

Thin film heterostructures for spintronics investigated by grazing incidence small angle scattering

JUNE 22 2021 | E. KENTZINGER

JÜLICH CENTRE FOR NEUTRON SCIENCE & PETER GRÜNBERG INSTITUT

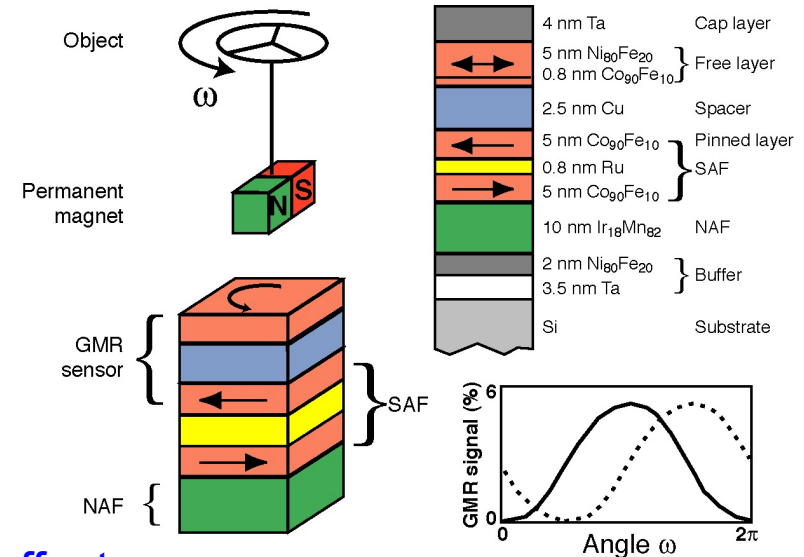
PLAN

- **Introduction: Spintronics**
- Method: Reflectivity, off-specular scattering, GISAS and neutron polarization analysis
- Simulation of specular reflectivity
- Simulation of polarized neutron reflectivity
- Simulation of GISAS intensities within the Distorted Wave Born Approximation
- Structural ordering in assemblies of magnetic nanoparticles
- Magnetic domain structure in FePd with perpendicular-to-plane anisotropy
- Take-Home Message

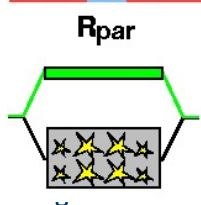
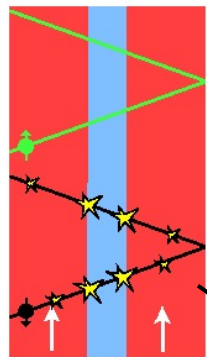
Magnetic thin films

- Ultra thin films
- Anisotropies at surfaces and interfaces
- Exchange bias
- Interlayer exchange coupling
- Giant magnetoresistance
- Tunnel magnetoresistance...

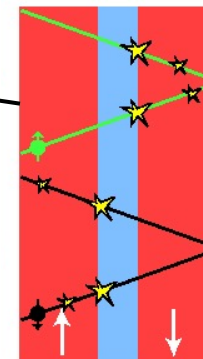
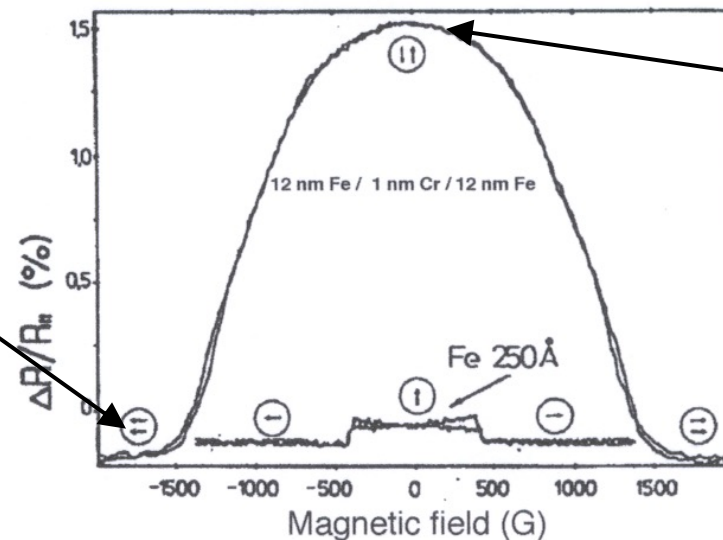
Sensor:



The Giant Magneto-Resistance effect:



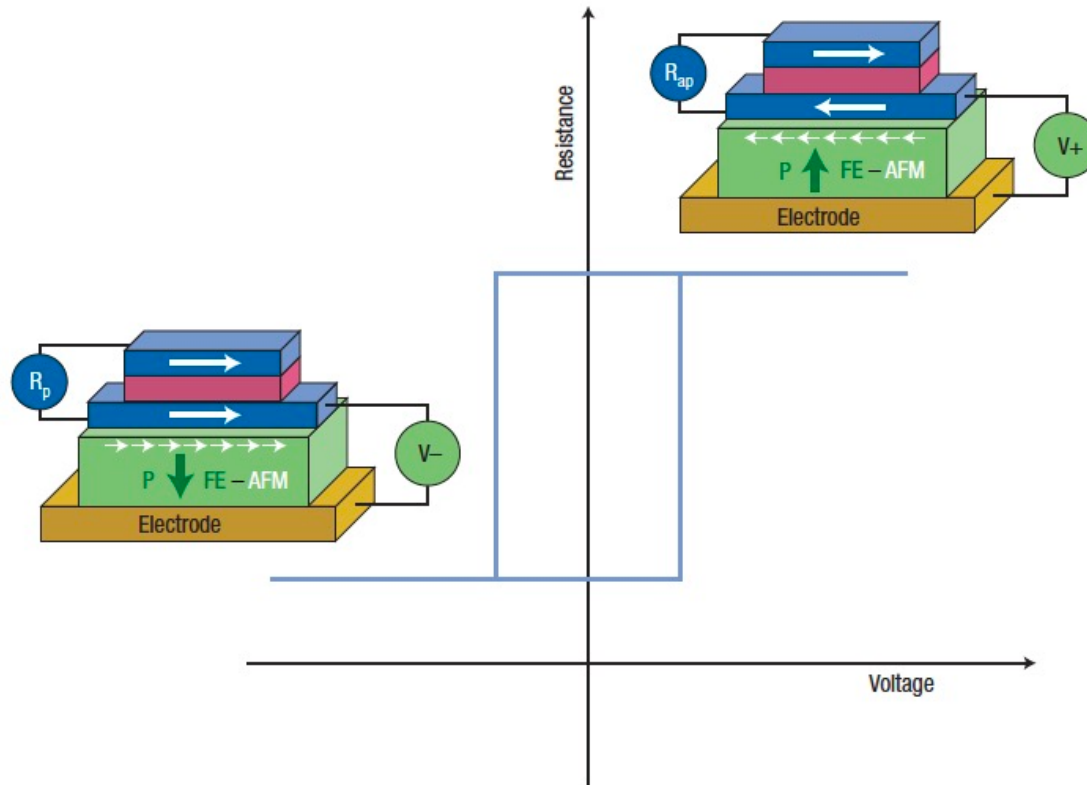
Imholt



New Field of science:
Spintronics

Nobel Prize 2007

The multiferroic RAM

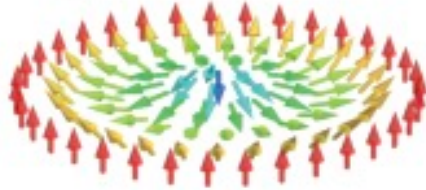


M. Bibes and A. Barthélémy, Nat. Mat. (2008)

Fast, dense, energy-efficient

Magnetic skyrmions in thin films

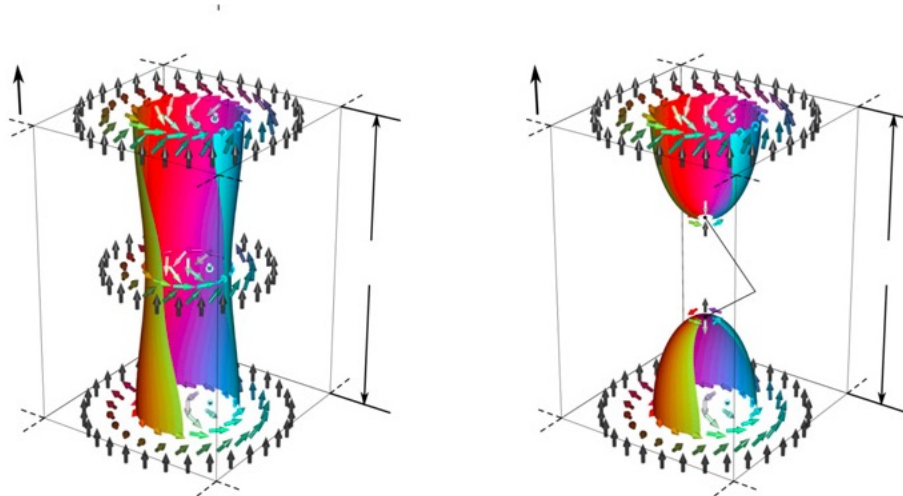
a Néel-type skyrmion



b Bloch-type skyrmion



A. Fert et al. Nature Reviews Materials (2017)



Rybakov et al, NJP (2016)

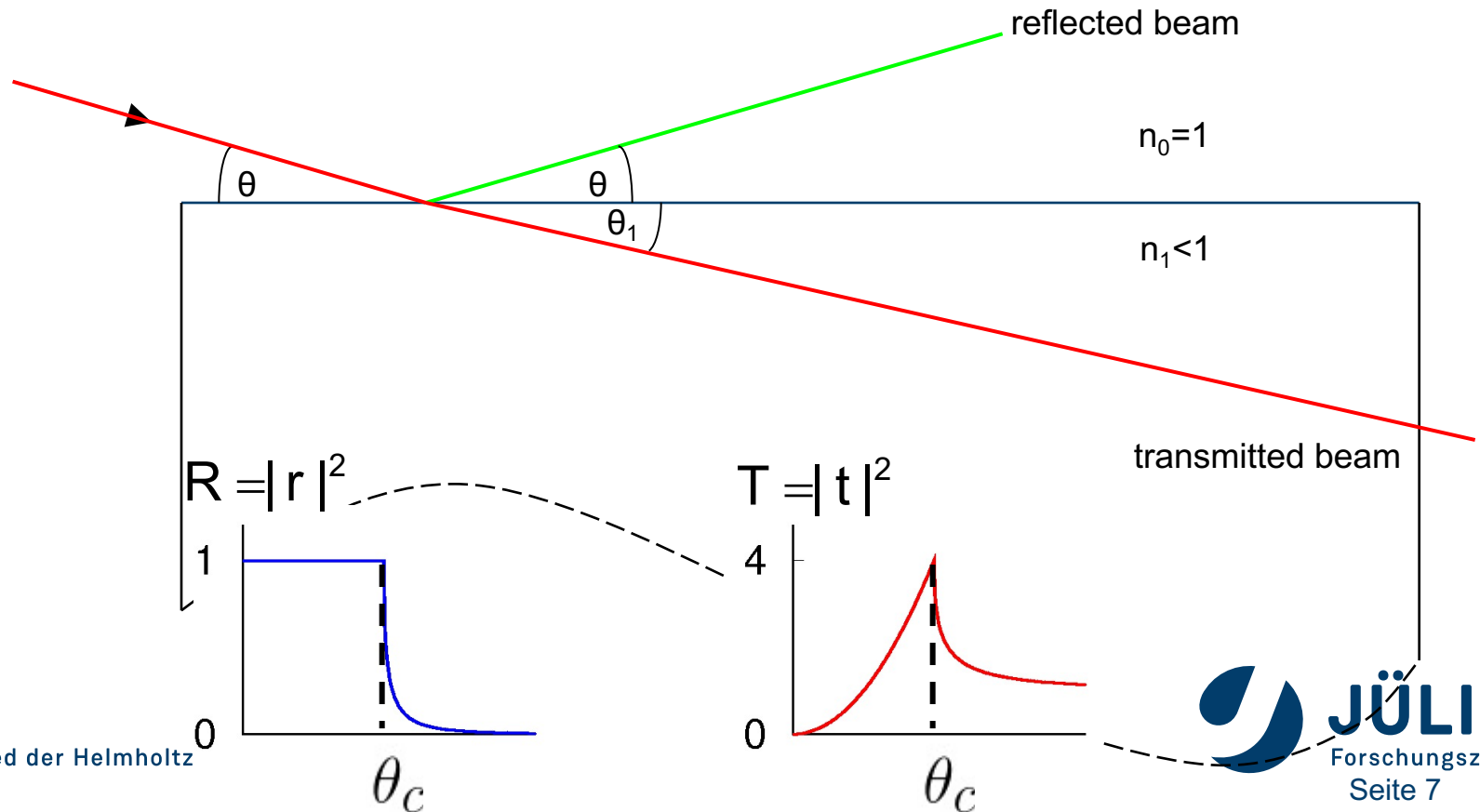
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SPECULAR REFLECTIVITY: FLAT SURFACE

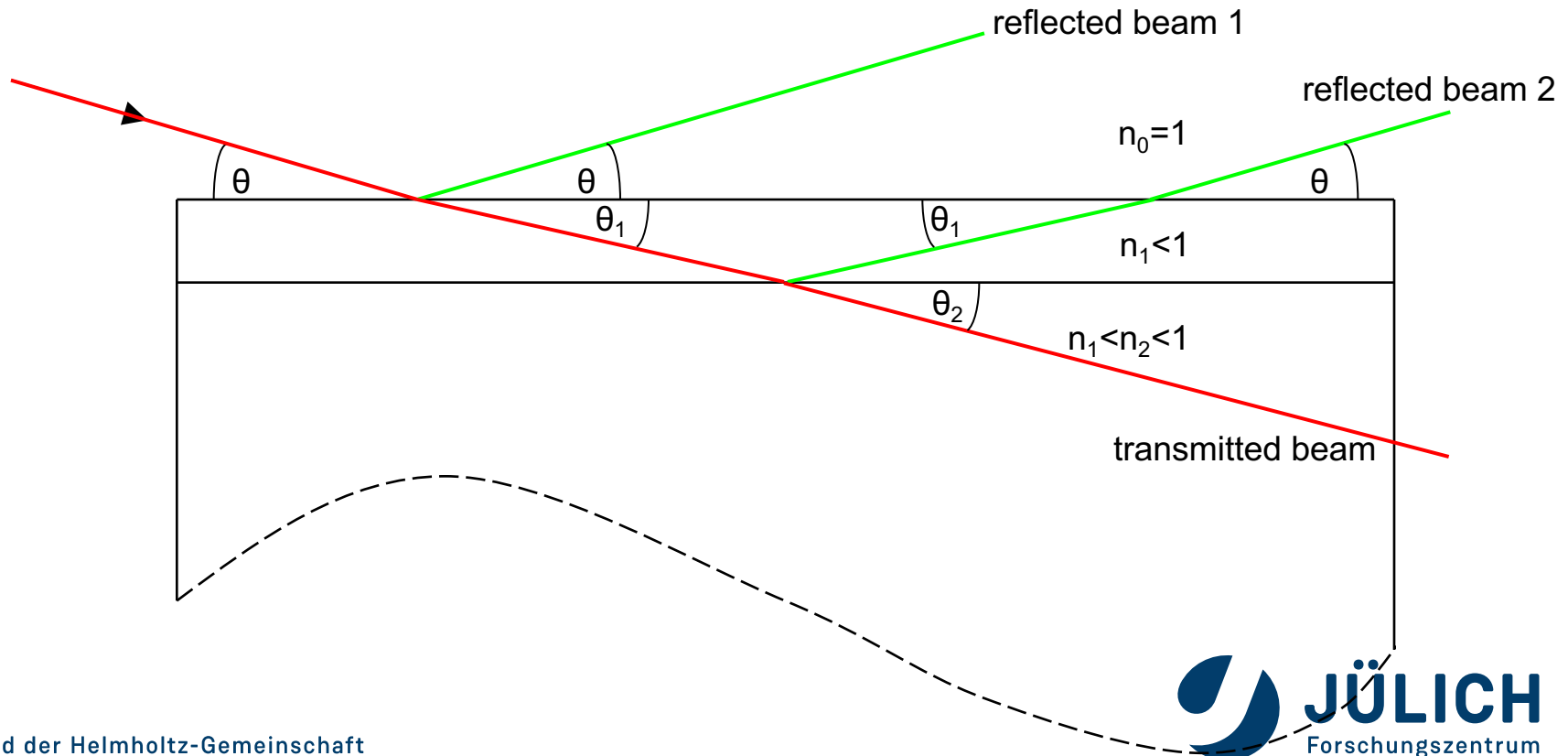
Refraction: $\theta_1 < \theta$, if $n_1 < n_0$

typically $n < 1$ for neutrons and x-rays



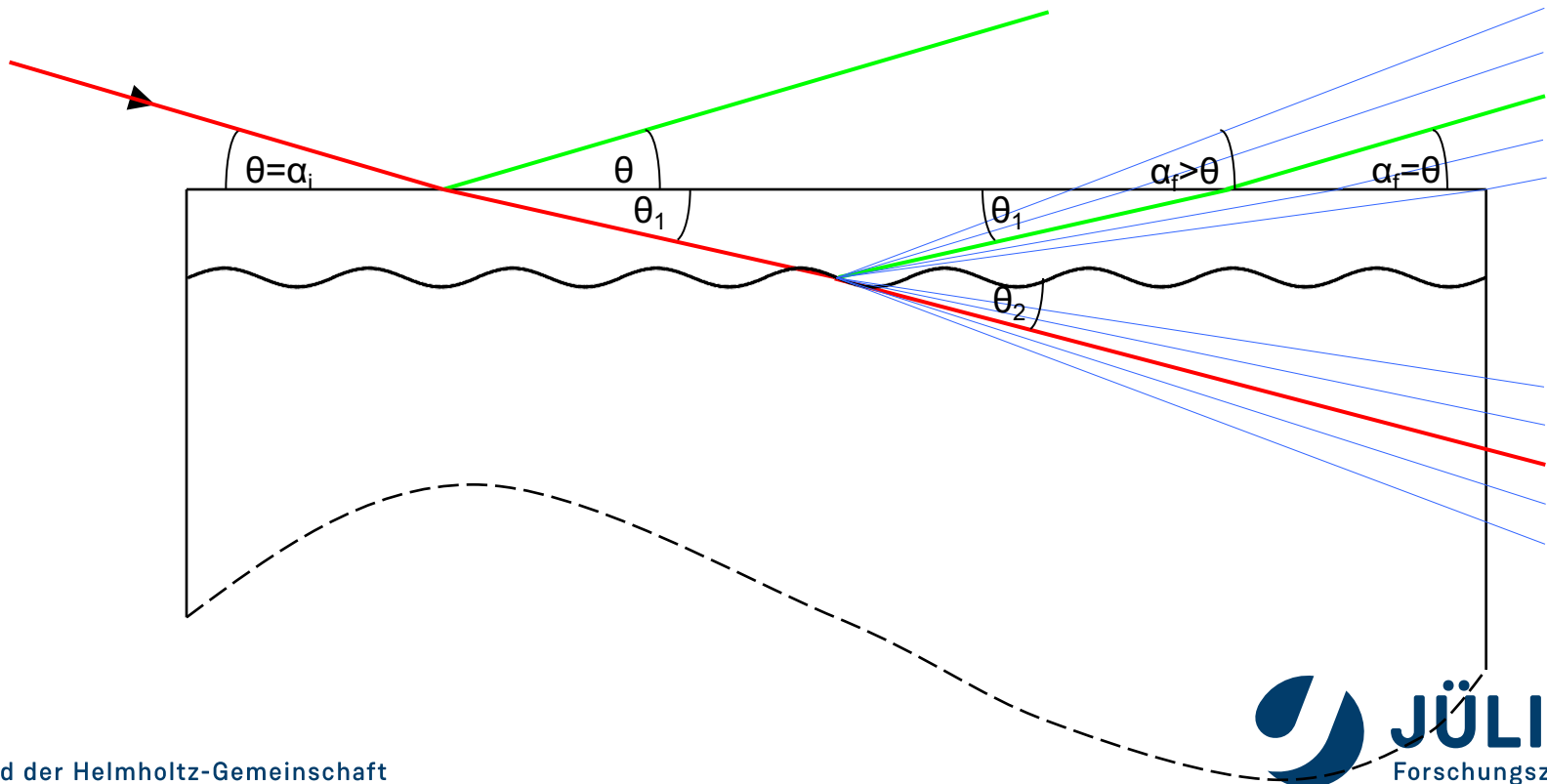
SPECULAR REFLECTIVITY: LAYER ON SURFACE

Interference between reflected beams
according to path length difference



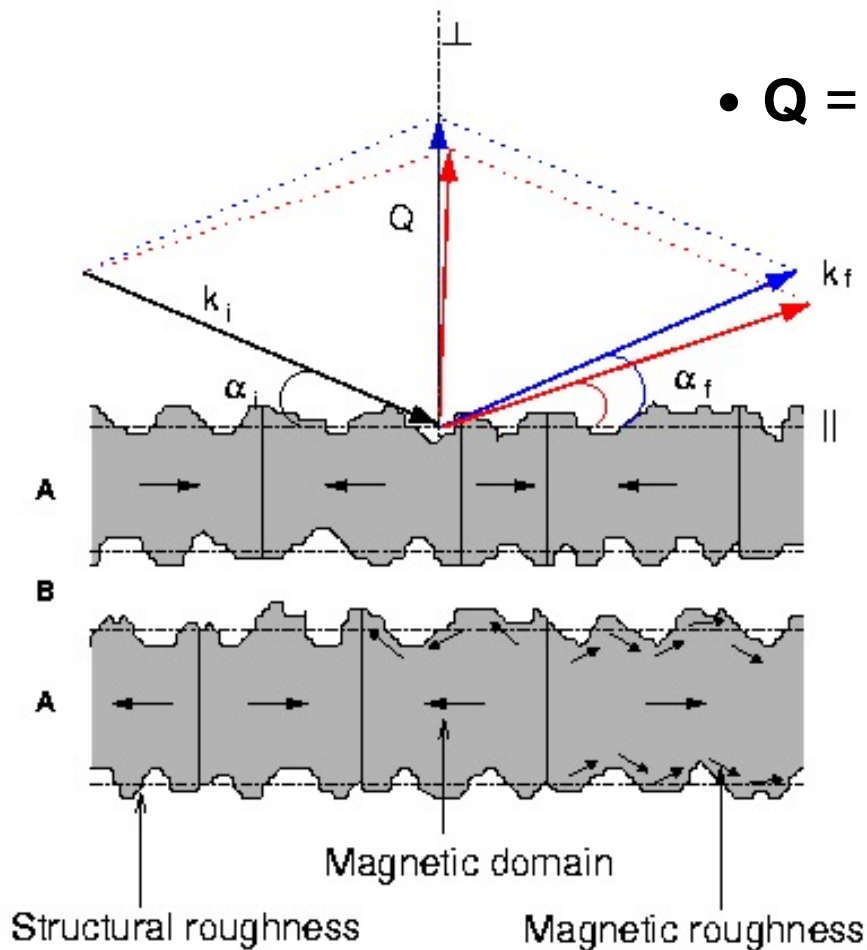
OFF-SPECULAR SCATTERING: ROUGH INTERFACE

Broken translational invariance
→ Off-specular (diffuse) scattering



REFLECTIVITY AND OFF-SPECULAR SCATTERING

- In-plane structure can be investigated by off-specular diffuse scattering:



- $Q = k_f - k_i$, $k = 2\pi / \lambda$

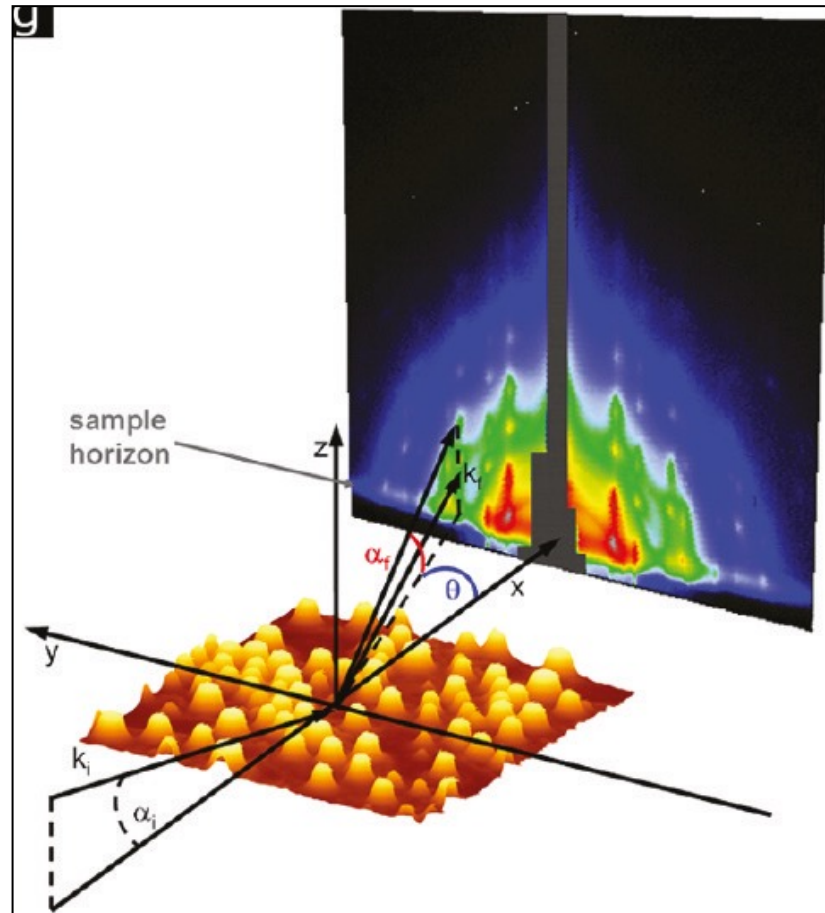
If $\alpha_f = \alpha_i$ then $Q_{||} = 0$:

- specular reflectivity
- average over lateral coordinates

If $\alpha_f \neq \alpha_i$ then $Q_{||} \neq 0$:

- off-specular scattering
- lateral correlations can be probed

Grazing Incidence Small Angle Scattering (GISAS)



$$Q = k_f - k_i = \begin{cases} Q_x \cong k.(\alpha_i^2 - \alpha_f^2 - \theta^2) \\ Q_y \cong k.\theta \\ Q_z \cong k.(\alpha_i + \alpha_f) \end{cases}$$

typically: $\begin{cases} 2\pi/Q_x \approx 1-20 \mu\text{m} \\ 2\pi/Q_y \approx 1-300 \text{ nm} \end{cases}$
(neutrons!)

S. Disch, E. Wetterskog et al. Nano Letters **11**, 1651 (2011)

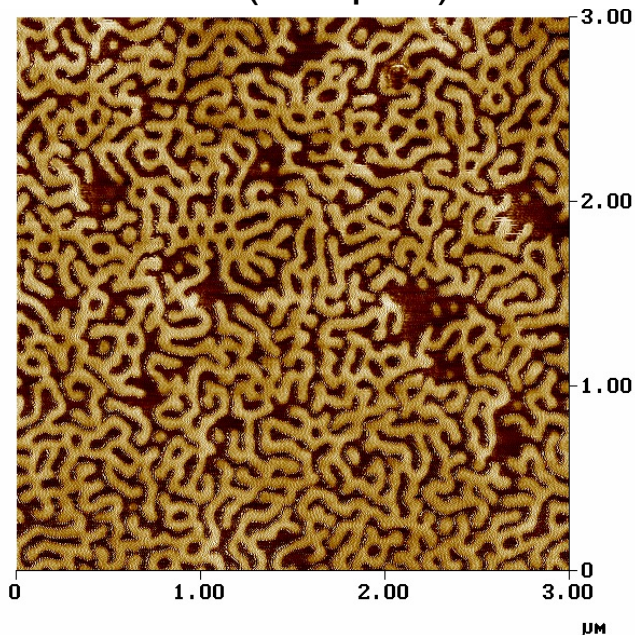
dPS-PB diblock copolymer

(D. Korolkov et al. J. Appl. Cryst. 45, 245 (2012))

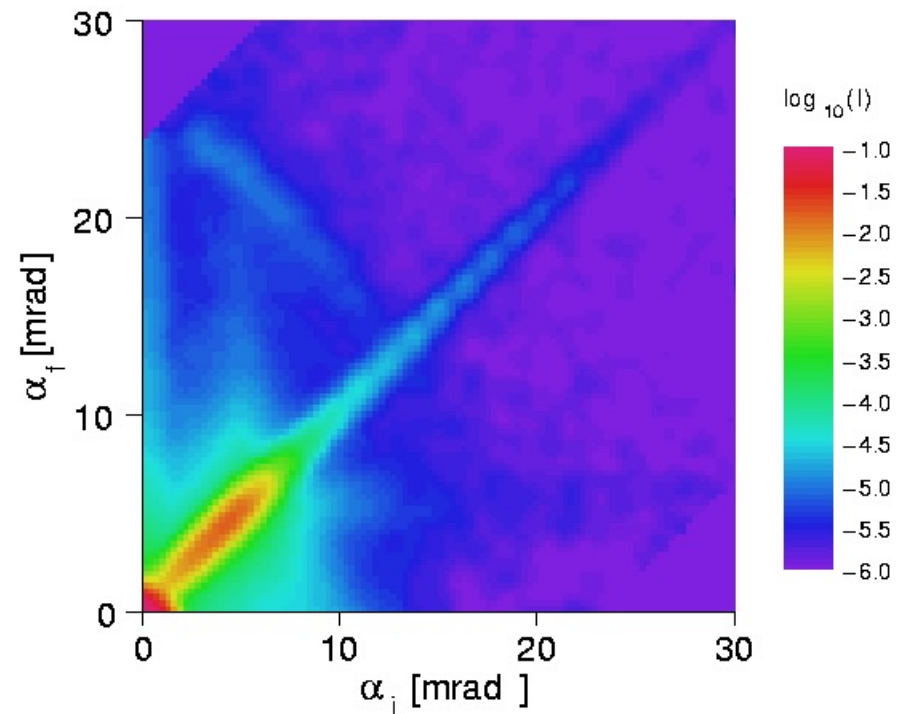
Polymer preparation:

- $f = 0.5 \rightarrow$ Lamellae
- High Mw $\rightarrow \perp$ ordering

AFM ($3 \times 3 \mu\text{m}^2$):



Off-specular on HADAS@Jülich
(now TREFF@MLZ)



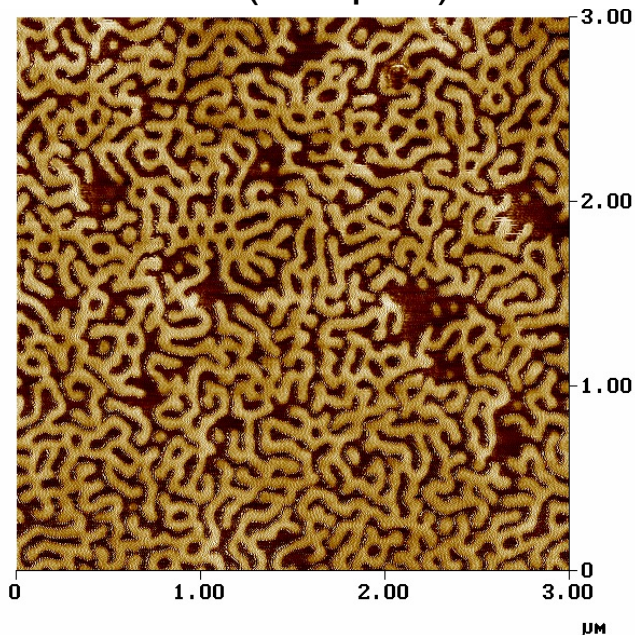
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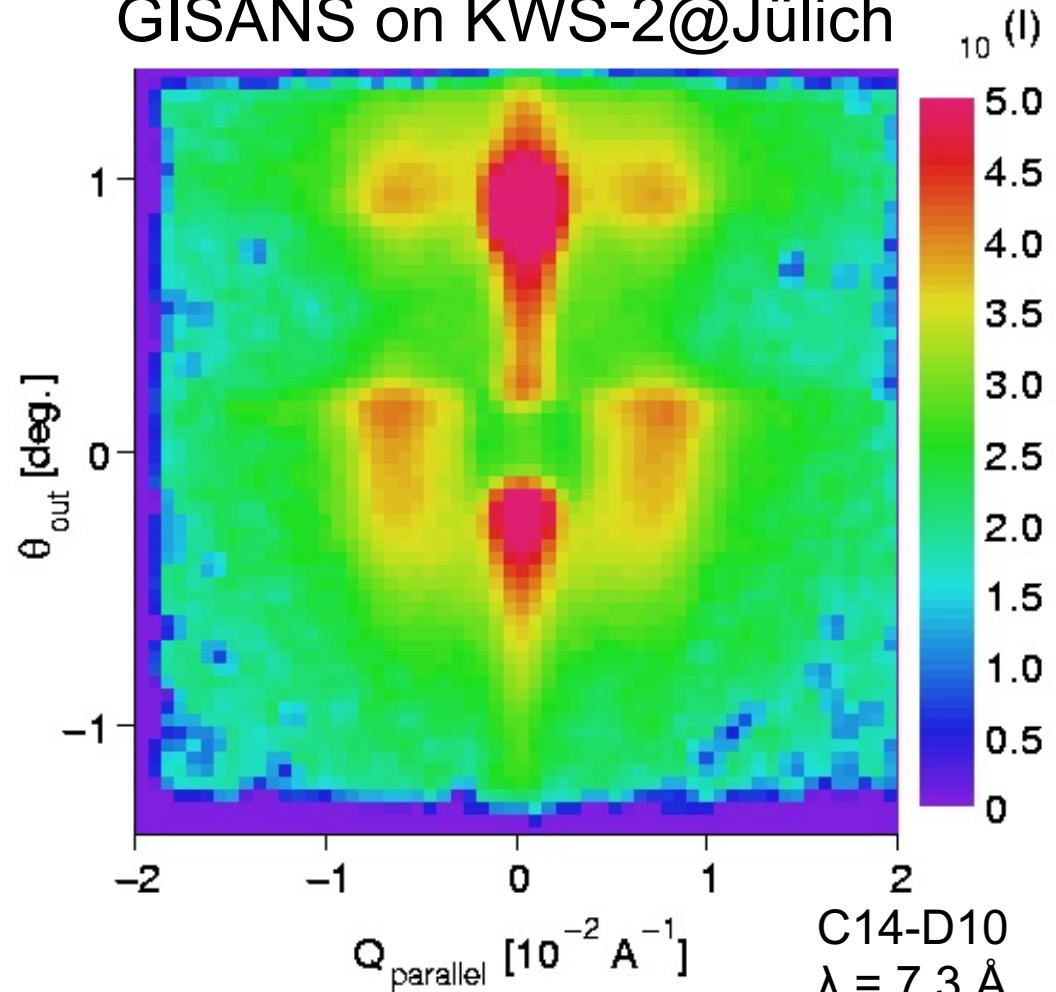
Polymer preparation:

- $f = 0.5 \rightarrow$ Lamellae
- High Mw $\rightarrow \perp$ ordering

AFM ($3 \times 3 \mu\text{m}^2$):



GISANS on KWS-2@Jülich



C14-D10
 $\lambda = 7.3 \text{ \AA}$

dPS-PB diblock copolymer

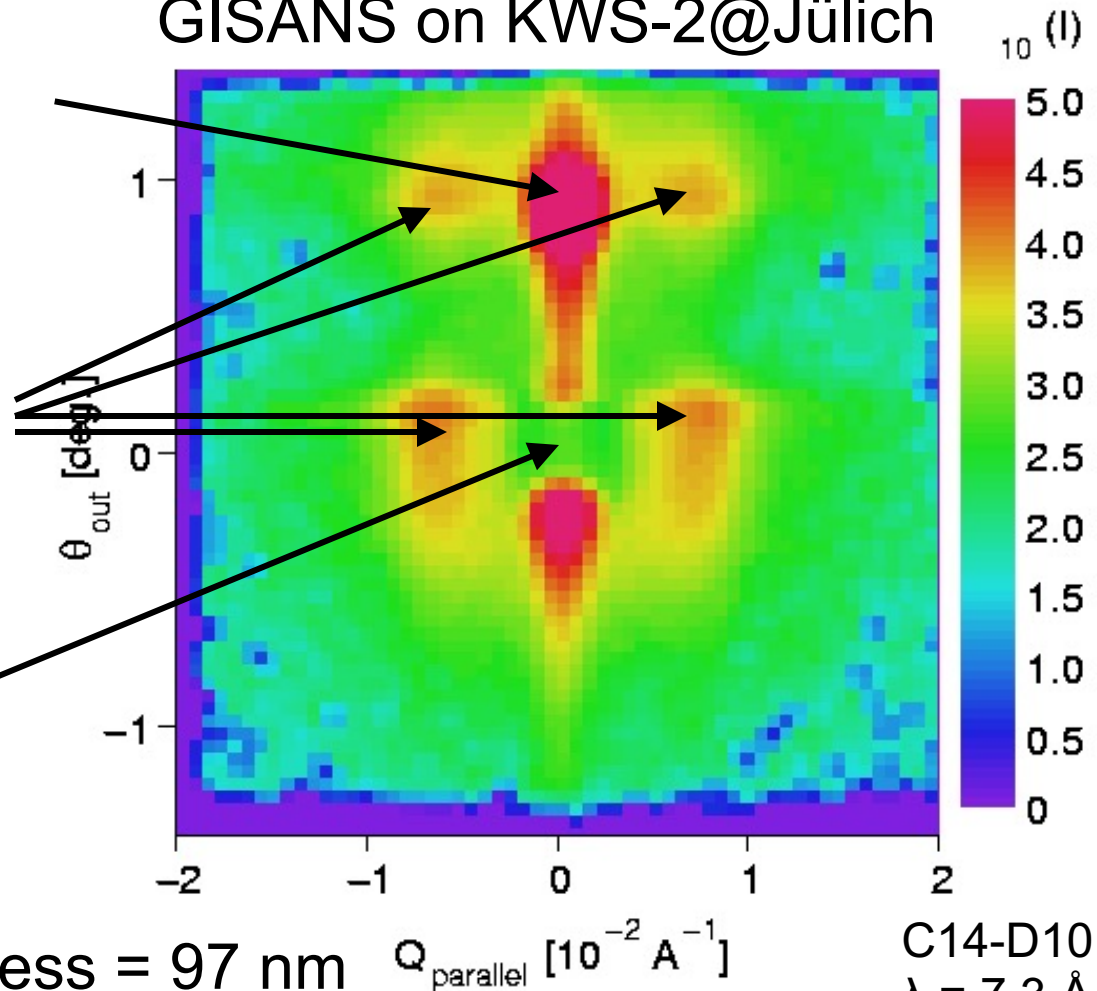
(D. Korolkov et al. J. Appl. Cryst. 45, 245 (2012))

GISANS on KWS-2@Jülich

Specularly reflected beam

Out-of-plane scattering
due to perpendicular
lamellae

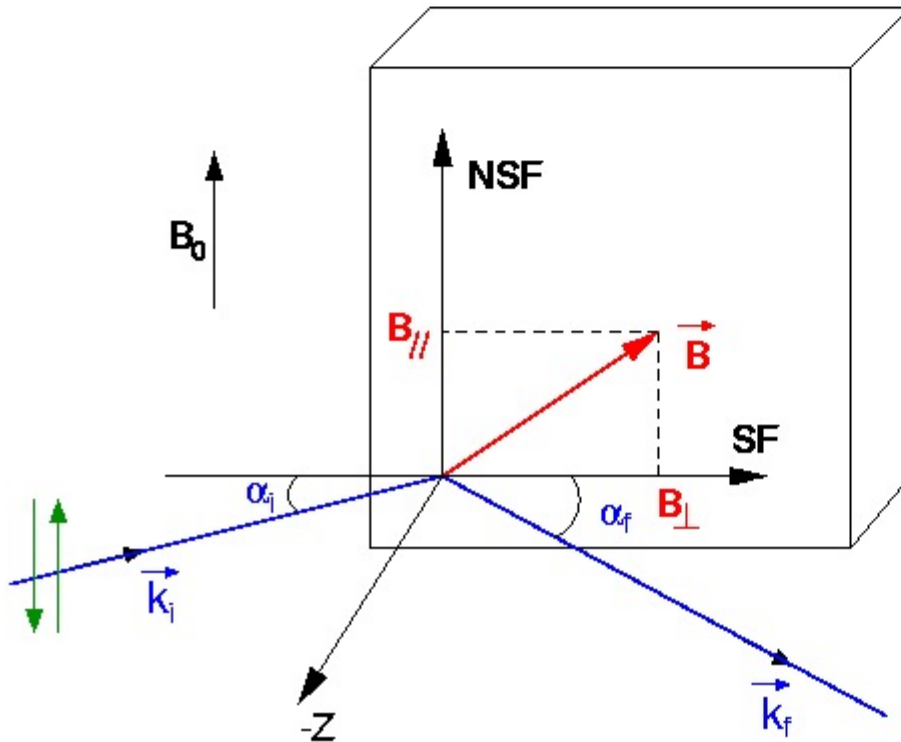
Direct beam covered by
beam stop



Polarization analysis

The neutron is a spin $\frac{1}{2}$ particle

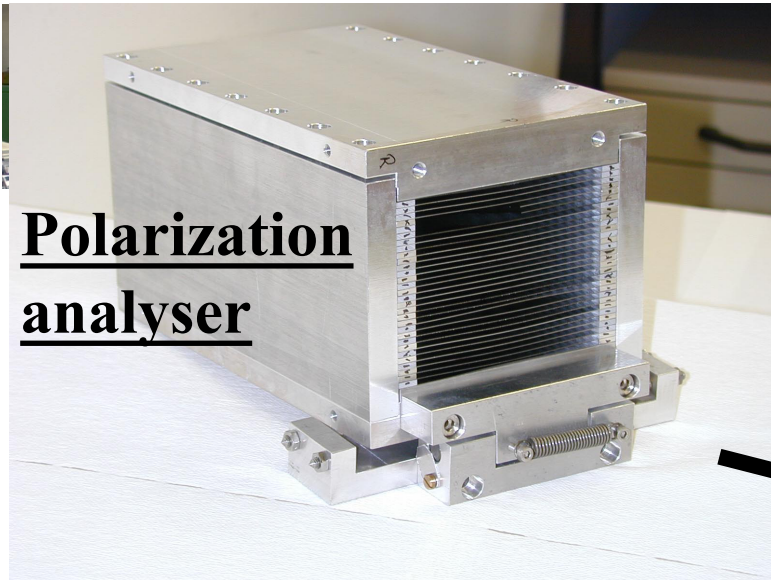
$$|\Psi_i(\mathbf{r})\rangle = \Psi_i^+(\mathbf{r})|+\rangle + \Psi_i^-(\mathbf{r})|-\rangle$$



$$I_{++}, I_{--} \Rightarrow \rho_i^N, B_{||}$$

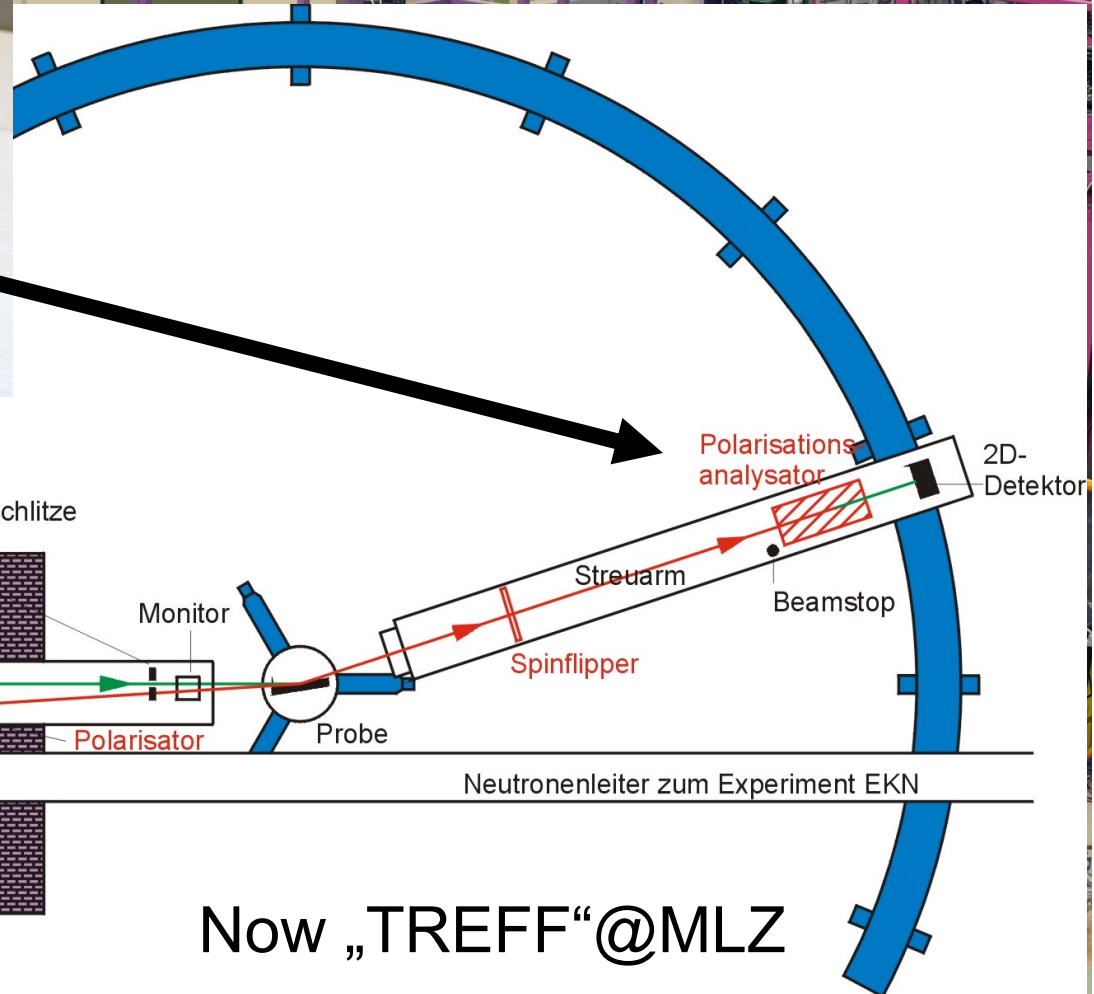
$$I_{+-}, I_{-+} \Rightarrow B_{\perp}$$

Polarization analysis... HADAS@Jülich (U. Rücker)



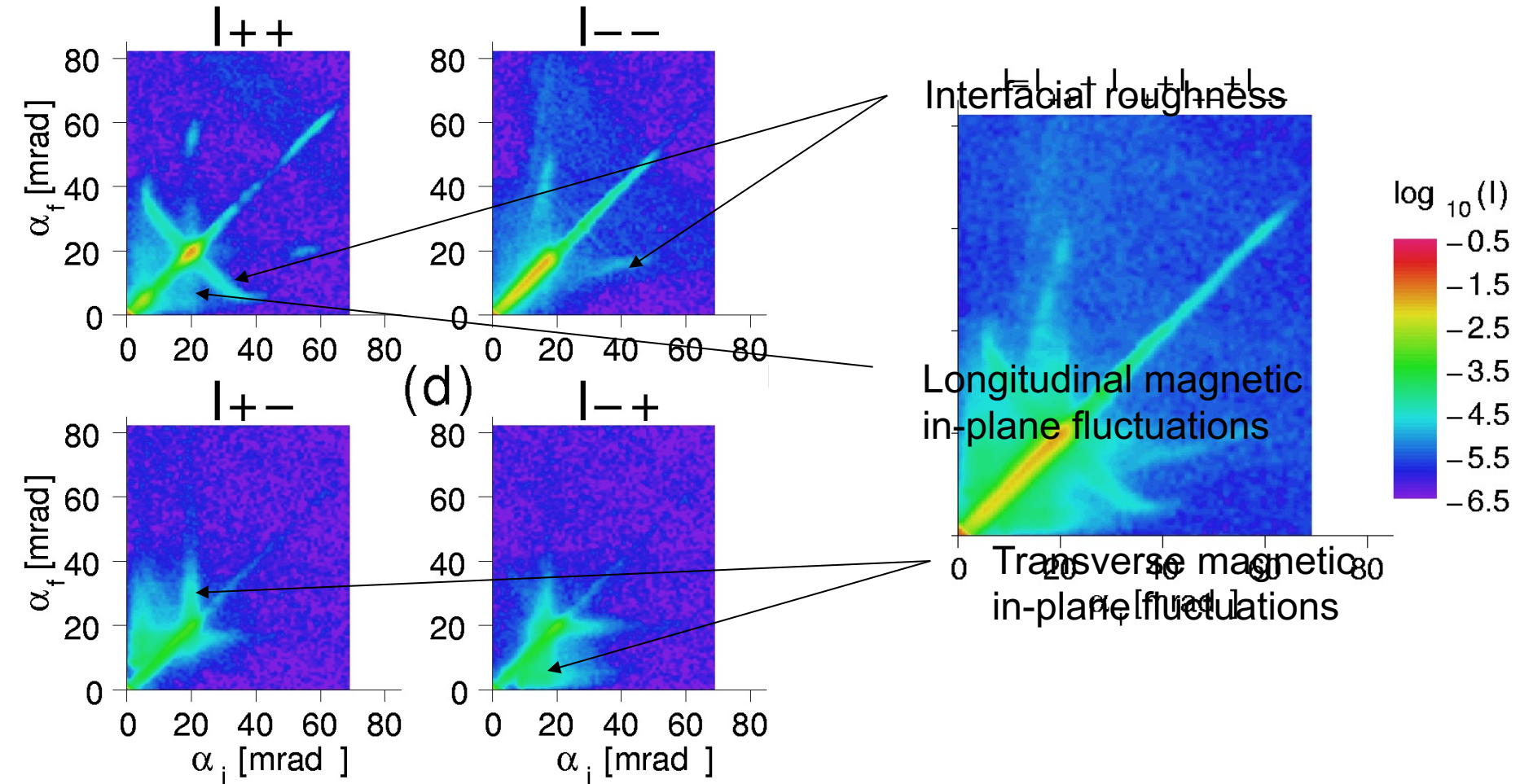
**Polarization
analyser**

Simultaneous measurement of reflectivity and off-specular scattering with polarization analysis



Now „TREFF“@MLZ

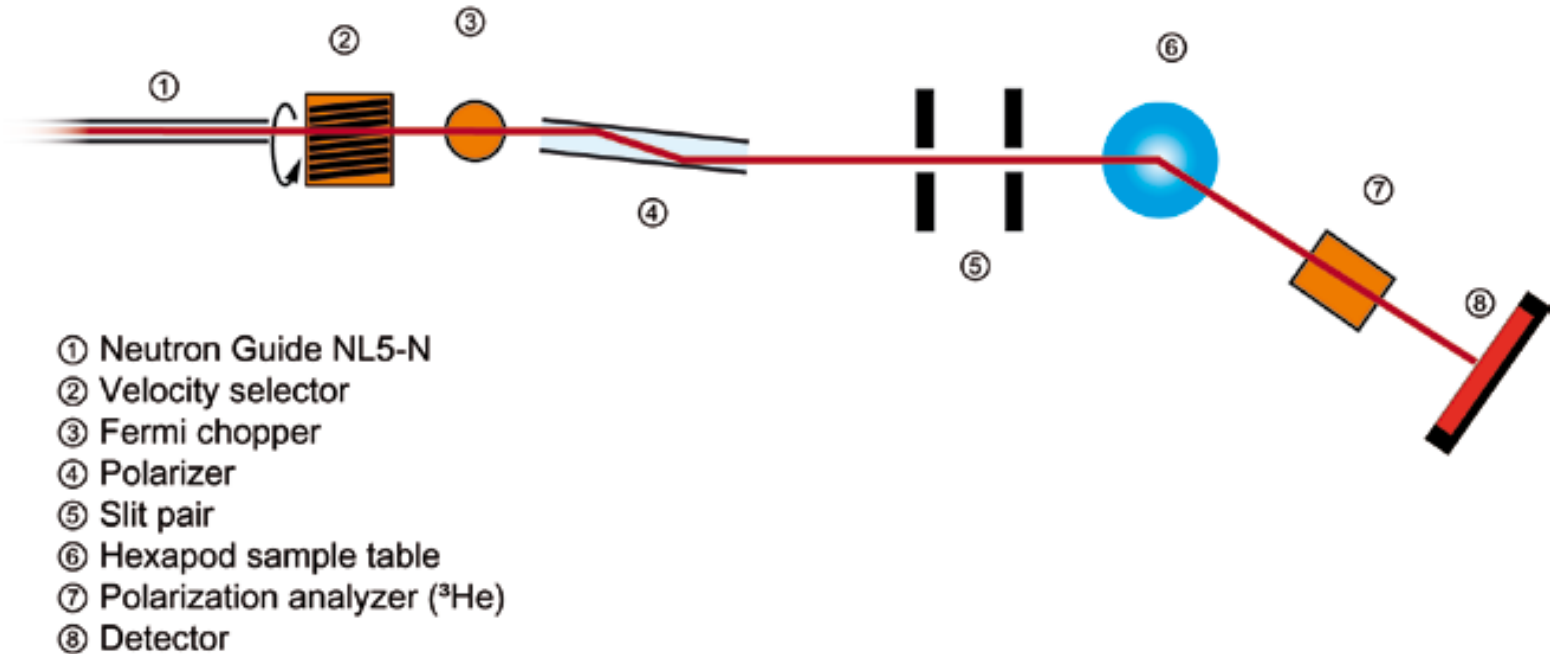
Remnant Polarizing Supermirror



With polarization analysis

Without analysis **ICH**
gszentrum

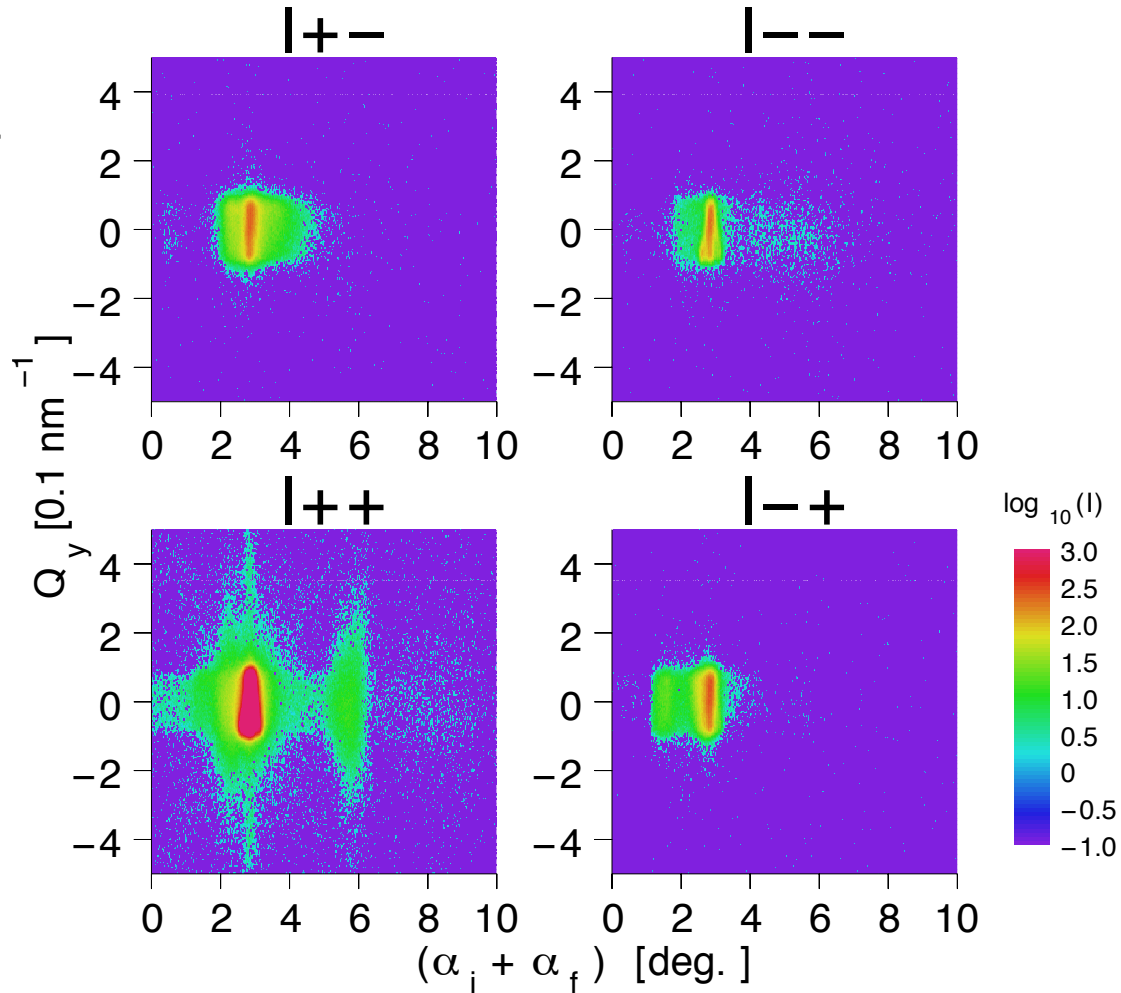
MARIA @ MLZ (S. Mattauch)



GISANS WITH POLARIZATION ANALYSIS @ MARIA

$$\alpha_i = 1.5^\circ$$

$$\mu_0 H = 4.4 \text{ mT}$$



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INTERACTION OF NEUTRONS AND X-RAYS WITH MATTER IN THE CONTINUUM LIMIT

Schrödinger equation for the wave function of the neutron:

$$\left[-\frac{\hbar^2}{2m} \Delta + V(\mathbf{r}) \right] \psi(\mathbf{r}) = E \psi(\mathbf{r}) \quad E = \hbar^2 k^2 / (2m)$$
$$k = 2\pi / \lambda$$

Interaction potential in layer 1:

$$V_1(\mathbf{r}) = V_1 = \frac{2\pi\hbar^2}{m} \rho$$

neutrons

$$\rho = \sum_j N_j (b'_j - i b''_j)$$

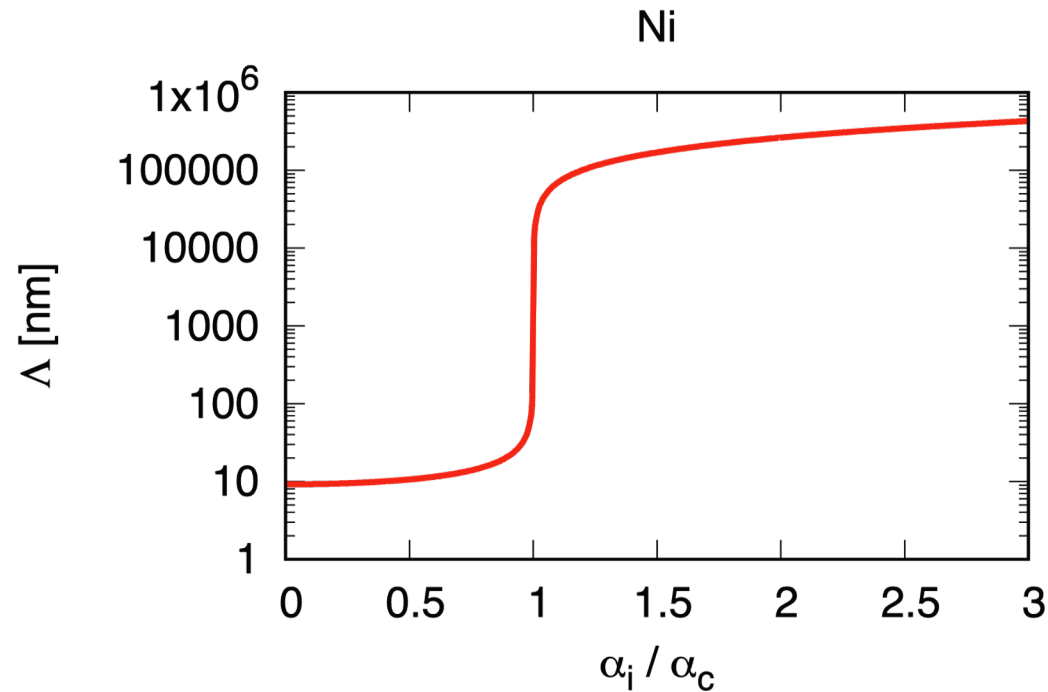
X-rays

$$\rho = r_0 \sum_j N_j (Z_j + f'_j - i f''_j)$$

$$r_0 = 2.82 \text{ fm}$$

Penetration depth in the substrate:

$$\Lambda = \frac{1}{k''_{z,1}}$$



(neutrons)

LAYERS ON SUBSTRATE

Wave function in layer l:

$$\Psi_l(\mathbf{r}) = e^{i\kappa\rho} (e^{i\mathbf{k}_z,1 \cdot \mathbf{z}} t_l + e^{-i\mathbf{k}_z,1 \cdot \mathbf{z}} r_l)$$

N layers

→ 2(N+2) coefficients have to be determined

Continuity of Ψ and $\partial\Psi/\partial\mathbf{z}$ at interfaces:

→ 2(N+1) equations

$$t_0 = 1$$

$$r_{N+1} = 0$$

➔ Solvable (recursively)

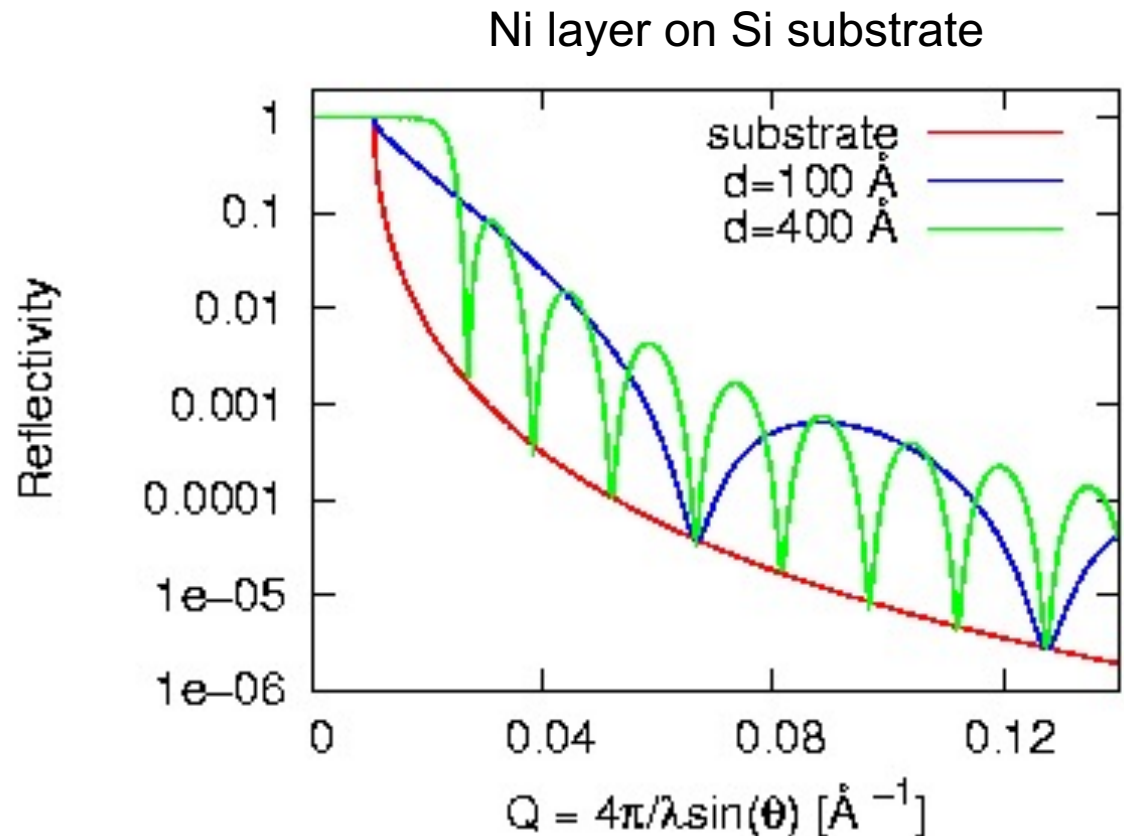
REFLECTIVITY FROM A LAYER ON SUBSTRATE

Optical path length difference
between beams reflected at
surface and interface:

$$\Delta = 2d \sin \theta$$

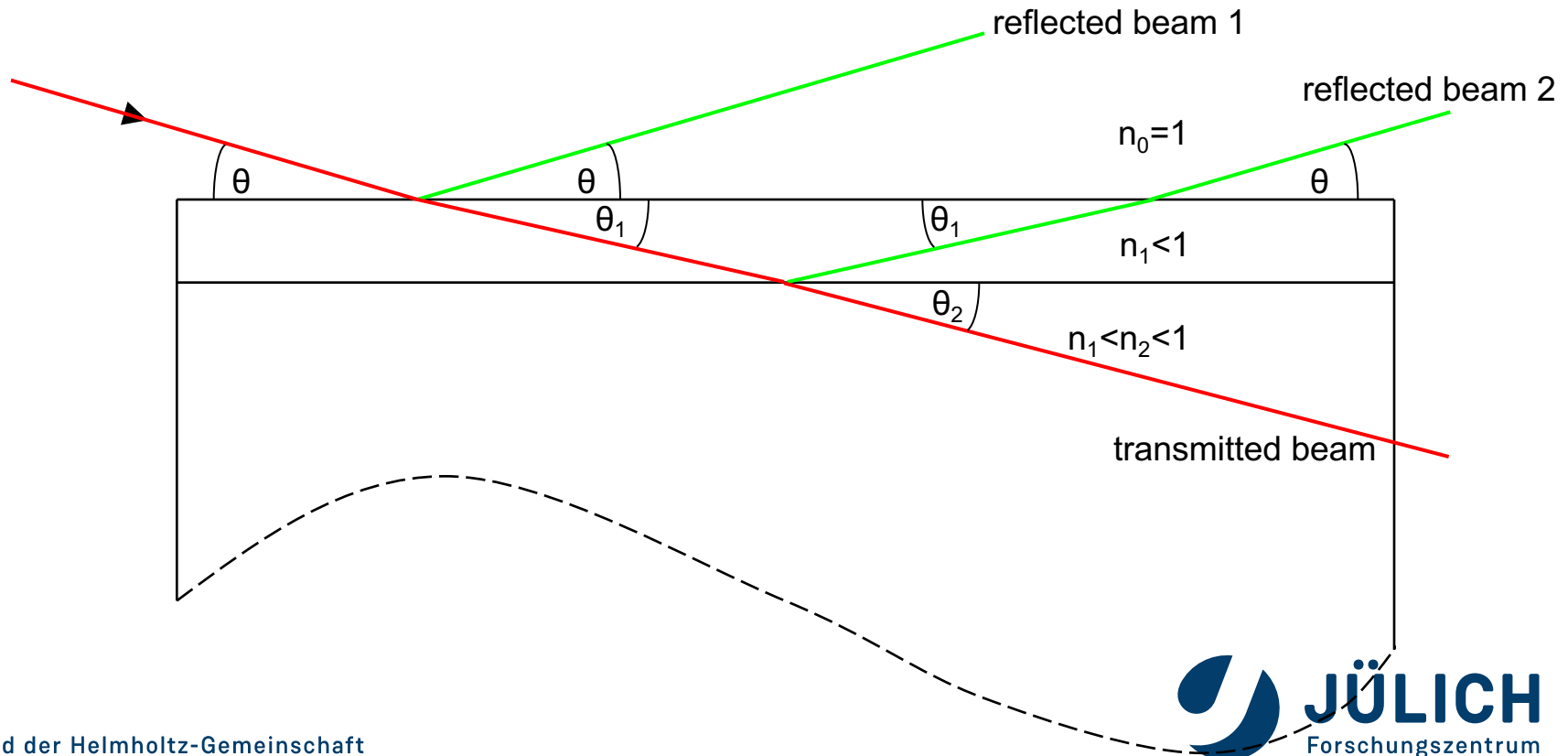
→ Distance between
interference maxima:

$$\delta Q \simeq \frac{2\pi}{d}$$



SPECULAR REFLECTIVITY: LAYER ON SURFACE

Interference between reflected beams
according to path length difference



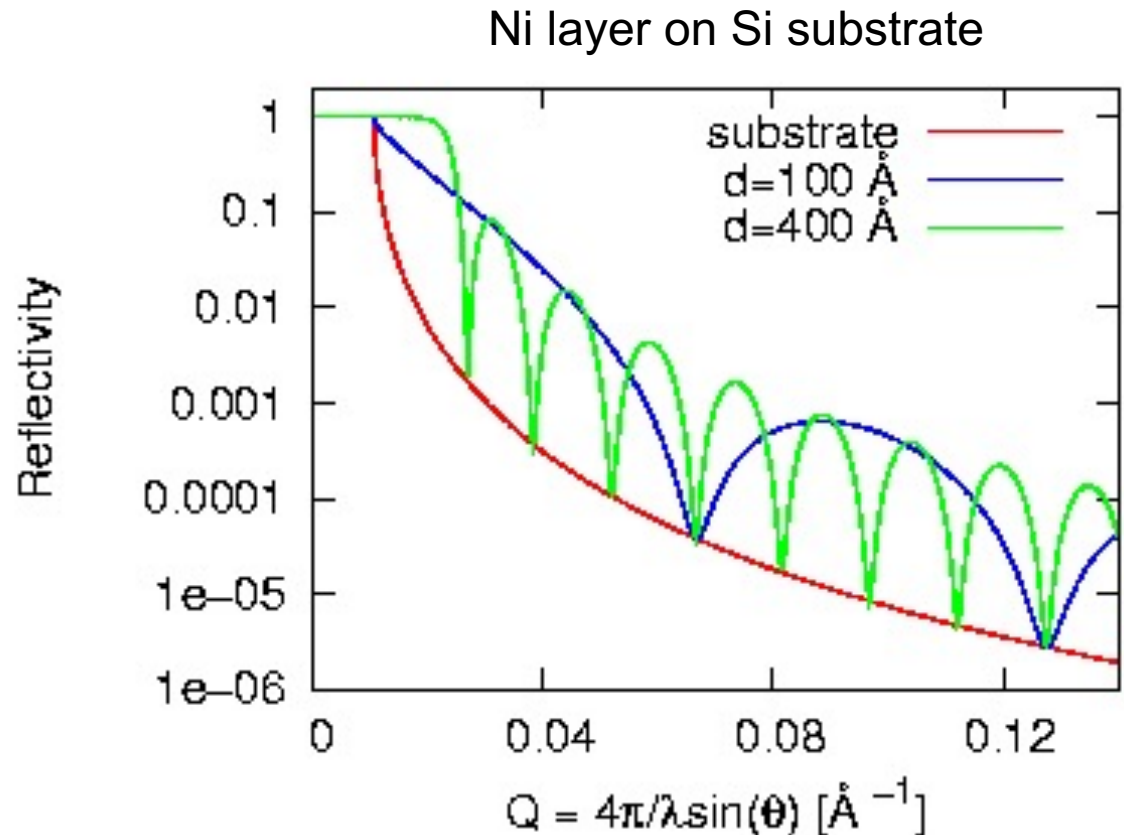
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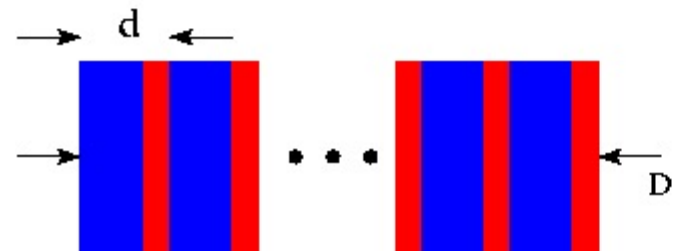
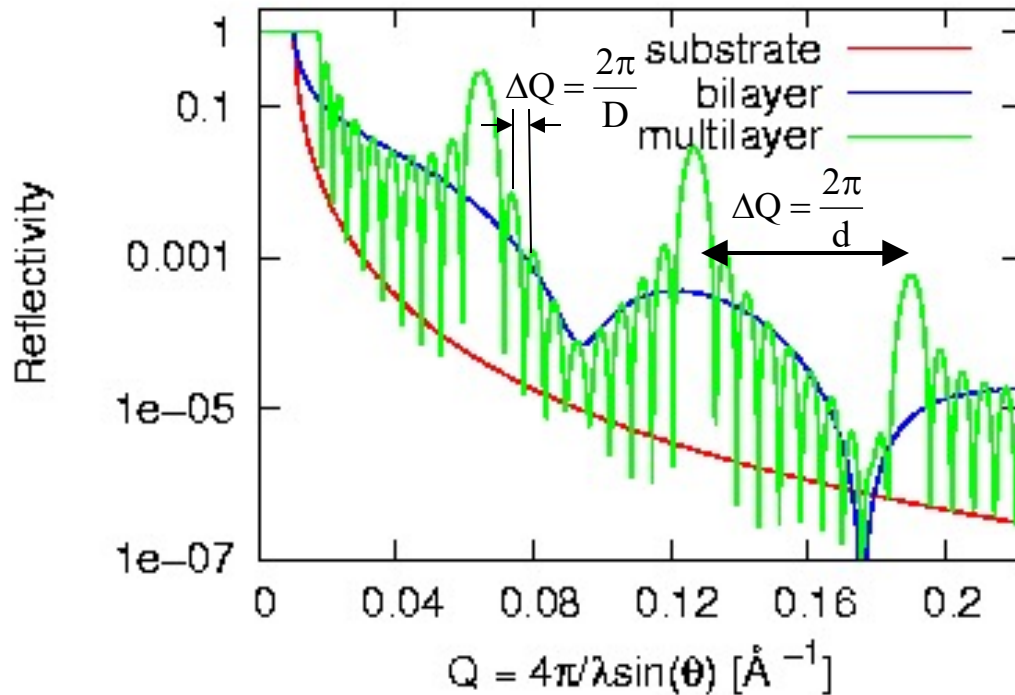
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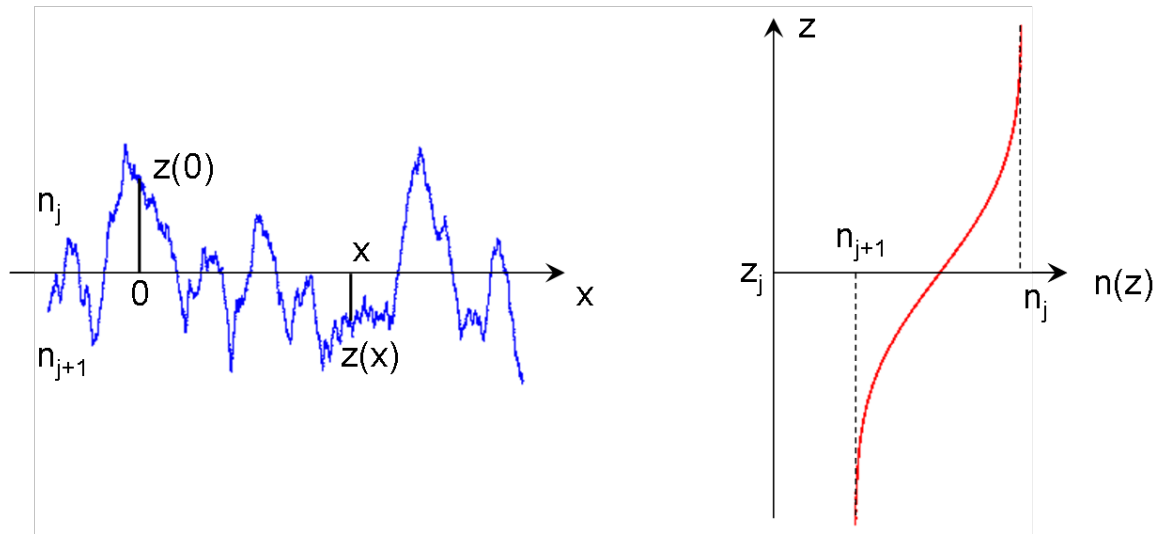


REFLECTIVITY FROM A MULTILAYER

10 Ni/Ti bilayers on Si



ROUGHNESS AND INTERDIFFUSION



z is a gaussian random variable with distribution function:

$$P(z) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{z^2}{2\sigma^2}\right),$$

→ Profile of refractive index between layers j and $j+1$:

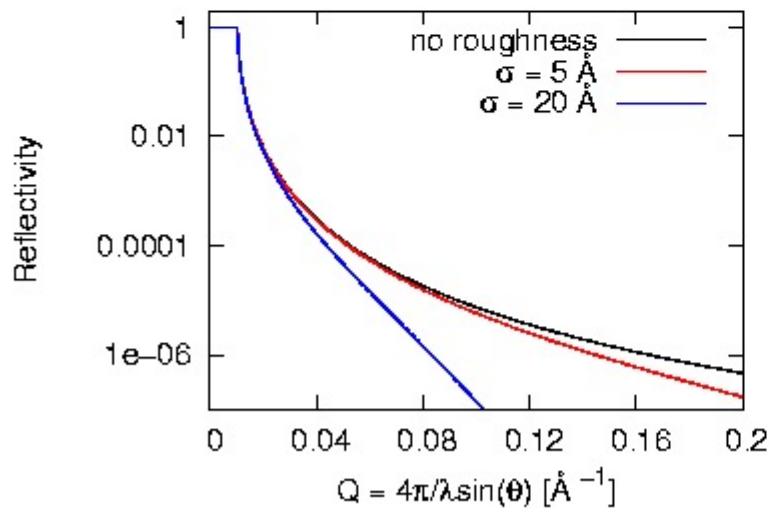
$$n(z) = \frac{n_j + n_{j+1}}{2} - \frac{n_j - n_{j+1}}{2} \operatorname{erf}\left(\frac{z - z_j}{\sqrt{2}\sigma_j}\right) \quad \text{with:} \quad \operatorname{erf}(z) = \frac{2}{\sqrt{\pi}} \int_0^z e^{-t^2} dt.$$

ROUGHNESS AND INTERDIFFUSION

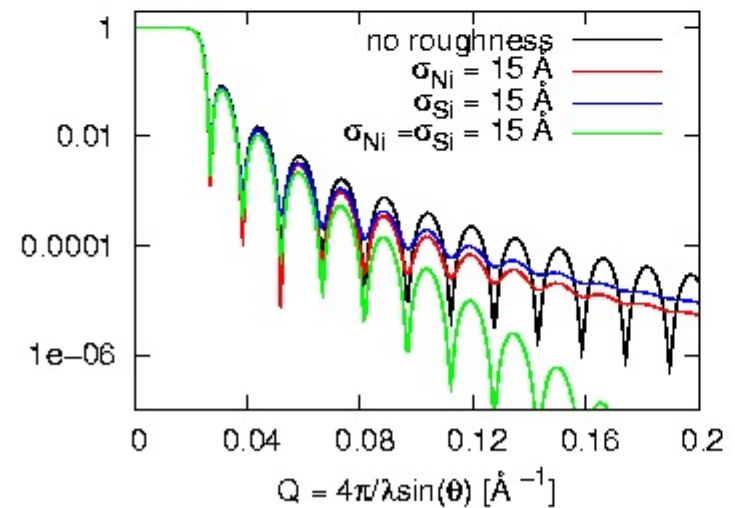
Reflectivity from such a surface obtained by averaging:

$$R_{rough} = R_{flat} \cdot \exp \left(-4\sigma_j^2 k_{zj} k_{zj+1} \right).$$

Si substrate



Ni on Si



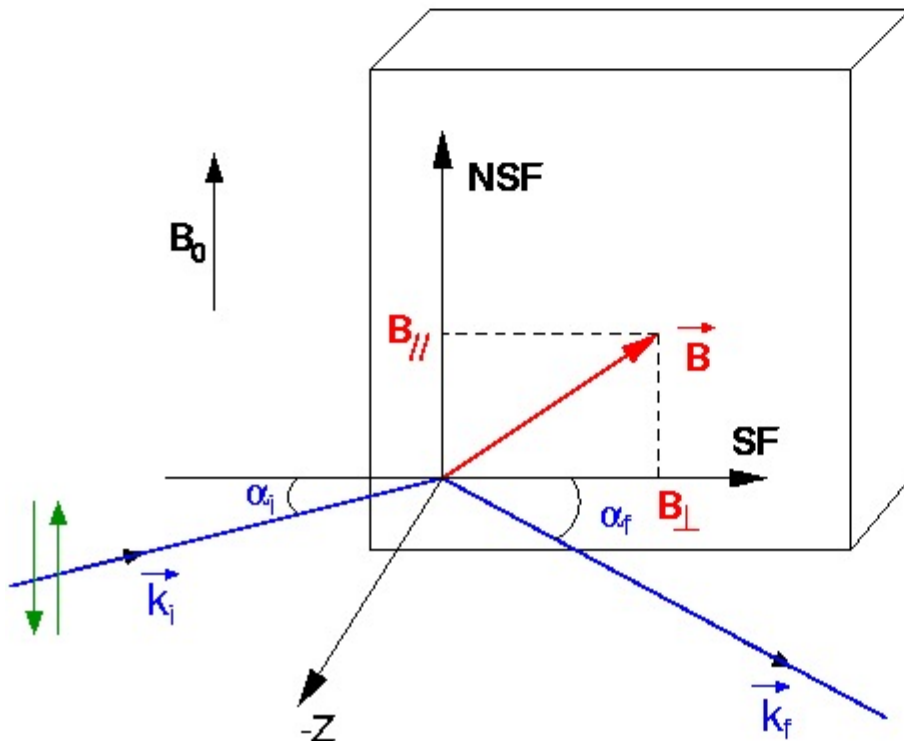
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Interaction of polarized neutrons with magnetic thin films

The neutron is a spin $\frac{1}{2}$ particle

$$|\Psi_i(\mathbf{r})\rangle = \Psi_i^+(\mathbf{r})|+\rangle + \Psi_i^-(\mathbf{r})|-\rangle$$



$$I_{++}, I_{--} \Rightarrow \rho_l^N, B_{\parallel}$$

$$\text{Mitglied de } I_{+-}, I_{-+} \Rightarrow B_{\perp}$$

$$\hat{V}_l^M = -\mu_n \hat{\sigma} \cdot \mathbf{B}_l$$

$\hat{\sigma}$ is the vector of Pauli matrices

Total interaction operator:

$$\hat{V}_l = \frac{2\pi \hbar^2}{m} \left(\rho_l^N \hat{1} + \rho_l^M \hat{\sigma} \cdot \mathbf{b}_l \right)$$

- ρ_l^N is the nuclear scattering length density
- ρ_l^M is the magnetic scattering length density

Schrödinger equation

$$|\Psi_l(\mathbf{r})\rangle = \Psi_l^+(\mathbf{r}) |+\rangle + \Psi_l^-(\mathbf{r}) |-\rangle = \begin{pmatrix} \Psi_l^+(\mathbf{r}) \\ \Psi_l^-(\mathbf{r}) \end{pmatrix}$$

$$\begin{aligned} \Psi_l^{+''}(z) + [k_{z,l}^2 - 4\pi(\rho_l^N + \rho_l^M b_{\parallel})] \Psi_l^+(z) + 4i\pi\rho_l^M b_{\perp} \Psi_l^-(z) &= 0 \\ \Psi_l^{-''}(z) + [k_{z,l}^2 - 4\pi(\rho_l^N - \rho_l^M b_{\parallel})] \Psi_l^-(z) - 4i\pi\rho_l^M b_{\perp} \Psi_l^+(z) &= 0 \end{aligned}$$

Non-spin-flip

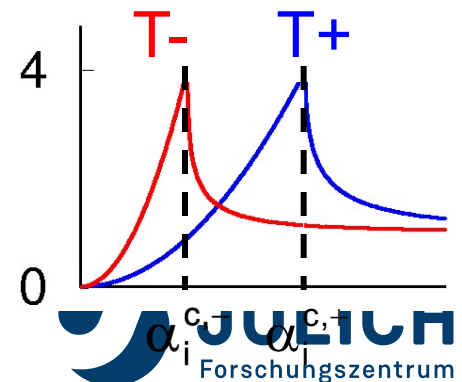
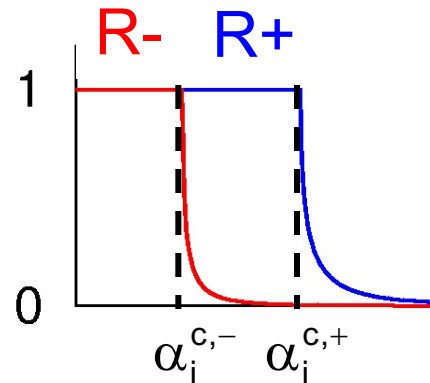
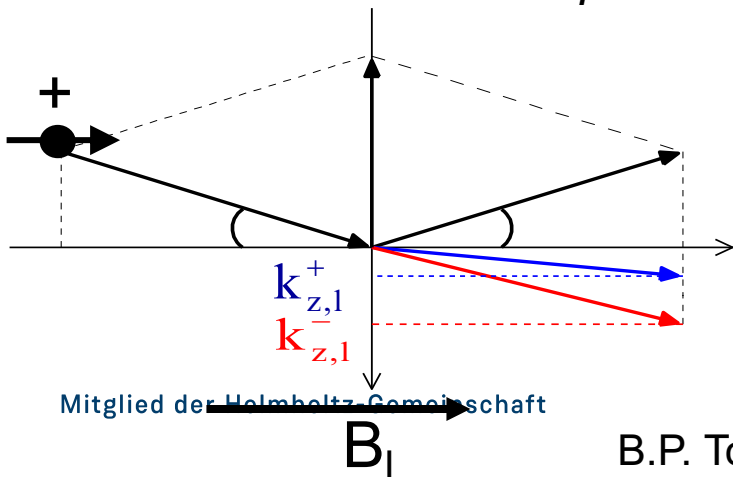
Spin-flip

Neutron wave function in layer I

$$|\Psi_I(\mathbf{r})\rangle = e^{i\mathbf{k}\cdot\mathbf{r}} (e^{i\hat{k}_{z,I}\cdot z} \hat{t}_I + e^{-i\hat{k}_{z,I}\cdot z} \hat{r}_I) |\Psi_0\rangle$$

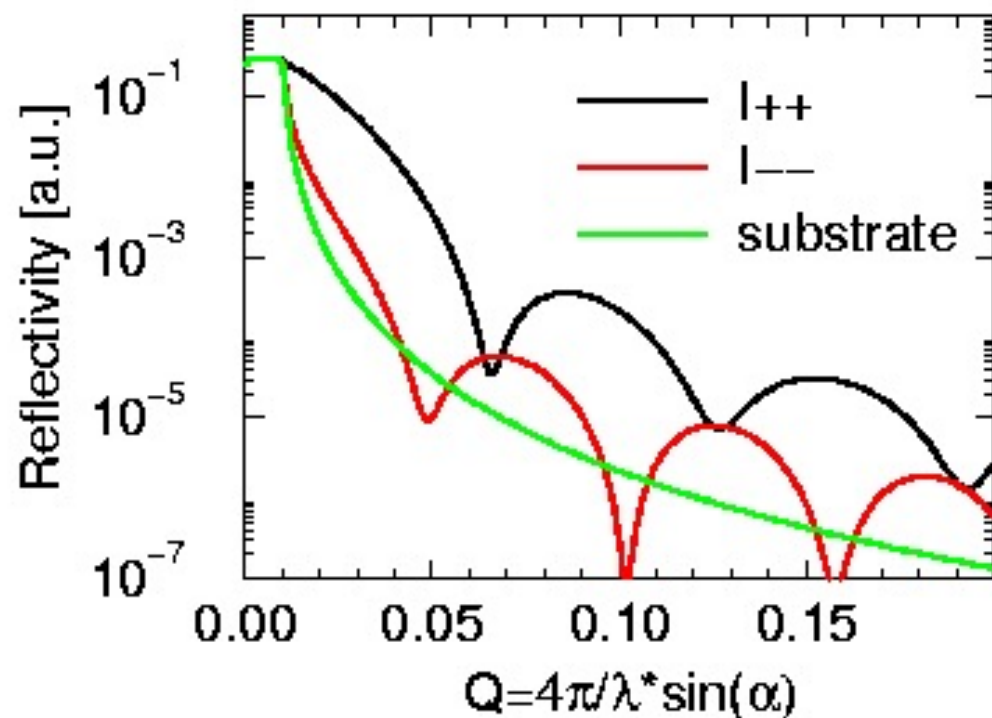
$$\hat{k}_{z,I} = \sqrt{\hat{k}_{z,0}^2 - 4\pi(\rho_I^N + \rho_I^M \hat{\sigma} \cdot \mathbf{b}_I)}$$

- $\hat{k}_{z,I}$, $\hat{t}_I$ and $\hat{r}_I$ have different eigenvalues depending on the projection of the spin on the quantization axis defined by $\mathbf{b}_I$ (i.e. $\hat{\sigma} \cdot \mathbf{b}_I = \pm 1$).

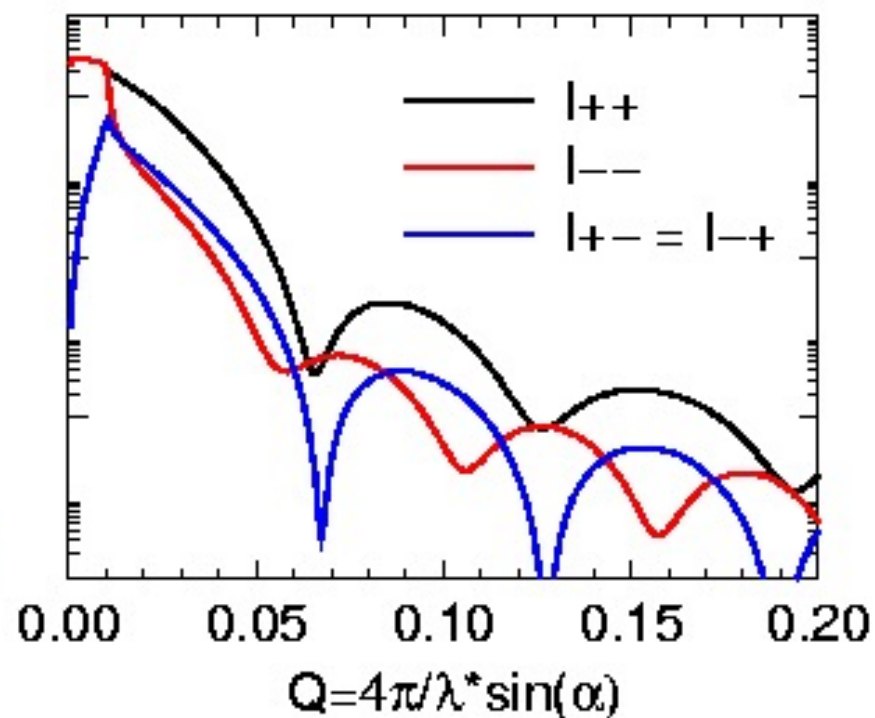


Magnetization collinear / non-collinear to B_0

$M_{\text{Fe}} // B_0$



$(M_{\text{Fe}}, B_0) = 45^\circ$

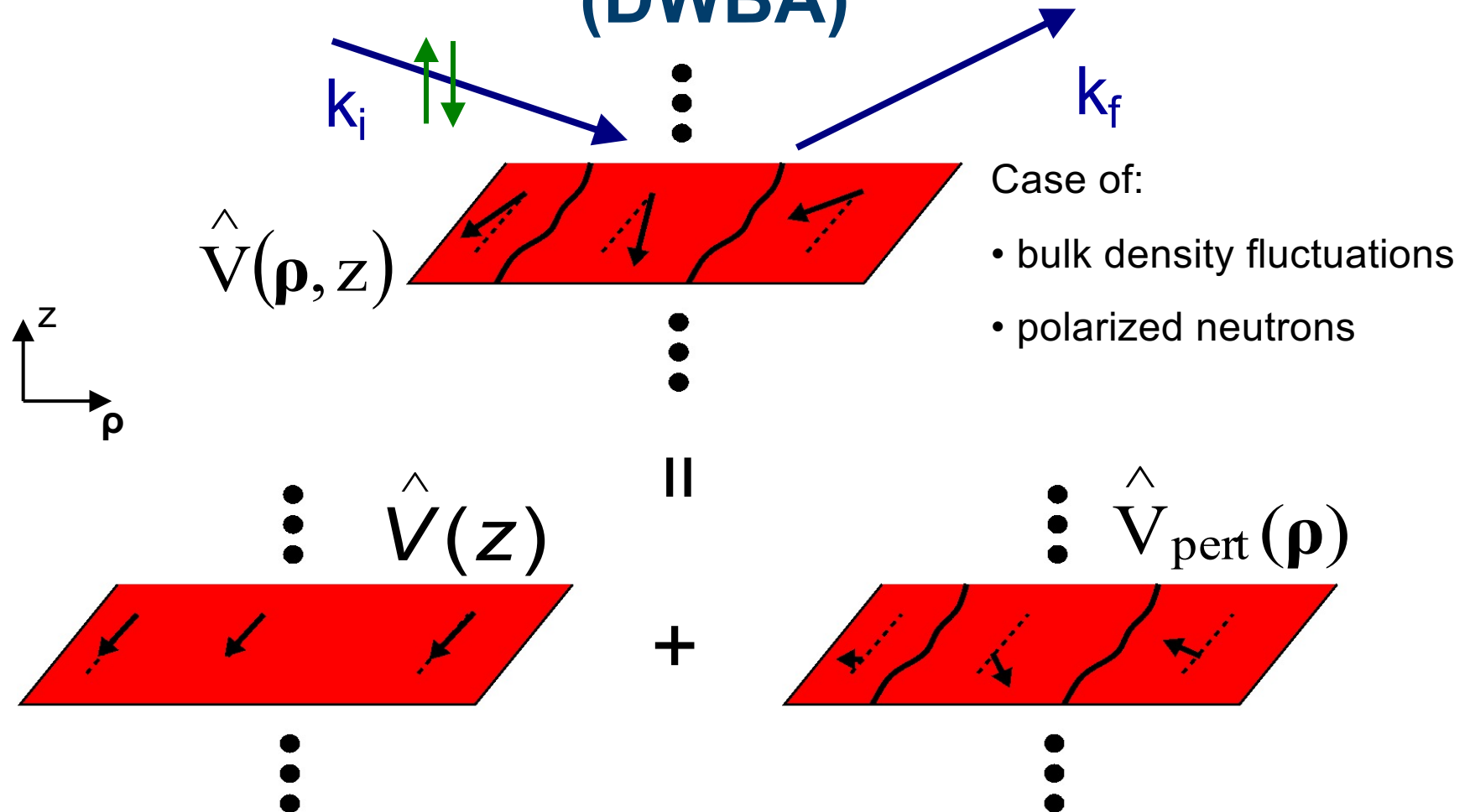


Fe (15 nm) / Au (2nm) on Si

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DISTORTED WAVE BORN APPROXIMATION (DWBA)



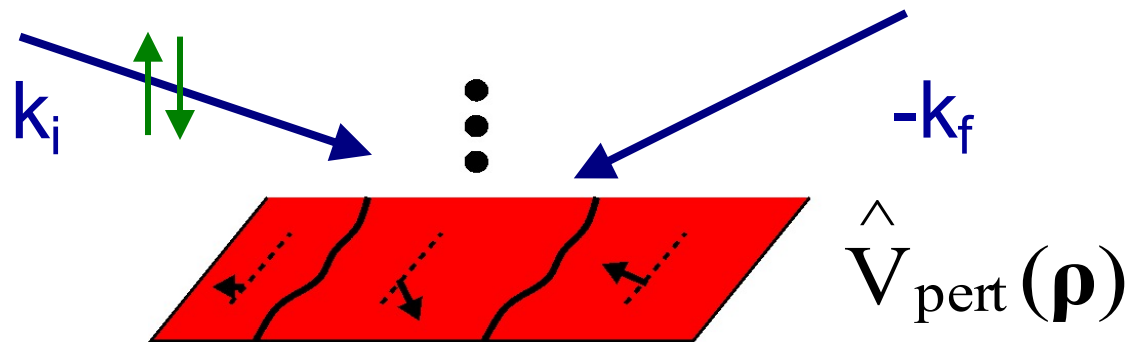
Leads to specular reflectivity

Mitglied der Helmholtz-Gemeinschaft

Leads to scattering

37

SCATTERING CROSS SECTION WITHIN DWBA



Average over the spin states

$$\frac{d\sigma}{d\Omega}(\mathbf{k}_i, \mathbf{k}_f) = \left\langle \left| \frac{m}{2\pi\hbar^2} \langle \Psi^f | \hat{V}_{pert} | \Psi^i \rangle \right|^2 \right\rangle$$

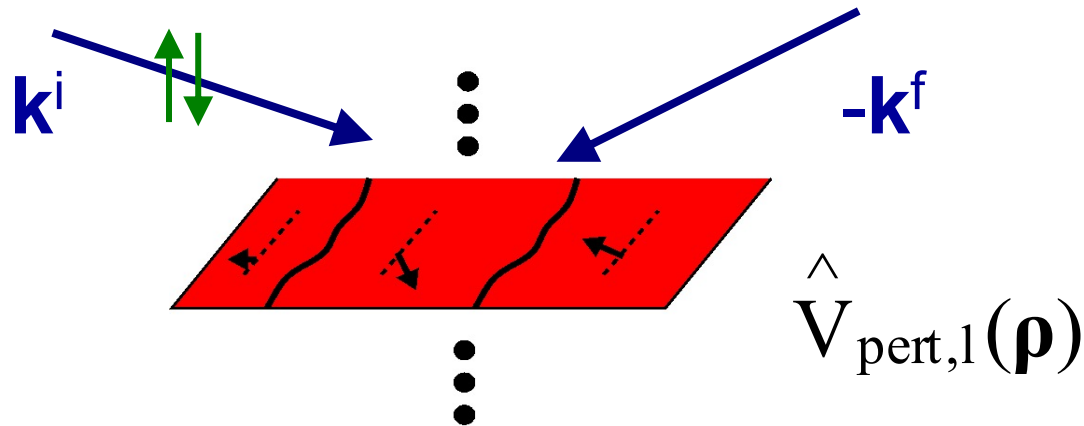
Configurational average

$\left\{ \begin{array}{l} |\Psi^i\rangle \\ \langle \Psi^f| \end{array} \right\}$
 are solutions of the Schrödinger equation for $\hat{V}(z)$

and incoming wave vectors

$$\left\{ \begin{array}{l} \mathbf{k}_i \\ -\mathbf{k}_f \end{array} \right\}$$

SCATTERING AMPLITUDE WITHIN DWBA



$$F(\mathbf{Q}_{\parallel}, k_{z,0}^f, k_{z,0}^i) = \sum_l \int dz \langle \Psi_0^f(\mathbf{k}^f, 0) | \cdot \hat{S}_l^{f+}(z) \cdot \hat{\mathbf{F}}_l(\mathbf{Q}_{\parallel}) \cdot \hat{S}_l^i(z) \cdot | \Psi_0^i(\mathbf{k}^i, 0) \rangle$$

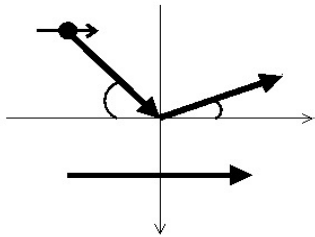
with: $\hat{\mathbf{F}}_l(\mathbf{Q}_{\parallel}) = -\frac{m}{2\pi\hbar^2} \int d\boldsymbol{\rho} e^{-i\mathbf{Q}_{\parallel} \cdot \boldsymbol{\rho}} \hat{V}_{\text{pert},l}(\boldsymbol{\rho})$ and $\mathbf{Q}_{\parallel} = \boldsymbol{\kappa}^f - \boldsymbol{\kappa}^i$

• $\hat{\mathbf{F}}_1(\mathbf{Q}_{\parallel})$: Fourier transform of the fluctuations. Like in BA!

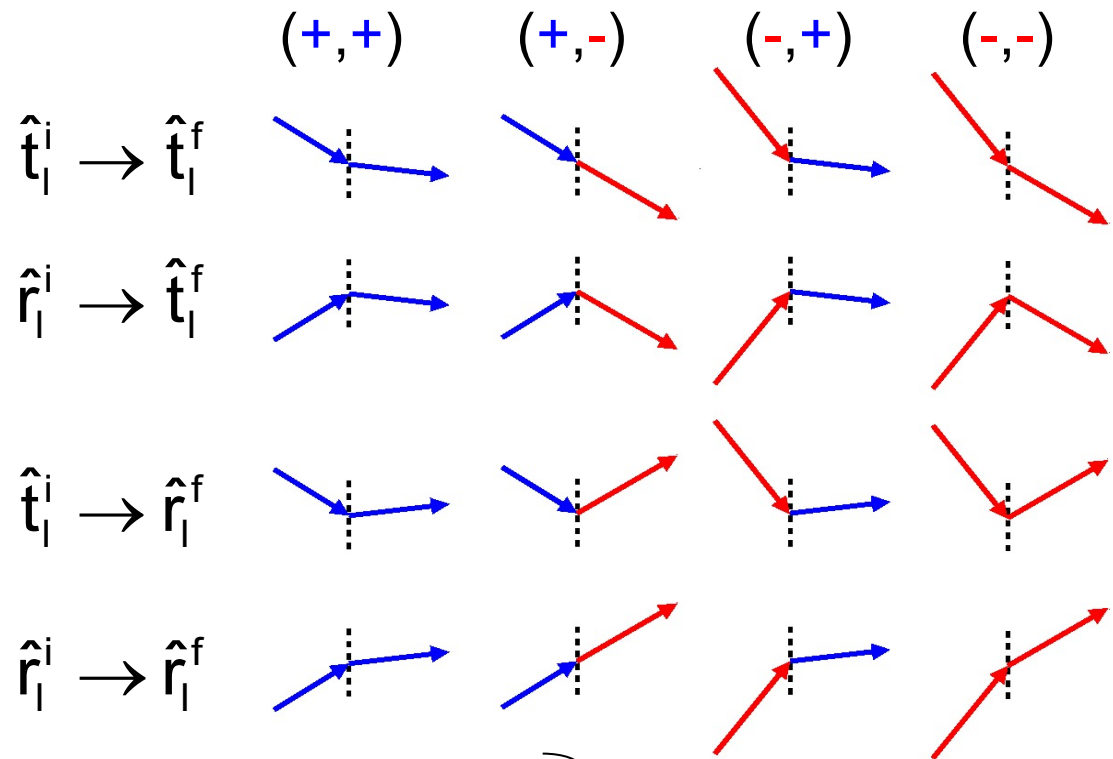
• $\hat{S}_1^i(z)$ and $\hat{S}_1^f(z)$ depend only on the reference potential $\hat{V}(z)$!

SCATTERING AMPLITUDE WITHIN DWBA

$$\left| \Psi_l^{i(f)}(\mathbf{r}) \right\rangle = e^{i\boldsymbol{\kappa}^{i(f)} \cdot \boldsymbol{\rho}} \left(e^{(\pm) i \hat{k}_{z,l}^{i(f)} \cdot z} \hat{\mathbf{t}}_l^{i(f)} + e^{(\mp) i \hat{k}_{z,l}^{i(f)} \cdot z} \hat{\mathbf{r}}_l^{i(f)} \right) \left| \Psi_0^{i(f)} \right\rangle$$



The scattering amplitude inside layer I decomposes virtually into 16 terms:



$\left. \begin{matrix} (+, +) \\ (-, -) \end{matrix} \right\}$ are illuminated by nuclear and longitudinal magnetic fluctuations
integrated by Helmholtz-Gemeinschaft

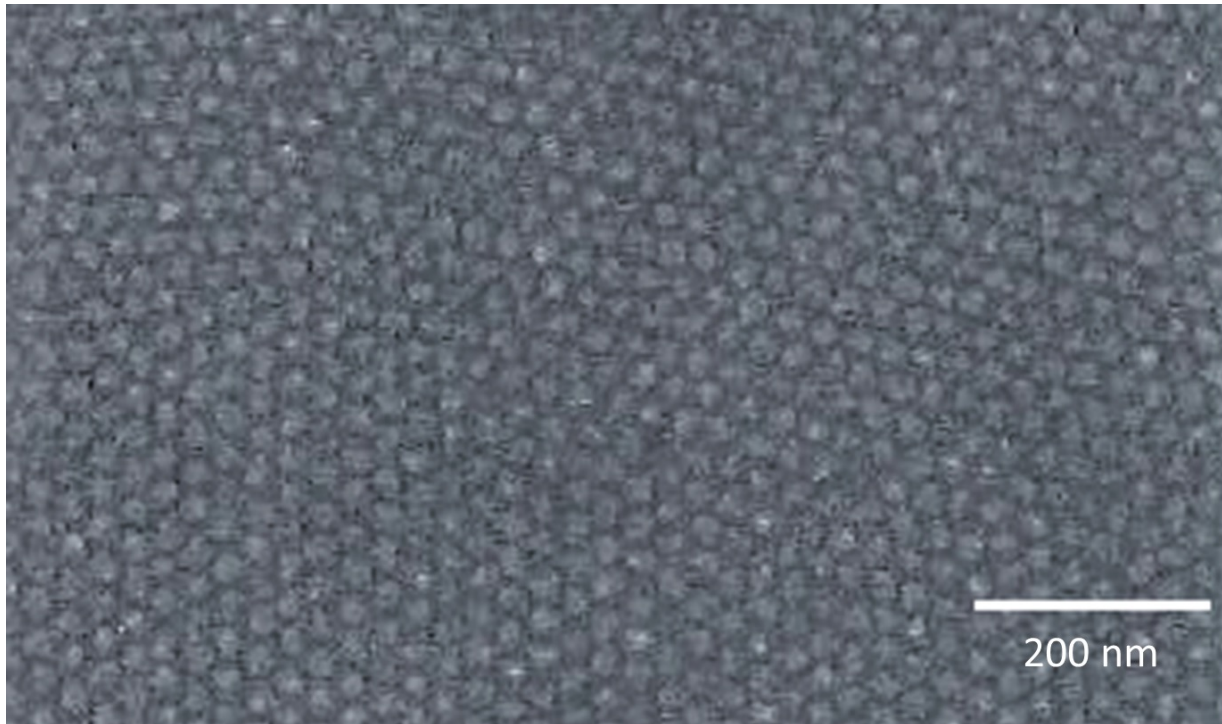
$\left. \begin{matrix} (+, -) \\ (-, +) \end{matrix} \right\}$ are illuminated by transverse magnetic fluctuations
Forschungszentrum

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- Take-Home Message

Ordered assembly of CoFe_2O_4 nanoparticles

(Asmaa Qdemat, JCNS)



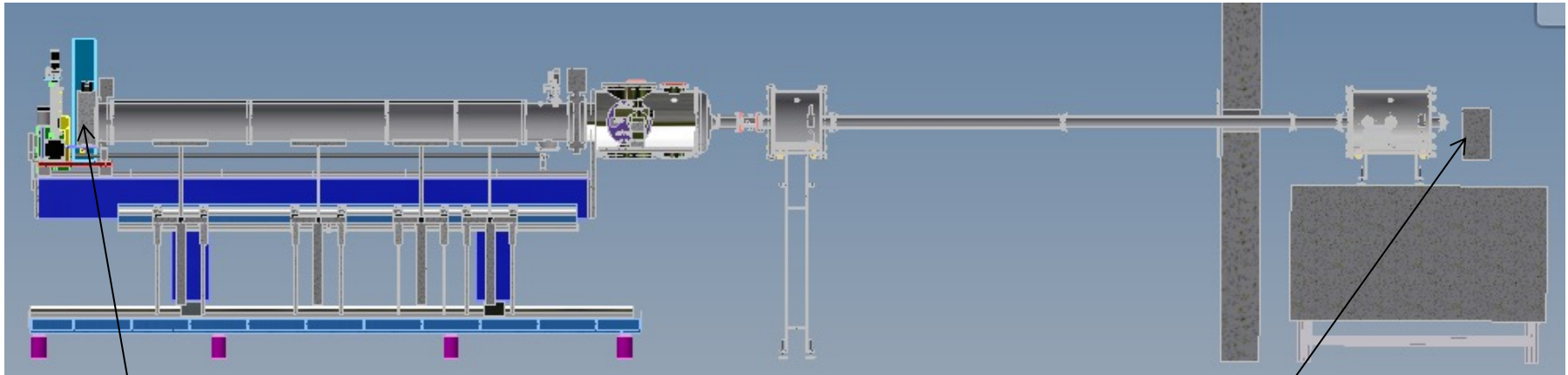
Giuseppe Portale
(Groningen, NL)

- Size ~ 20 nm
- Hexagonal ordering

GALAXI@JCNS

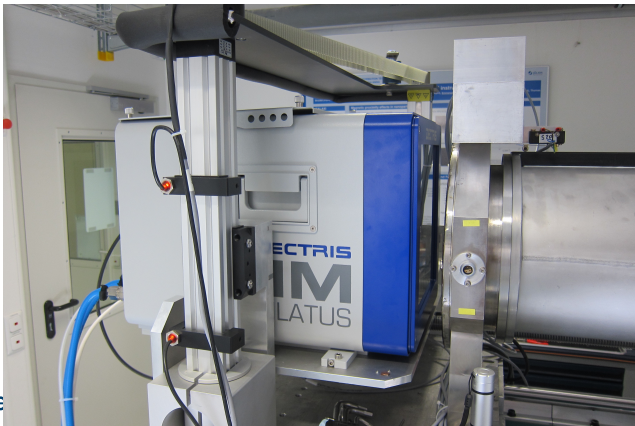
- High brilliance laboratory GISAXS diffractometer

Ulrich Rücker

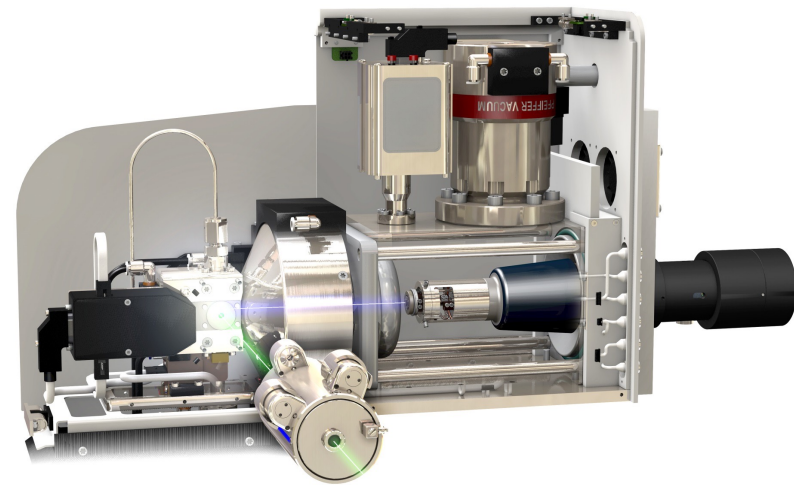


1M Pilatus from Dectris

Metaljet from Bruker AXS



Mitglie

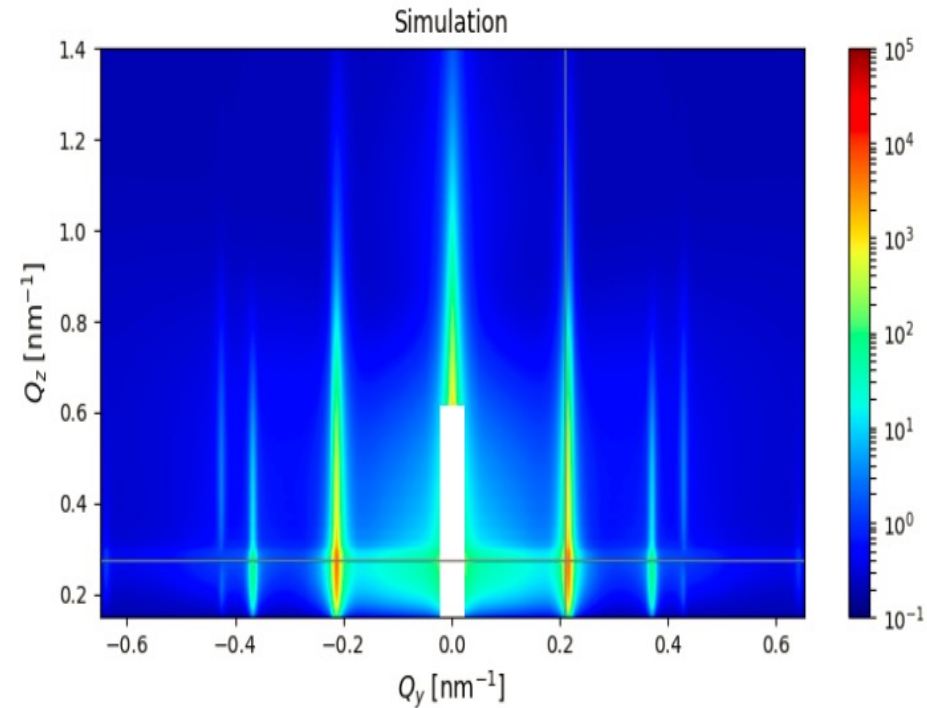
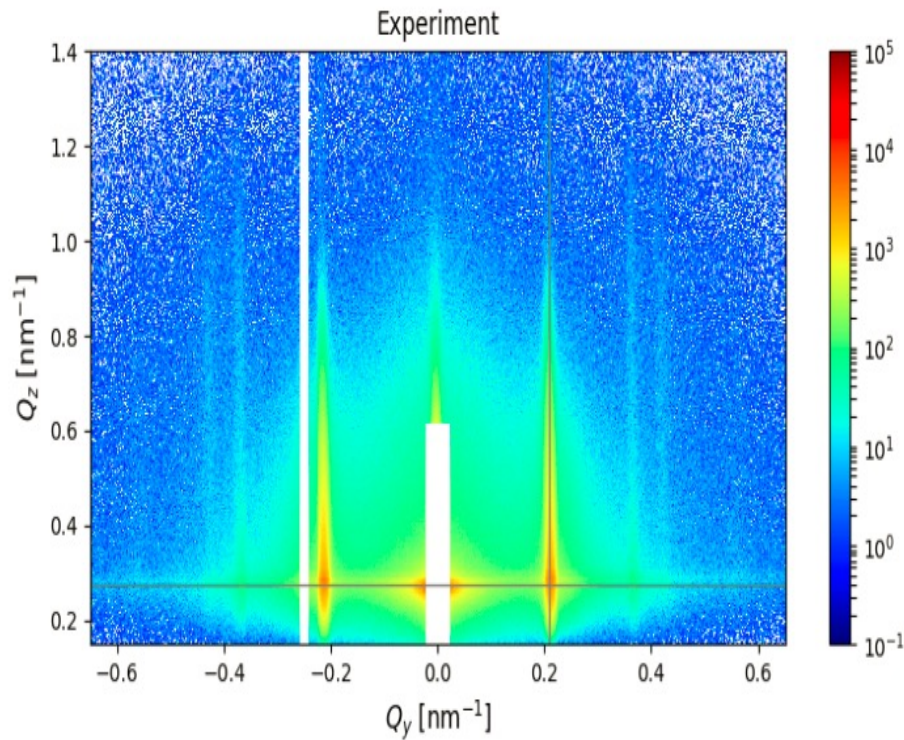
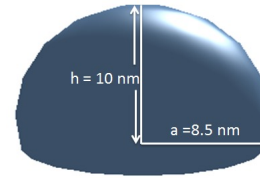


1stnn=28 nm

a=8.5 nm

h=10 nm

$$\alpha_i = 0.17^\circ$$



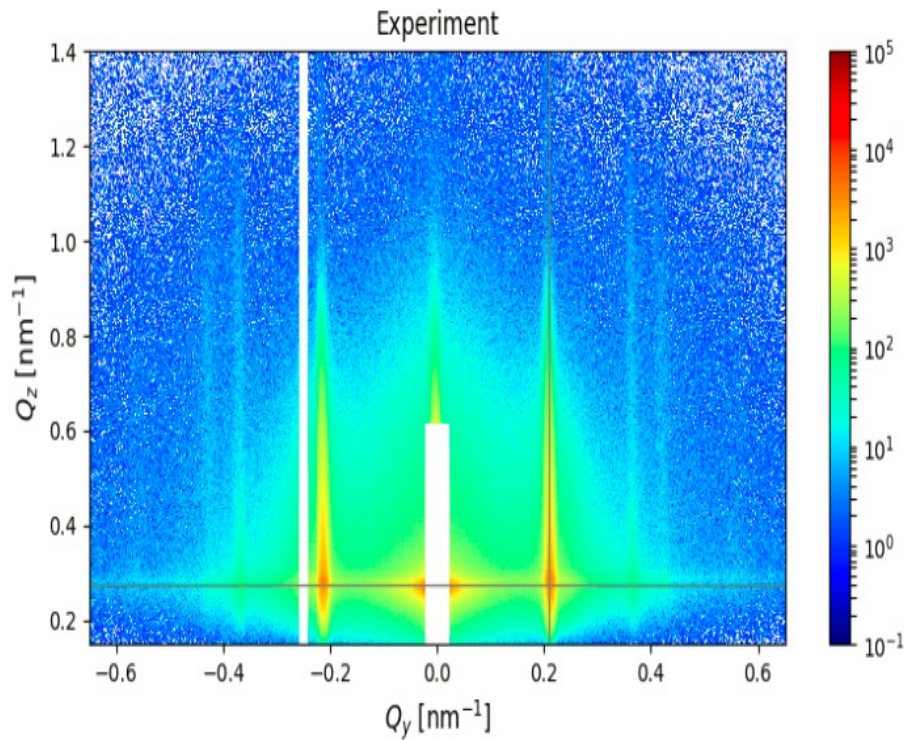
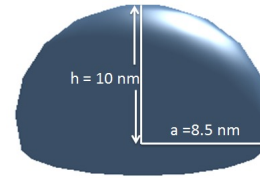
www.bornagainproject.org

1stnn=28 nm

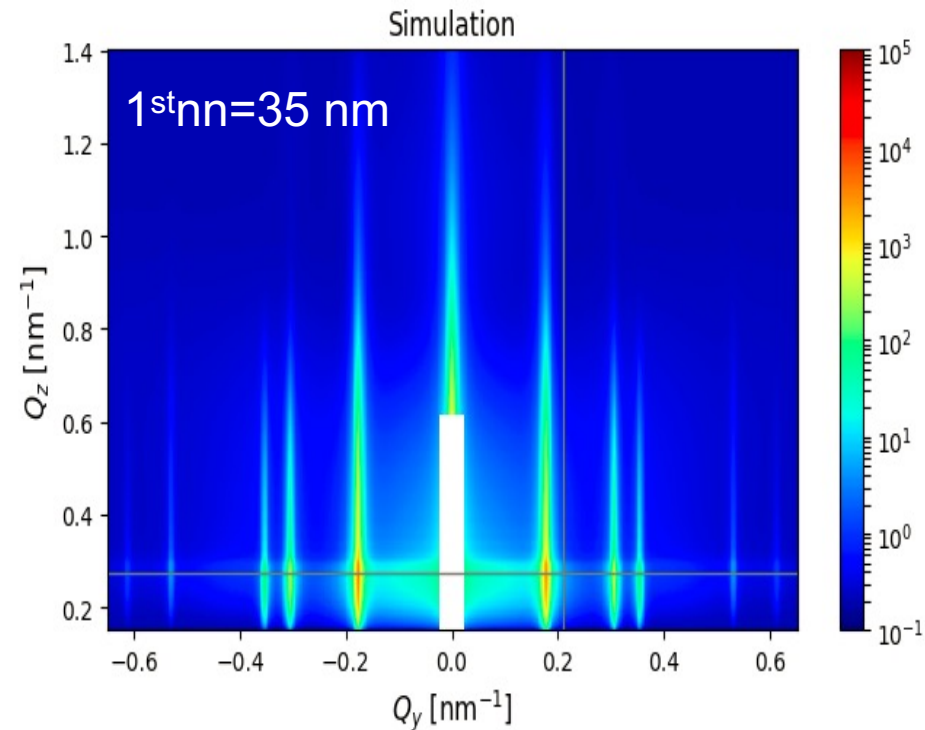
a=8.5 nm

h=10 nm

$$\alpha_i = 0.17^\circ$$



GALAXI



BornAgain program :

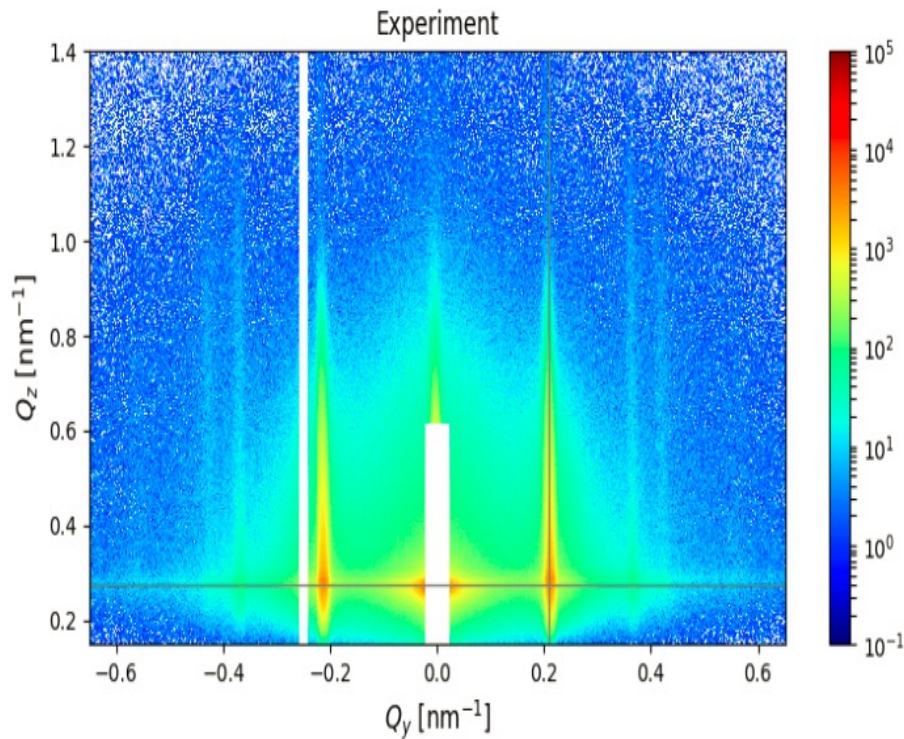
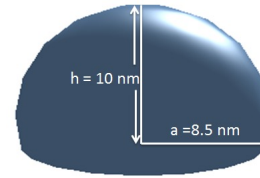
www.bornagainproject.org

1stnn=28 nm

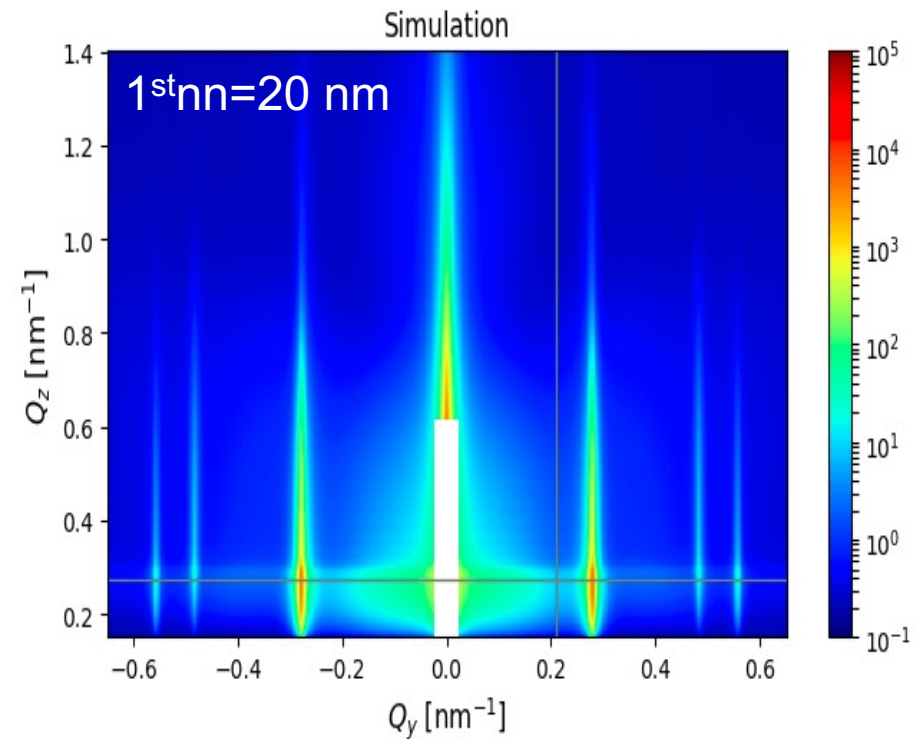
a=8.5 nm

h=10 nm

$$\alpha_i = 0.17^\circ$$



GALAXI



BornAgain program :

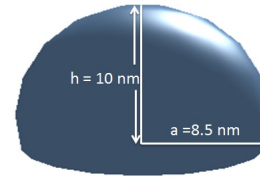
www.bornagainproject.org

1stnn=28 nm

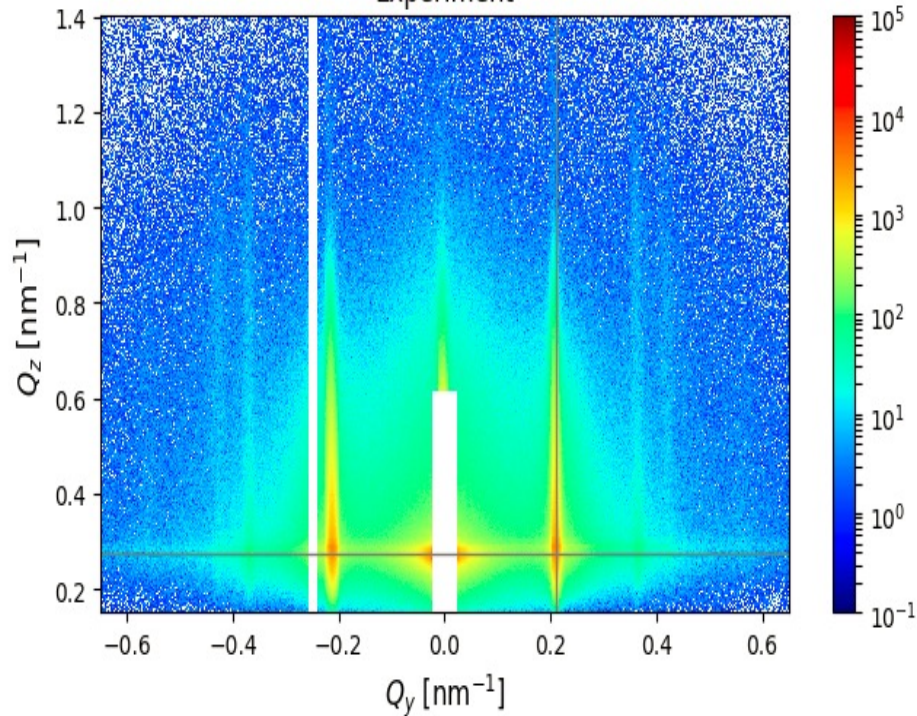
a=8.5 nm

h=10 nm

$$\alpha_i = 0.17^\circ$$

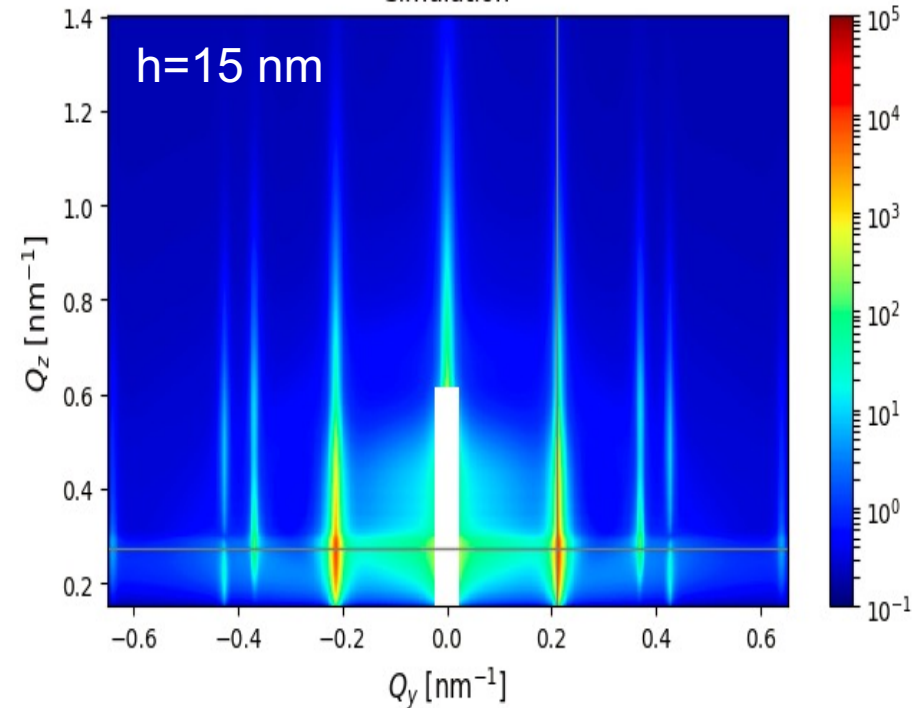


Experiment



GALAXI

Simulation



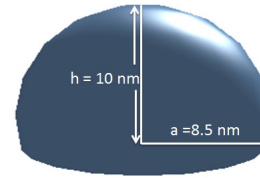
BornAgain program :

www.bornagainproject.org

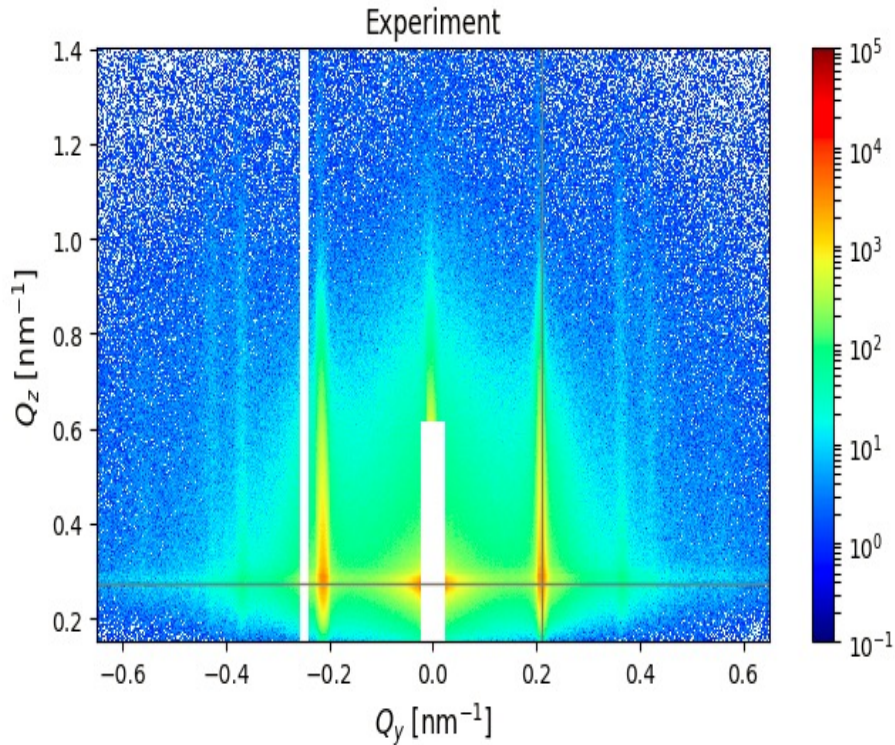
1stnn=28 nm

a=8.5 nm

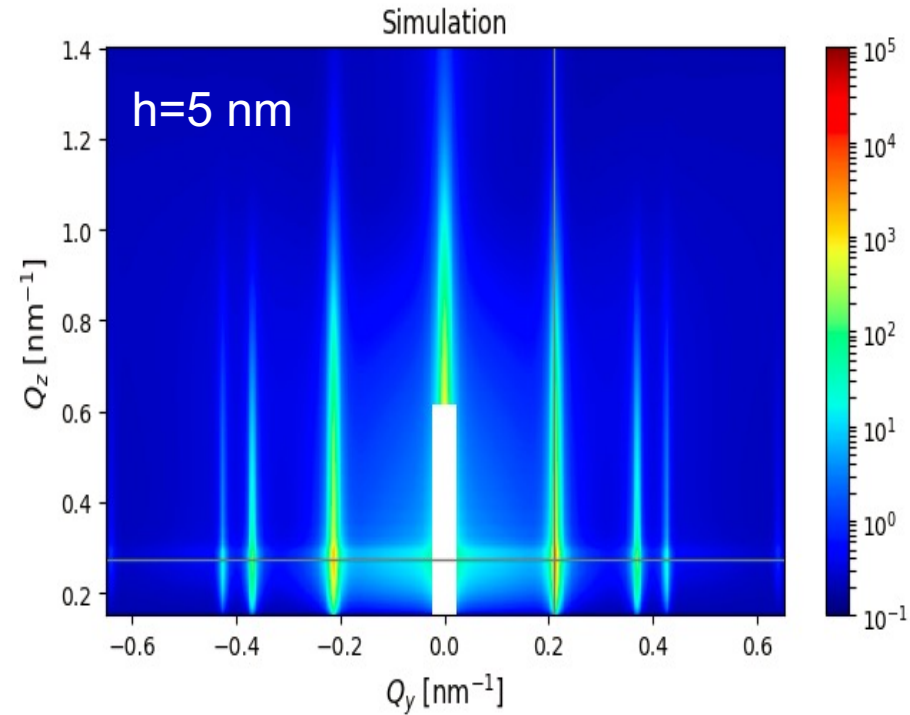
h=10 nm



$$\alpha_i = 0.17^\circ$$



GALAXI



BornAgain program :

www.bornagainproject.org

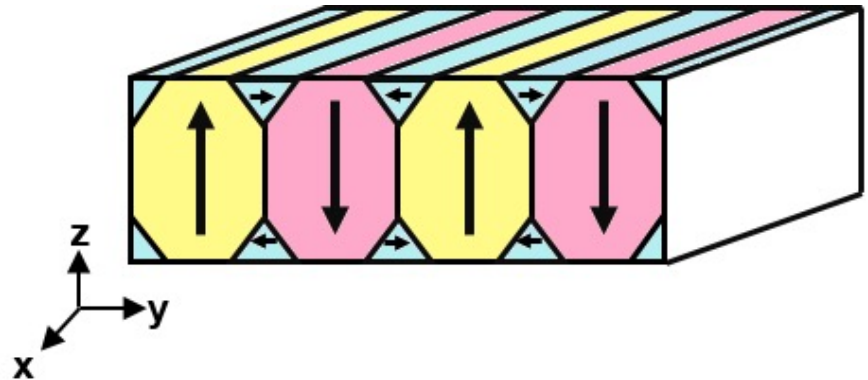
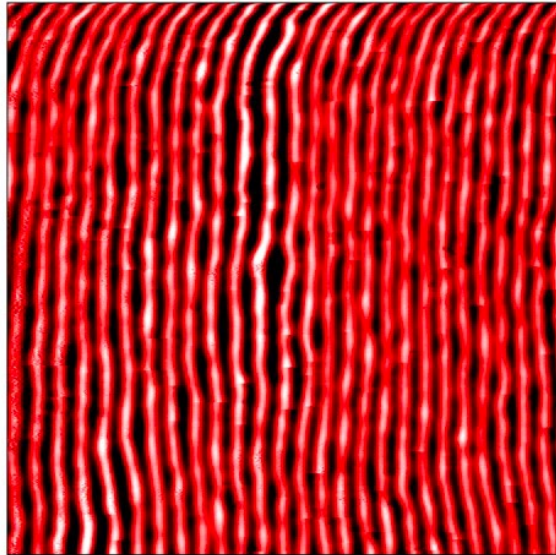
PLAN

- Introduction: Spintronics
- Method: Reflectivity, off-specular scattering, GISAS and neutron polarization analysis
- Simulation of specular reflectivity
- Simulation of polarized neutron reflectivity
- Simulation of GISAS intensities within the Distorted Wave Born Approximation
- Structural ordering in assemblies of magnetic nanoparticles
- **Magnetic domain structure in FePd with perpendicular-to-plane anisotropy**
- Take-Home Message

FePd layer with perpendicular magnetic anisotropy

(Annika Stellhorn, JCNS)

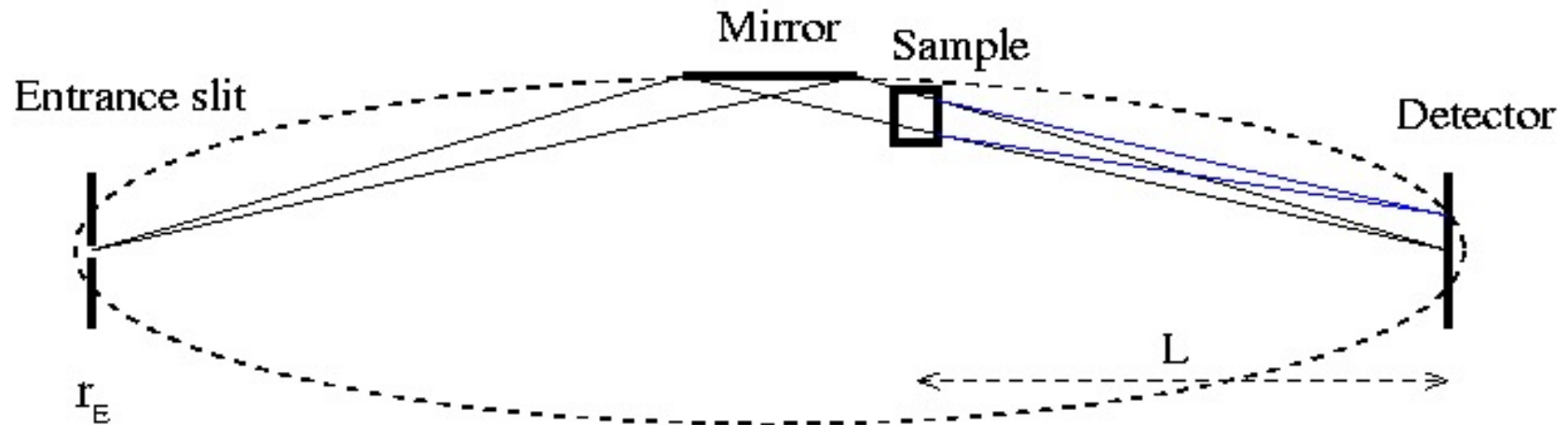
MFM



A. Stellhorn et al. JMMM **476**, 483 (2019)

KWS-3 @ MLZ (V. Pipich)

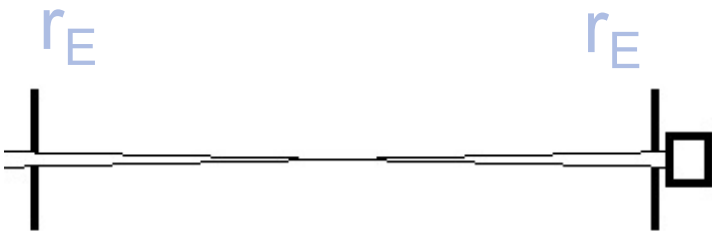
1:1 imaging of an entrance aperture onto a detector by reflection on an ellipsoidal mirror



$$\delta Q = \frac{k \cdot r_E}{L} \quad \left(k = \frac{2\pi}{\lambda} \right) \quad \left. \begin{array}{l} k = 0.5 \text{ \AA}^{-1} \\ r_E = 2 \text{ mm} \\ L = 10 \text{ m} \end{array} \right\} \leadsto \delta Q = 10^{-4} \text{ \AA}^{-1}$$

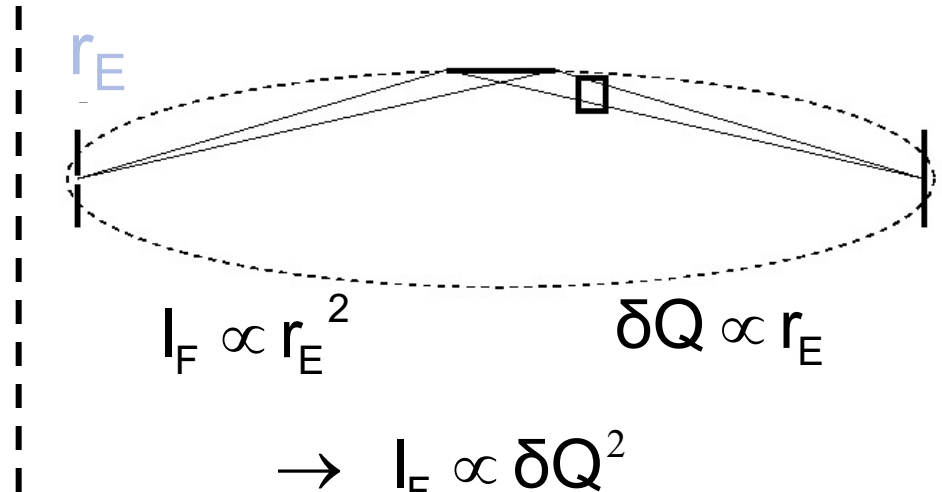
→ Any physical issue at the μm length scale!

Comparison: Pinhole / Focusing mirror SANS



$$I_P \propto r_E^4 \quad \delta Q \propto r_E$$

$$\rightarrow I_P \propto \delta Q^4$$



$$I_F \propto r_E^2 \quad \delta Q \propto r_E$$

$$\rightarrow I_F \propto \delta Q^2$$

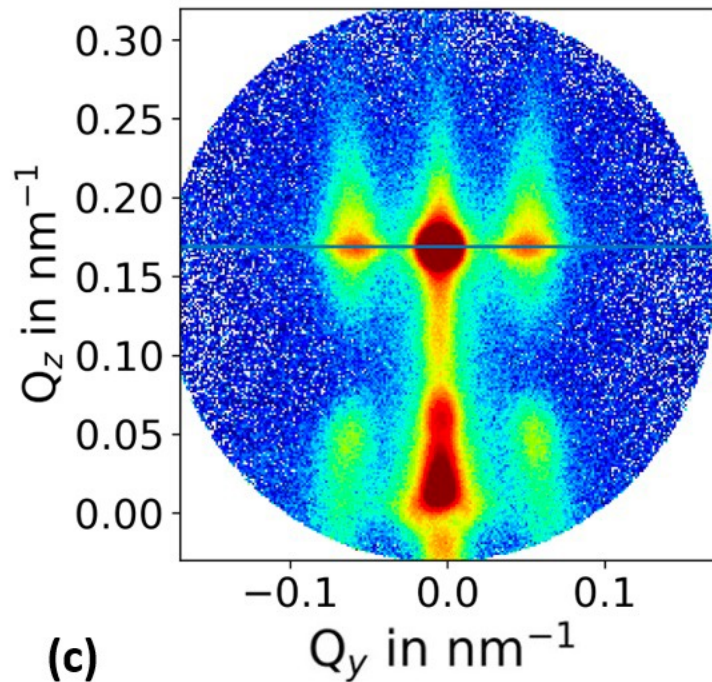
Therefore: $\frac{I_F}{I_P} \propto \frac{1}{\delta Q^2}$

$$\frac{I_F}{I_P} > 1 \quad \text{für} \quad \delta Q < 7.6 \times 10^{-3} \text{ \AA}^{-1}$$

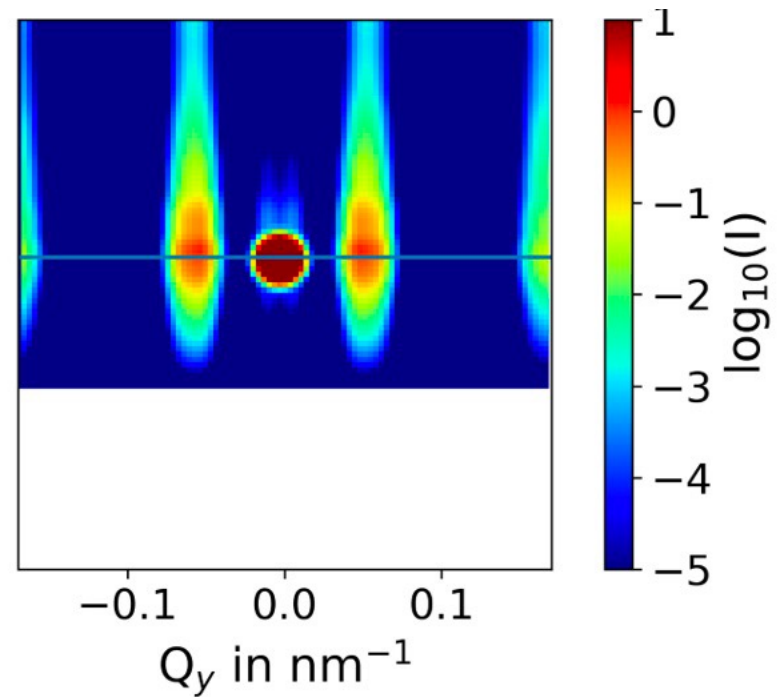
The KWS-3 Mirror



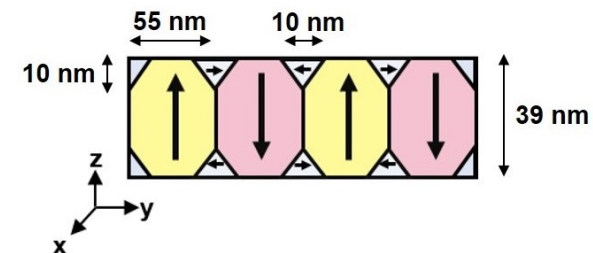
GISANS @ KWS-3



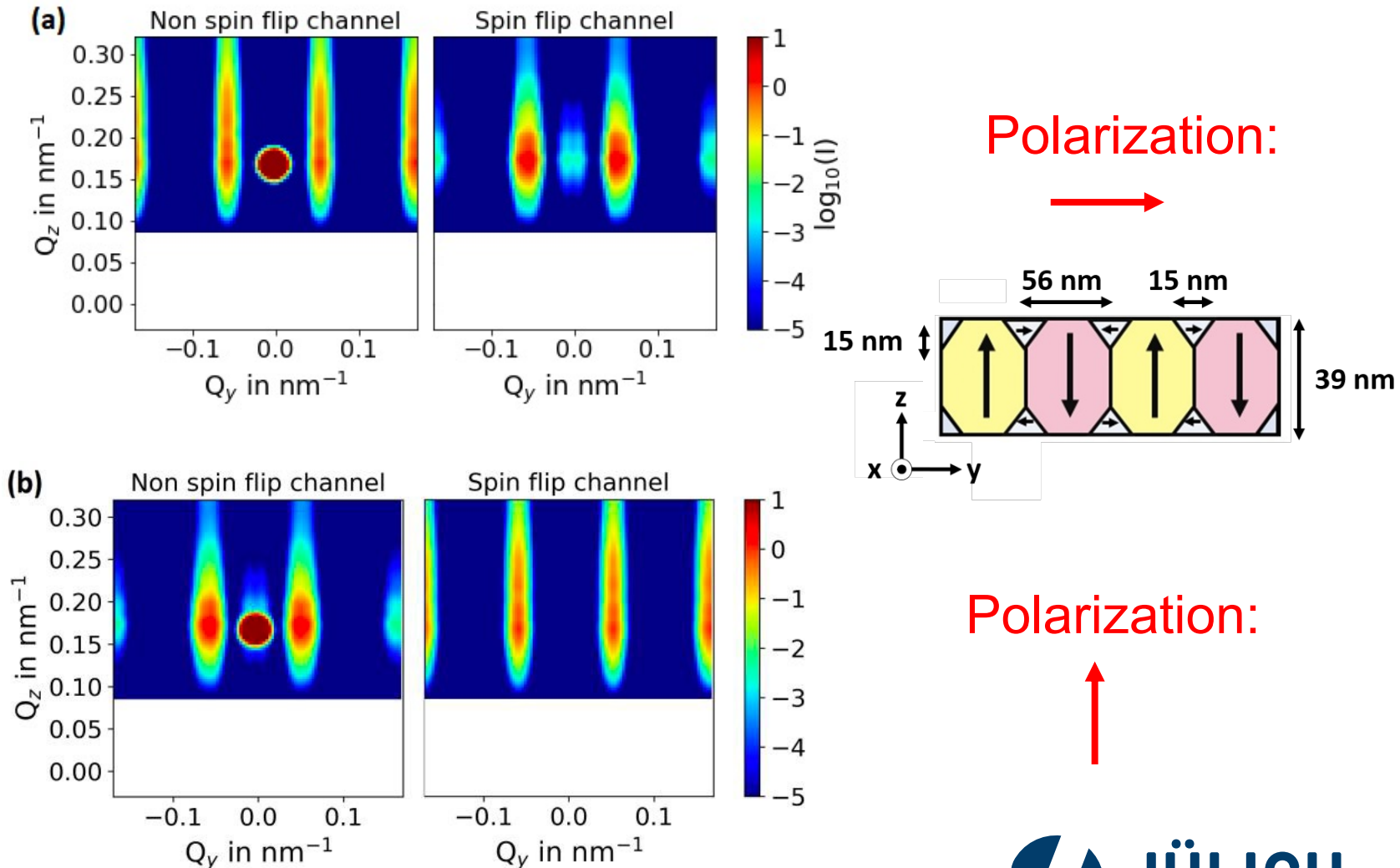
DWBA simulation



A. Stellhorn et al. *JMMM* **476**, 483 (2019)



3D vector magnetometry by GISANS with polarization analysis (DWBA simulations)



PLAN

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- Magnetic domain structure in FePd with perpendicular-to-plane anisotropy
- **Take-Home Message**

TAKE-HOME MESSAGE

- **Reflectometry from thin films:**
 1. Scattering length density profile along their depth
 2. Information averaged over in-plane coordinates
- **Grazing Incidence Small Angle Scattering:**
 1. In-plane correlations can be investigated
 2. Depth-resolution accessed by interpreting signal as a function of angles of incidence and exit
- **Neutron polarization analysis:**
 - 3D vector magnetometry

THANKS TO...

The Students:

- Asmaa Qdemat
- Annika Stelhorn
- Tanvi Bhatnagar
- Patrick Schöffmann

Simulations:

- Marina Ganeva

The Instrument Scientists:

- Anirban Sarkar
- Vitaliy Pipich
- Stefan Mattauch
- Ulrich Rücker
- Earl Babcock

Everything:

- Thomas Brückel

...AND YOU FOR YOUR ATTENTION