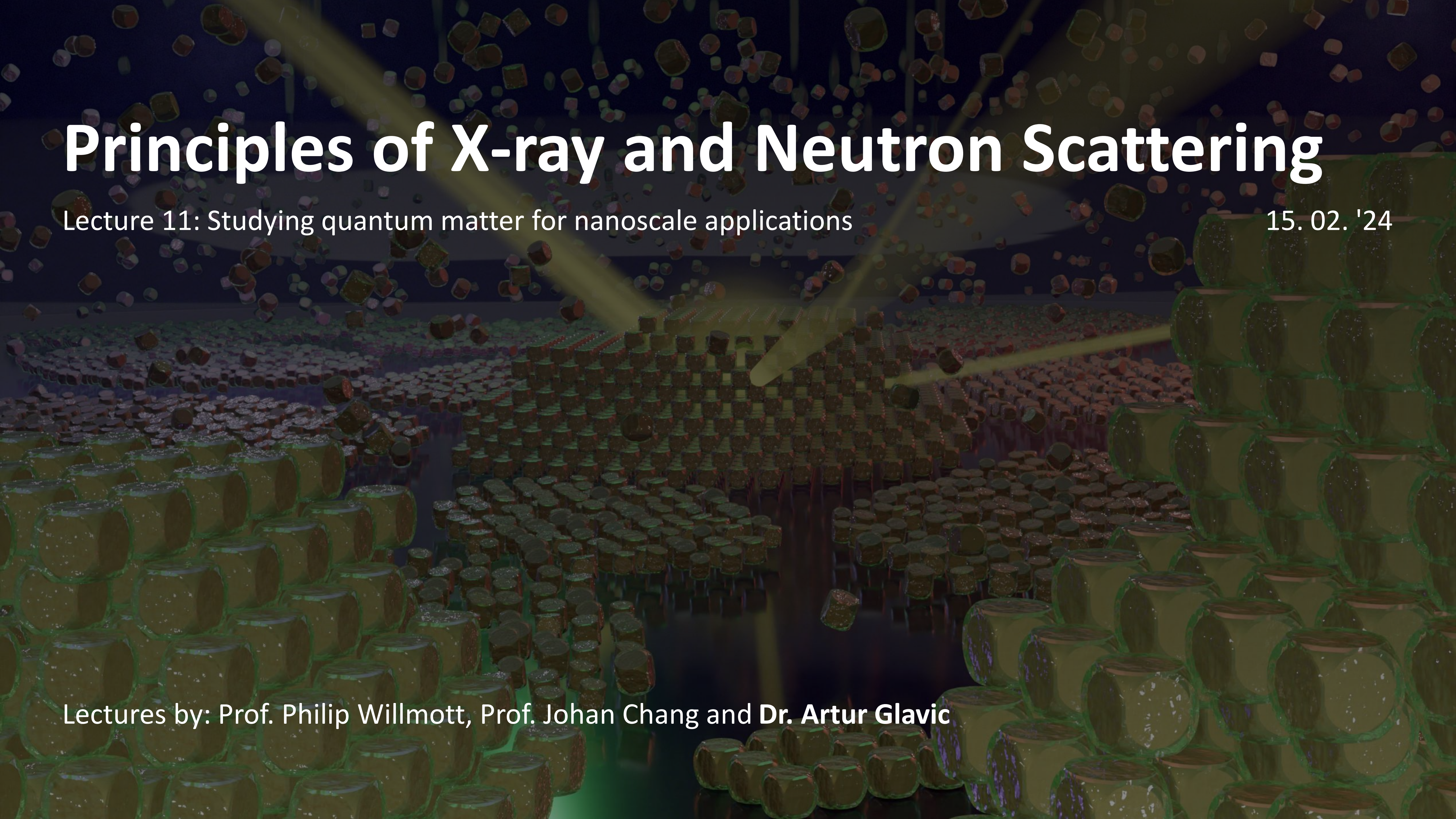


Principles of X-ray and Neutron Scattering

Lecture 11: Studying quantum matter for nanoscale applications 15. 02. '24

Lectures by: Prof. Philip Willmott, Prof. Johan Chang and **Dr. Artur Glavic**

Course Outline

Monday	Tuesday	Wednesday	Thursday	Friday
Lecture 1 10-10h45 Philip	**Lecture 4** 10-10h45 Philip	**Lecture 7** 10-10h45 Artur	**Lecture 10** 10-10h45 Artur	**Lecture 13** 10-10h45 Johan
Lecture 2 11-11h45 Philip	**Lecture 5** 11-11h45 Philip	**Lecture 8** 11-11h45 Artur	**Lecture 11** 11-11h45 Artur	**Lecture 14** 11-11h45 Johan
Lunch - Mensa	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**
Lecture 3 13h00-13h45 Philip	**Lecture 6** 13h00-13h45 Philip	**Lecture 9** 13h00-13h45 Artur	**Lecture 12** 13h00-13h45 Artur	**Lecture 15** 13h00-13h45 Johan
		Exercise Class 14h30-16		Exercise Class 14h30-16

X-ray scattering

Neutron Scattering

Resonant x-ray scattering

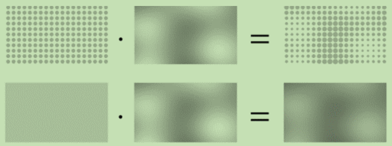
Neutron Lectures:

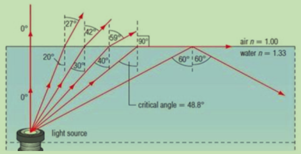
7: Neutrons & Scattering to Determine Structure
8: Inelastic Neutron Scattering to Investigate Dynamics
9: Magnetic Scattering
10: Neutron Polarization Analysis
11: Studying quantum matter for nanoscale applications
12: Neutron Instrument Development

Lecture 11: Studying quantum matter for nanoscale applications

Theoretical Background

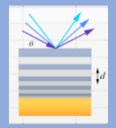
- Continuum description of matter
- Dynamic effects at grazing incidence

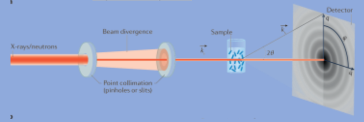


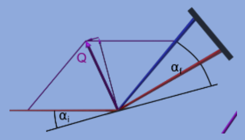


Practical Implementation

- Neutron guides and focusing optics
- SANS and reflectometry instruments







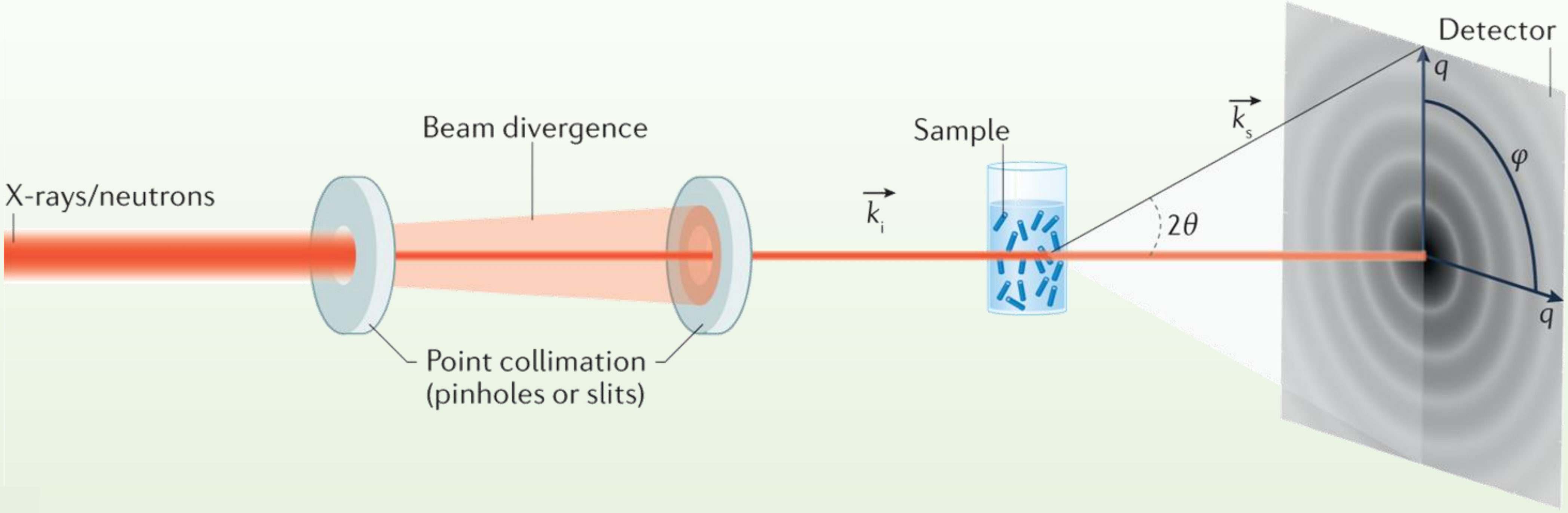
Example Application

- Surface spin canting in magnetic nanoparticles
- GISANS on frustrated artificial spins

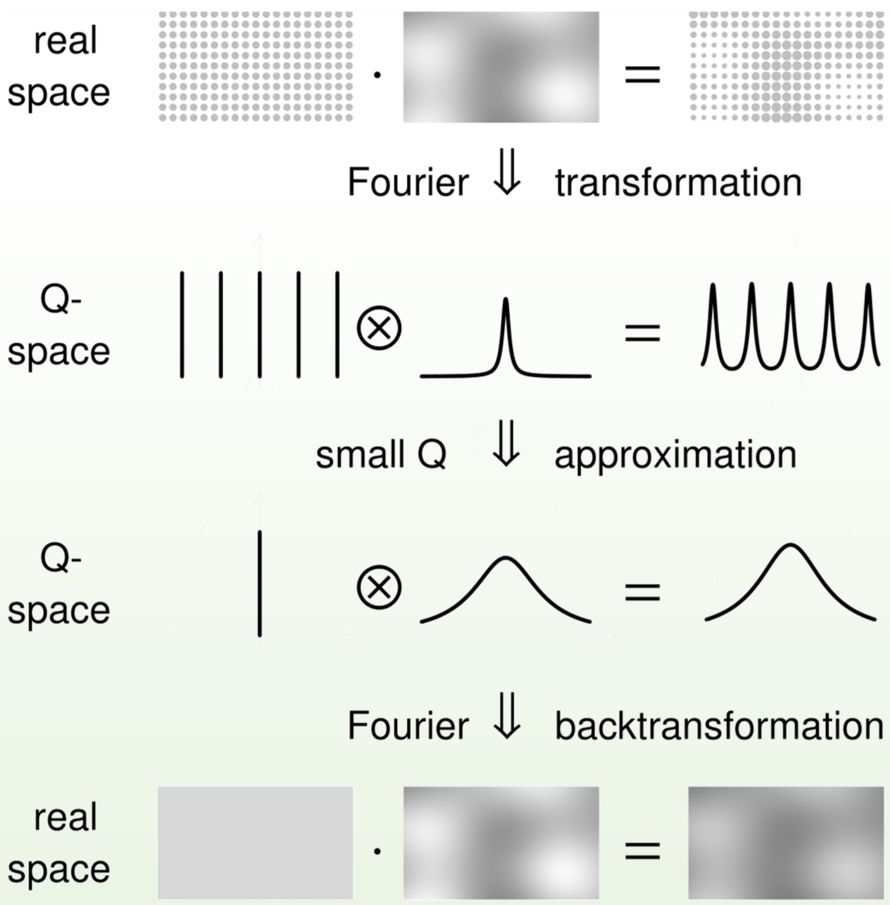
Small Angle (Neutron) Scattering – SA(N)S

- Interest in structures with sizes 1-100 nm
- Very large compared with neutron wavelength (10x-1000x)
- Due to properties of Fourier transform the relevant signal is at small Q ➔ small scattering angle

- Instruments require very good angular resolution (long collimation and distant detector)
- Wavelength resolution is less important and can be relaxed to regain intensity



Continuum Description for SAS



- Q-rang limited to < 0.1 $Å^{-1}$
- Convolution theorem;
  signal is given by nm-size variation in scattering potential
- Material described as continuum with average material dependent scattering length density (SLD) parameter

$$\rho_n = \rho_{FU} \sum_{l=1}^{N_{FU}} b_l \cdot n_l$$

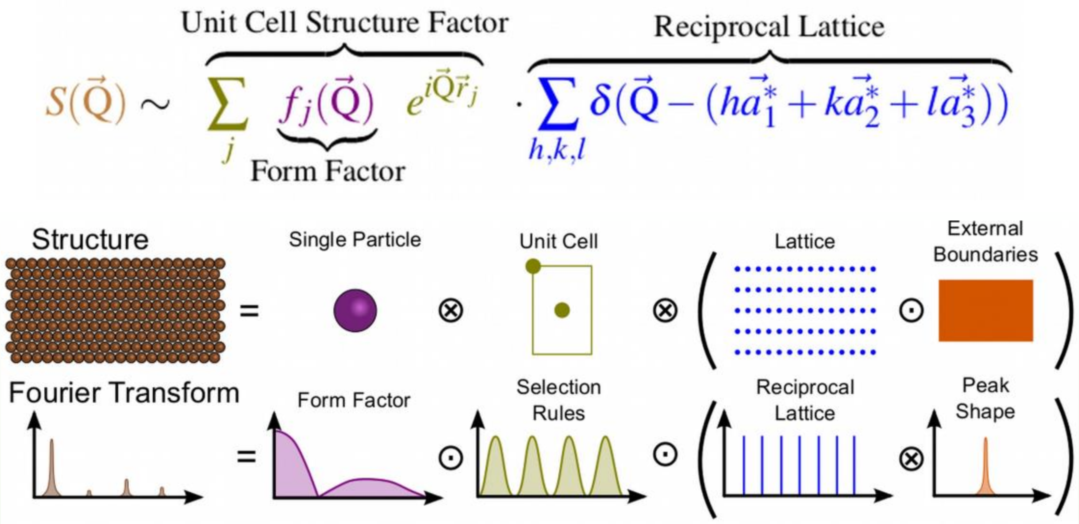
- X-ray atomic or neutron magnetic form factor decay at much larger Q-values
  ➔ continuum description only depends on one complex number to fully describe the scattering cross-section

$$\rho_m = b_H \cdot M$$

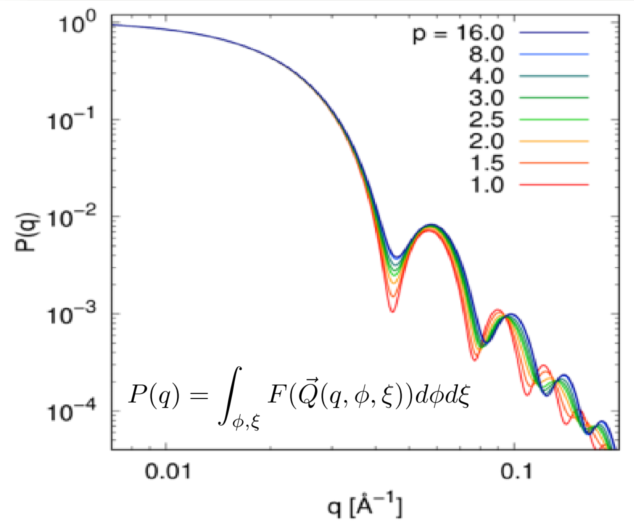
$$b_H = \frac{\gamma r_0}{2\mu_B} = 2.7 \text{ fm}$$

$$\rho_x = \rho_{FU} r_e \sum_{l=1}^{N_{FU}} f_i(E) \cdot n_l$$

Structure Factor in SAS



- Structure factor only applies for organized systems
- Independent particles behave as incoherent scatterers and can be described by their form factor alone



- The form factor is the Fourier transform of the particle shape scaled by the contrast between particle and surrounding medium

$$f_j(\vec{Q}) = \Delta\rho_j F_j(\vec{Q}) = \int \Delta\rho_j(\vec{r}) e^{i\vec{Q}\vec{r}} d^3r$$

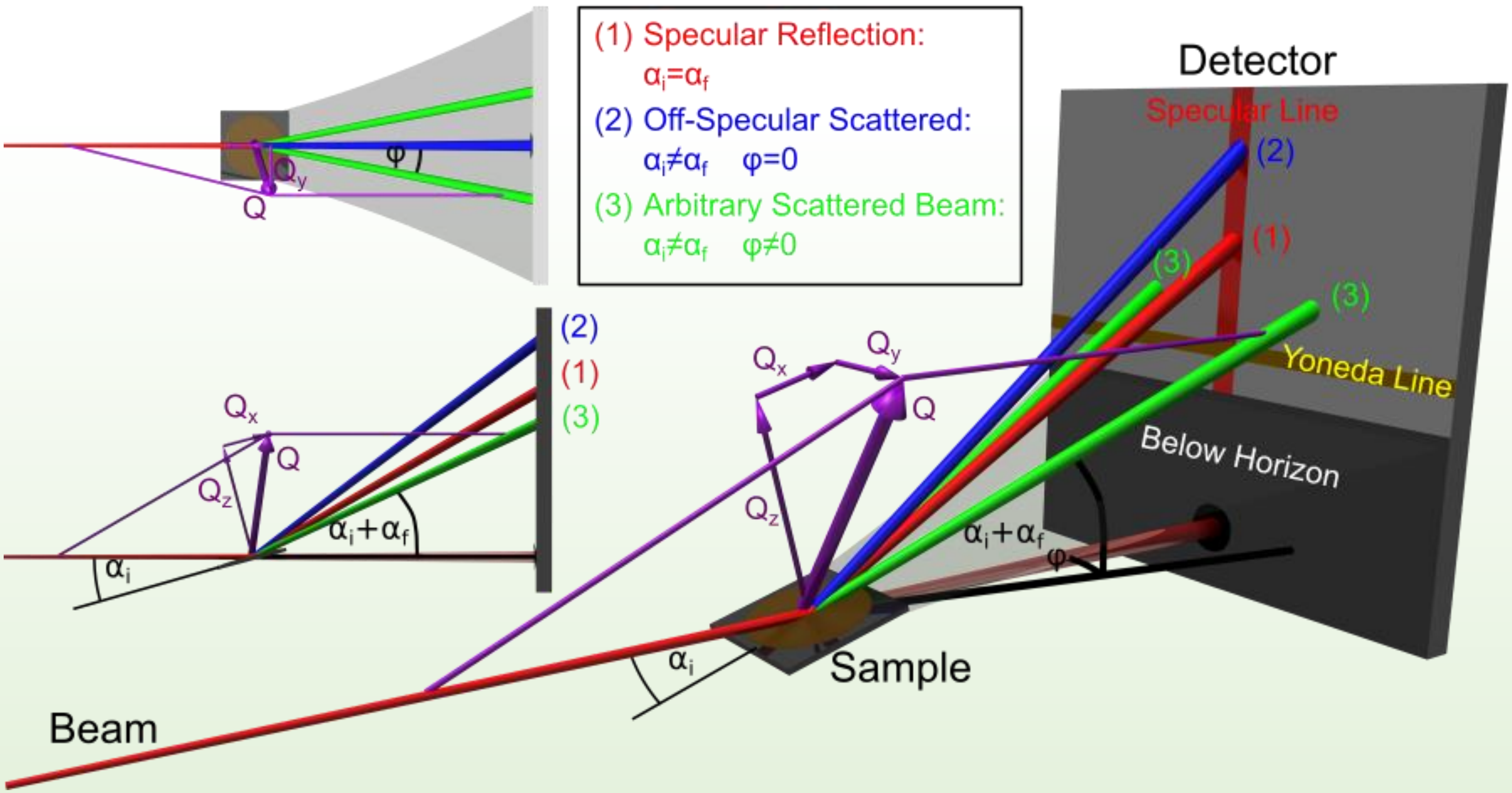
- nm-size particles are not identical like atoms, size/shape distribution has to be taken into account
- Analysis programs include form factors for typical shapes

$$F_{sphere}(\vec{Q}, R) = 4\pi R^3 \frac{\sin QR - QR\cos QR}{(QR)^3}$$

$$F_{cube}(\vec{Q}, a) = a^3 \mathrm{sinc}(q_x a)\mathrm{sinc}(q_y a)\mathrm{sinc}(q_z a)$$

Effects at grazing incidence

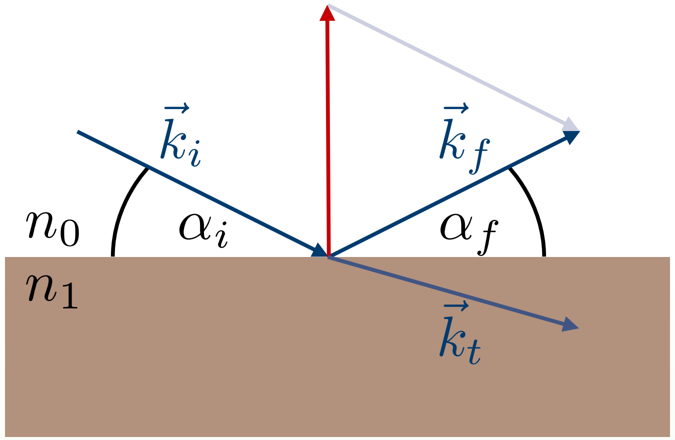
Geometry and naming conventions



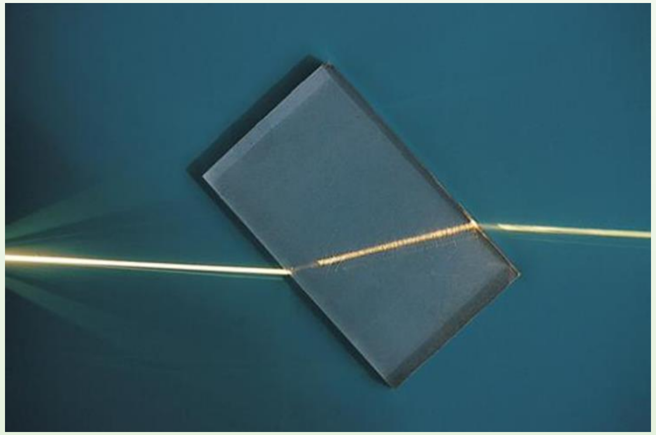
Effects at grazing incidence

At each interface:

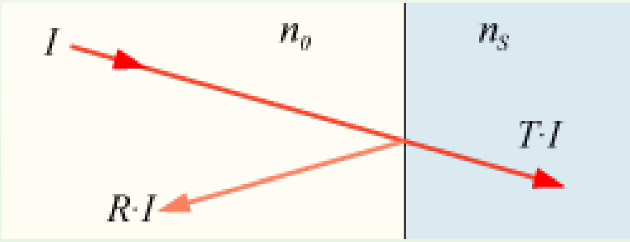
reflection



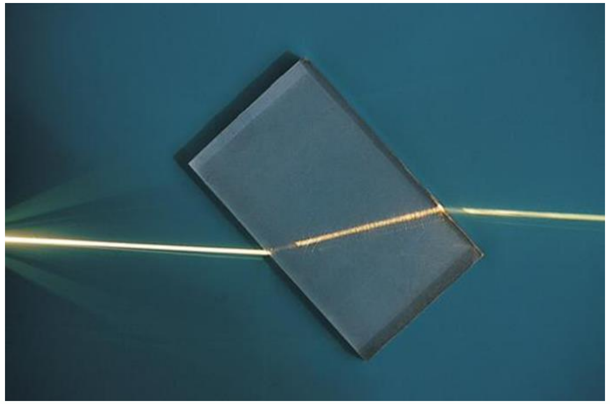
refraction



transmission



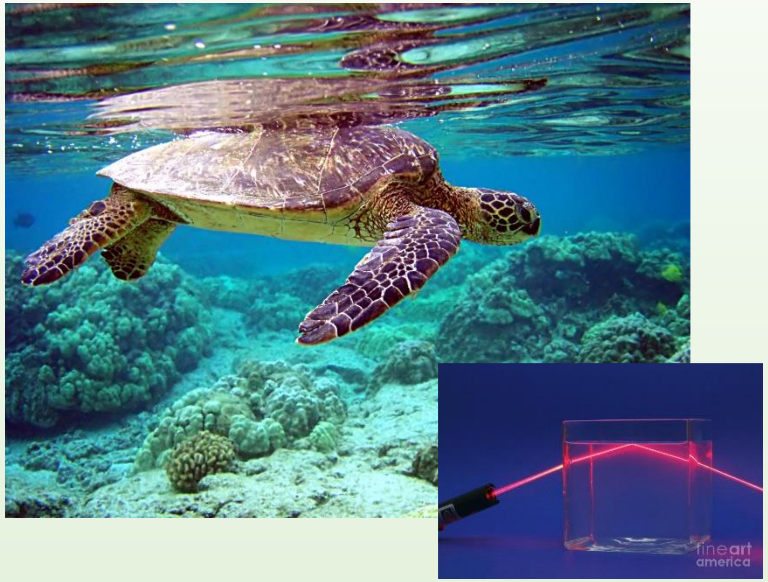
Refraction



By definition, the refractive index is: $$n_i^2 = \frac{k_i^2}{k_0^2}$$

Interaction can’t change the in-plane component of wave vector, leads to Snell’s law:

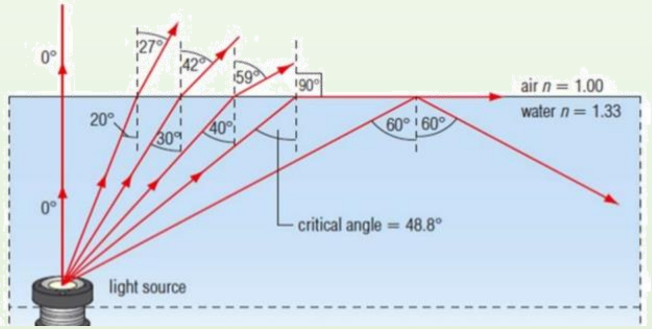
$$\left.\begin{aligned} k_{x,t} &= k_{x,i} \\ n_1 k_t &= k_0 \end{aligned}\right\} \cos\alpha_j = \frac{k_x}{n_j k_0} \Rightarrow \boxed{\frac{\cos\alpha_i}{\cos\alpha_t} = \frac{n_1}{n_0}}$$

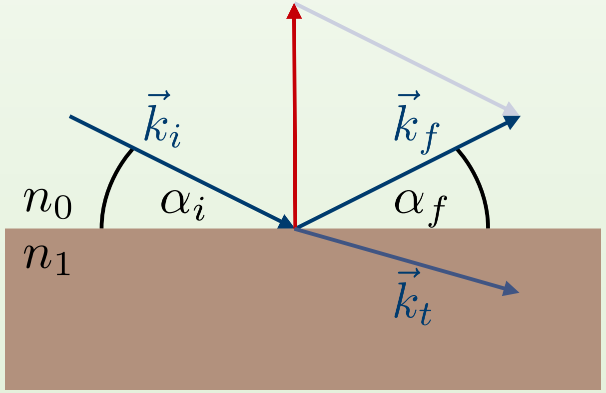


On side with larger n; critical angle of total reflection: $$\cos\alpha_c = \frac{n_1}{n_0}$$

Wave vector z-component in the medium:

$$k_{z,t} = k_t \sin\alpha_t = n_1 k_0 \sin\alpha_t$$





Reflection from a single interface

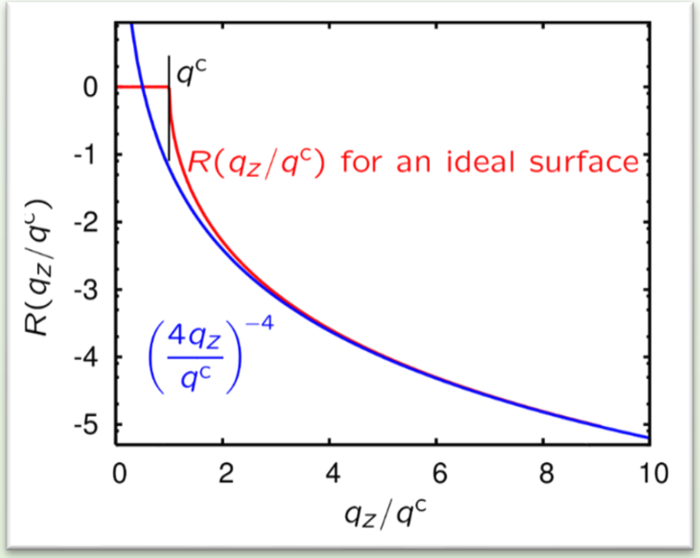
Reflectivity determined from reflectance: $R = |r_{0,1}|^2$

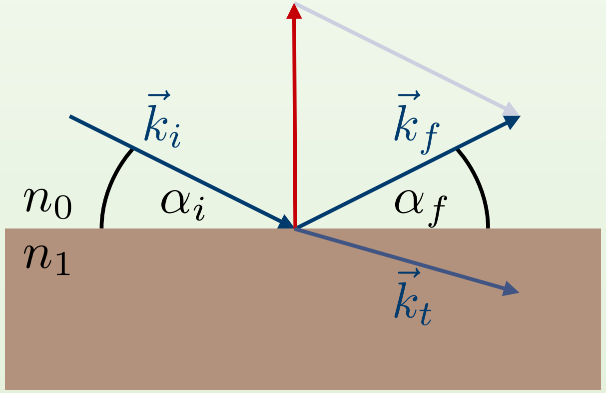
using the reciprocal space vector

$$q_z = \frac{2\pi}{\lambda}(\sin\alpha_i + \sin\alpha_f) = \frac{4\pi}{\lambda}\sin\alpha_i$$

one finds that the reflectivity is:

$$R(q_z) = \left|\frac{1-\sqrt{1-(q_c/q_z)^2}}{1+\sqrt{1-(q_c/q_z)^2}}\right|^2$$





Reflection from multiple interfaces

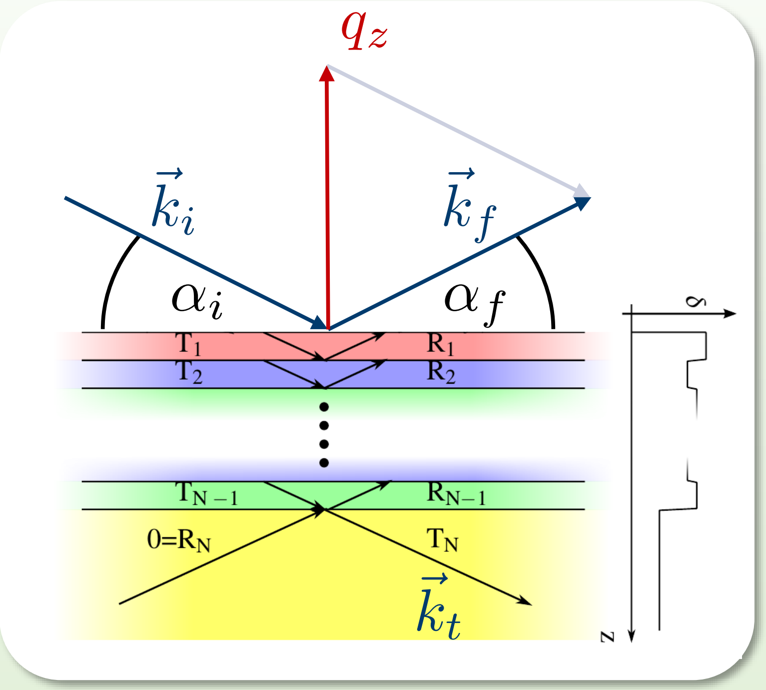
Interfering parts from all interfaces, but general relation for each layer:

$$X_j = \frac{R_j}{T_j} = e^{-2ik_{z,j}z_j} \frac{r_{j,j+1} + X_{j+1}e^{2ik_{z,j+1}z_j}}{1 + r_{j,j+1}X_{j+1}e^{2ik_{z,j+1}z_j}}$$

Together with two boundary conditions

$R_N = 0$ and $T_0 = 1$

can be solved analytically for any number of interfaces.



Referred to as Parratt's formalism, which yields a iterative solution for any number of layers that is "exact"

Refractive Index for Neutrons

Can be derived from Schrödinger's equation:

$$n_j = \sqrt{1 - \frac{V_j}{E}} \approx 1 - \frac{V_j}{2E} \quad := 1 - \delta + i\beta$$

Nuclear: $V_l^{Fermi} = b_l \frac{2\pi}{m_n} \delta(\vec{r})$

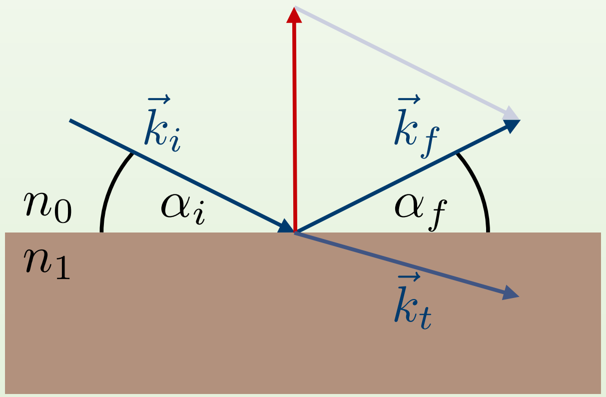
Magnetic: $V^m = \vec{\mu}\vec{B}_\perp$

Related to scattering length density (SLD):

$$n_j = 1 - \frac{\lambda^2}{2\pi}(\rho_n + \rho_m)$$

Typical values for δ are 10^{-5} for x-rays and 10^{-6} for neutrons

➔ *n* is very close to 1

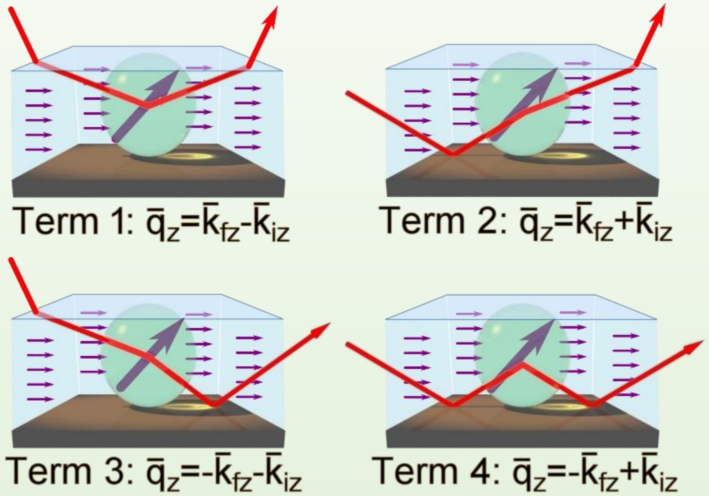


Scattering within the plane

The Distorted Wave Born Approximation (DWBA)

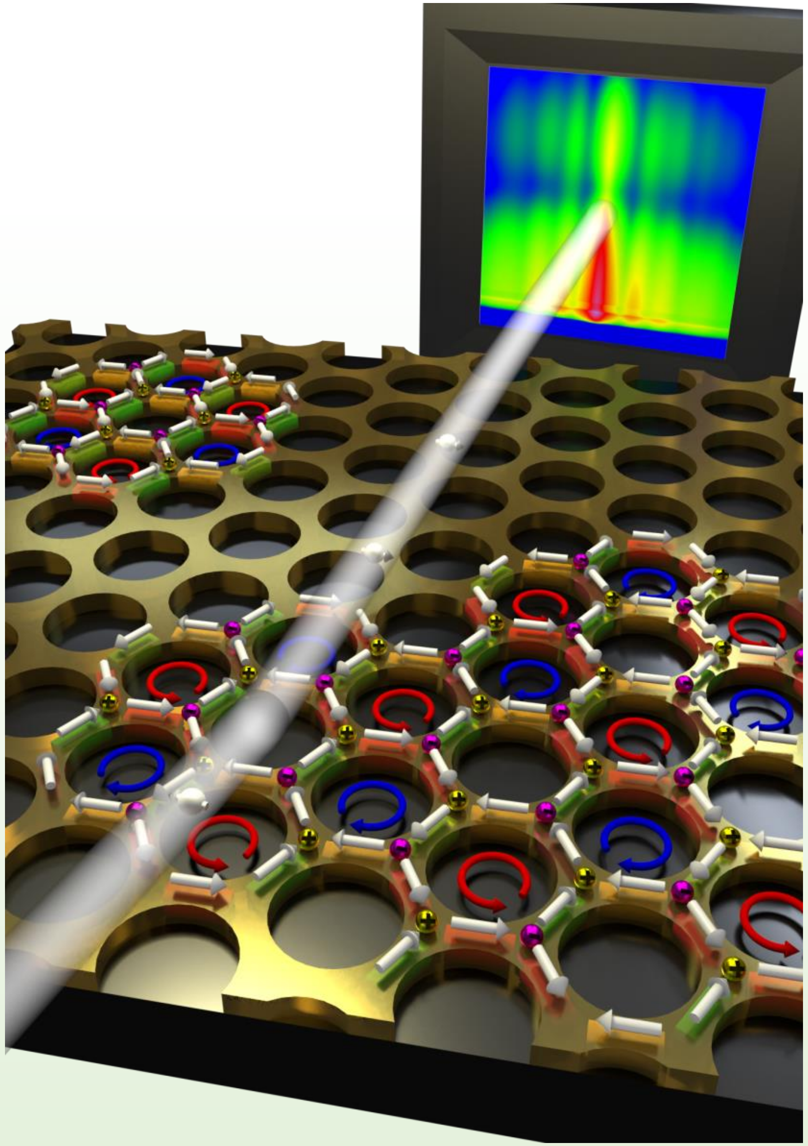
- Use optical formalism (Parratt) to describe strong dynamic effect
- In each layer perform Born approximation for the in-plane scattering of the difference potential that fulfills small scattering condition
- Need to account for all possible initial and final wave directions that may be present due to reflection below

$$
\begin{aligned}
\Rightarrow F_{DWBA}\left(Q_{||}, k_{i,z}, k_{f,z}\right) = \quad & F\left(Q_{||}, k_{f,z} - k_{i,z}\right) \\
& + r_i F\left(Q_{||}, k_{f,z} + k_{i,z}\right) \\
& + r_f F\left(Q_{||}, -k_{f,z} - k_{i,z}\right) \\
& + r_i r_f F\left(Q_{||}, -k_{f,z} + k_{i,z}\right)
\end{aligned}
$$



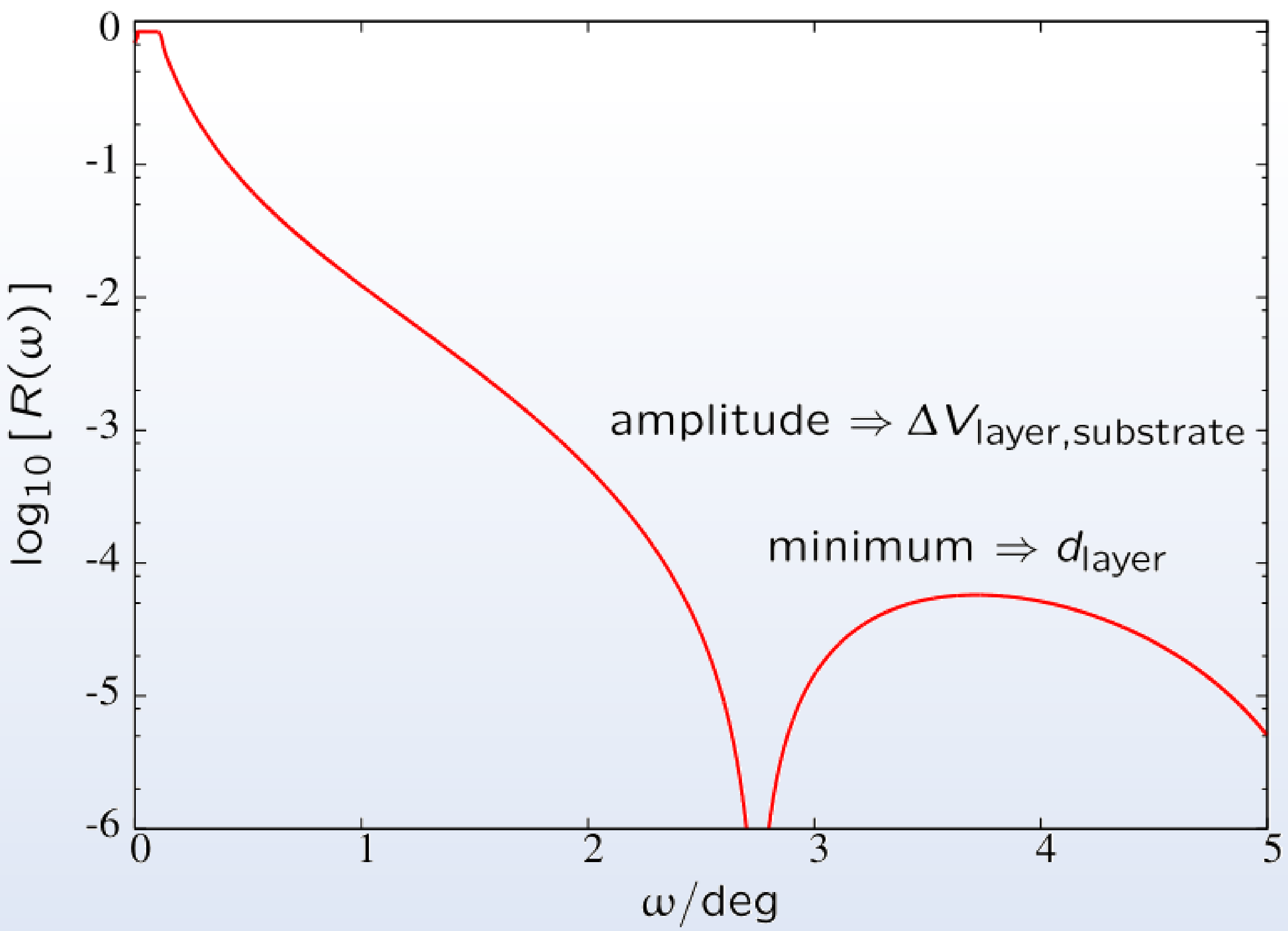
Relevant for off-specular and GISAS:

- Roughness between layers
- Magnetic domains
- Structured samples
- Embedded particles



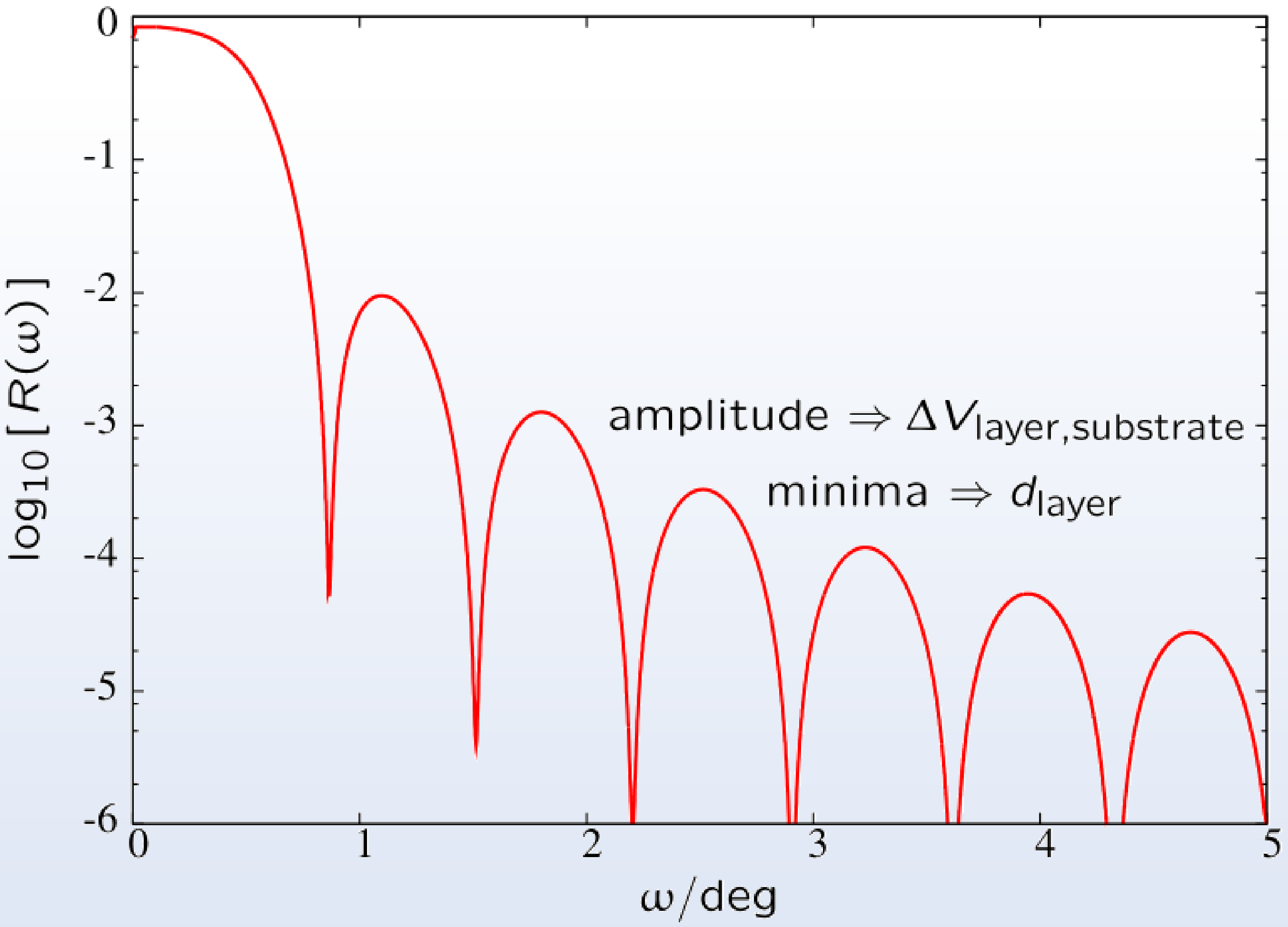
NR from homogeneous layers

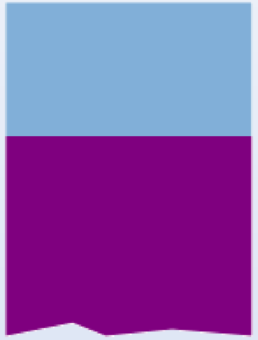
Simple example of single layer reflectivity:



NR from homogeneous layers

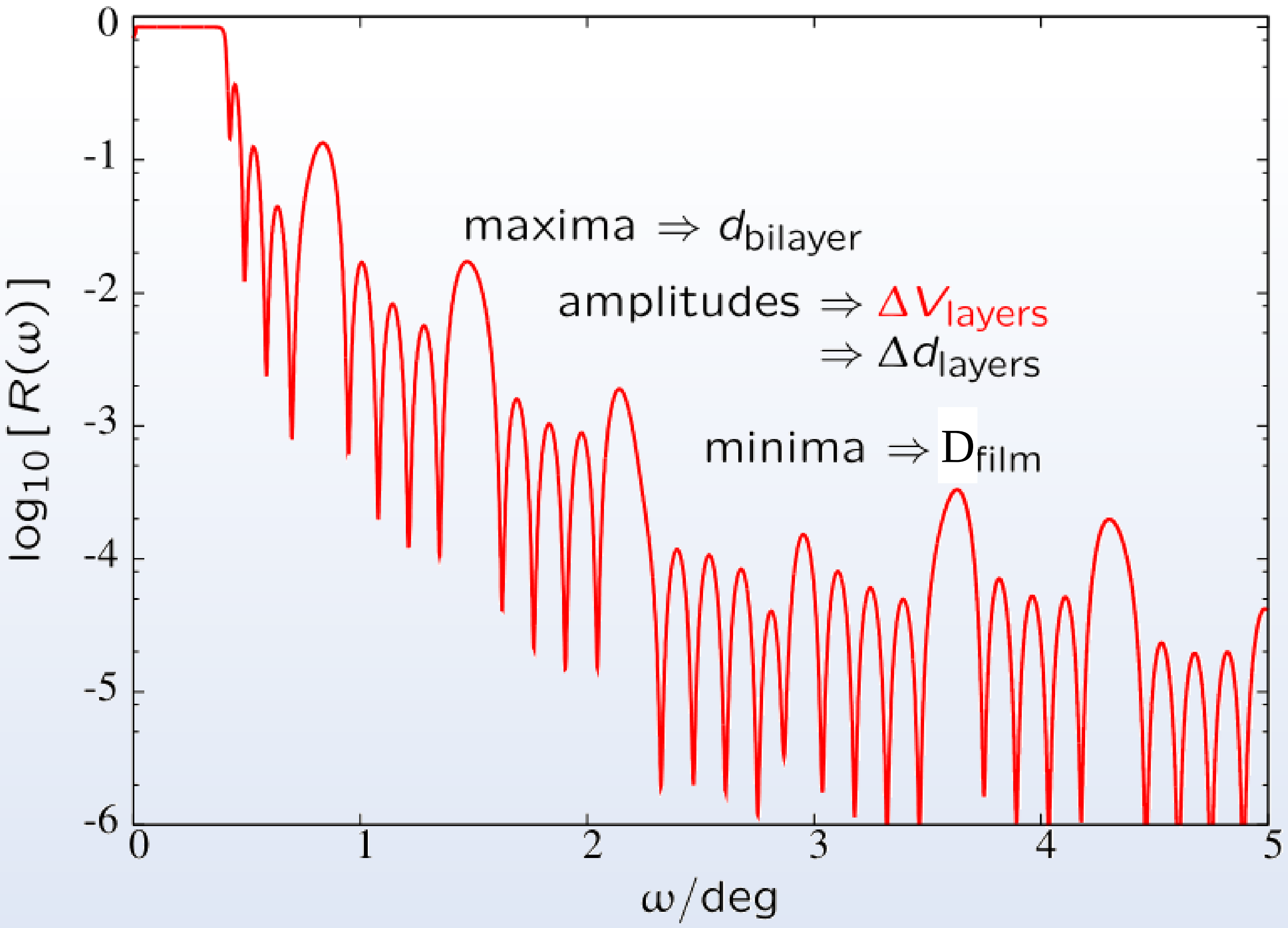
Simple example of single layer reflectivity:

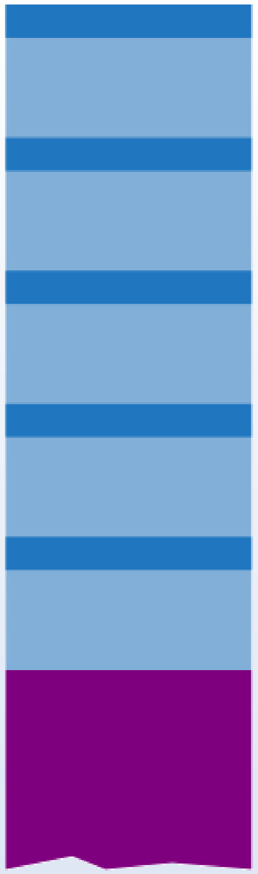




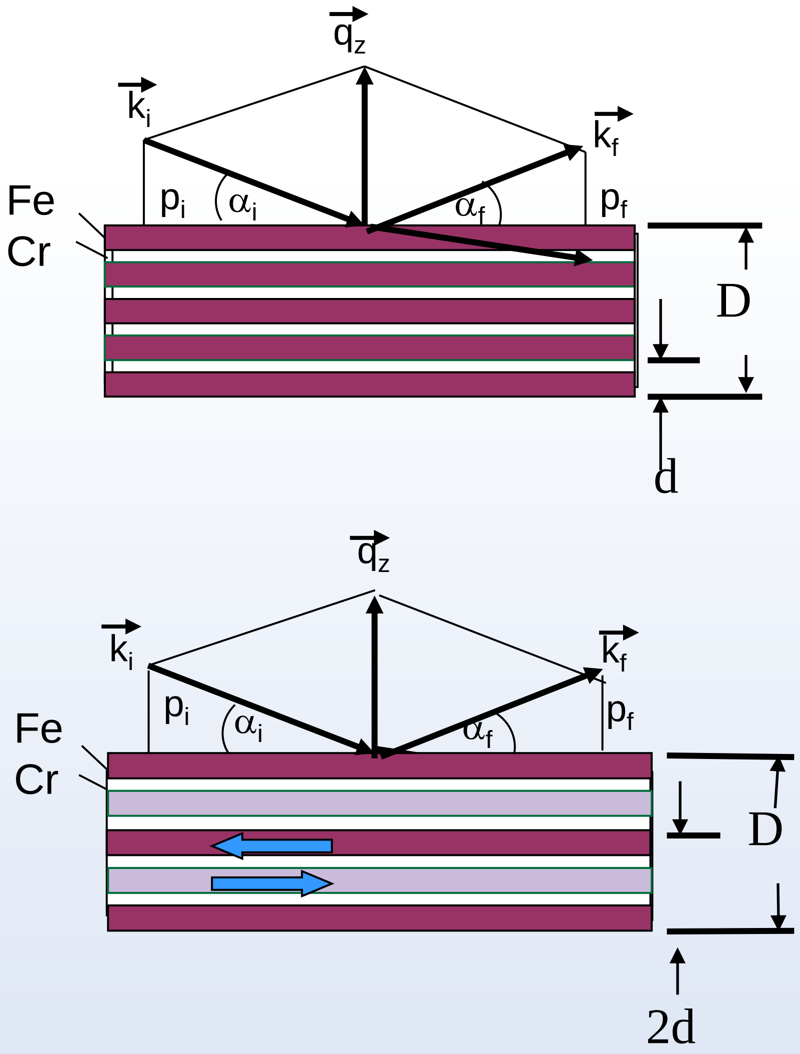
NR from homogeneous layers

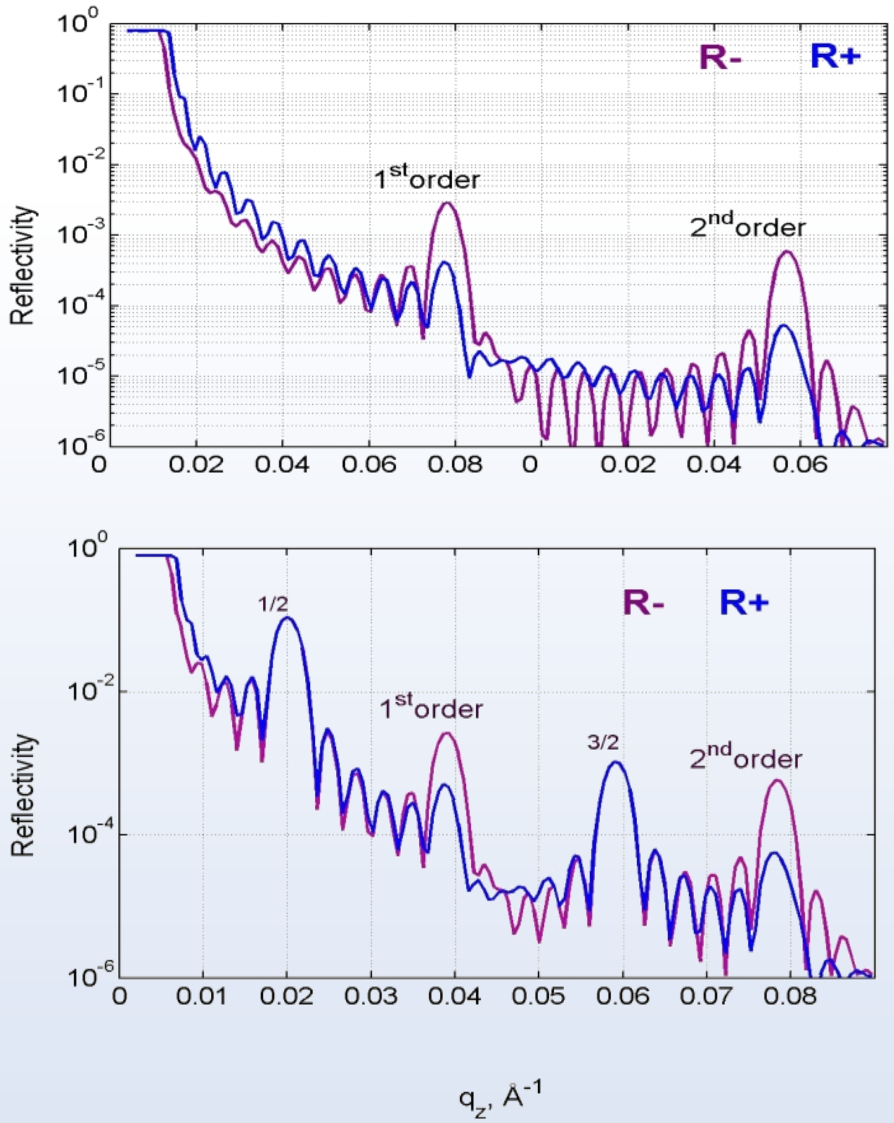
Simple example of multilayer reflectivity:



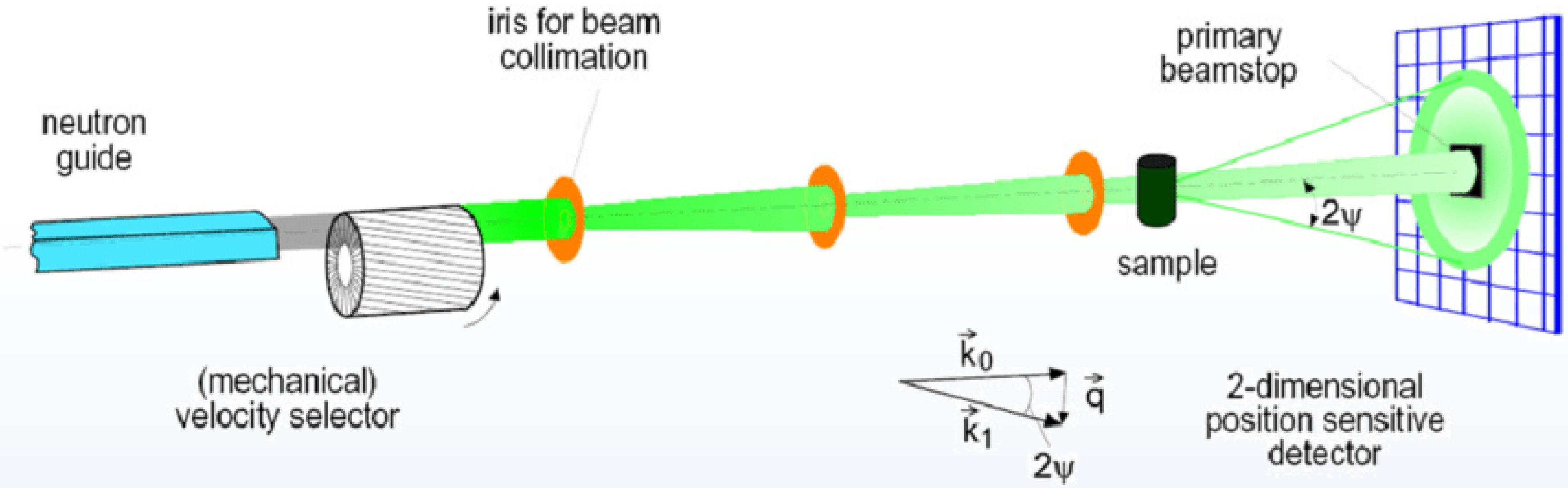


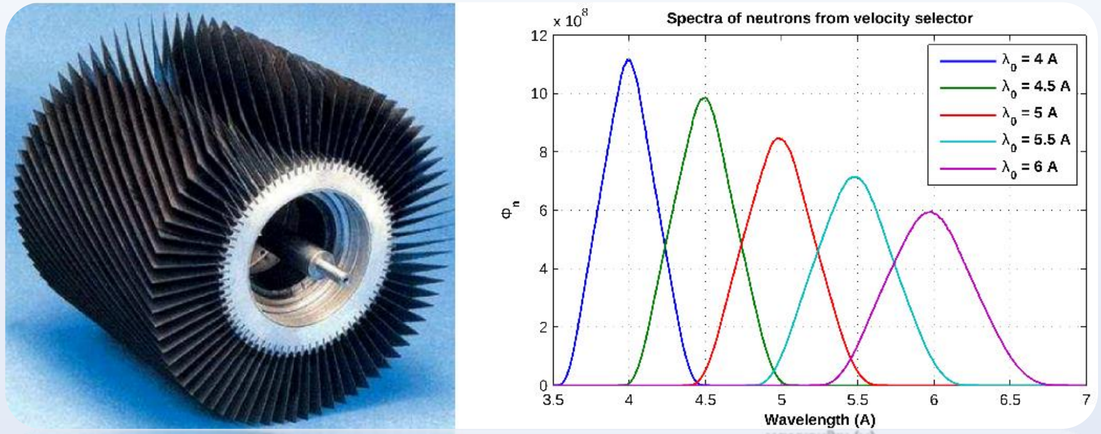
Polarized NR from magnetic layers



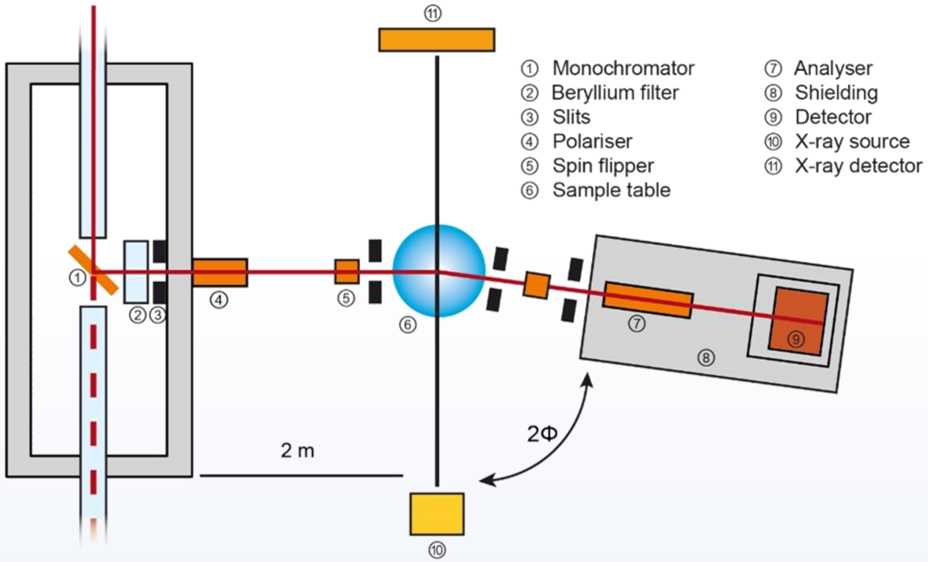


SANS instruments

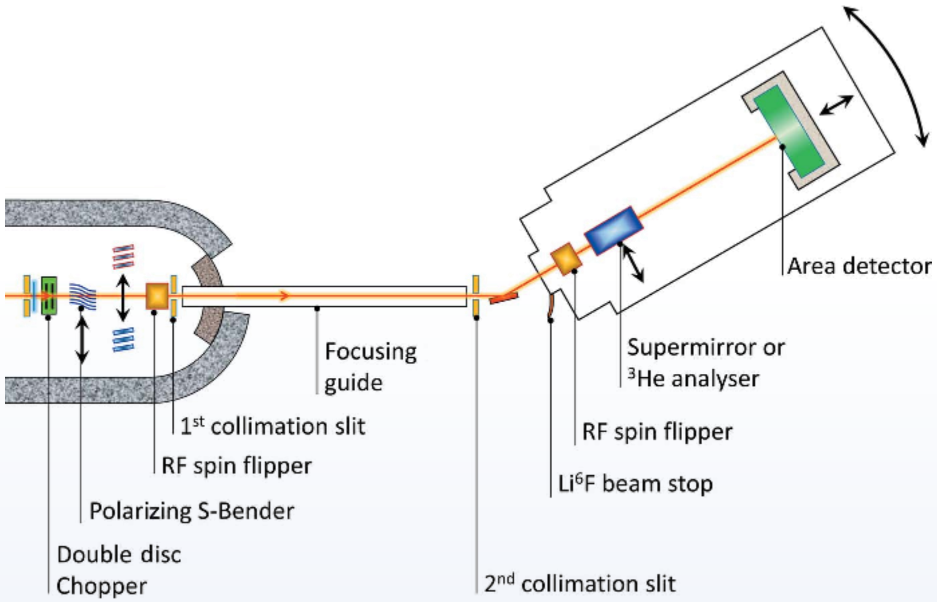




Neutron Reflectometers

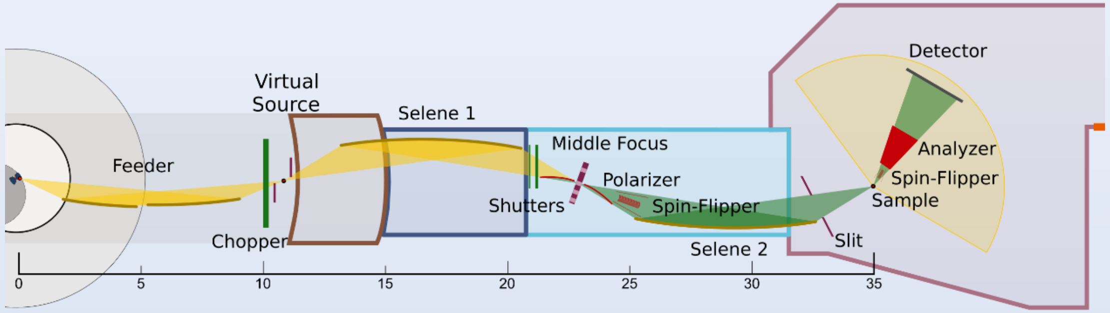


Traditional Monochromatic (NREX@MLZ)

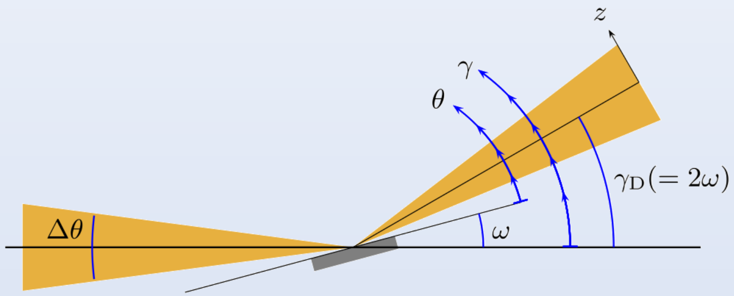


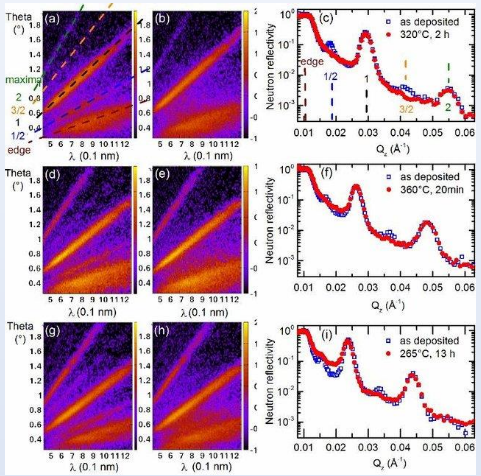
Traditional ToF (D17@ILL)

Focusing (Estia@ESS)

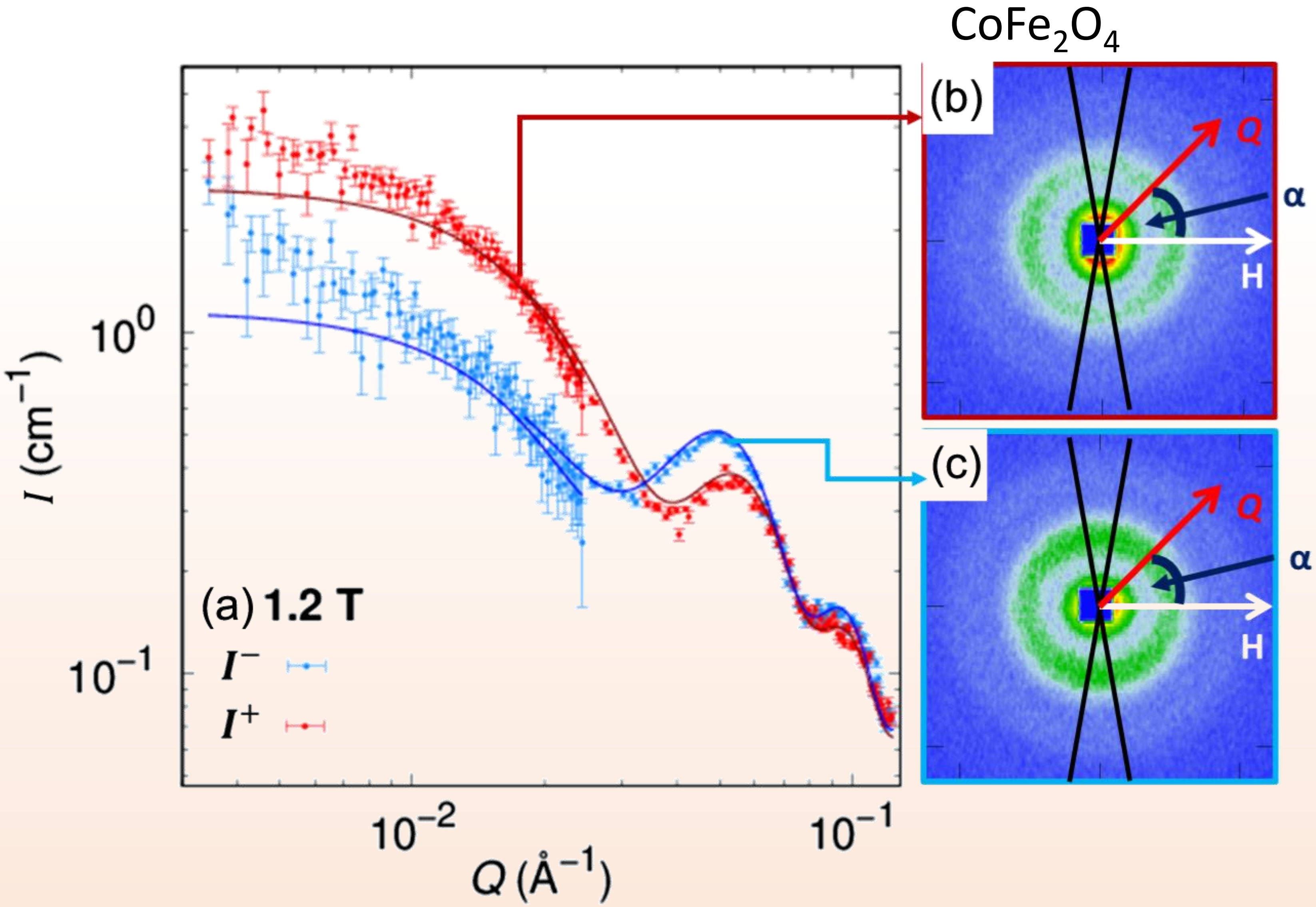


- Pure specular reflectivity
- Beam focused on small sample
- Detector determines reflection angle





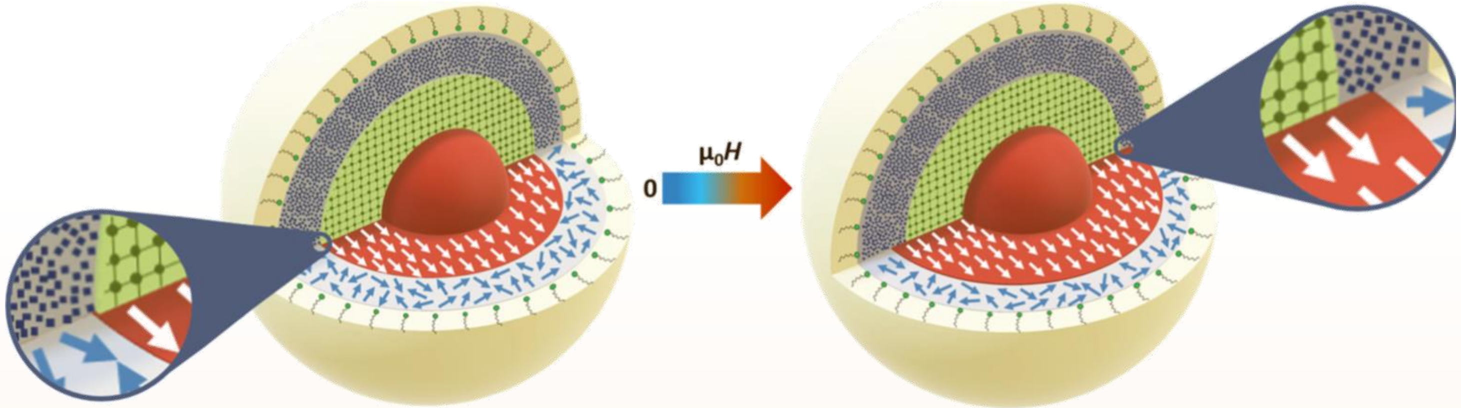
SANS on Magnetic Nanoparticles

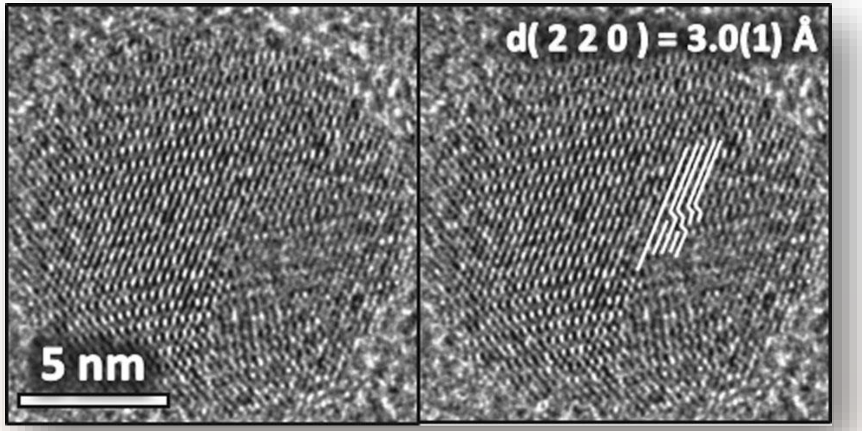


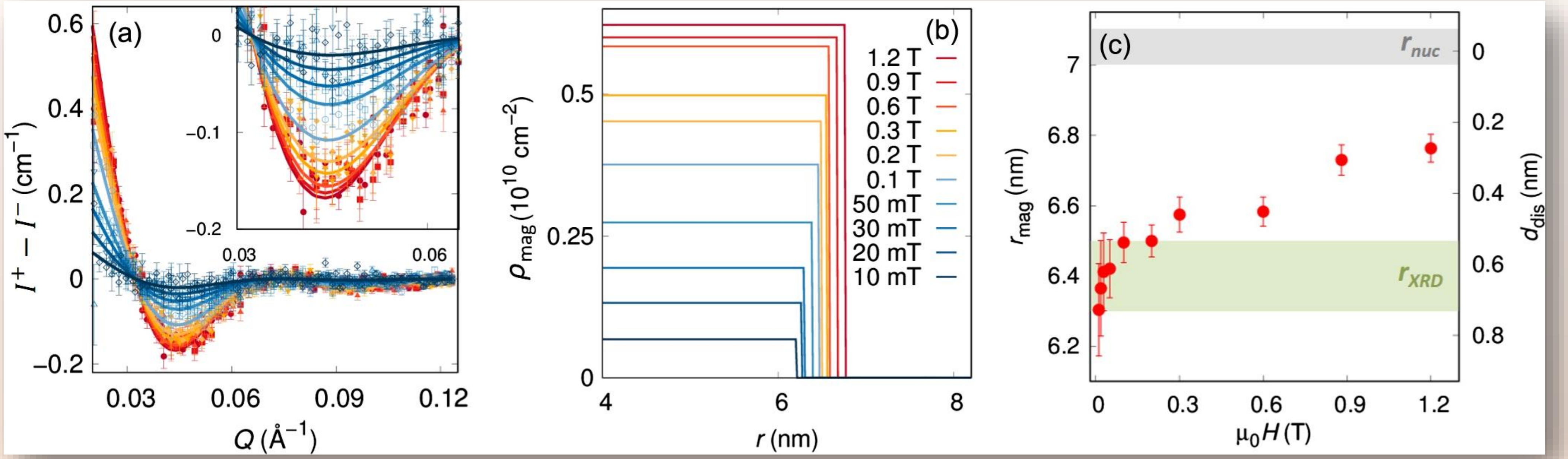
Form factor is a sphere of magnetic core and non-magnetic shell

PHYSICAL REVIEW X 10, 031019 (2020)

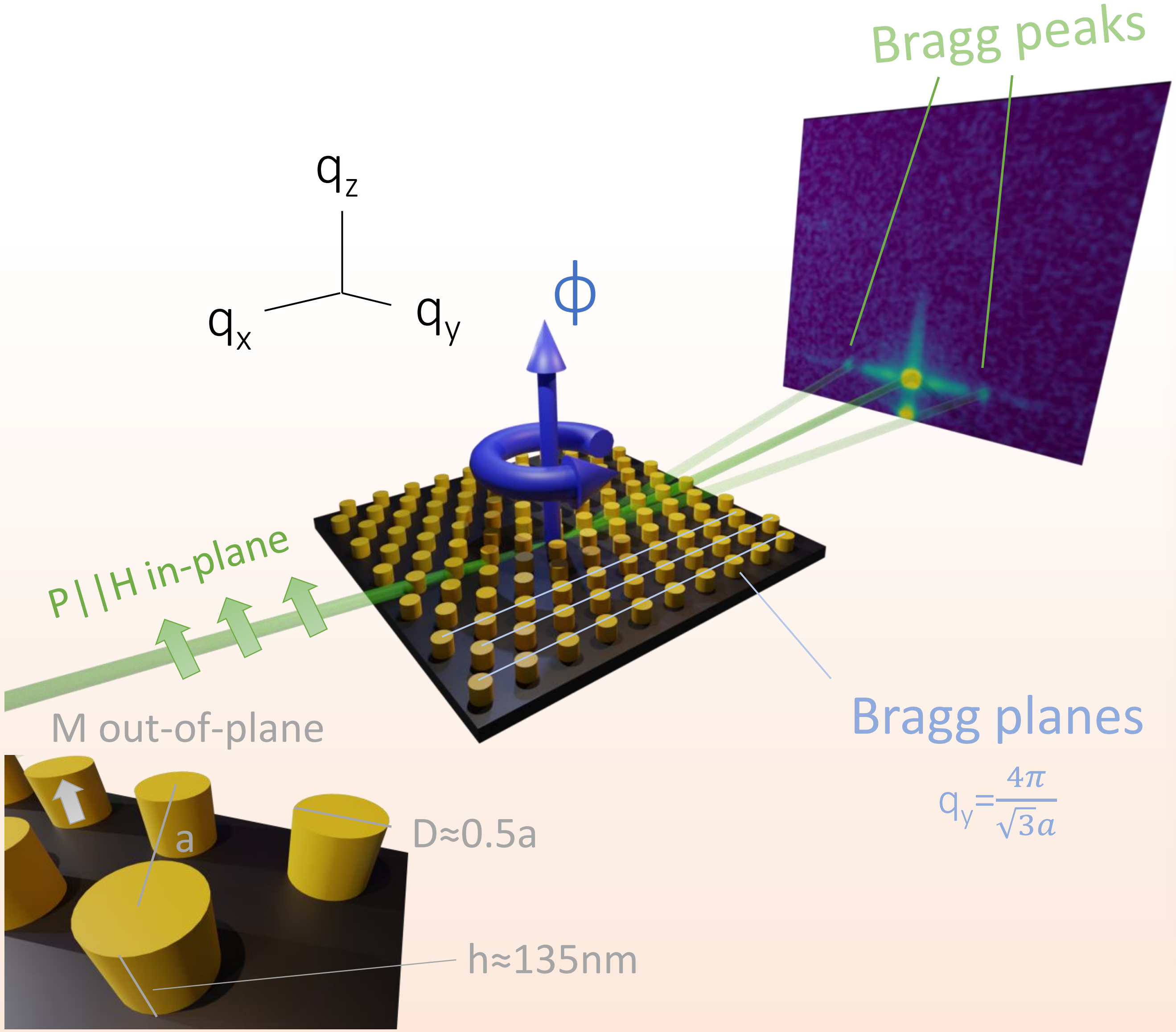
Polarized SANS on Magnetic Nanoparticles

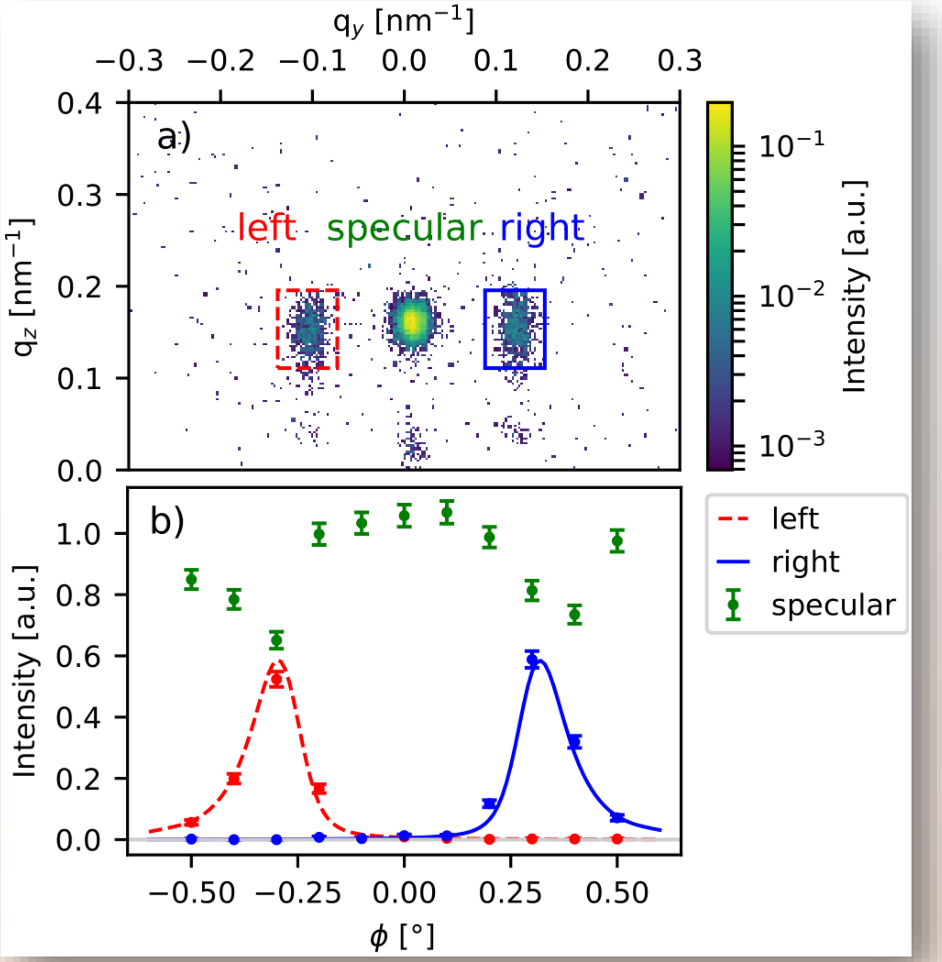






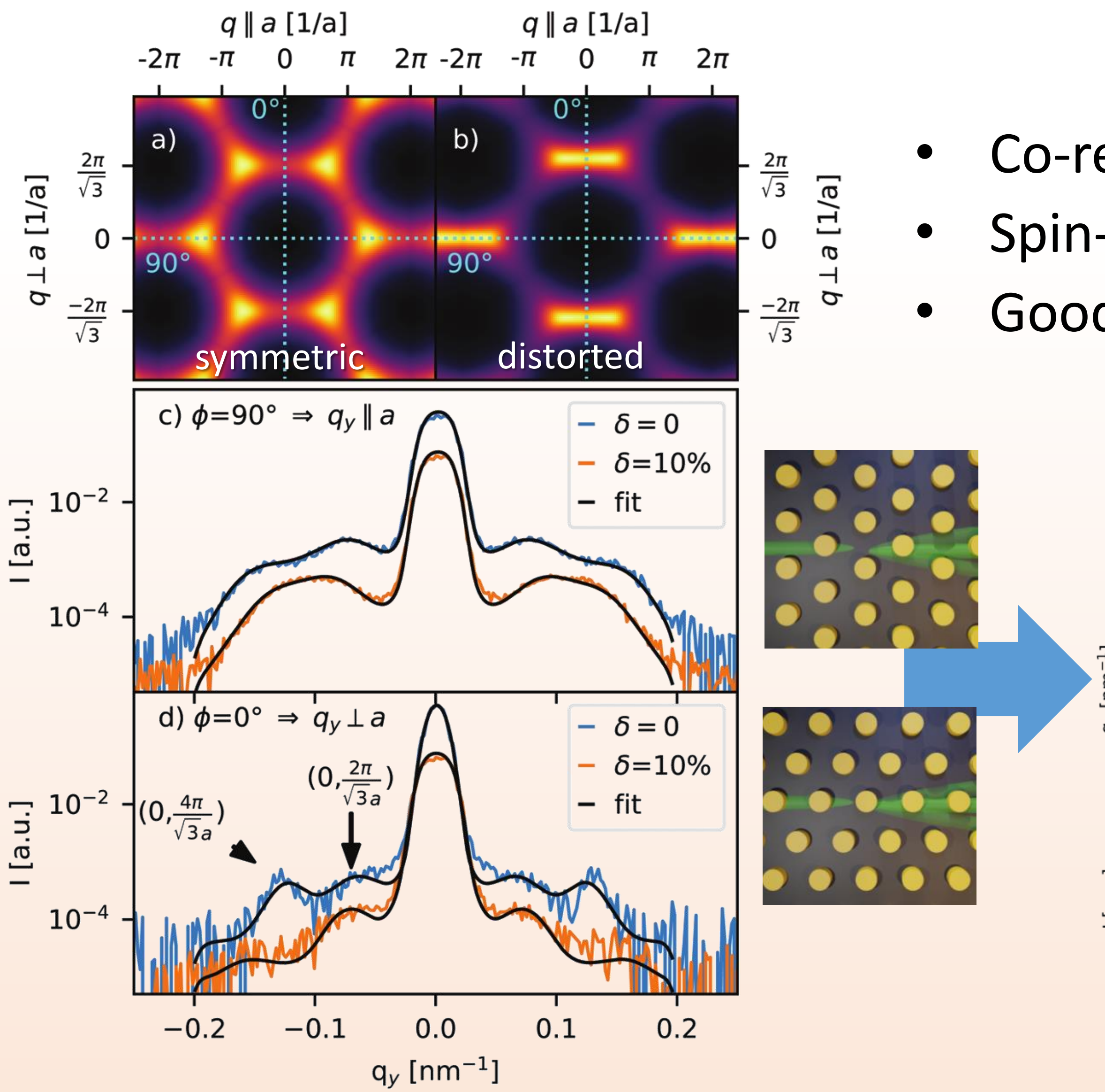
Frustrated artificial spins on triangular lattice



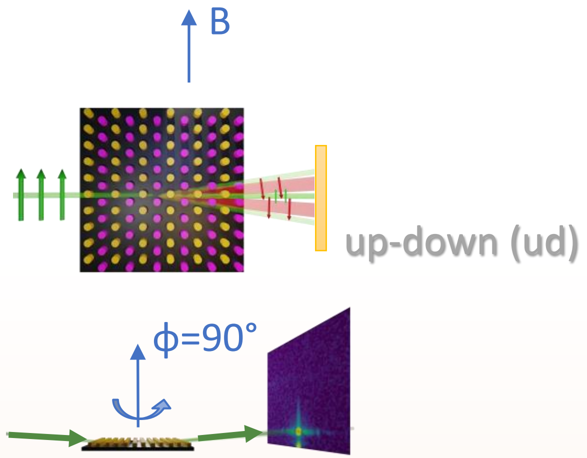


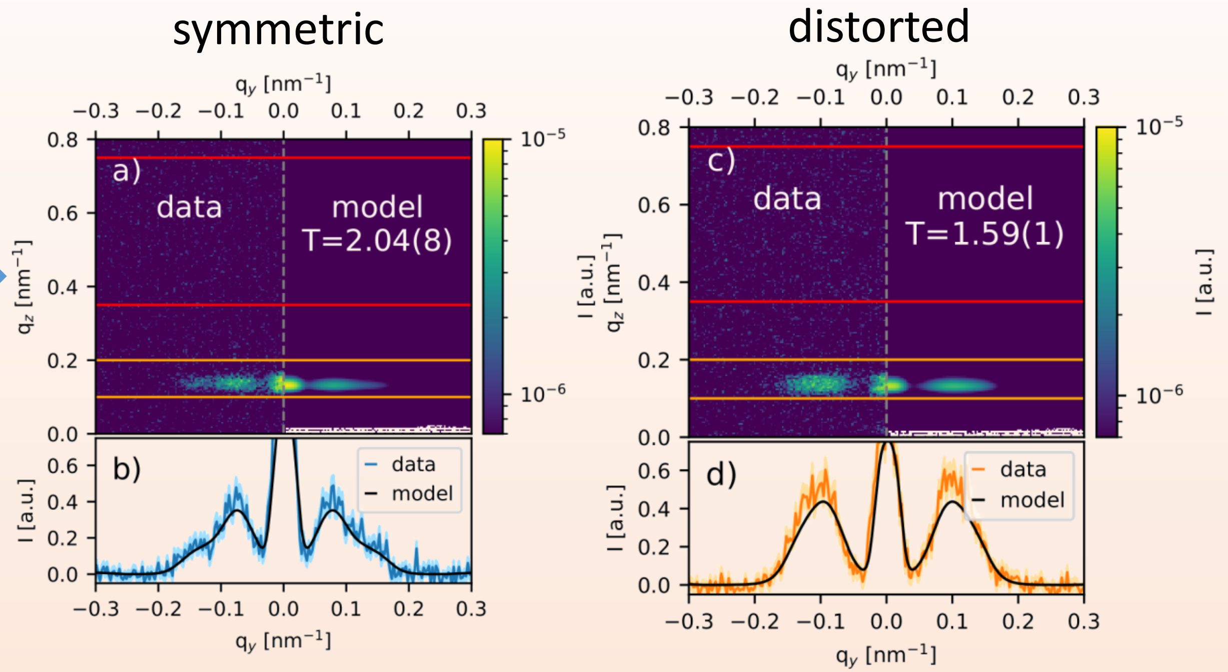
P. Pip, A. Glavic, et al. , *Nanoscale Horizons* 6, 474-481 **(2021)**

Frustrated artificial spins on triangular lattice



- Co-refined ϕ=0° & 90°
- Spin-flip later simulated
- Good agreement with data

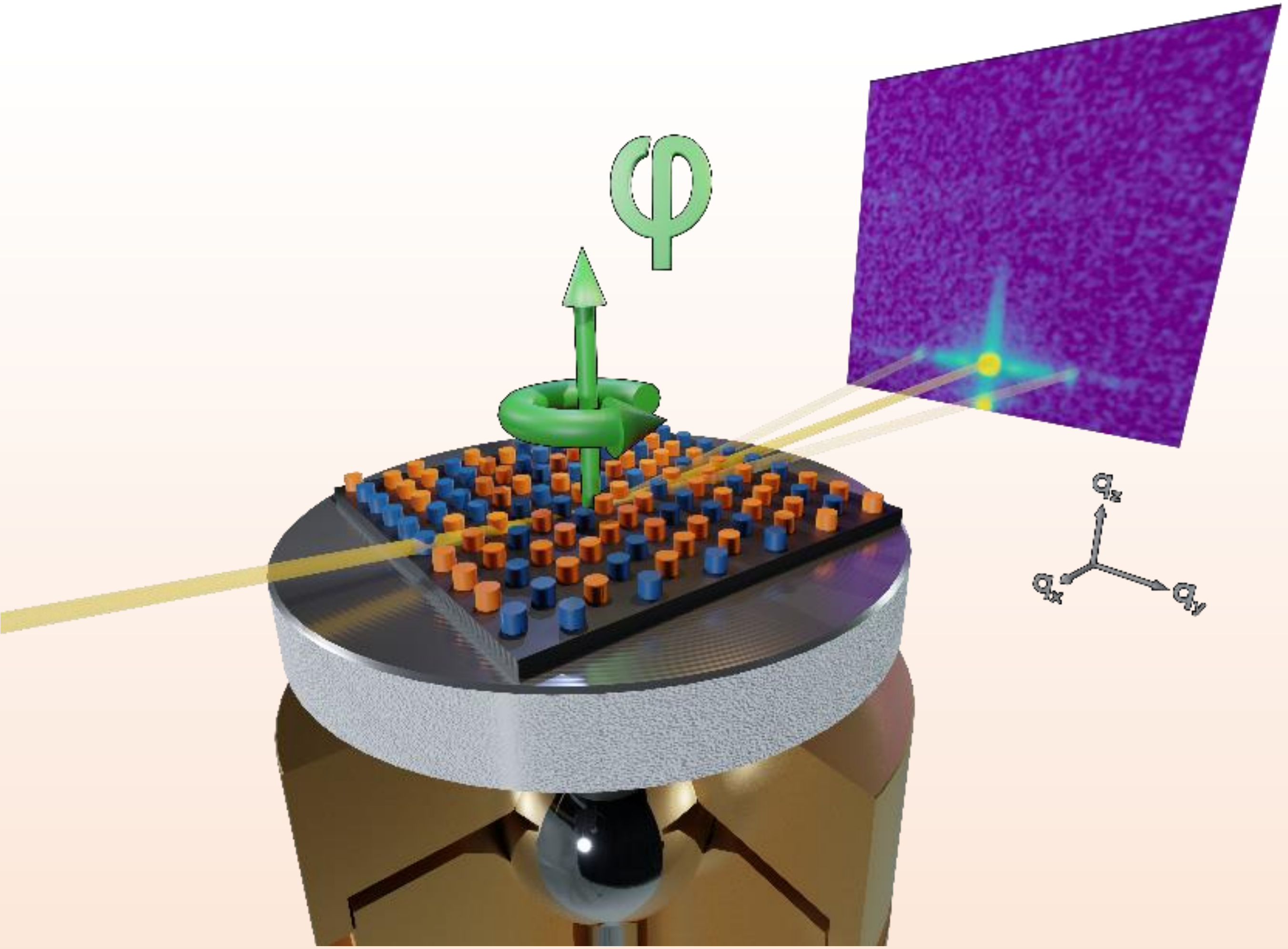




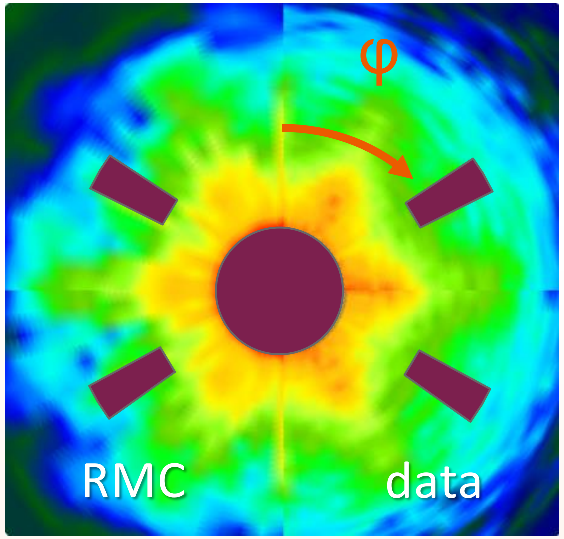
P. Pip, A. Glavic, et al. , *Nanoscale Horizons* 6, 474-481 **(2021)**

Frustrated artificial spins on triangular lattice

Follow up experiment at PSI



symmetric



distorted