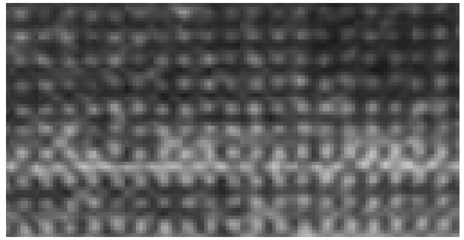
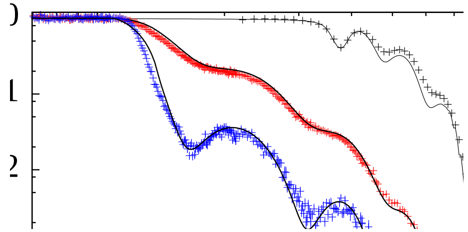
jochen stahn
laboratory for neutron scattering
PSI Paul Scherrer Institut

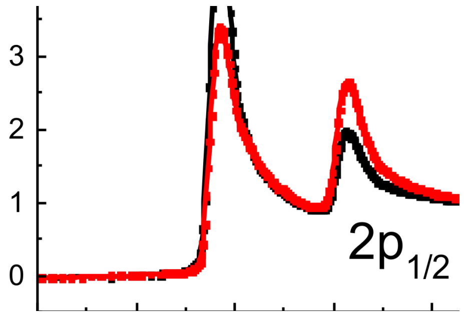
introduction to polarised neutron and resonant x-ray reflectometry

PSI summer school on condensed matter research
Zuoz, 8. – 13. august 2010
magnetic phenomena 12. 08. 2010

- motivation — magnetically dead layers
  - $\longrightarrow$ reflectometry — $\Rightarrow \rho(z)$
  - $\longrightarrow$ neutrons, γ — $\Rightarrow \mathbf{B}$
- reflectometry in general — only specular, no absorption
  - $\longrightarrow$ index of refraction — $n = 1 - \delta$
  - $\longrightarrow$ Fresnel reflectivity
  - $\longrightarrow$ multiple interfaces — matrix method
- neutron reflectivity
  - $\longrightarrow$ experimental set-up
  - $\longrightarrow$ measurement — Fe/Si, FeSi/GaAs interfaces
- resonant γ reflectivity
  - $\longrightarrow$ absorption — $n = 1 - \delta - i\beta$
    - $\circ$ intro to XMCD
  - $\longrightarrow$ experimental set-up
  - $\longrightarrow$ measurement — Pt/Co
- $\sum$

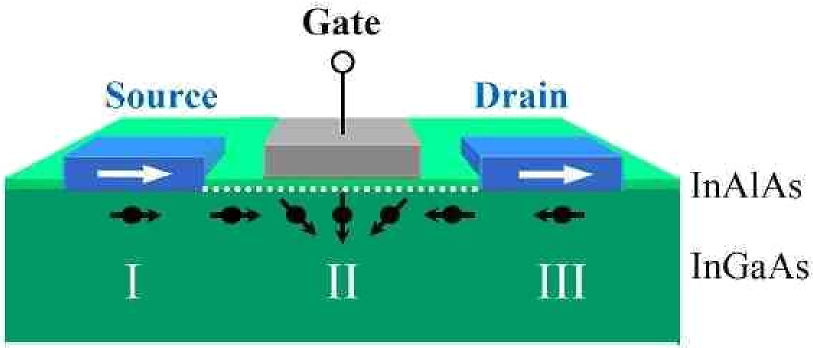






. . . the damn magnetically dead layers . . .

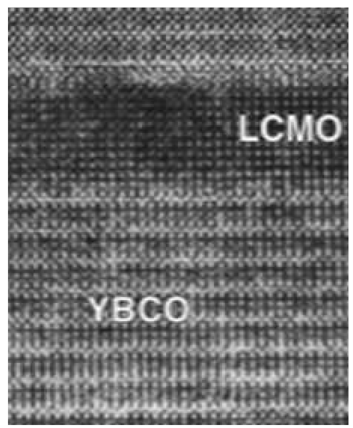
- down-scaling ⇒ thin magnetic films
  e.g. magnetic data storage



- spin polarised electron injection
  e.g. spin-injection in a spin-transistor

- conflicting properties at interfaces

$$\text{e.g. interface } \frac{\text{ferro} - \text{magnet}}{\text{superconductor}}$$



analogy to visible light

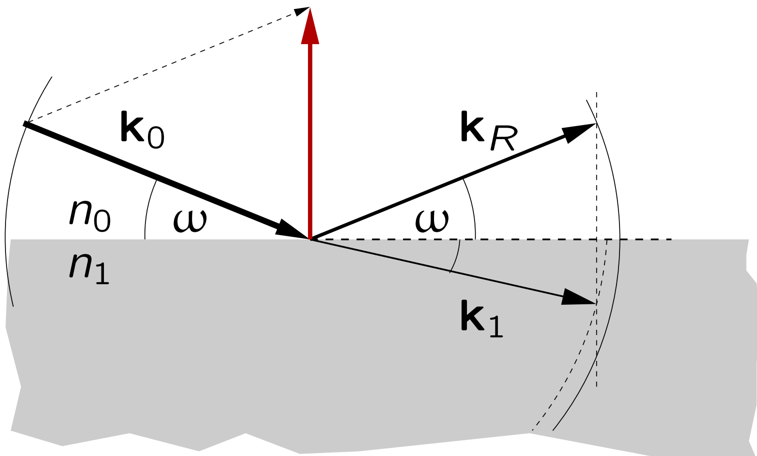
flat surfaces partly reflect light
→ picture of the boot

some media also transmit light
→ ground below the water

parallel interfaces
→ colorful soap bubbles

scientist's explanation:

- ○ index of refraction,
- ○ Fresnel reflectivity,
- ○ transmittance,
- ○ interference,
- ○ bla bla bla . . .

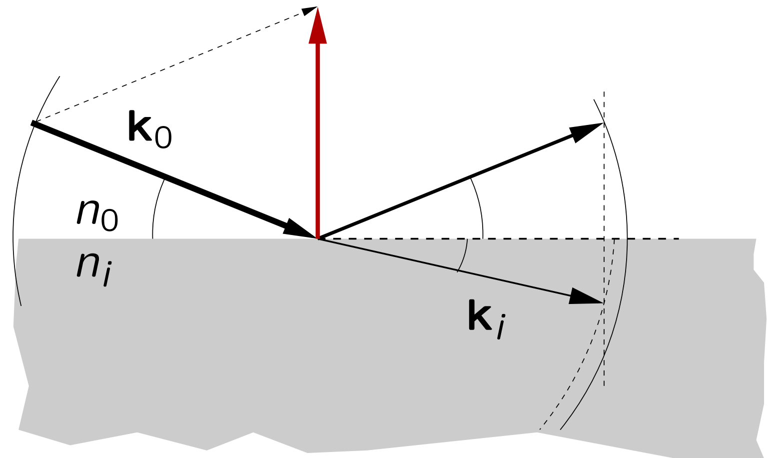


plane wave in a medium i:

$$\frac{\hbar^2}{2m}\frac{\mathrm{d}^2}{\mathrm{d}r^2}A\,\mathrm{e}^{\mathrm{i}k_i r} + (E-V_i)\,A\,\mathrm{e}^{\mathrm{i}k_i r} = 0$$

$$\frac{\hbar^2}{2m}(-k_i^2)\mathrm{e}^{\mathrm{i}k_i r} + (E-V_i)\,\mathrm{e}^{\mathrm{i}k_i r} = 0$$

$$\Rightarrow k_i^2 = (E-V_i)\frac{2m}{\hbar^2}$$



$$n_i^2 = \frac{k_i^2}{k_0^2}$$ by definition

$$= \frac{E-V_i}{E}$$ with $V_0 = 0$ (vacuum)

$$n_i = \sqrt{1-V_i/E}$$

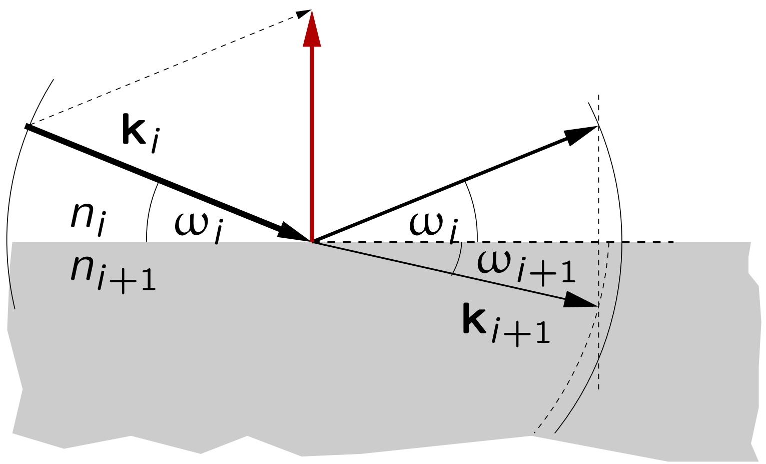
$$\approx 1 - V_i/2E$$ for $V_i \ll E$

$$:= 1 - \delta$$ $n_i - 1 \propto V_i \quad \Rightarrow$ **what is V_i?**

what is the reflected intensity?

assumptions:

- one interface, only
- ideally flat and sharp
- homogeneous in x and y
  $\Rightarrow$ only normal (z) components are relevant



continuity conditions for a plane wave impinging on the interface $i, i+1$:

$$\Psi_{z,i} = \Psi_{z,i+1}$$

$$\frac{\mathrm{d}}{\mathrm{d}z}\Psi_{z,i} = \frac{\mathrm{d}}{\mathrm{d}z}\Psi_{z,i+1}$$

with

$$\Psi_{z,j} = A_j^{\uparrow}\mathrm{e}^{\mathrm{i}k_{z,j}z} + A_j^{\downarrow}\mathrm{e}^{-\mathrm{i}k_{z,j}z}$$

$$\begin{aligned} k_{z,j} &= k_j \sin\omega_j \\ &= n_j k_0 \sin\omega_j \end{aligned}$$

reflectance

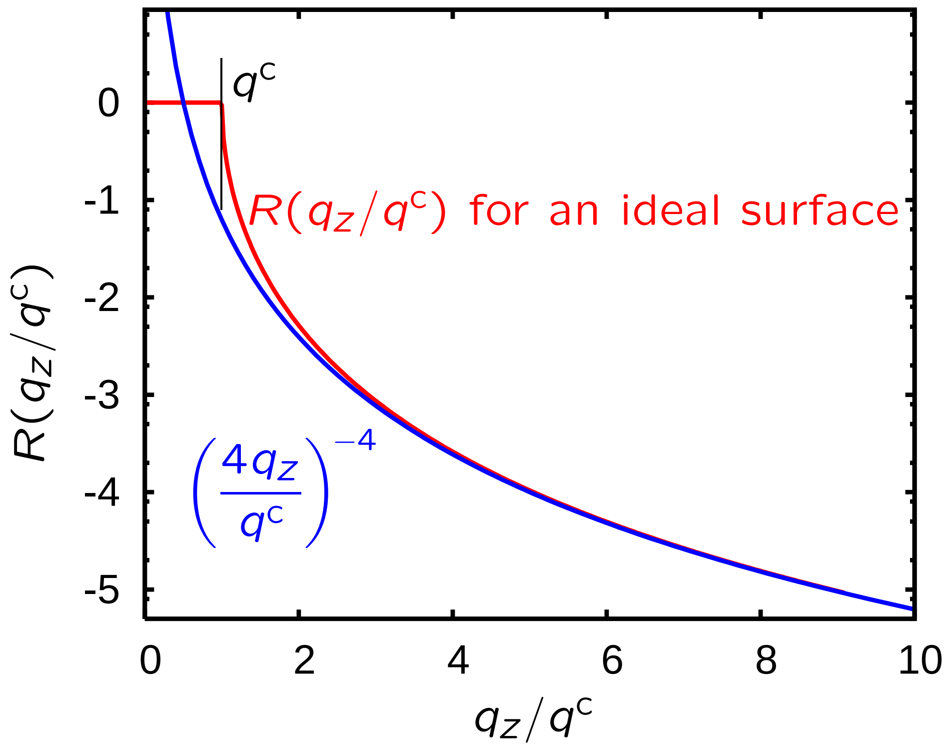
$$\begin{aligned} r_{i,i+1} &= \frac{A_i^{\uparrow}}{A_i^{\downarrow}} \\ &\vdots \\ &= \frac{n_i \sin\omega_i - n_{i+1}\sin\omega_{i+1}}{n_i \sin\omega_i + n_{i+1}\sin\omega_{i+1}} \end{aligned}$$

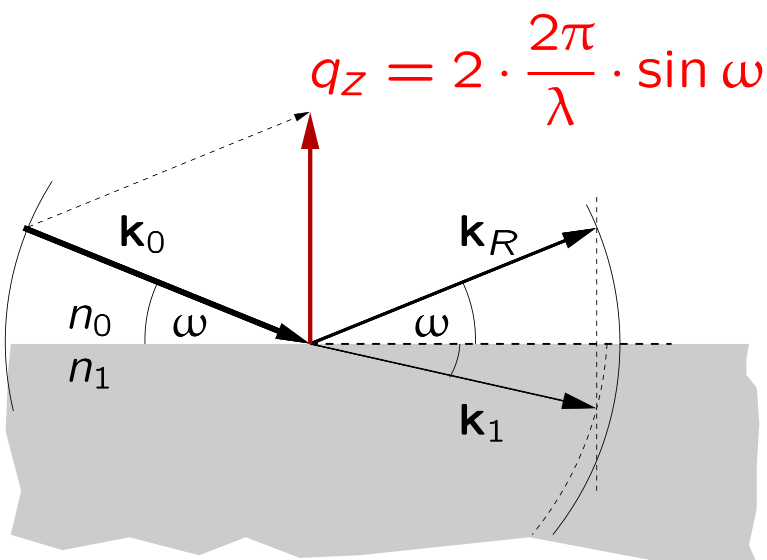
reflectance for $\omega_{i+1} > 0$

$$r_{i,i+1} = \frac{n_i \sin\omega_i - n_{i+1} \sin\omega_{i+1}}{n_i \sin\omega_i + n_{i+1} \sin\omega_{i+1}}$$

transmittance for $\omega_{i+1} > 0$

$$t_{i,i+1} = \frac{2\, n_i \sin\omega_i}{n_i \sin\omega_i + n_{i+1} \sin\omega_{i+1}}$$





air/solid interface for $q_z > q^c$

$$r_{0,1} = \frac{1 - \sqrt{1 - (q^c/q_z)^2}}{1 + \sqrt{1 - (q^c/q_z)^2}}$$

$$R(q_z) = |r_{0,1}(q_z)|^2$$

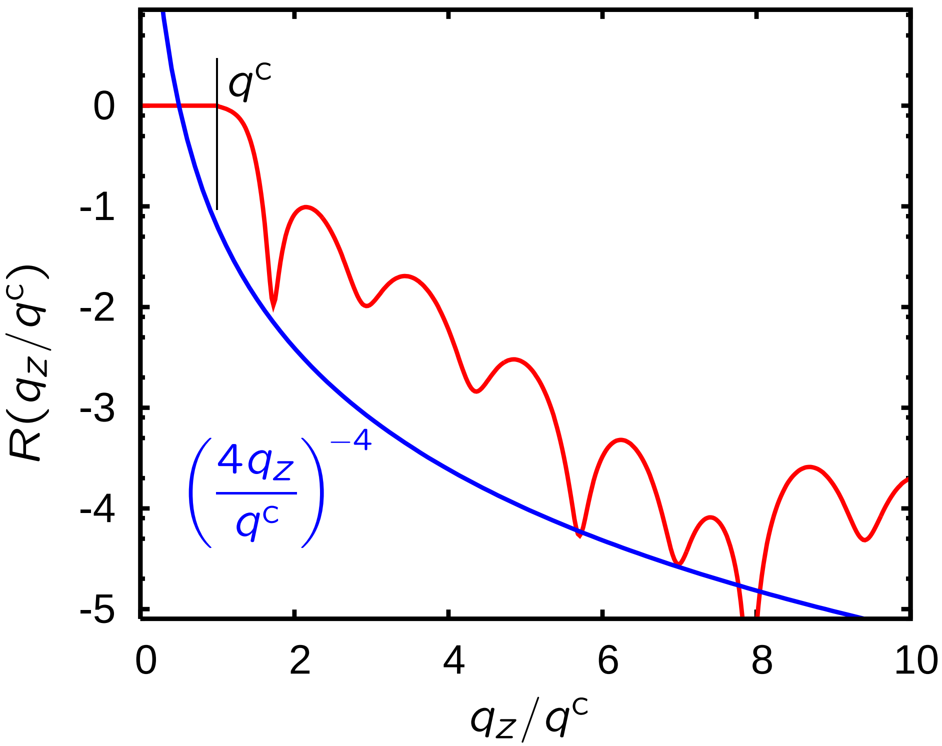
reflected intensity of a multilayer

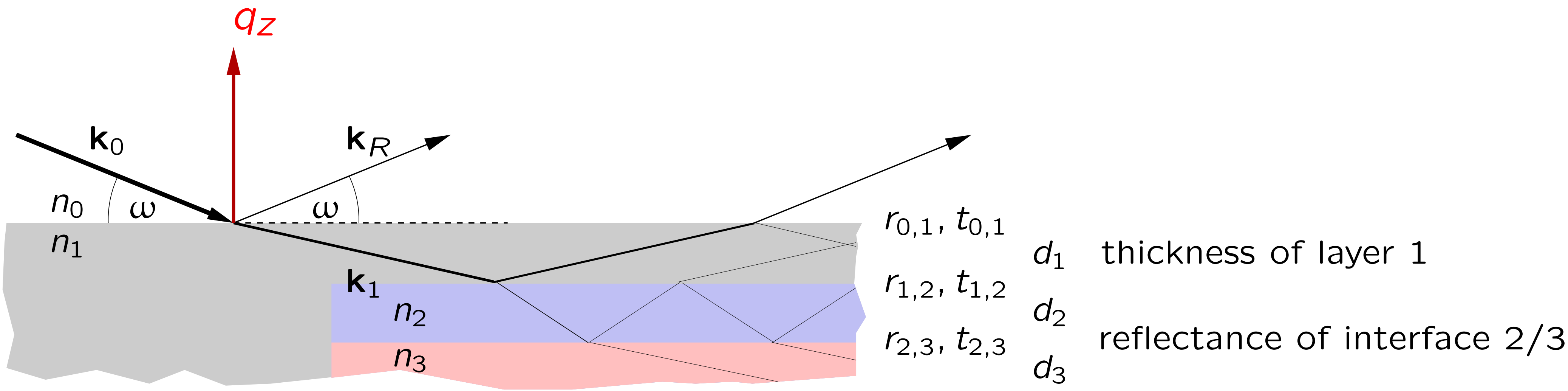
several parallel interfaces:

interference of all waves

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

what is $r(q_z)$ of a multilayer?





what is $r(q_z)$ of a multilayer?

$$\Psi_0(0) = \begin{pmatrix} A_0^{\uparrow} \\ A_0^{\downarrow} \end{pmatrix}$$

free choice of phase

$$= \begin{pmatrix} 1/t_{0,1} & r_{0,1}/t_{0,1} \\ r_{0,1}/t_{0,1} & 1/t_{0,1} \end{pmatrix} \begin{pmatrix} A_1^{\uparrow} \\ A_1^{\downarrow} \end{pmatrix}$$

continuity condition

$$= \mathbf{I}_{0,1} \begin{pmatrix} e^{ik_{z,1}d_1} & 0 \\ 0 & e^{-ik_{z,1}d_1} \end{pmatrix} \begin{pmatrix} A_1^{\uparrow} e^{-ik_{z,1}d_1} \\ A_1^{\downarrow} e^{ik_{z,1}d_1} \end{pmatrix}$$

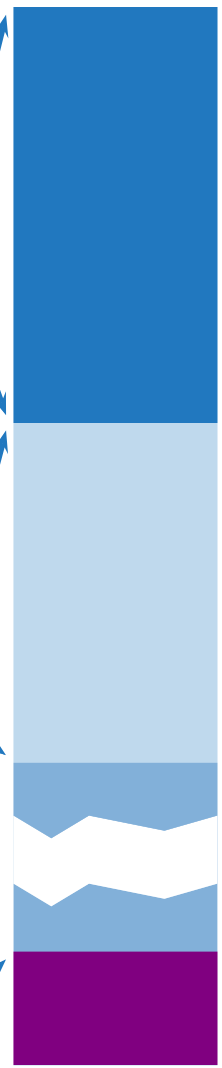
phase factor

$$= \mathbf{I}_{0,1}\mathbf{T}_1 \begin{pmatrix} 1/t_{1,2} & r_{1,2}/t_{1,2} \\ r_{1,2}/t_{1,2} & 1/t_{1,2} \end{pmatrix} \begin{pmatrix} A_2^{\uparrow} e^{-ik_{z,1}d_1} \\ A_2^{\downarrow} e^{ik_{z,1}d_1} \end{pmatrix}$$

$$= \mathbf{I}_{0,1}\mathbf{T}_1\mathbf{I}_{1,2} \begin{pmatrix} e^{ik_{z,2}d_2} & 0 \\ 0 & e^{-ik_{z,2}d_2} \end{pmatrix} \begin{pmatrix} A_2^{\uparrow} e^{-ik_{z,2}(d_1+d_2)} \\ A_2^{\downarrow} e^{ik_{z,2}(d_1+d_2)} \end{pmatrix}$$

$$\vdots$$

$$:= \mathbf{M} \begin{pmatrix} A_{\mathrm{substr}}^{\uparrow} e^{-ik_{z,\mathrm{substr}} \sum_i d_i} \\ A_{\mathrm{substr}}^{\downarrow} e^{ik_{z,\mathrm{substr}} \sum_i d_i} \end{pmatrix}$$

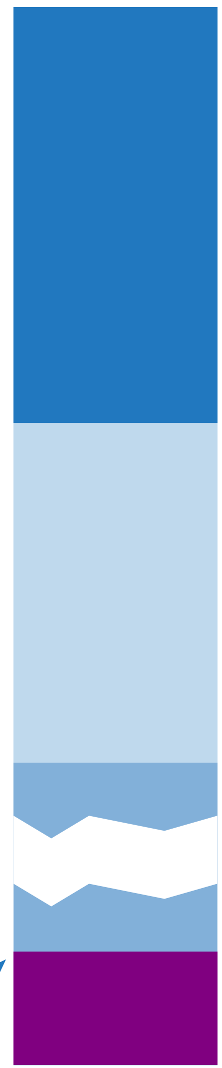


what is $r(q_z)$ of a multilayer?

$$\Psi_0(0) = \begin{pmatrix} A_0^{\uparrow} \\ A_0^{\downarrow} \end{pmatrix} = \mathbf{M} \begin{pmatrix} 0 \\ A_{\text{substr}}^{\downarrow}\, e^{\mathrm{i} k_{z,\text{substr}} \sum_i d_i} \end{pmatrix}$$

$$r(q_z) = A_0^{\uparrow}/A_0^{\downarrow} = \frac{M_{1\,2}\, A_{\text{substr}}^{\downarrow}\, e^{\mathrm{i} k_{z,\text{substr}} \sum_i d_i}}{M_{2\,2}\, A_{\text{substr}}^{\downarrow}\, e^{\mathrm{i} k_{z,\text{substr}} \sum_i d_i}} = \frac{M_{1\,2}(q_z)}{M_{2\,2}(q_z)}$$

there is no upcoming wave



calculation of $M_{1\,2}(q_z)$ and $M_{2\,2}(q_z)$ is trivial . . .

. . . if all n_i and d_i are known!

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

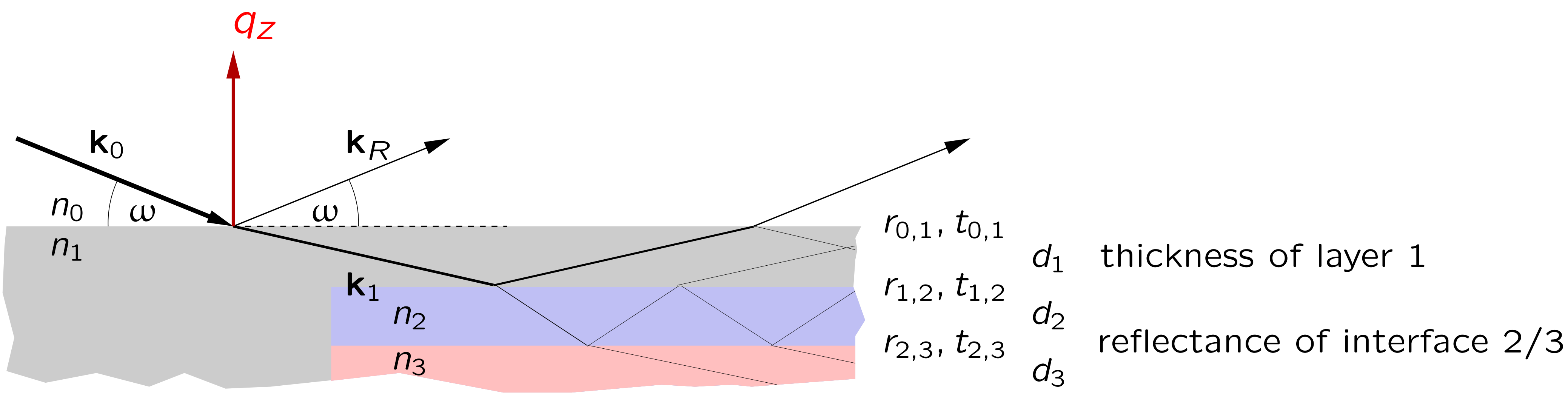
$\Rightarrow$ all phase information is lost

$\Rightarrow$ one way road:

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

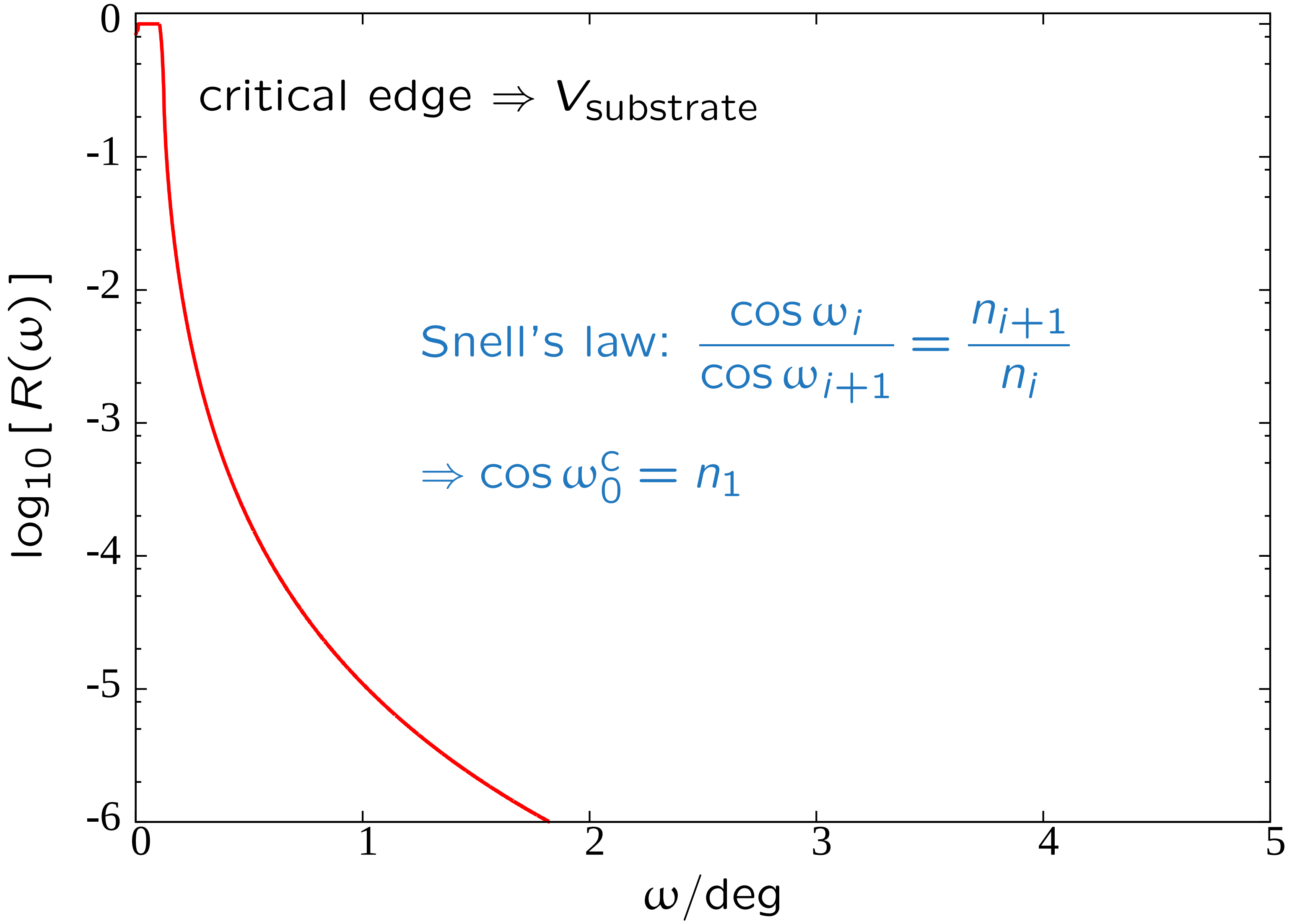
real effects to be taken into account:

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

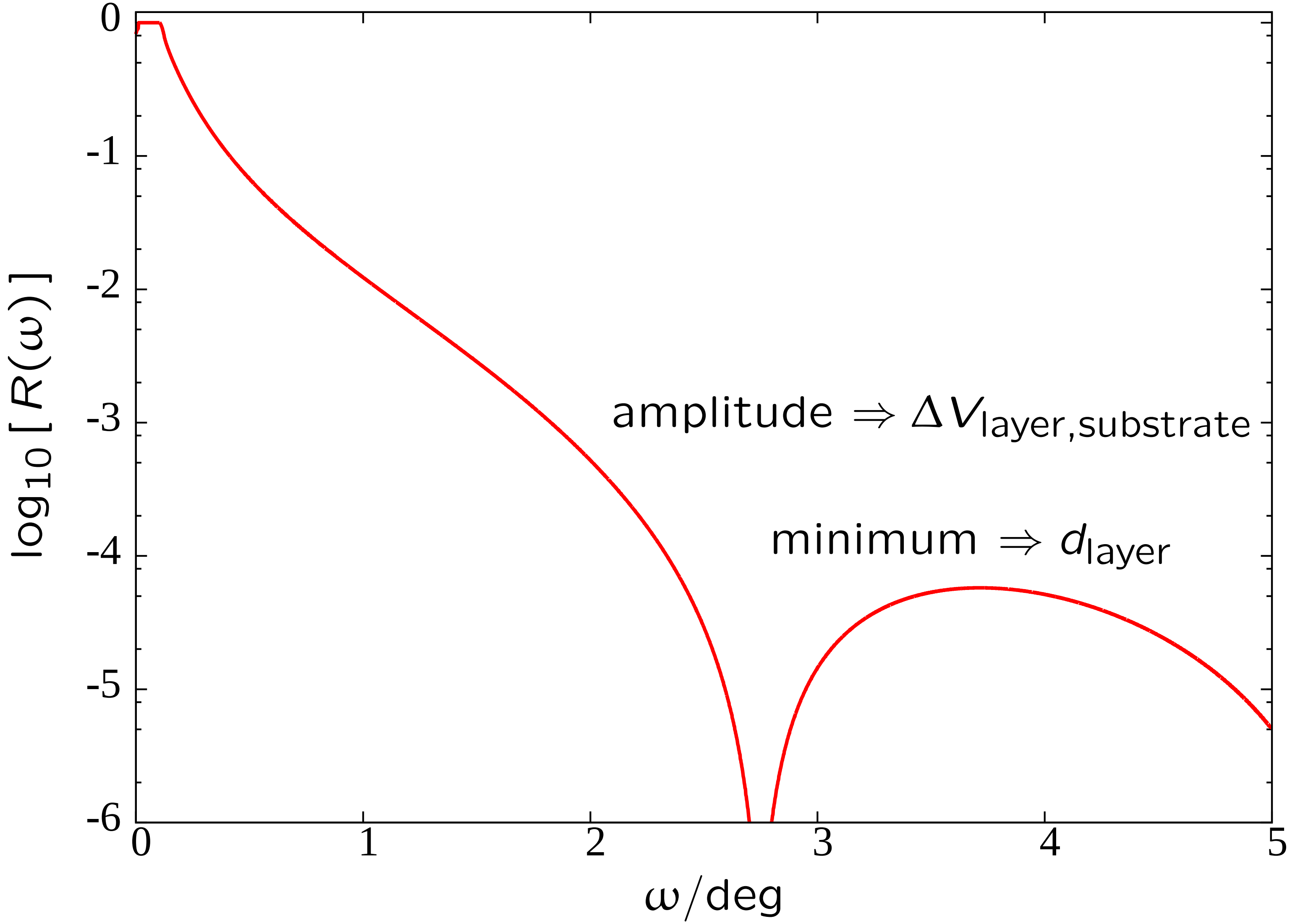


simulated reflectivity

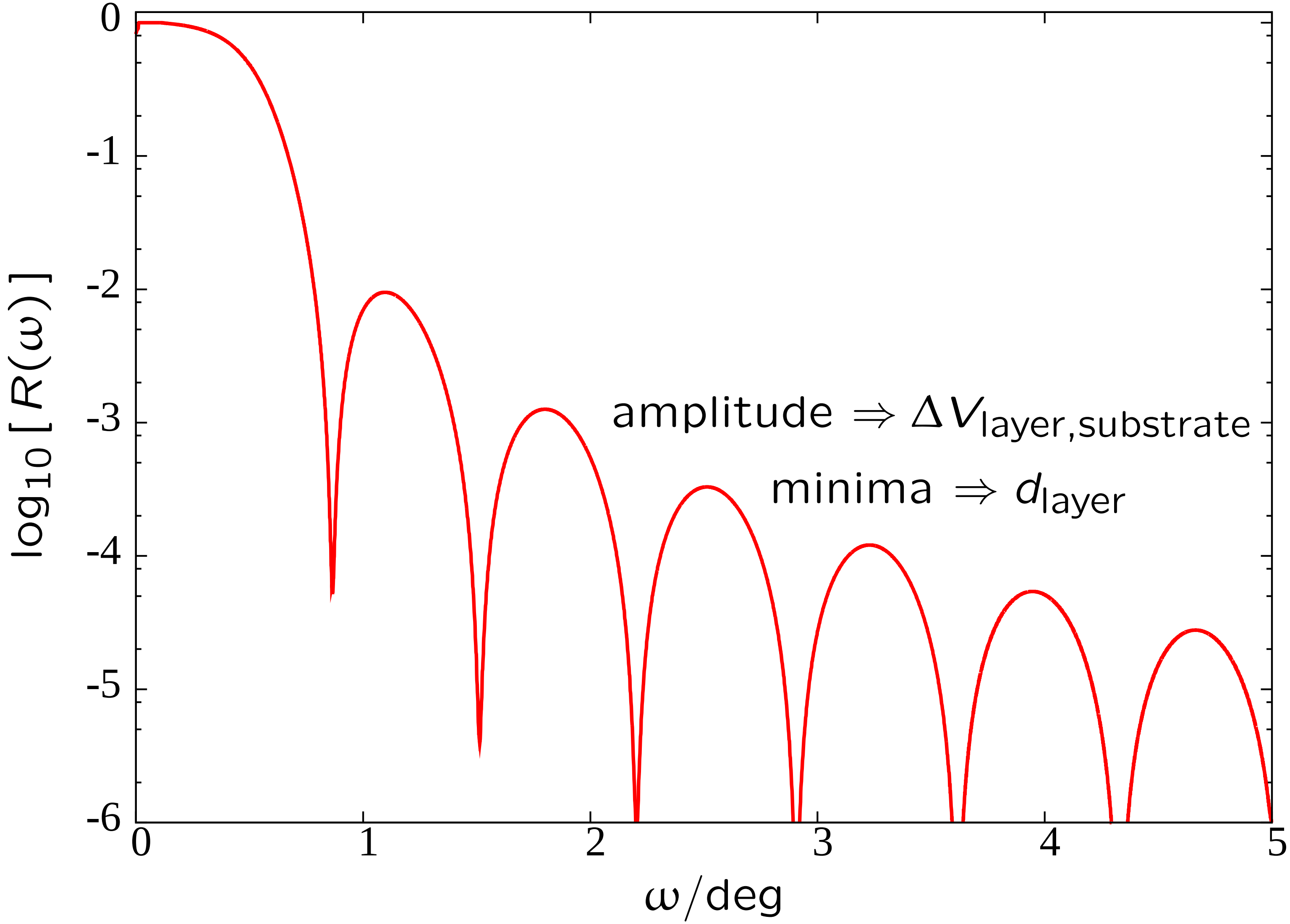
. . . of a surface

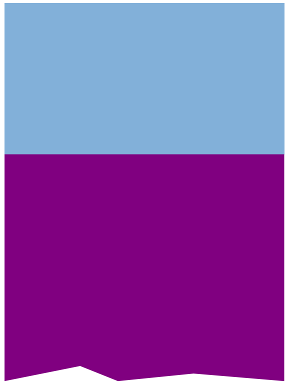


. . . of a thin layer

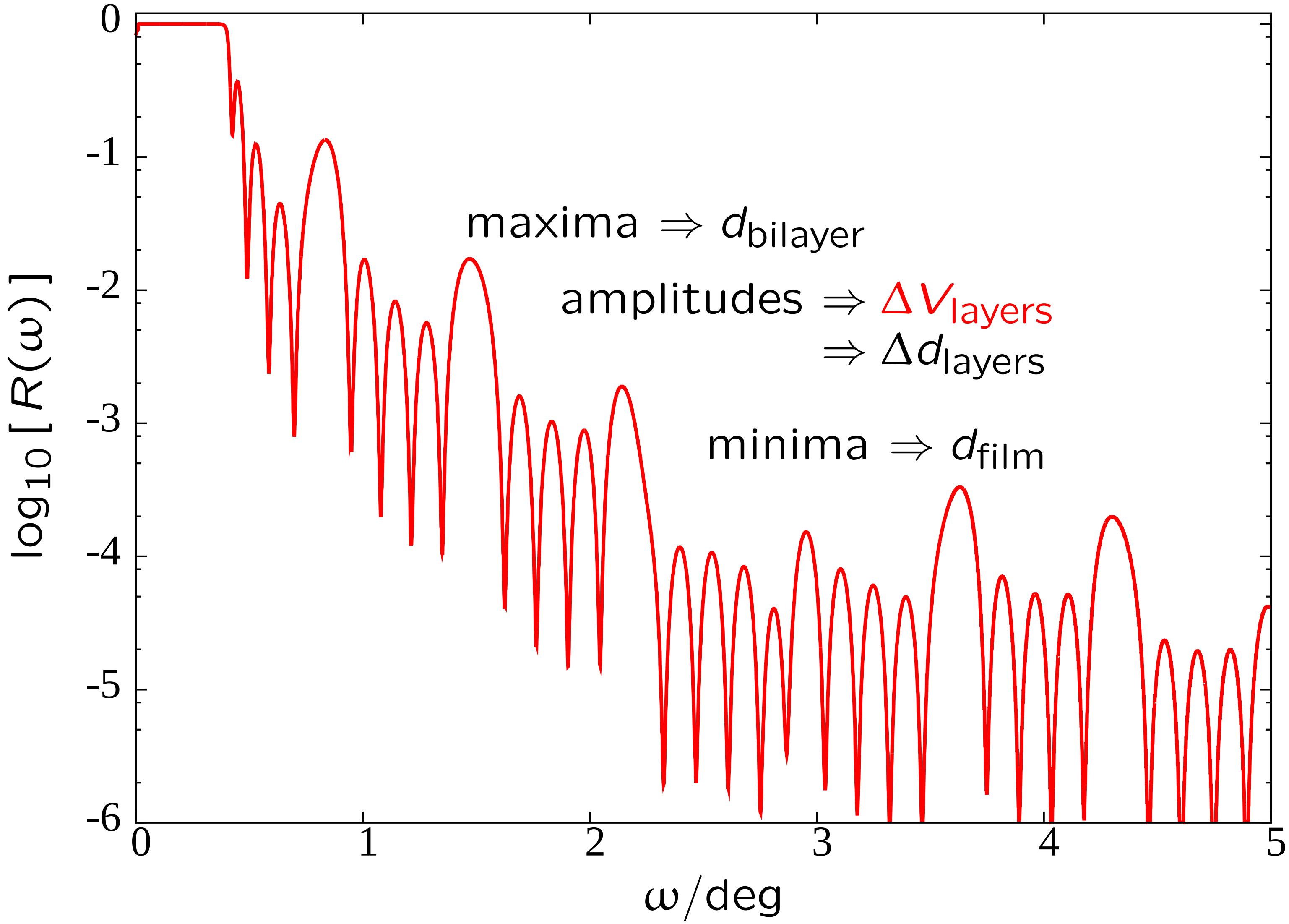


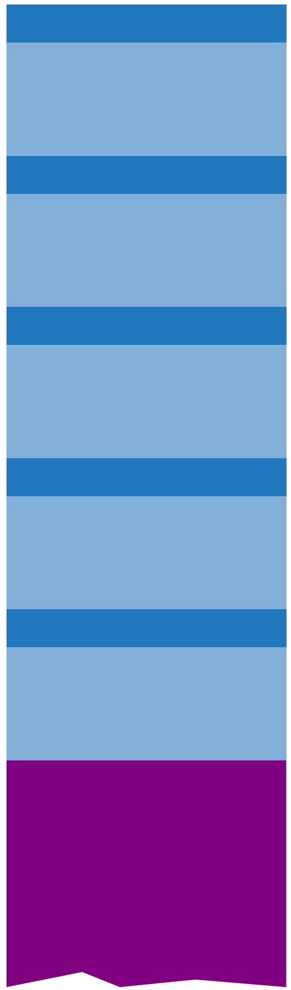
. . . of a thick layer





. . . of a periodic multilayer





what is V_i for neutrons?

interaction neutron / nucleus j

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

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

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

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

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

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

$\boldsymbol{\mu} \uparrow\uparrow \mathbf{B} \Rightarrow V^{\text{m}} = +\mu B$
$\boldsymbol{\mu} \uparrow\downarrow \mathbf{B} \Rightarrow V^{\text{m}} = -\mu B$
$\boldsymbol{\mu} \perp \mathbf{B} \Rightarrow$ spin-flip scattering

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

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

some numbers

$$\delta = 1 - n = \frac{\lambda^2}{2\pi}(\rho^b + \rho^m)$$

Ni: $\rho^b = 9.4 \cdot 10^{-6}\,\text{Å}^{-2}$

$\Rightarrow \delta^{nuc} = 3.75 \cdot 10^{-5}, \lambda = 5\,\text{Å}$ $\delta \ll 1$

$\Rightarrow \omega^c \approx 0.5^\circ$ small angles of incidence!

Fe: $\rho^m \approx 6 \cdot 10^{-6}\,\text{Å}^{-2}$

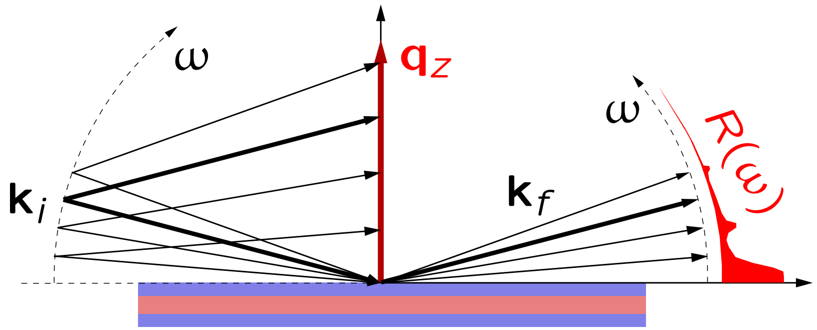
$\Rightarrow \delta^m \approx 2.4 \cdot 10^{-5}, \lambda = 5\,\text{Å}$ $\rho^m \approx \rho^b$

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

angle-dispersive set-up

variation of ω with fixed λ

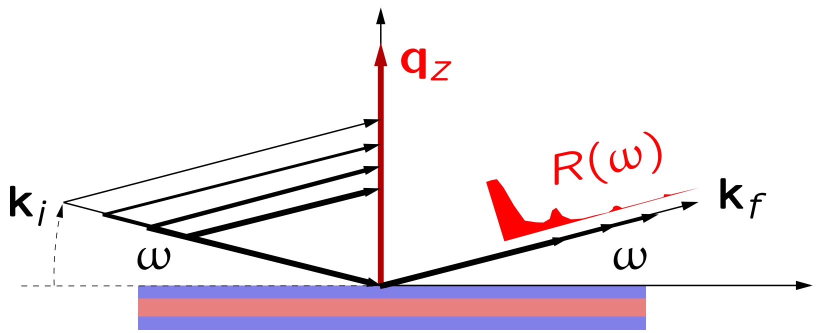
detection under 2ω



energy-dispersive set-up

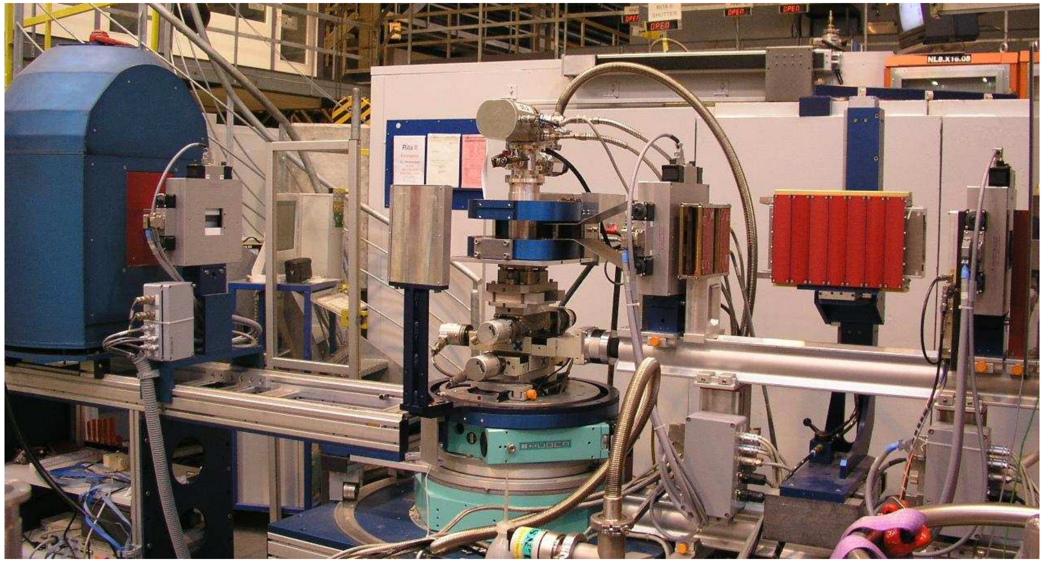
variation of λ with fixed ω

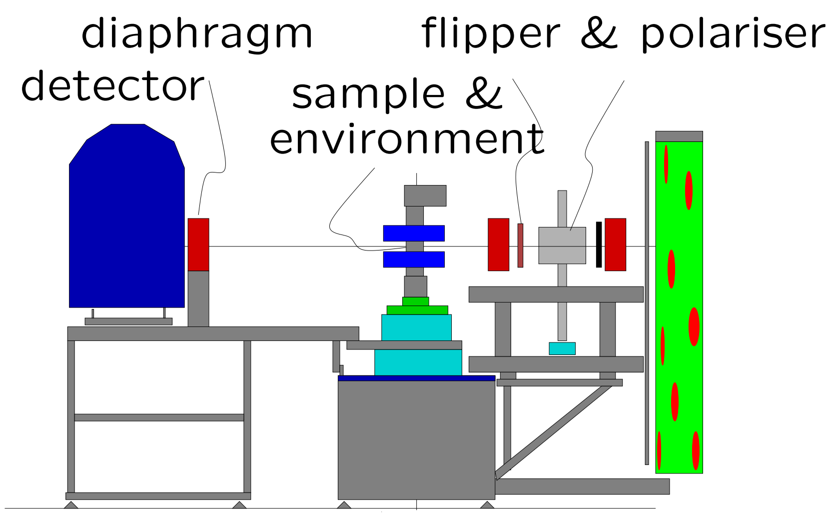
detection via time-of-flight

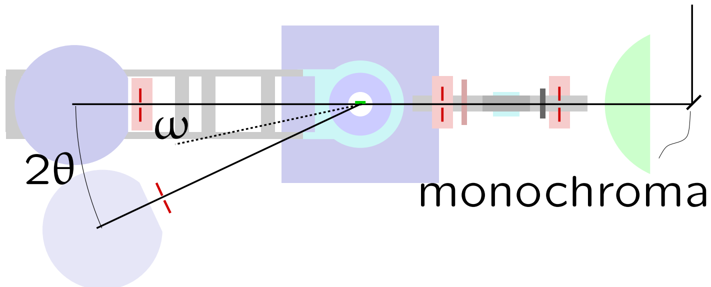


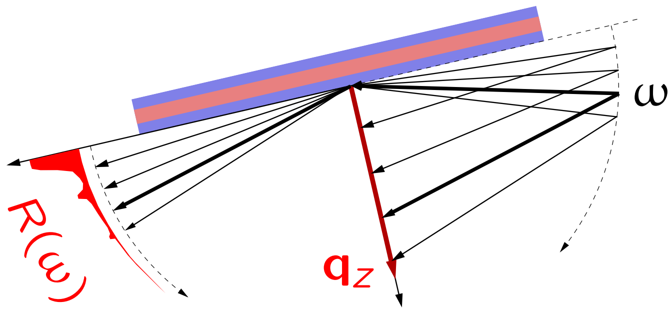
neutron reflectometer

instrument: morpheus at SINQ



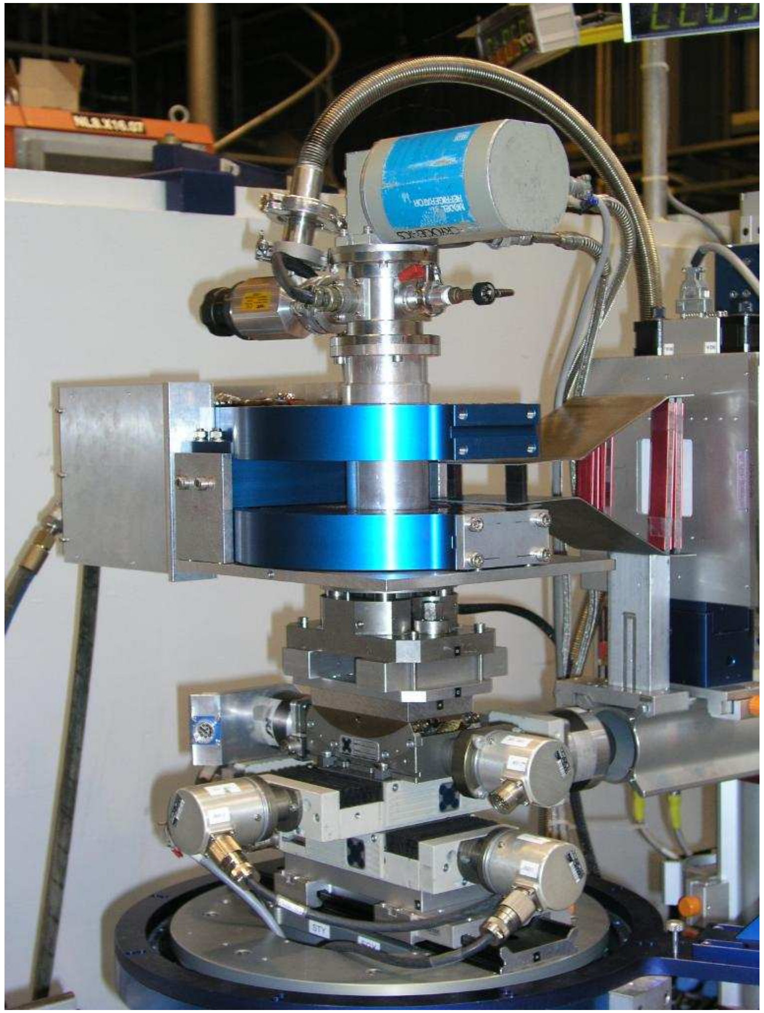






cooling with a
closed cycle refrigerator $8\,\mathrm{K} < T < 300\,\mathrm{K}$

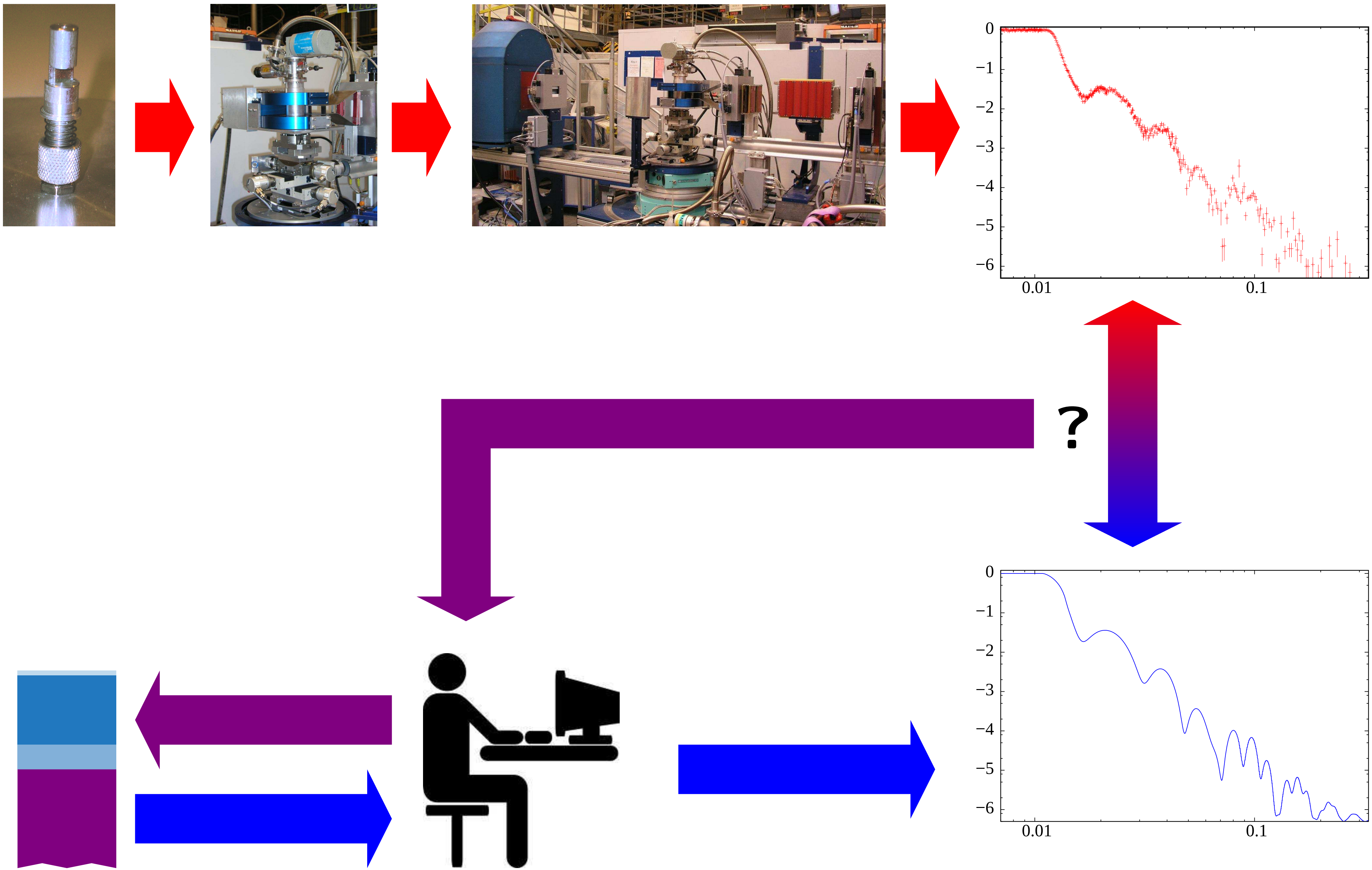
application of an external magnetic field with
Helmholtz coils $-1000\,\mathrm{Oe} < H < 1000\,\mathrm{Oe}$



tilt- and
translation stages
for alignment

and **sample**

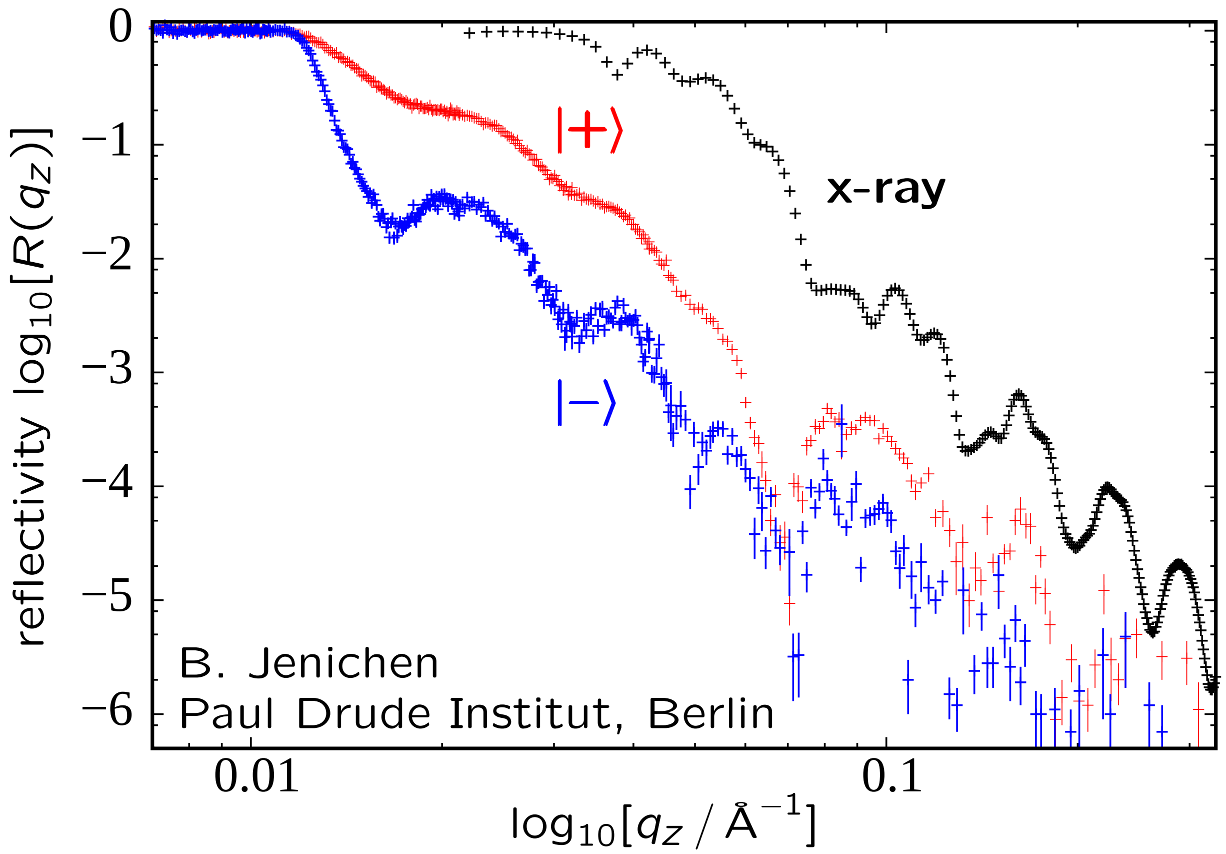
from the sample to $\rho(z)$

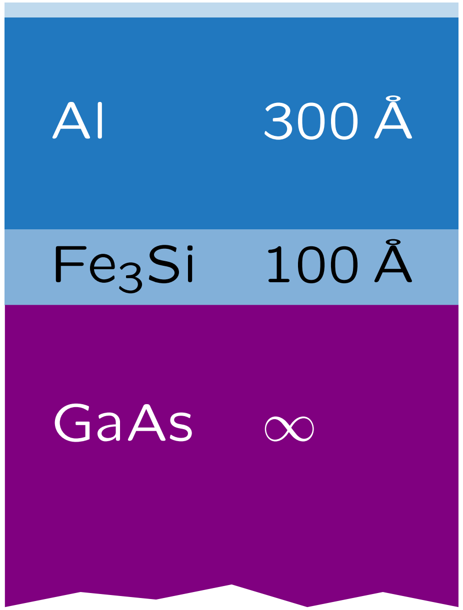


reflectometry with
non-resonant **x-rays** and
polarised neutrons

on a

Fe_3Si film on GaAs





sample size: $5 \times 5\,mm^2$
measurement time: 24 h neutron
1 h x-ray

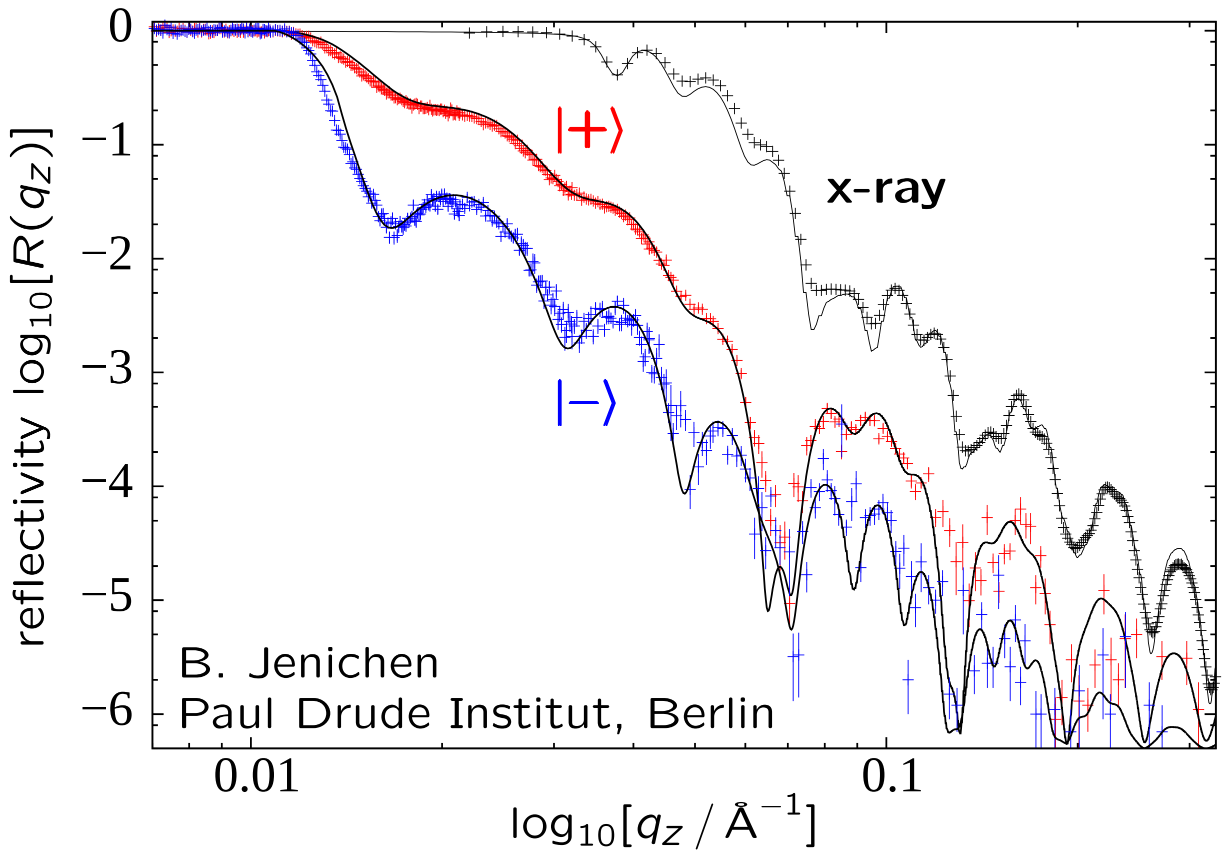
reflectometry with

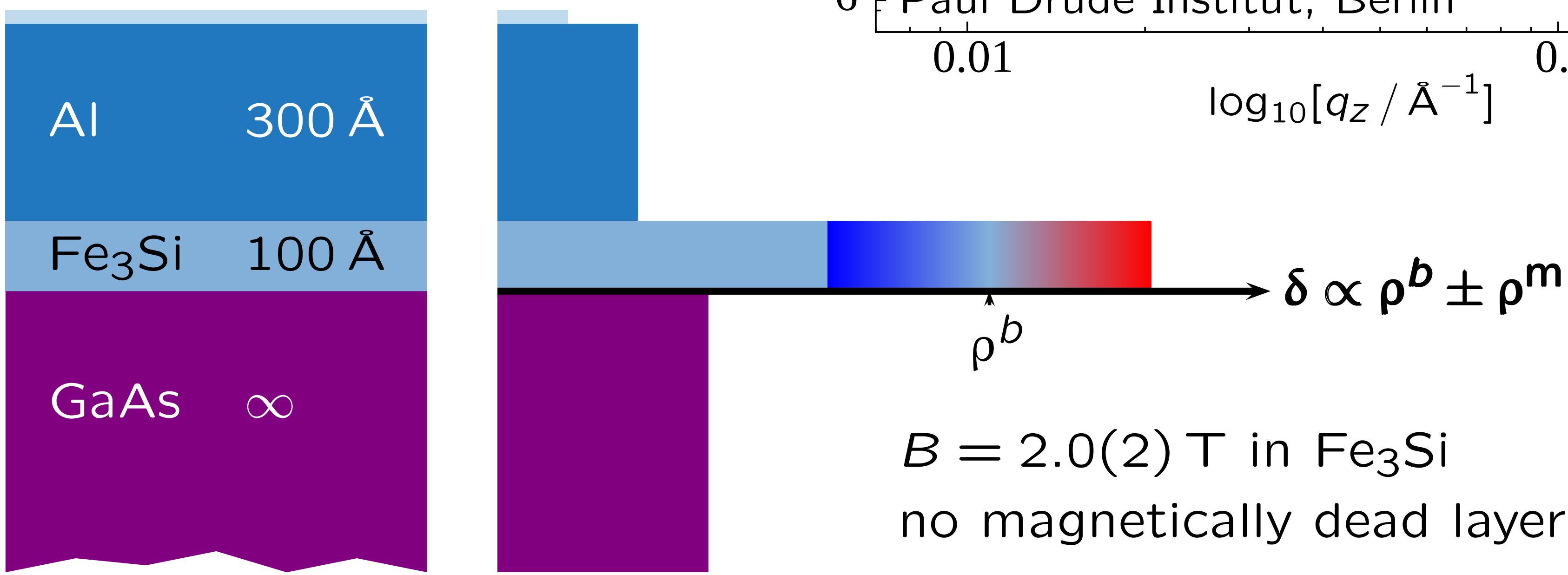
non-resonant **x-rays** and

polarised neutrons

on a

Fe_3Si film on GaAs





$B = 2.0(2)$ T in Fe_3Si

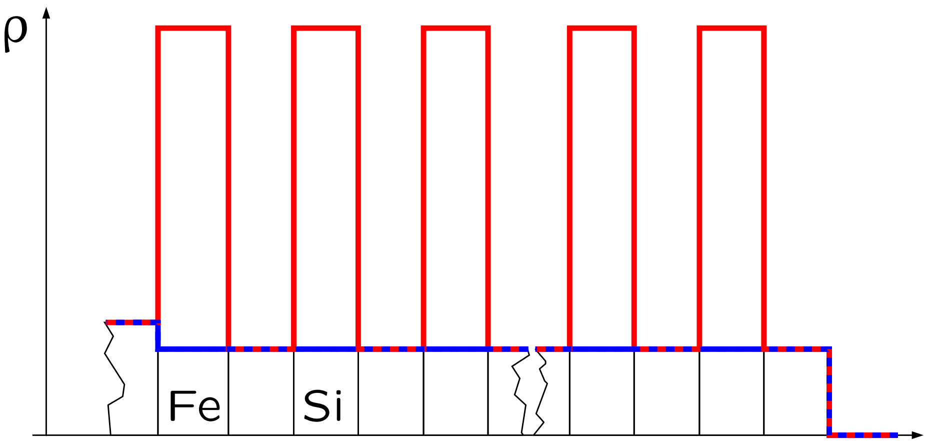
no magnetically dead layer detectable

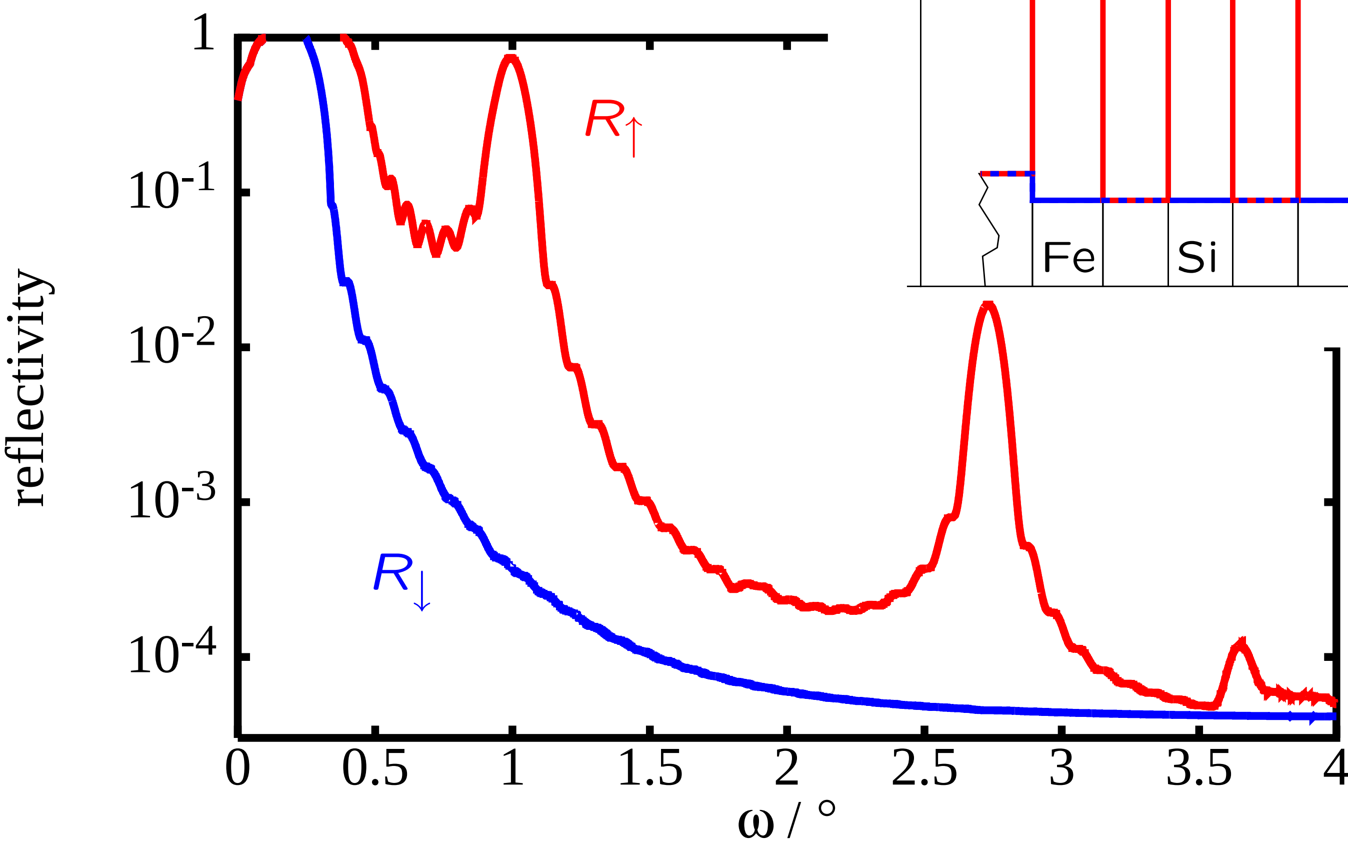
Fe/Si multilayer (a neutron polariser)

ideal case:

$$\rho^{b}_{\mathrm{Fe}} + \rho^{\mathrm{m}}_{\mathrm{Fe}} \gg \rho_{\mathrm{Si}}$$

$$\rho^{b}_{\mathrm{Fe}} - \rho^{\mathrm{m}}_{\mathrm{Fe}} = \rho_{\mathrm{Si}}$$

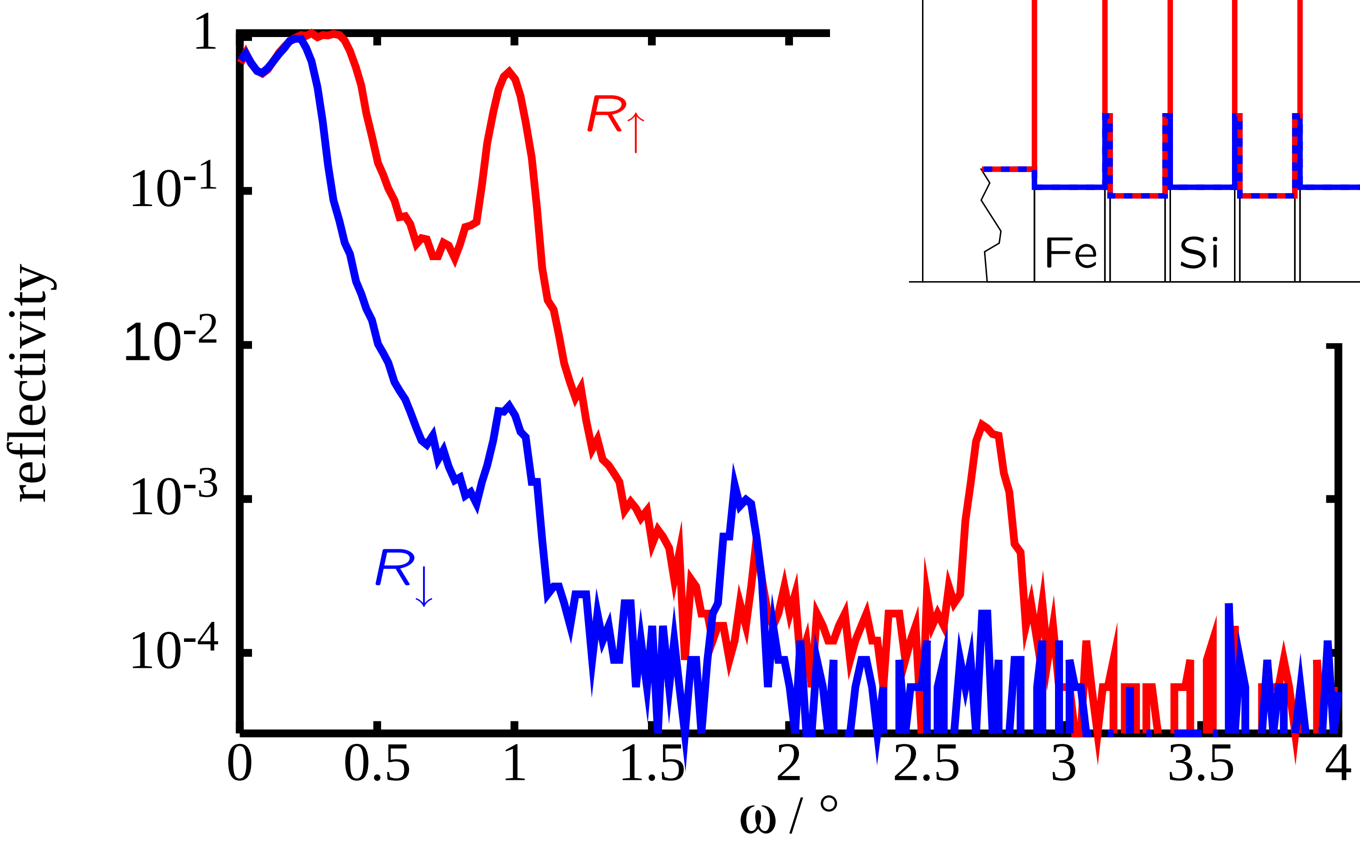


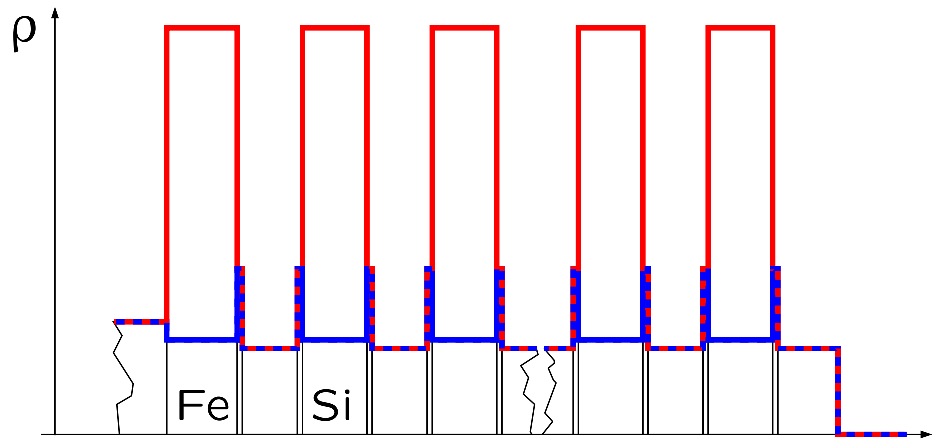


Fe/Si multilayer (a neutron polariser)

reality: interdiffusion leads to 5 Å thin magnetically dead layers

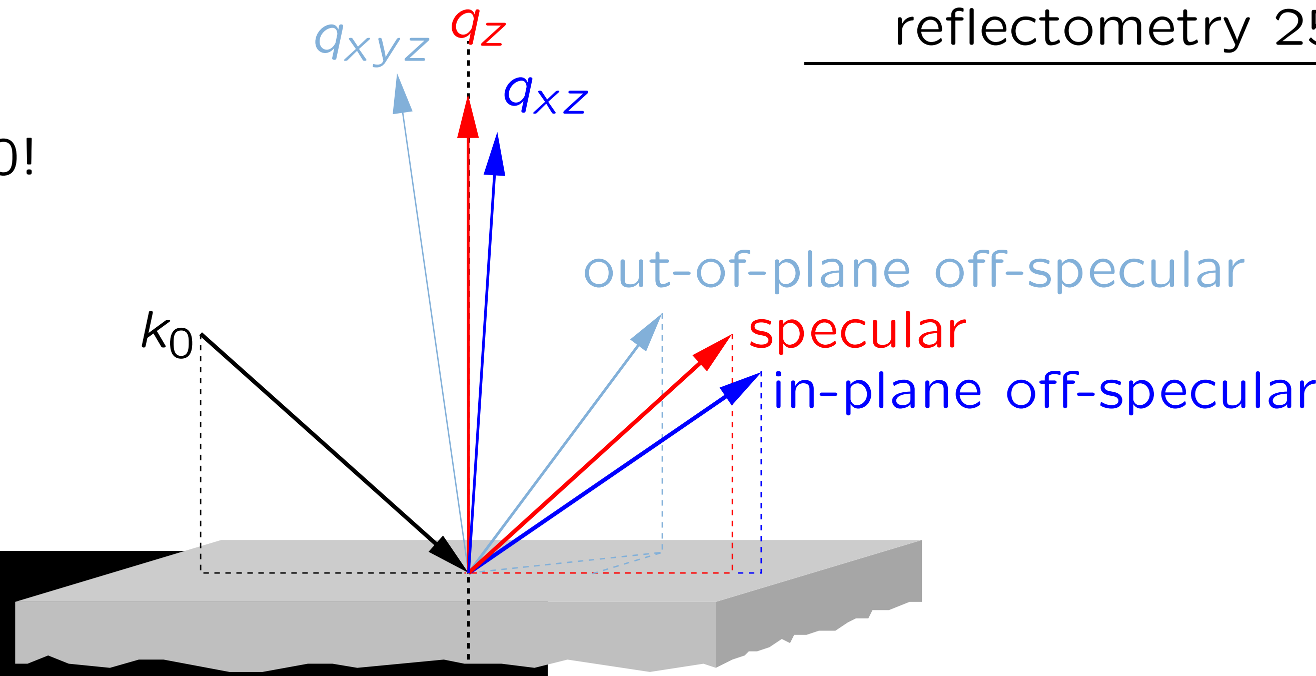
sample size: $70 \times 50\,\mathrm{mm}^2$
measurement time: 1 h

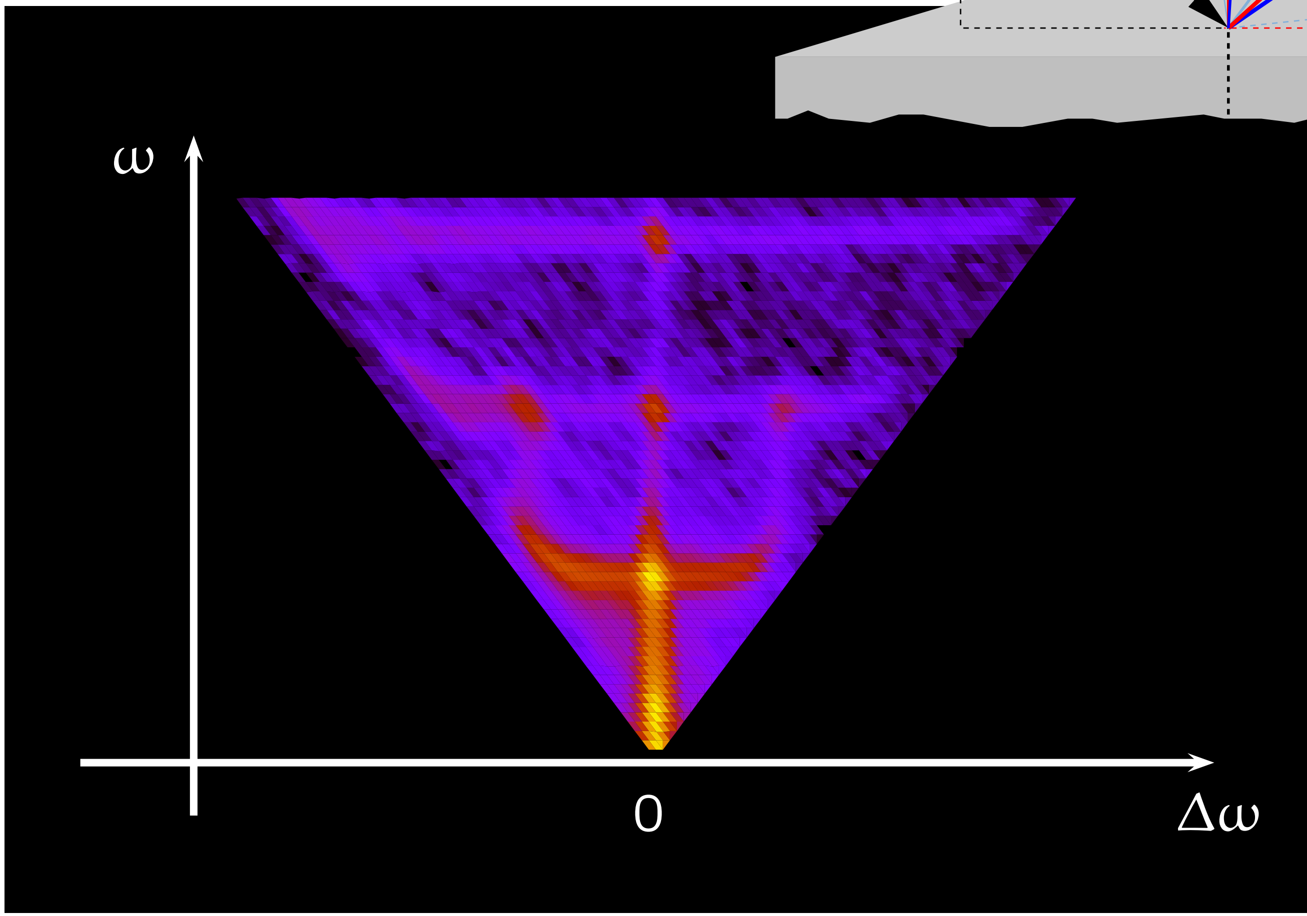




off-specular scattering

$\rho = \rho(x, z) \Rightarrow R(q_x \neq 0) \neq 0!$





Ni/Ti multilayer
(non magnetic!)

resolution in x: $\approx 0.01^\circ$
resolution in y: $> 1^\circ$
$\Rightarrow$ integrated over y

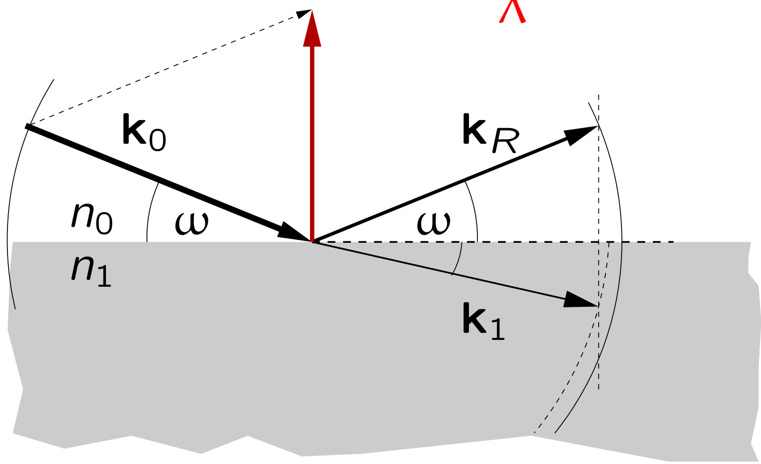
specular polarised reflectometry

probes magnetic and structural depth profiles

with

atomic to sub-μm resolution

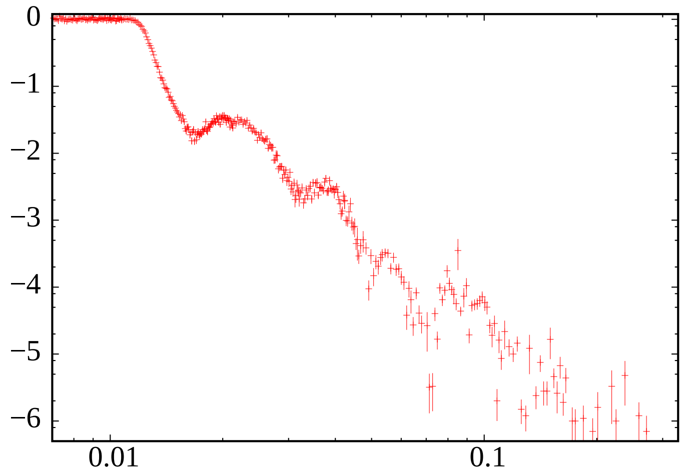
$$q_z \propto \frac{\sin\omega}{\lambda}$$



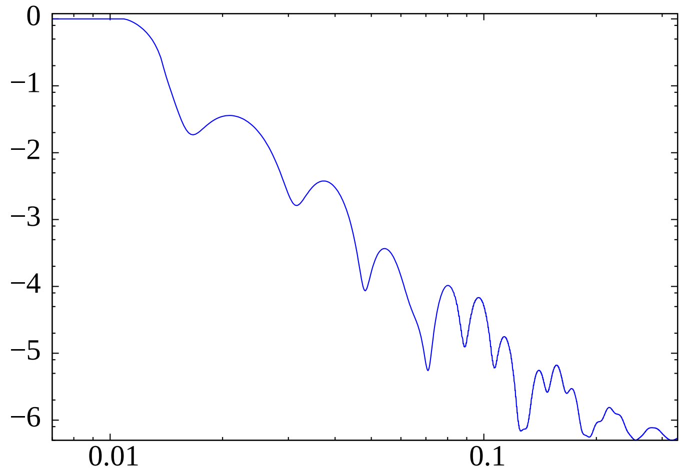
$$n = n(\rho^b, \boldsymbol{\mu}\mathbf{B})$$

in-plane magnetism

isotope selective

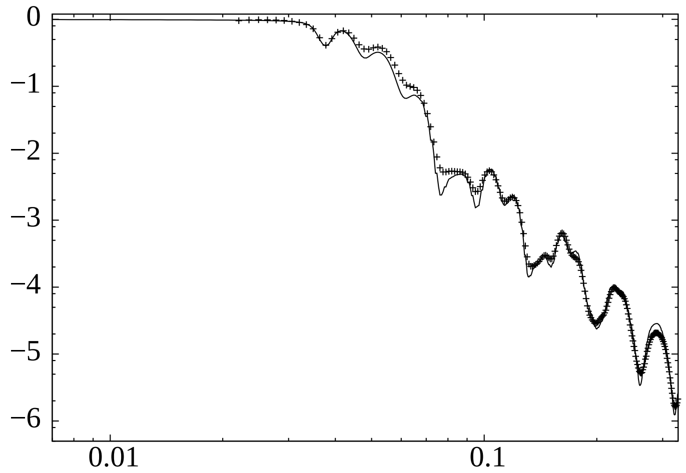


data analysis by comparison to calculated profile(s)



off resonance: $\frac{\rho_{x-ray}^{magnetic}}{\rho^{electron}} < 10^{-2}$

(neutrons: $\rho^m \approx \rho^b$)



but:

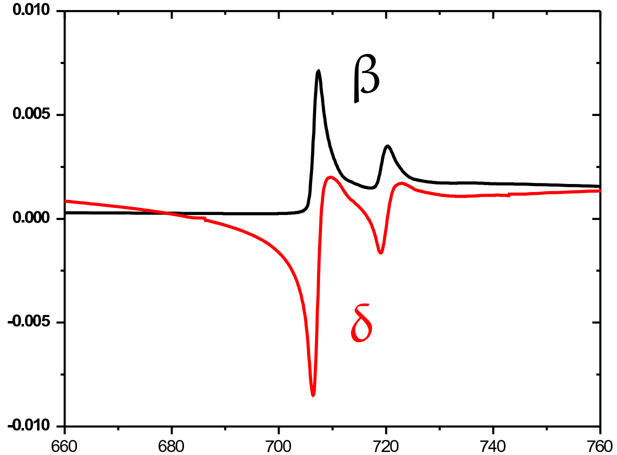
$n = 1 - \delta - i\beta$

and at resonances (close to absorption edges):

δ and β depend on the – photon energy

– photon polarisation

– density of states (DOS)

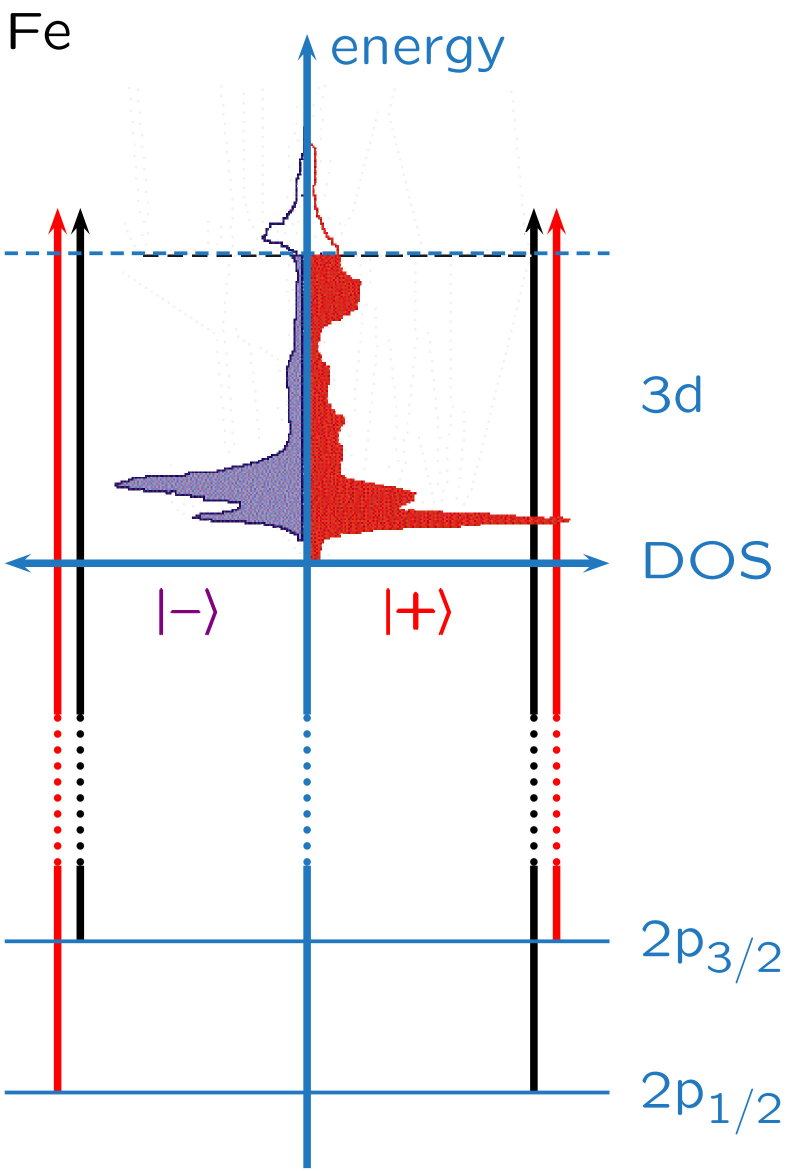


PRL **61**, 1245 (1988)

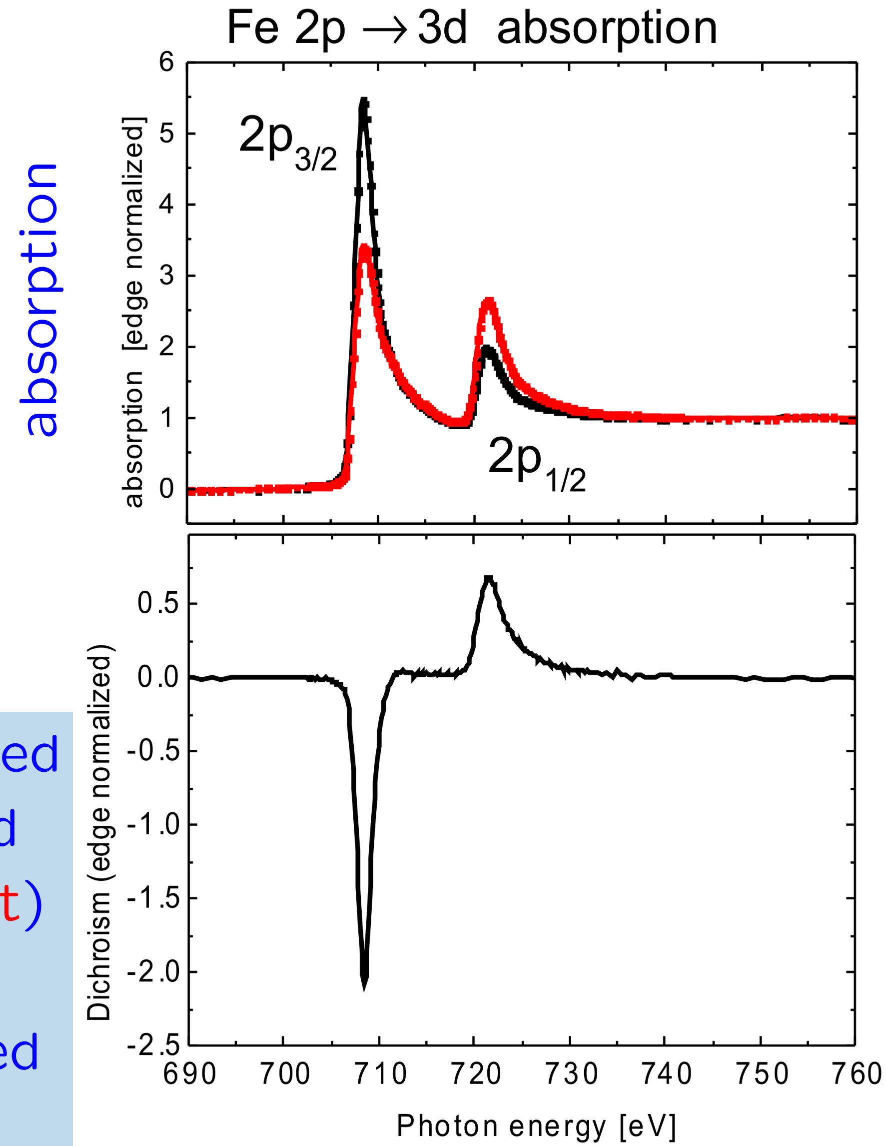
used by XMCD (x-ray magnetic circular dichroism) to probe atomic magnetic states

$\Rightarrow$ XMCD + reflectometry $\longrightarrow$ depth-resolved magnetic information

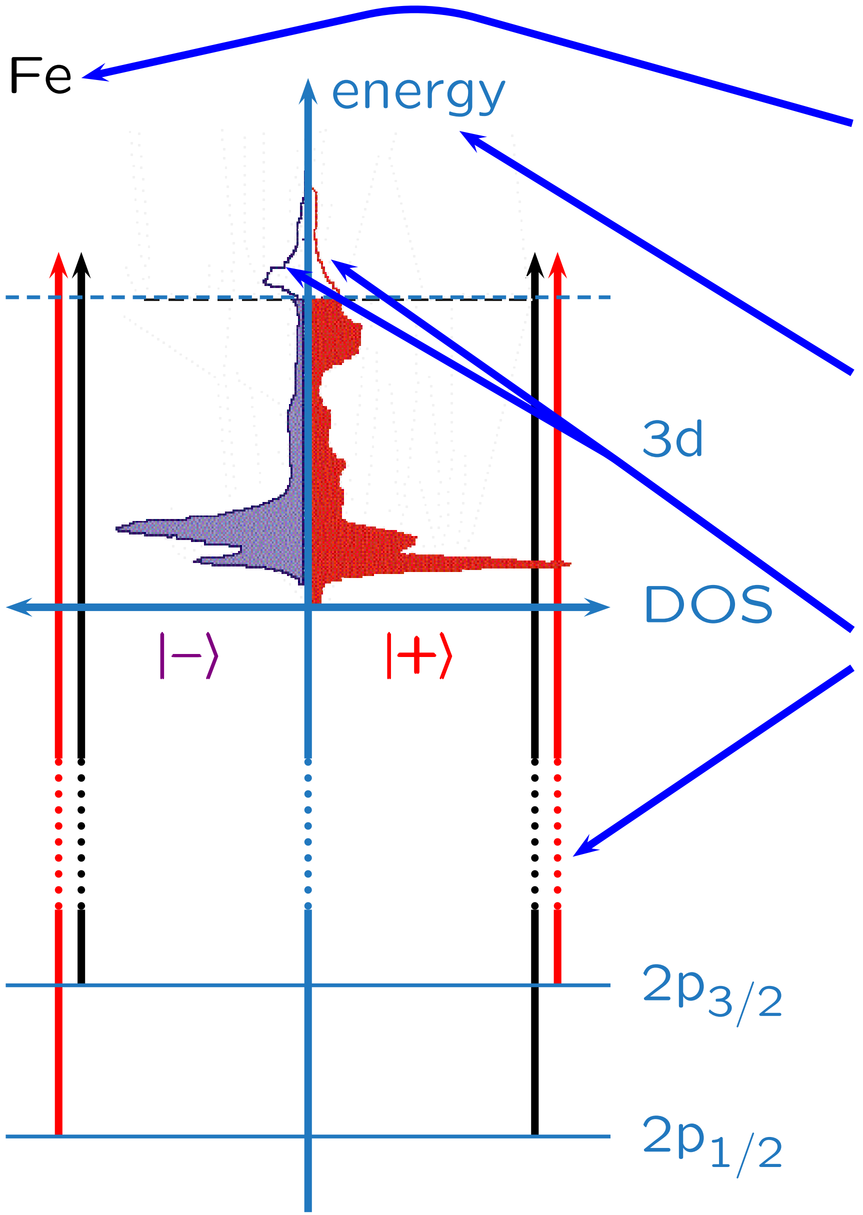
x-ray magnetic circular dichroism



transition allowed for $\Delta l = \pm 1$ and
$\Delta m = +1$ (right)
$\Delta m = -1$ (left)
circular polarised light



x-ray magnetic circular dichroism



- absorption energy depends on element

  ⇒ element specific
- tunable x-ray energy needed

  ⇒ synchrotron
- absorption cross section depends on DOS and transition rules

  ⇒ detection of magnetism

  separation of spin and orbit

XMCD + reflectometry

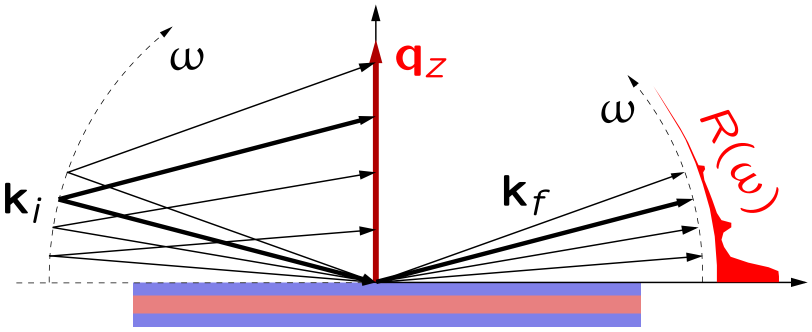
reflectometry measurements

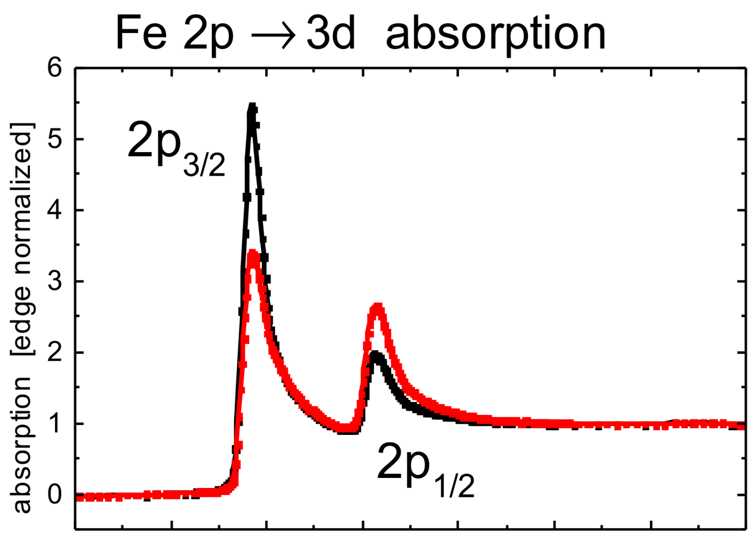
- at both absorption peaks
- with both circular polarisations

+

XMCD and absorption measurements

to get optical constants





data interpretation:

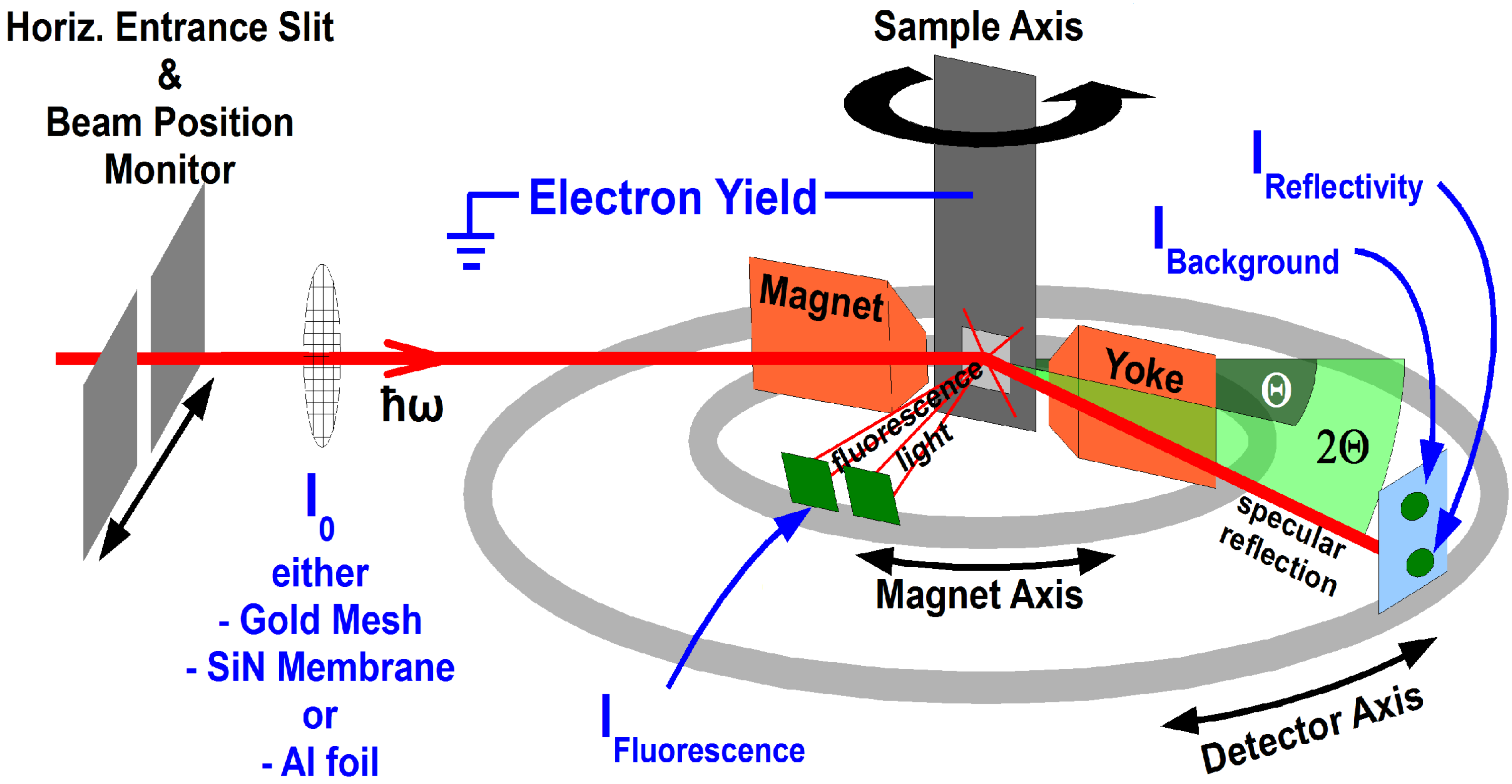
analogue to n-reflectometry

but with $n = 1 - \underbrace{\delta - i\beta}$

including:

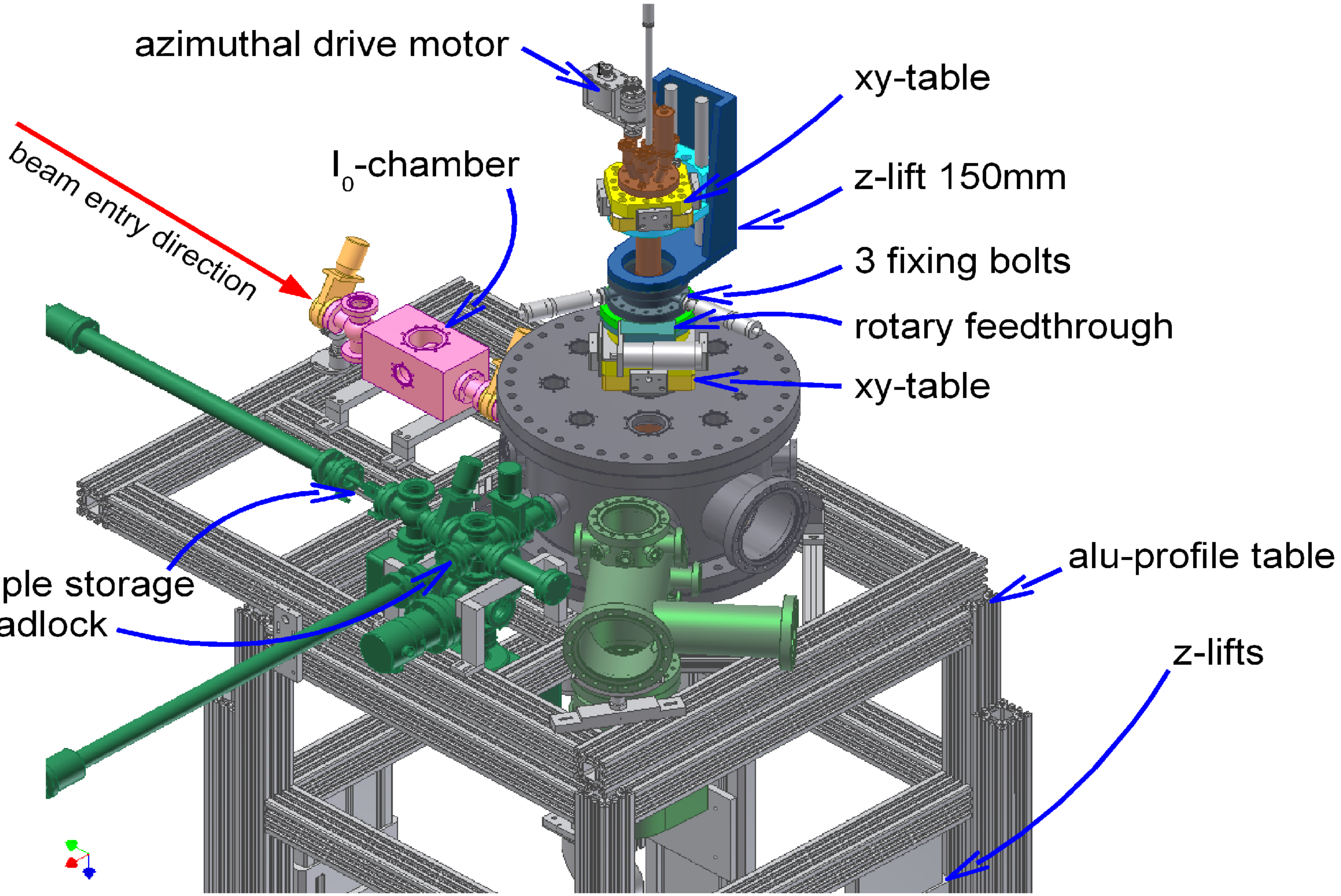
- coherent dispersion + absorption
- resonant scattering
- magnetic contributions

schematic set-up of the experiment . . .



by E. Göring, Max-Planck Institut für Metallforschung, Stuttgart

... and how it looks at the beamline UE56-2_PGM at BESSY-II, Berlin



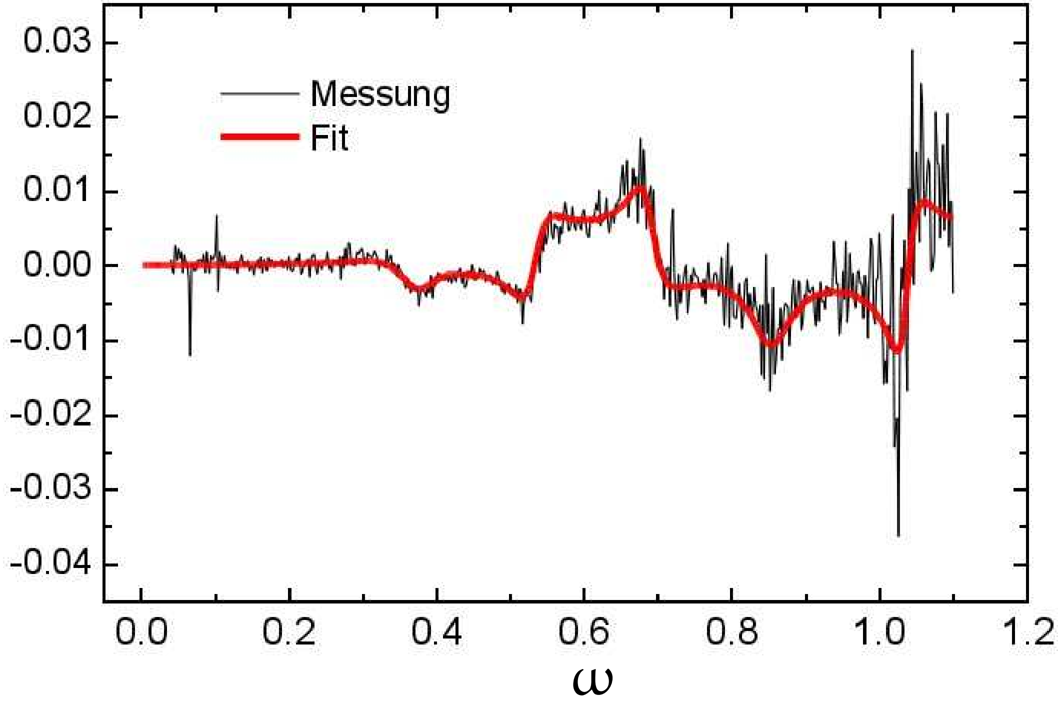
by E. Göring, Max-Planck Institut für Metallforschung, Stuttgart

experiment:
magnetism at the Co/Pt interface
(by E. Göring, MPI Stuttgart)

Pt L_3-edge
$\Rightarrow$ only M in Pt can be seen!

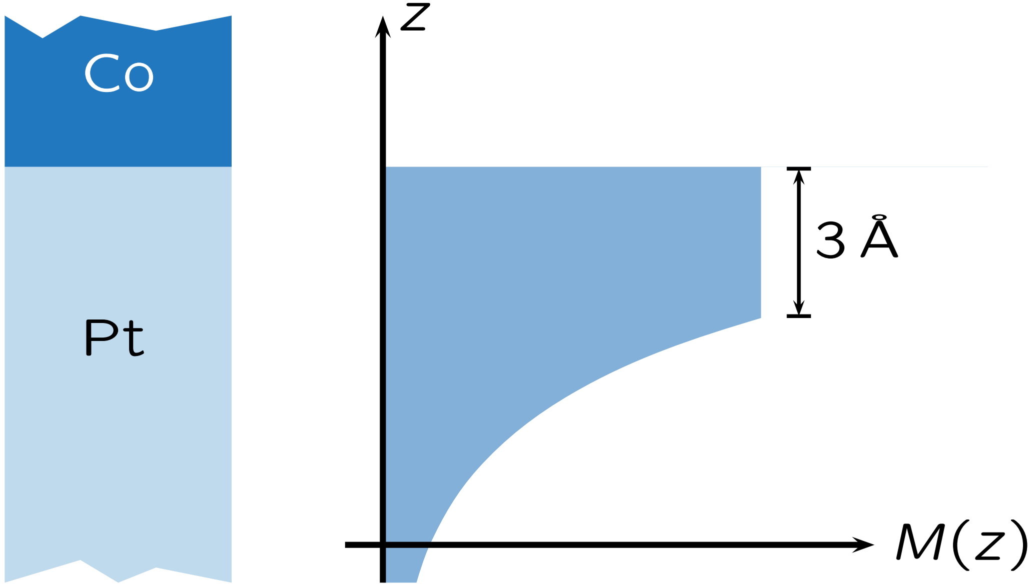
fit of the normalised difference

$$\frac{R^+ - R^-}{R^+ + R^-}$$

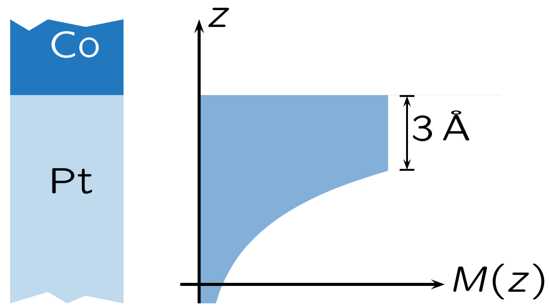


$M = 0.21(4)\,\mu_B$ per Pt atom

exponential decay



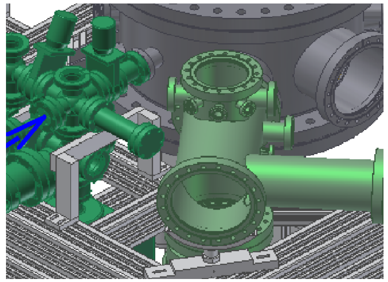
summary resonant x-rays



- \+ element specific
- \+ separation of spin and orbit momentum
- \+ sensitive to sup-atomic layers
- \+ depth selective

+− absorption limits penetration depth

$\Rightarrow$ balance of signal strength and penetration depth
$\Rightarrow$ restricted to the surface region ($<$ 1000 Å)



− needs ultra high vacuum
$\Rightarrow$ restriction of space and external parameters

\+ short measuring time (20 min)

− difficult simulation:
knowledge of the optical constants

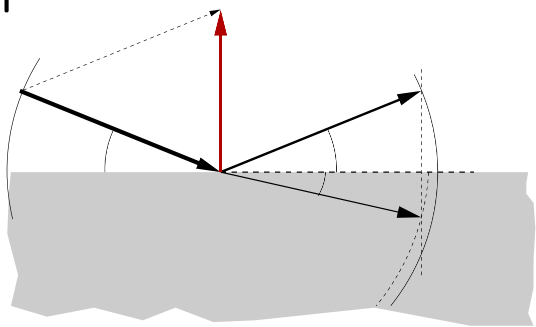
reflectometry

probes depth-profile of some potential

averages laterally

⇒ ideal for layered systems

data analysis by modelling

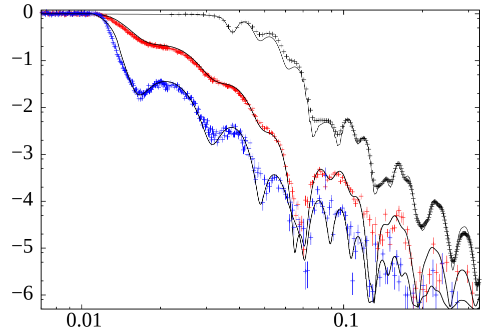


with neutrons

resolution: atom to sub-μm

isotope selective

detects in-plane magnetic induction



with x-rays

resolution: atom to sub-μm

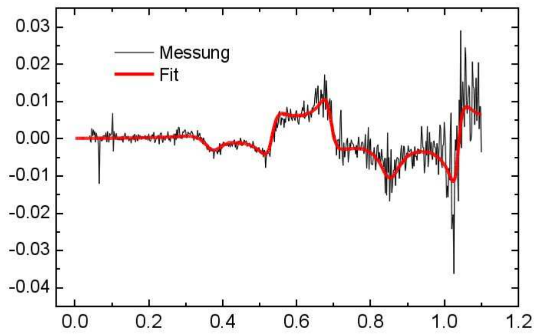
detects electron density

. . . in resonance

detects magnetic states of atoms

element specific

separation of spin and orbit



reflectometry, in general :

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X-ray and Neutron Reflectivity
Lect. Notes Phys. 770 (Springer 2009)

U. Pietsch, V. Holý, T. Baumbach:
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(Springer 2004)

. . . on magnetic systems

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C. R. Physique **8**, 763-776 (2007)

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