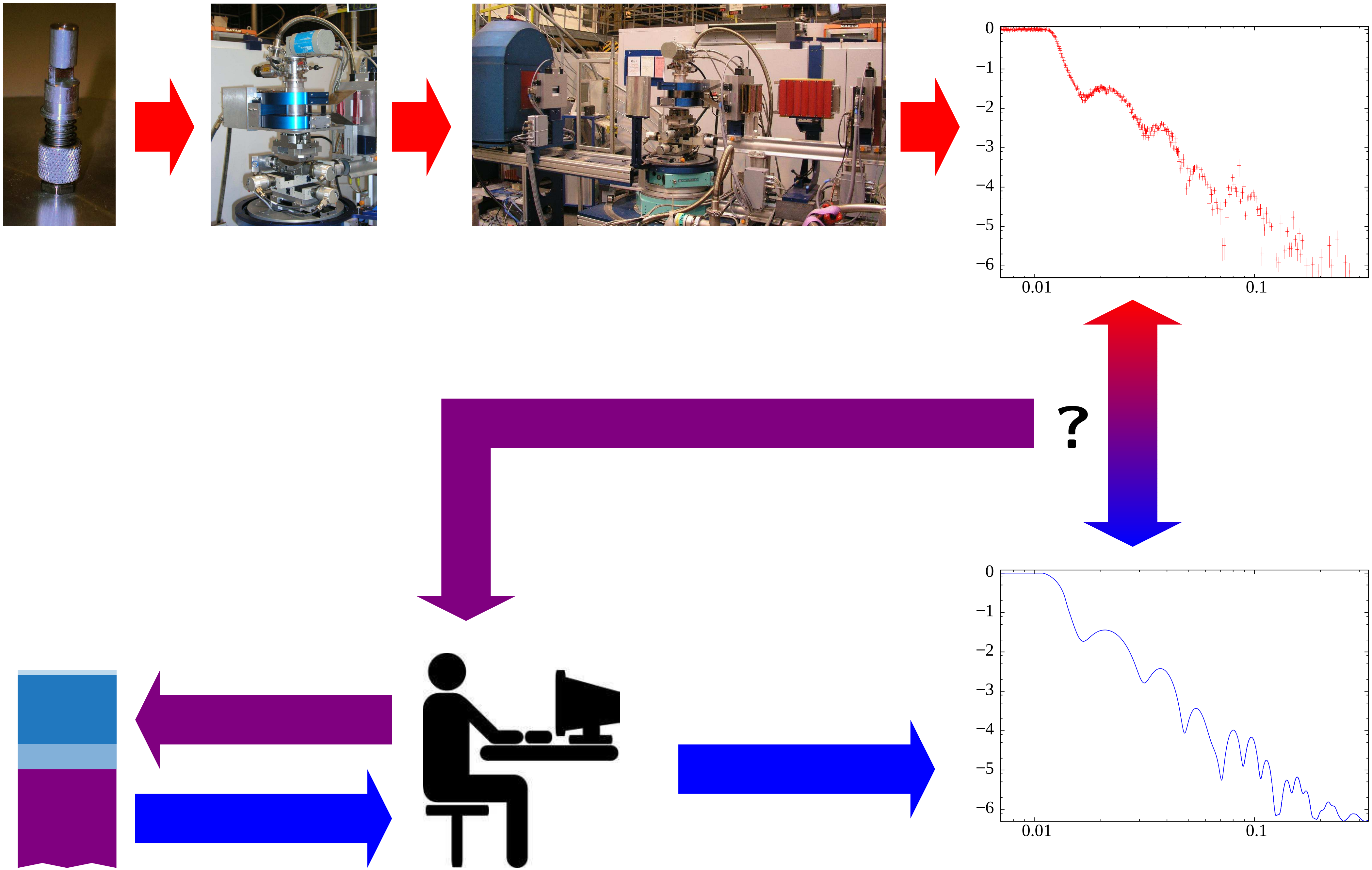
tilt- and
translation stages
for alignment

application of an external magnetic field with
Helmholtz coils $-1000\,\mathrm{Oe} < H < 1000\,\mathrm{Oe}$

or
usage of a cryo-magnet $1.4\,\mathrm{K} < T < 300\,\mathrm{K}$
$-50\,000\,\mathrm{Oe} < H < 50\,000\,\mathrm{Oe}$

and **sample**

from the sample to a profile



magnetic signal almost as strong as nuclear one

only $B_\perp$ is probed

no element sensitivity

high **depth resolution** (down to 0.1 nm)

strongly **model-based**

penetration depth 1 000 nm

resolution limit 500 nm

limited q_Z-range accessible

extreme sample environments are *no* problem

time-consuming data analysis

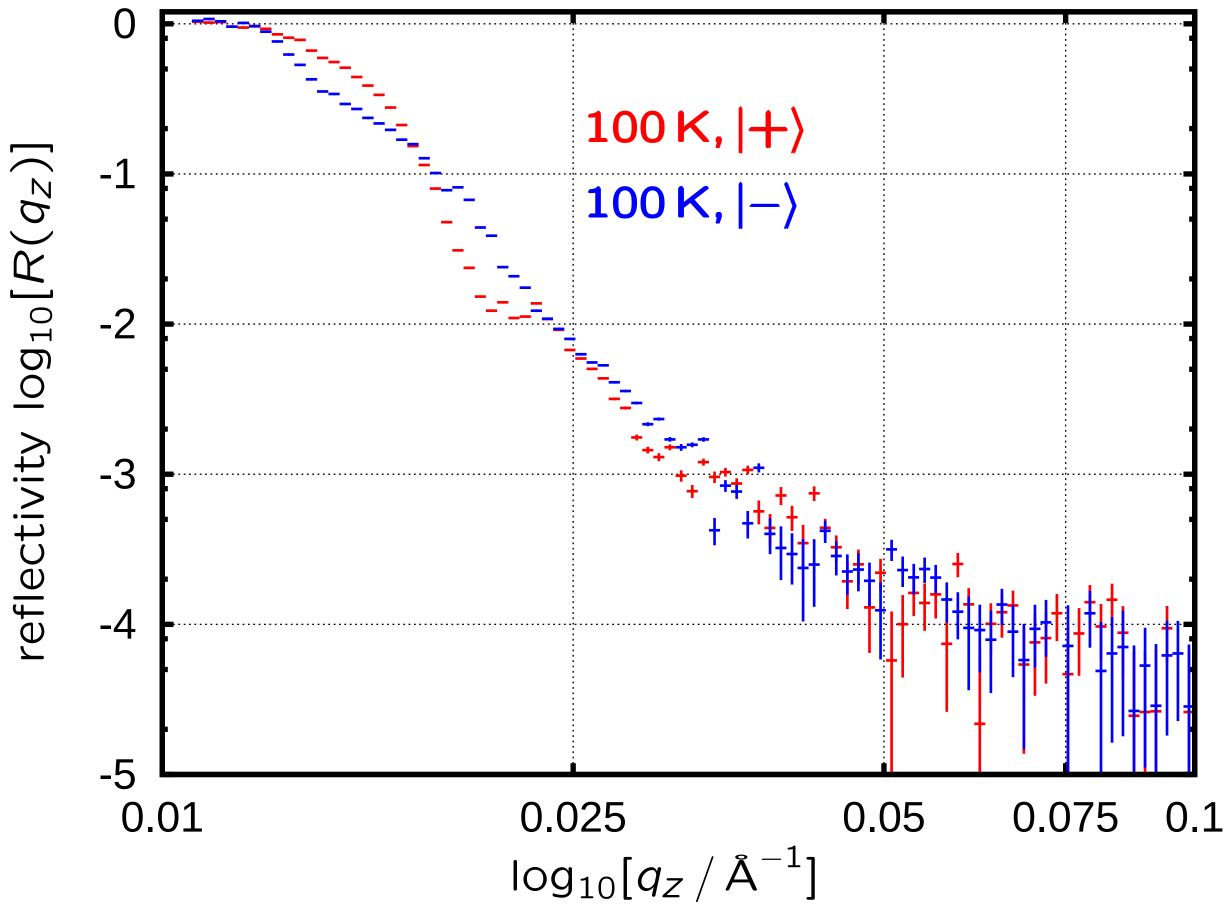
supporting methods needed

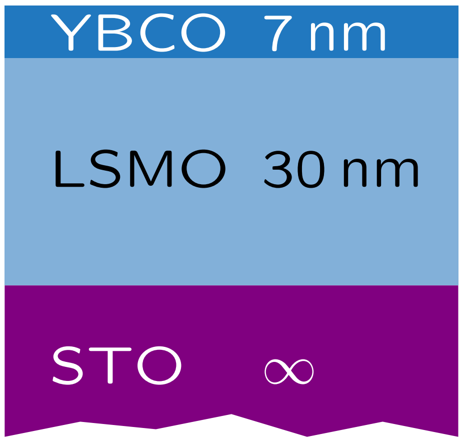
bi-layer: STO / LSMO / YBCO

Y. Sassa, M. Radovic, Oct. 2009

sample size: $10 \times 5\,\mathrm{mm}^2$

measurement time: 6 h





measurements: LSMO/YBCO interfaces

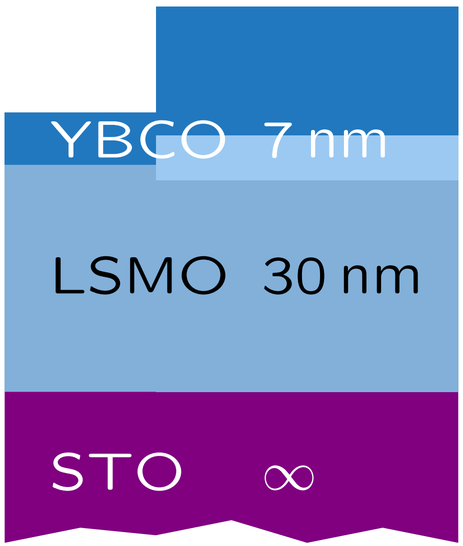
bi-layer: STO / LSMO / YBCO

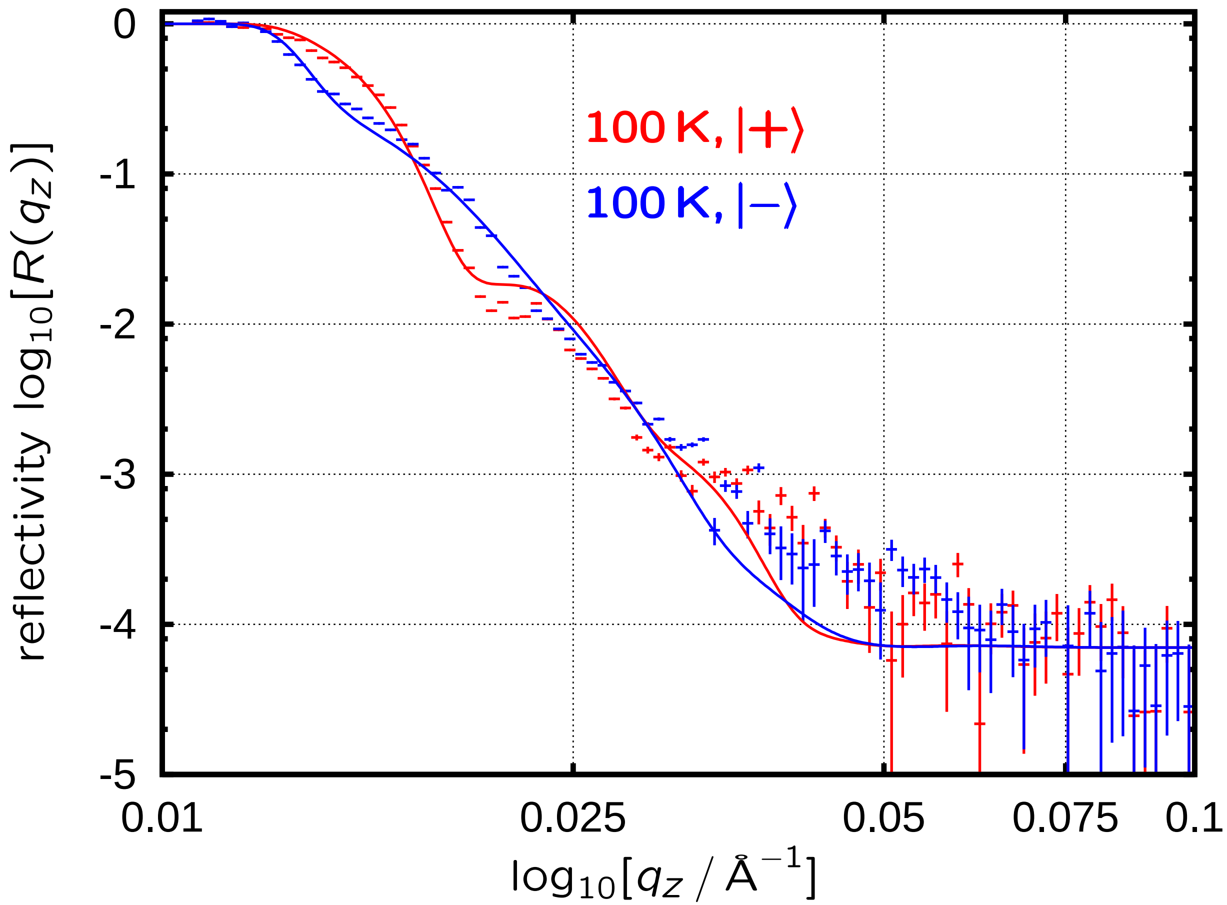
Y. Sassa, M. Radovic, Oct. 2009

sample size: $10 \times 5\,\mathrm{mm}^2$

measurement time: 6 h

fit-time: 8 h





simulation

free parameters:

- thicknesses
- magnetisation

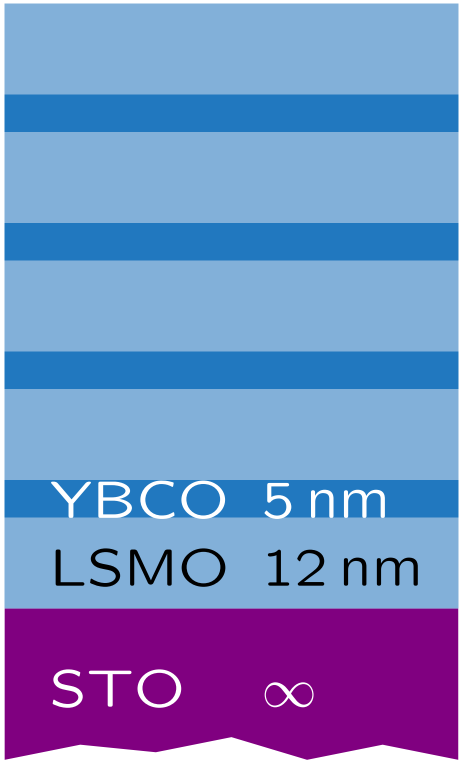
fit is not satisfactory!

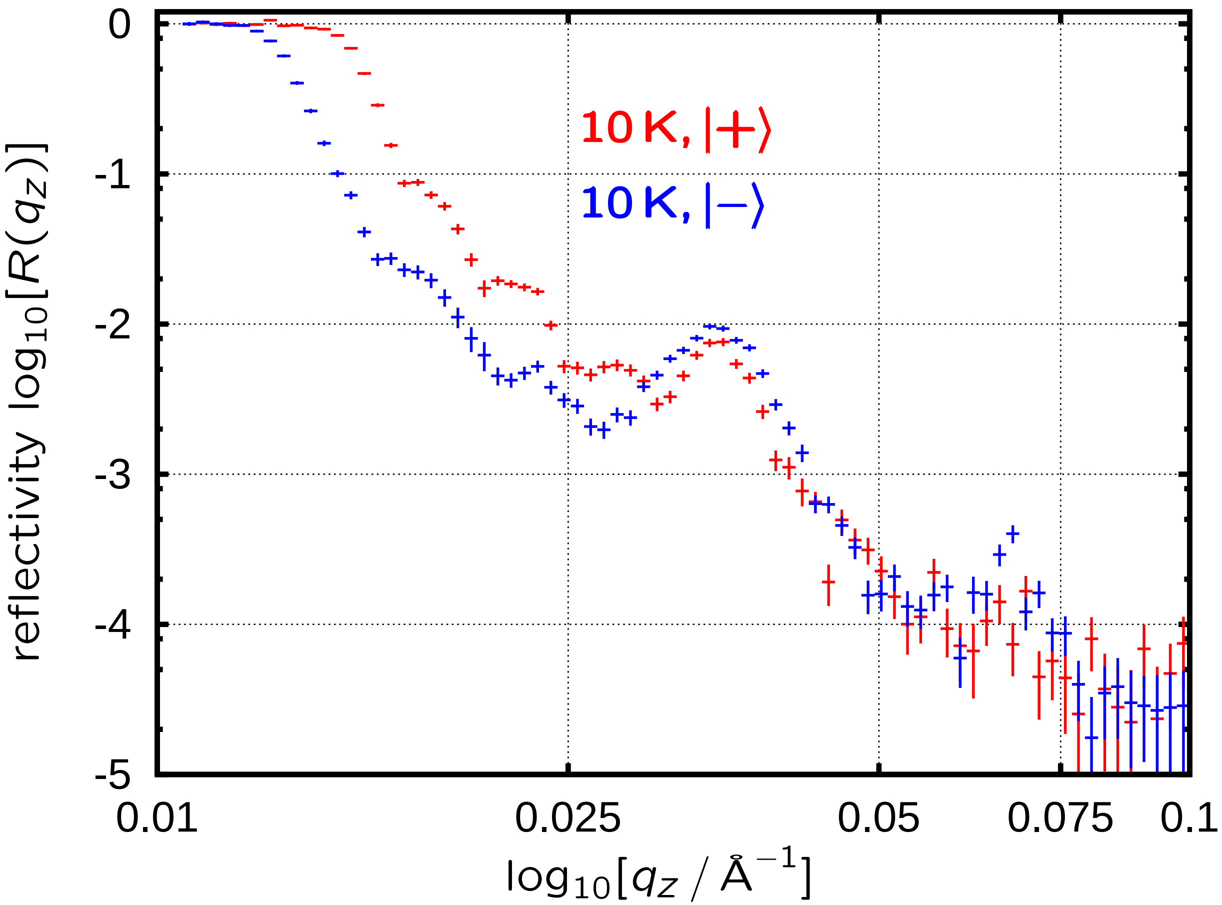
multi-layer: STO / [LSMO / YBCO]$_4$ / LSMO ELLA 10-018

M. Radovic, May 2010

sample size: $5 \times 5\,\mathrm{mm}^2$

measurement time: 18 h





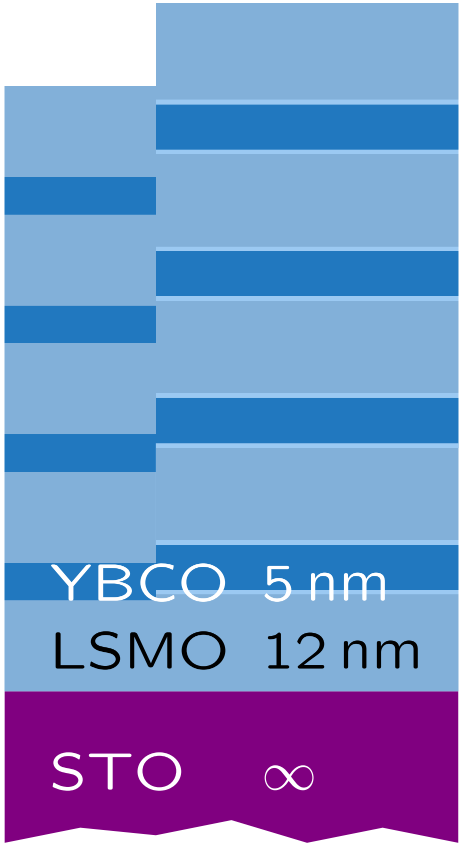
multi-layer: STO / [LSMO / YBCO $]_4$ / LSMO ELLA 10-018

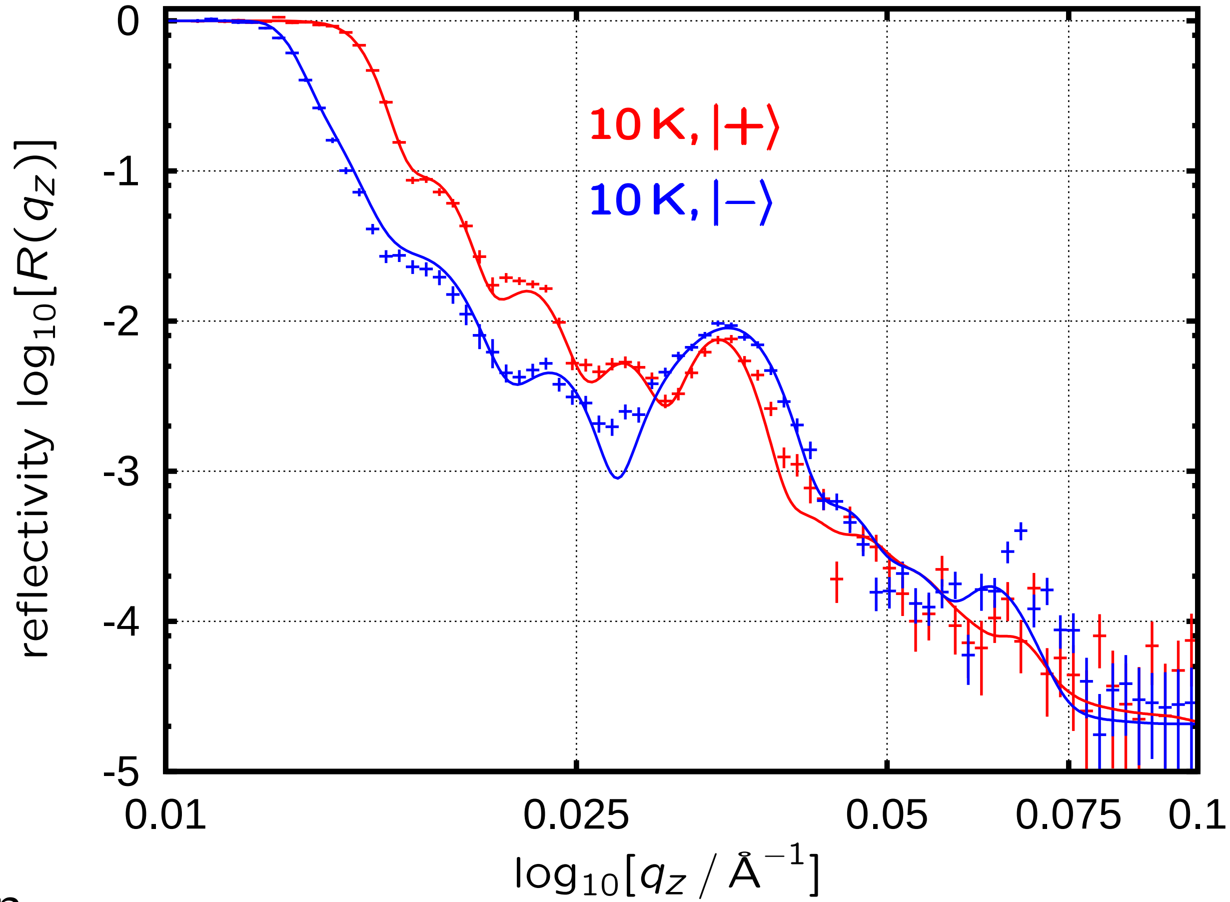
M. Radovic, May 2010

sample size: $5 \times 5\,\mathrm{mm}^2$

measurement time: 18 h

fit-time: 12 h





simulation

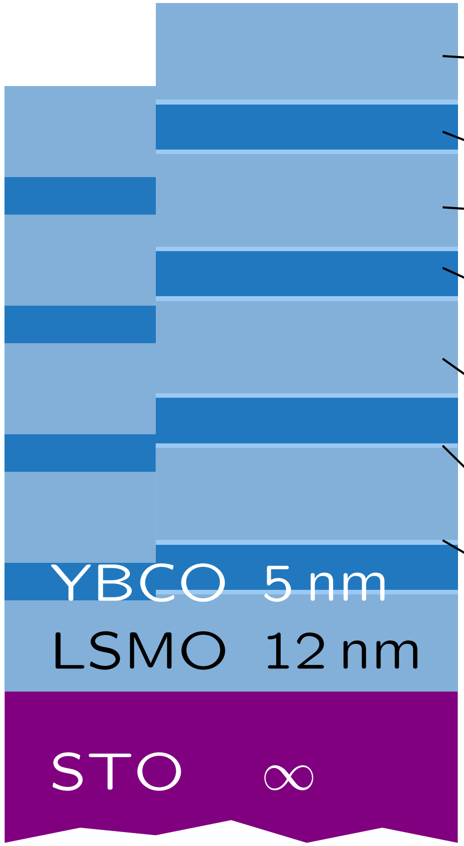
free parameters:

- thicknesses
- magnetisation
- magnetically dead layers

fit is good

multi-layer: STO / [LSMO / YBCO $]_4$ / LSMO ELLA 10-018

M. Radovic, May 2010



top layer: density reduced by ≈ 20%

thicknesses increased by ≈ 10%

YBCO: density reduced by ≈ 4%

LSMO: magnetisation 2.0 μ_B/Mn

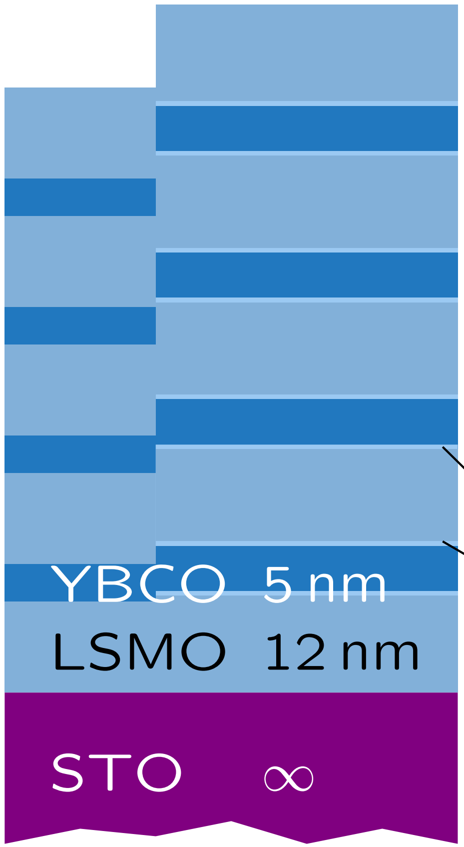
LSMO: magnetically dead layers ≈ 0.6 nm

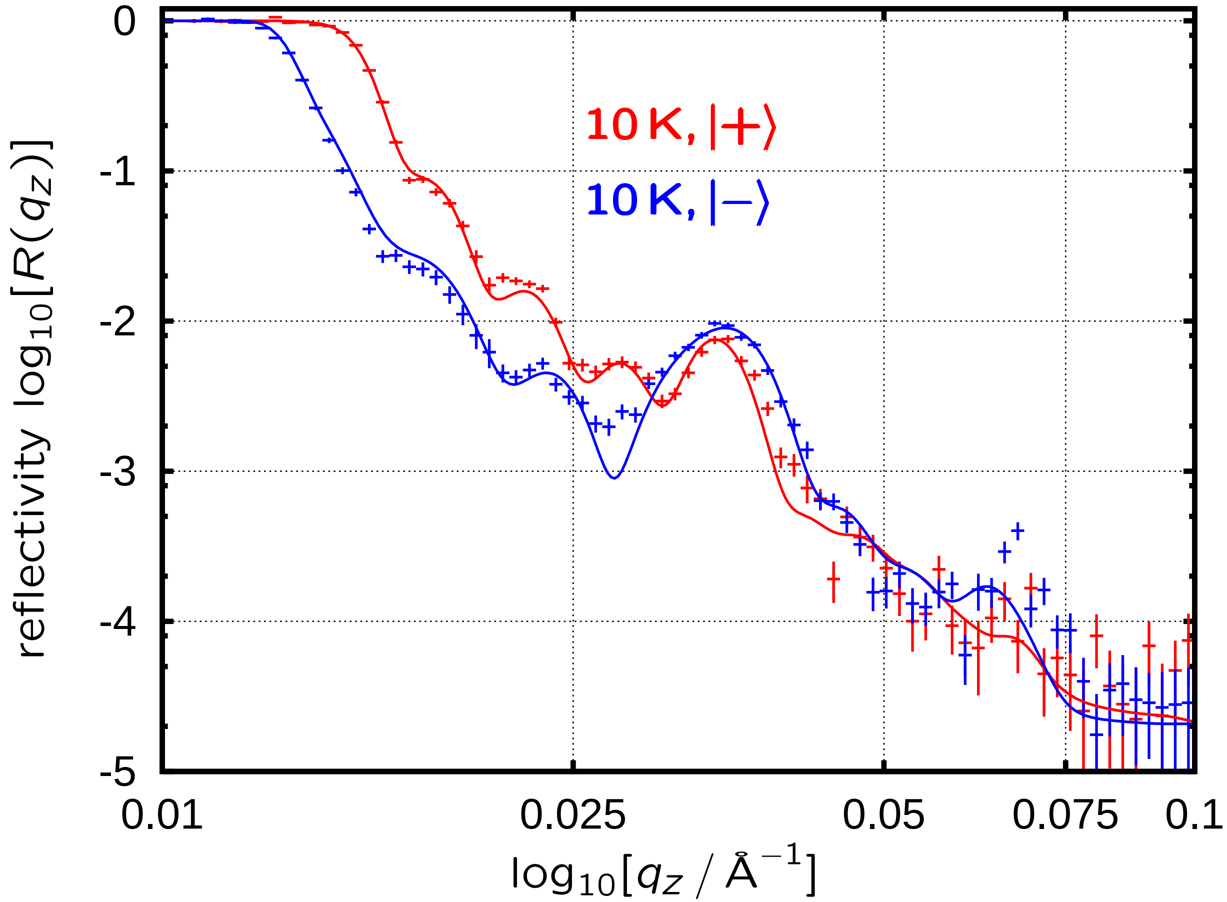
measurements: LSMO/YBCO interfaces

multi-layer: STO / [LSMO / YBCO $]_4$ / LSMO

ELLA 10-018

M. Radovic, May 2010

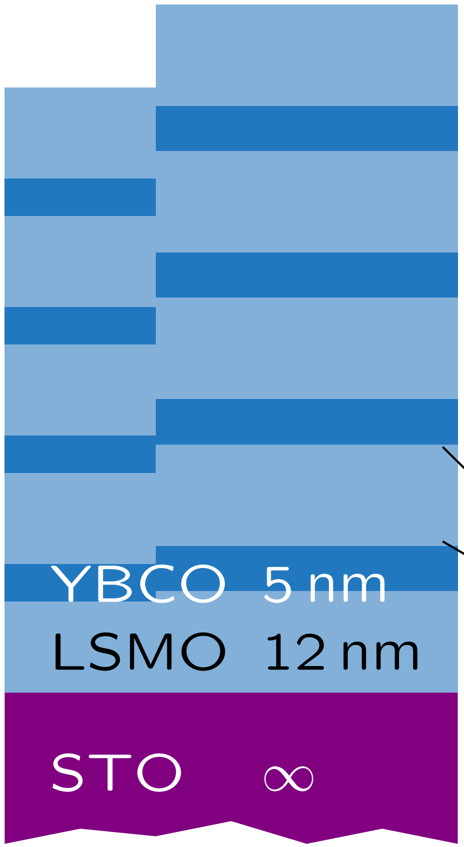


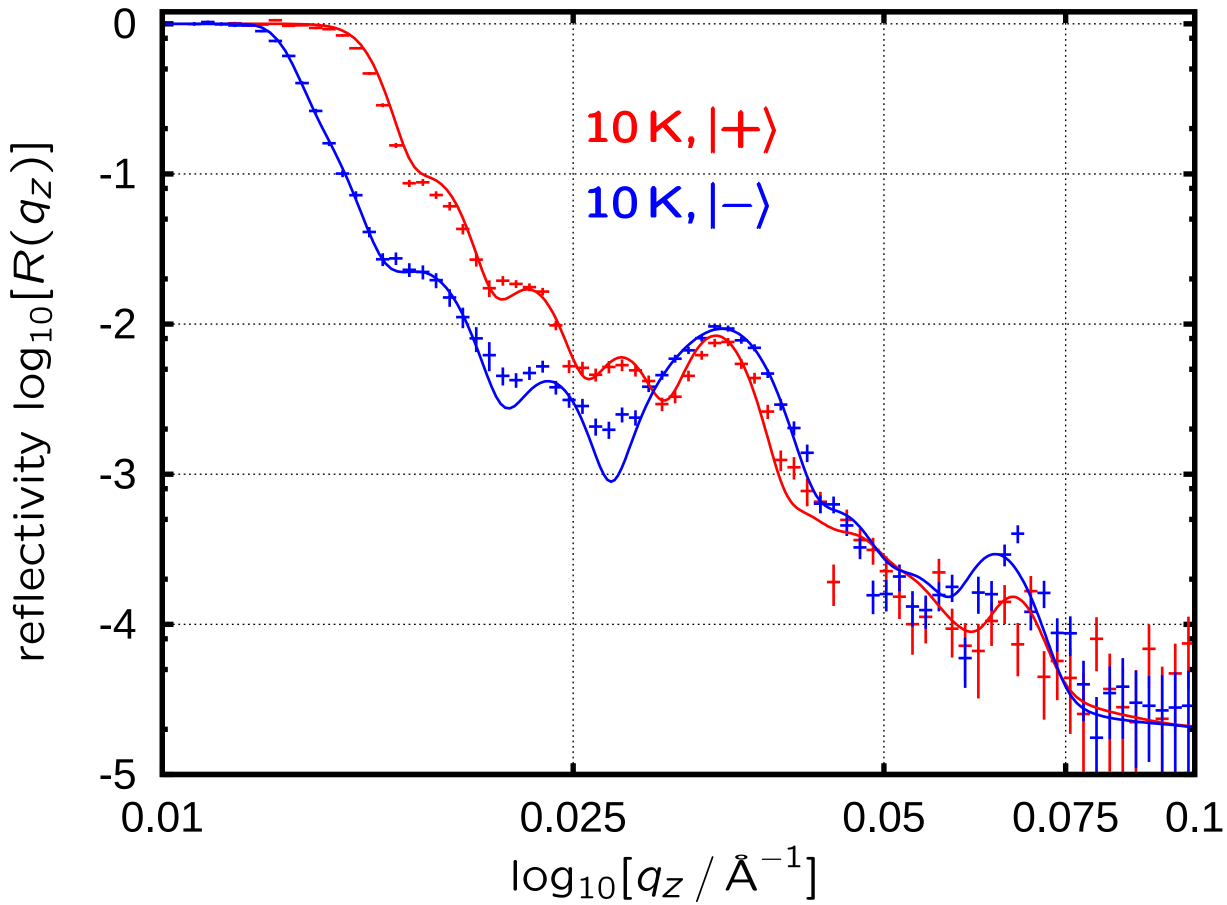


LSMO: magnetically dead layers ≈ 0.6 nm

multi-layer: STO / [LSMO / YBCO $]_4$ / LSMO ELLA 10-018

M. Radovic, May 2010





LSMO: no magnetically dead layers

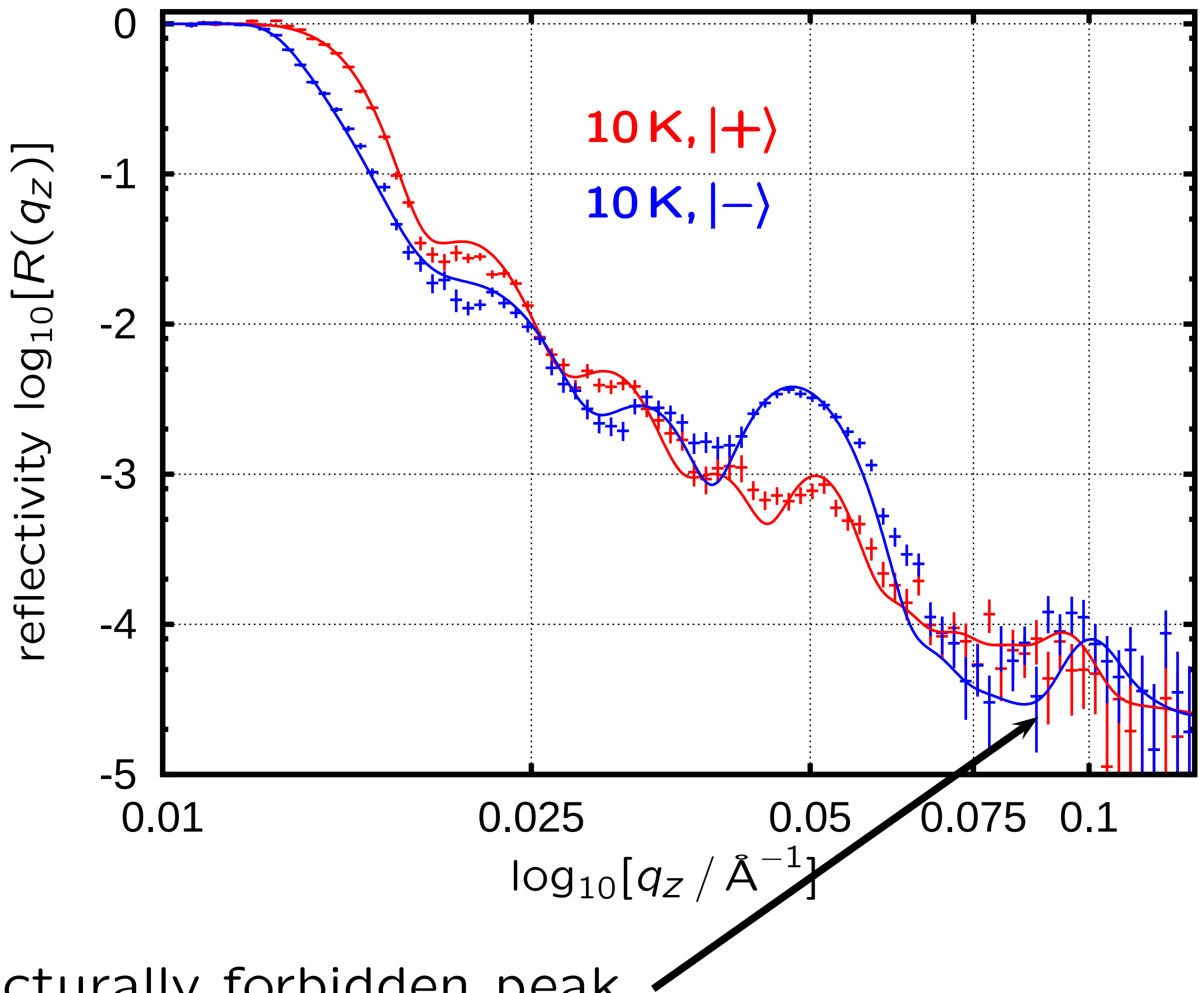
⇒ higher q_z & better statistics needed

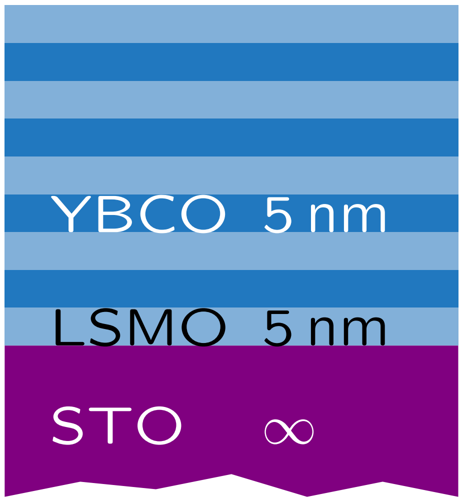
multi-layer: STO / [LSMO / YBCO $]_4$ / LSMO ELLA 10-021

M. Radovic, May 2010

sample size: $5 \times 5\,\mathrm{mm}^2$

measurement time: 18 h





structurally forbidden peak

$\Rightarrow$ magnetic profile breaks symetry

magnetically dead layer of $\approx 0.6\,\mathrm{nm}$

to be fitted with a more complex model

to take along:

- PNR probes $\rho(z)$ where $\rho = \rho(\text{composition}, B_\perp)$

∘ atomic depth resolution

∘ lateral integration over several μm

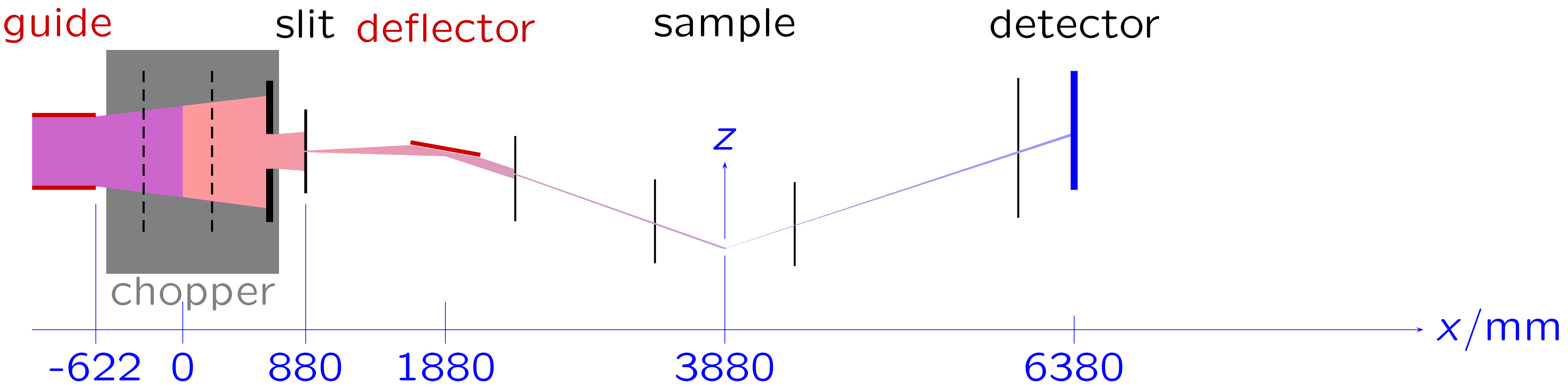
- data analysis via comparison with model

  ⇒ no unique solution

  ⇒ PNR is a *team-player*

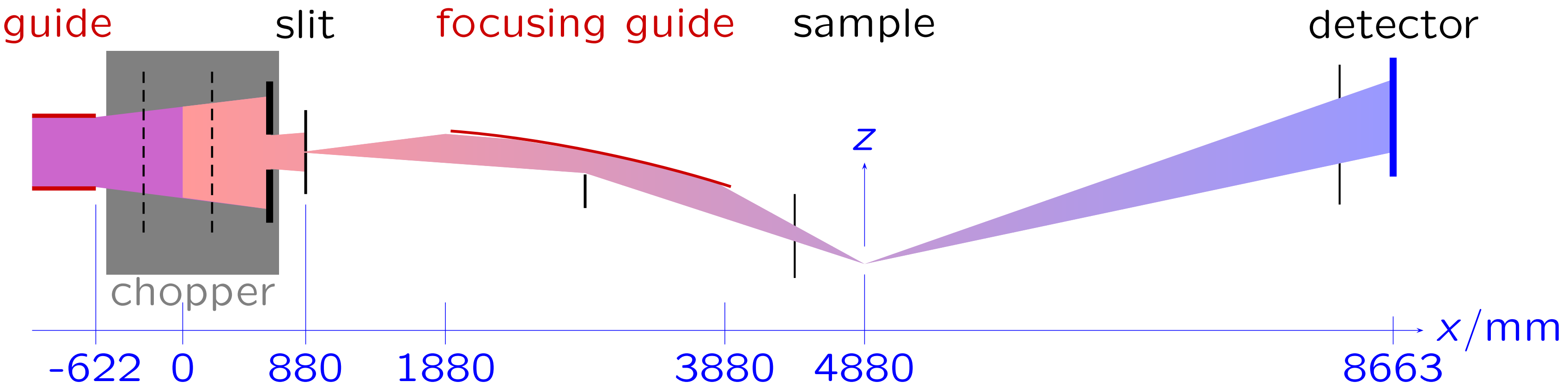
	probes	depth	
ARPES	surface	1 nm	
RIXS	bulk	100 nm	element specific
PNR	interfaces	1 000 nm	$\rho(z)$, $B_\perp$
XR	interfaces	100 nm	$\rho(z)$

conventional TOF set-up on Amor:



- Δq defined by flight-path length and slits
- energy-dispersive

selene set-up on Amor:



- Δq defined by flight-path length and position-sensitive detector
- energy- and angle-dispersive