INSTITUT LAUE LANGEVIN

Nanoscience with neutrons – small angle scattering and neutron reflectivity

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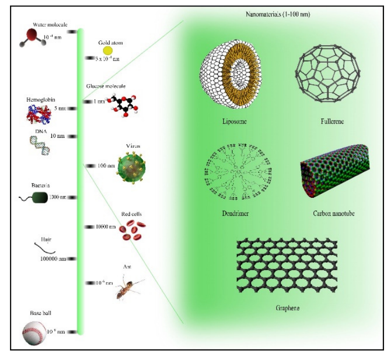
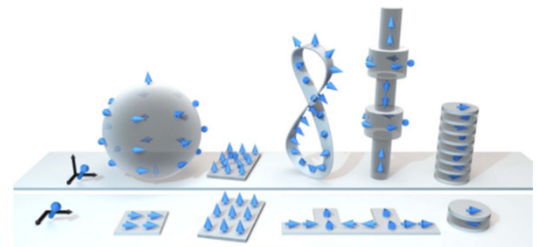
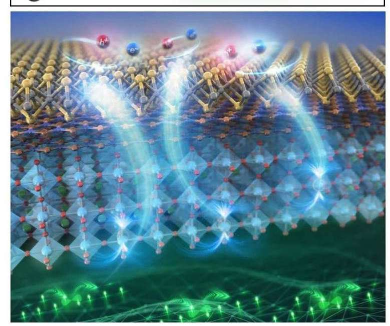
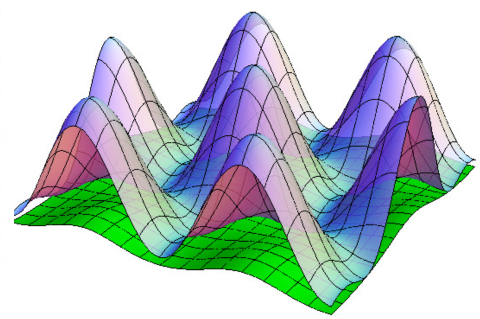
What is nanoscience anyway?



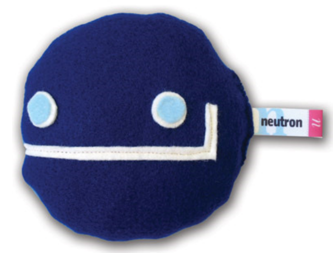






And neutrons, what are they good for?

→ They keep your atomic nuclei stable.



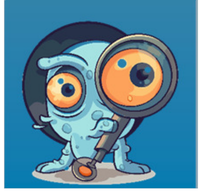
→ They can see magnetism:

- → Same magnitude as structural scattering
- → Simple magnetic interaction: dipolar,sensitive to B → easy to model

→ They scatter from the atomic nucleus, not the electron cloud, clean structure, weak scattering → easy to model

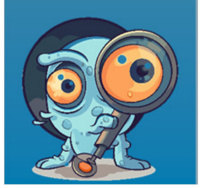
→ They see Hydrogen, Li and other light elements really well

→ They probe the whole sample (and the substrate and the sample holder)

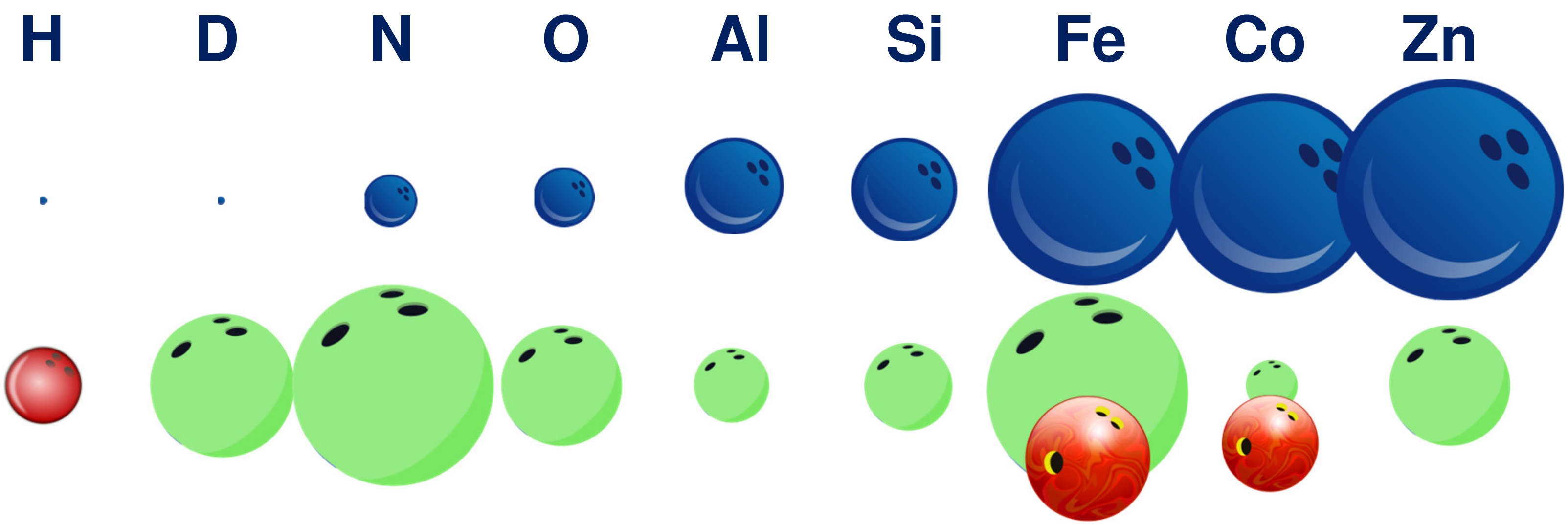


And neutrons, what are they good for?

- → They keep your atomic nuclei stable.
- → **They can see magnetism.**
- → They scatter from the atomic nucleus, not the electron cloud.
- → **They can see Hydrogen, Deuterium and Li really, really well.**
- → **They probe the whole sample, including structures deep inside the sample (and the substrate and the sample holder).**

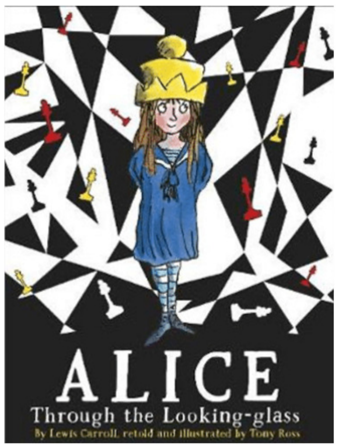


Why neutrons? Neutron scattering strength varies more or less randomly from element to element, and isotope to isotope.

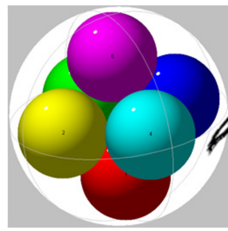


How can I look at nanoscale objects with neutrons?

To measure average distance you need elastic, coherent scattering, generally in reciprocal space. This is true also for SANS and NR

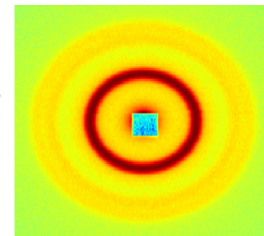


Real space



Do some scattering

Reciprocal space



$$\mathbf{d} = \frac{2\pi}{\Delta q}$$

$$2\mathbf{d}\sin\theta = n\lambda, \mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$$

$$\mathbf{k} = \frac{2\pi}{\lambda} \qquad q = \frac{4\pi \sin\theta}{\lambda}$$

Reciprocal space [edit]

Reciprocal space (also called k-space) provides a way to visualize the results of the Fourier transform of a spatial function. It is similar in role to the frequency domain arising from the Fourier transform of a time dependent function; reciprocal space is a space over which the Fourier transform of a spatial function is represented at spatial frequencies or wavevectors of plane waves of the Fourier transform. The domain of the spatial function itself is often referred to as real space. In physical applications, such as crystallography, both real

Wikipedia

elastic = no change in energy

coherent = characteristic lenghtscale

nanometres!

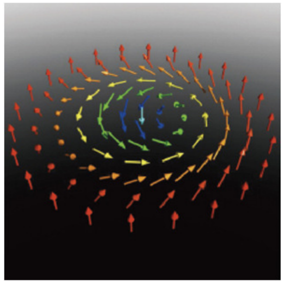
So for a nanoscale scattering experiment:

$$2d \sin\theta = n\lambda$$

d, distance probed, nm
largish compared to λ

Wavelengths of Probe, of the order of 0.1 - 2 nm. Of the order of atomic spacing. **FIXED**

λ fixed, d large:
θ must be small!



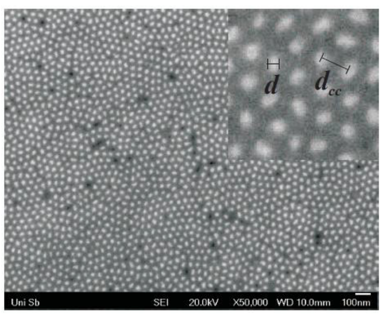
Do I need a special setup to measure small angles?

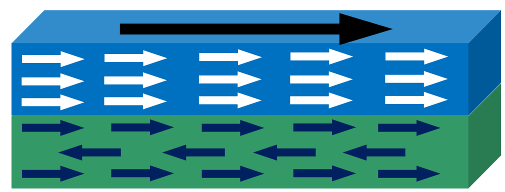
Nanoscale scattering experiments measure small angles; which scattering geometry works best?

Nano-**structures** → elastic scattering → $\mathbf{2d \sin \theta} = n\lambda$, $\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$
Lengths scales are large → scattering angles are small

Two possible scattering geometries to capture small angles:

→ **Transmission:** particle like structures, like nanoparticles, polymers, superconducting flux line lattices, precipitates
→ **Reflection:** planar structures, like thin films, interfaces, multilayers, gratings, neutron guides, supermirrors





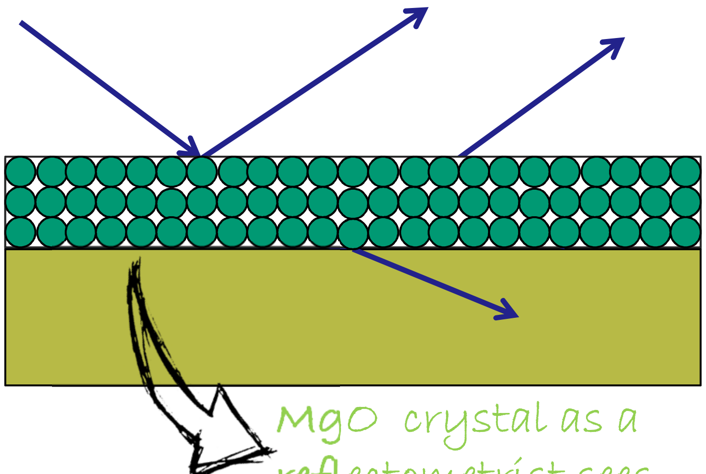
How does a small angle experiment differ from a normal diffraction experiment?

Small angles → small q → length scale probed is large! We are no longer sensitive to individual atoms, but to the average scattering power per unit volume – the **scattering length density (SLD) ρ.**

MgO crystal as a crystallographer sees it

nucleus-neutron interaction (Born approximation):

$$V_n = \frac{2\pi\hbar^2}{m_n} b\delta(\mathbf{r})$$ Point like



MgO crystal as a reflectometrist sees it

$$V_n(z) = \frac{2\pi\hbar^2}{m_n}\rho$$ (assume V independent of x and y)

$$\rho = \sum_i N_i b_i$$

N_i – number density for species i

b_i – bound coherent scattering length for species i

How do I calculate a scattering length density?

How to calculate the SLD for crystalline materials.

$$\rho = \sum_i N_i\, b_i$$

N_i – number density for species i
b_i – bound coherent scattering length for species i

formula units per unit cell:
1 – simple
2 – bcc
4 – fcc
6 - hcp

Example MgO:

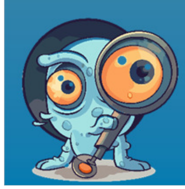
b_{Mg} = 5.38fm b_O = 5.80fm → b_{MgO} = 11.2 fm
f.u./unit cell = 4
$V_{unit\ cell} = a^3 = 4.22Å^3 = 74.99Å^3$

$$N_{MgO} = \frac{4}{V_{unit\ cell}} = 0.053Å^{-3}$$

$$\boldsymbol{\rho_{MgO} = N_{MgO} b_{MgO} = 5.96\ 10^{-6}Å^{-2}}$$

https://www.ncnr.nist.gov/resources/activation/
http://icsd.cds.rsc.org/

ICSD



How do I calculate an SLD?

How to calculate the SLD using the density.

$$\rho = \sum_i N_i \, b_i$$

N_i – number density for species i

b_i – bound coherent scattering length for species i

Example H_2O:

1fm = 10^{-5} Å

b_H = -3.74 fm b_O = 5.80 fm → b_{H2O} = -1.68 fm

density = 1 g/cm^3, $m_{atomic,\,H2O}$ = 18 g/mol

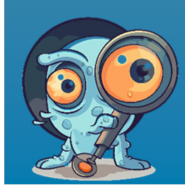
N_A = 6.022x10^{23} particles/mol

1cm = 10^8 Å, 1cm^3 = 10^{24} Å

$$N_{H2O} = \frac{density\ x\ NA}{m_{atomic}} = \frac{1\frac{g}{cm^3}\,x\,6.022x1023\,\frac{particles}{mol}}{18\,\frac{g}{mol}}$$

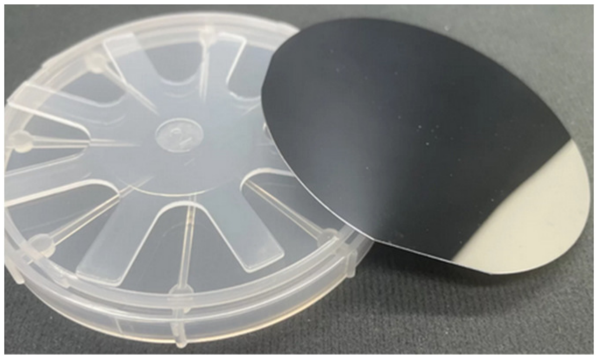
$= 0.033$ Å^{-3}

$\rho_{H2O} = N_{H2O} b_{H2O} = -0.56\ 10^{-6}$Å^{-2}



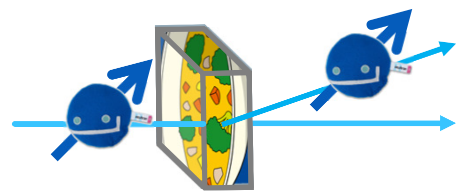
So, the SLD determines the scattering?

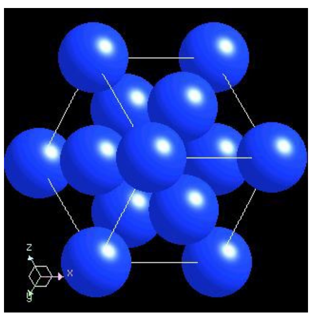
Having an SLD is not enough – you need contrast to see scattering. Contrast: changes in the SLD on the lenghtscale to which we are sensitive = differences in SLD.



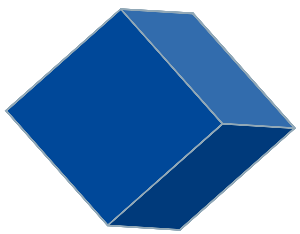
$$\rho = \sum_i N_i \, b_i$$

N_i – number density for species i
b_i – bound coherent scattering length for species i

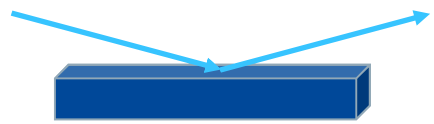




Si crystal – atomic resolution



Si crystal – SLD

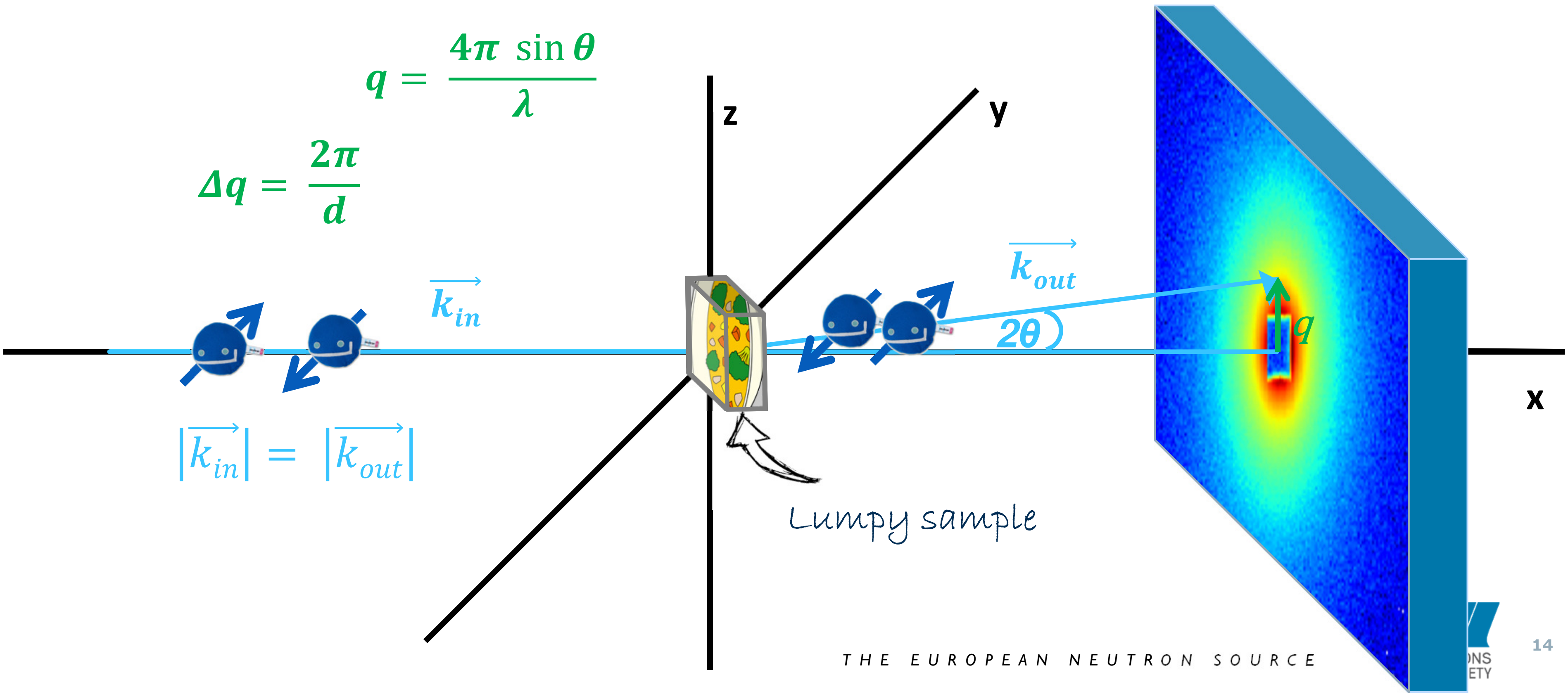


Two possible scattering geometries to capture small angles:

→ **Transmission:** particle like structures, like nanoparticles, polymers, superconducting flux line lattices, precipitates

→ **Reflection:** planar structures, like thin films, interfaces, multilayers, gratings, neutron guides, supermirrors

The neutron beam transmits through the sample and scatters away from the direct beam at small angles.



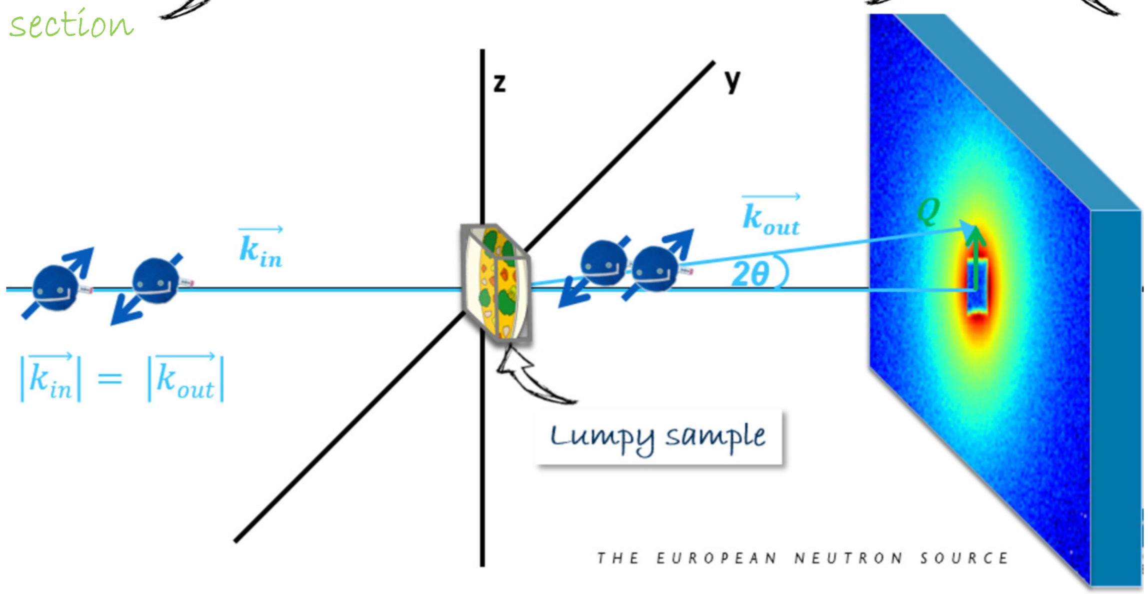
The principles of elastic (neutron) scattering apply also to small angle (neutron) scattering.

$$I(q) = \frac{d\Sigma}{d\Omega}(q) = \frac{X}{V}(\Delta\rho V_p)^2\, P(q)S(q)$$

macroscopic, differential scattering cross section

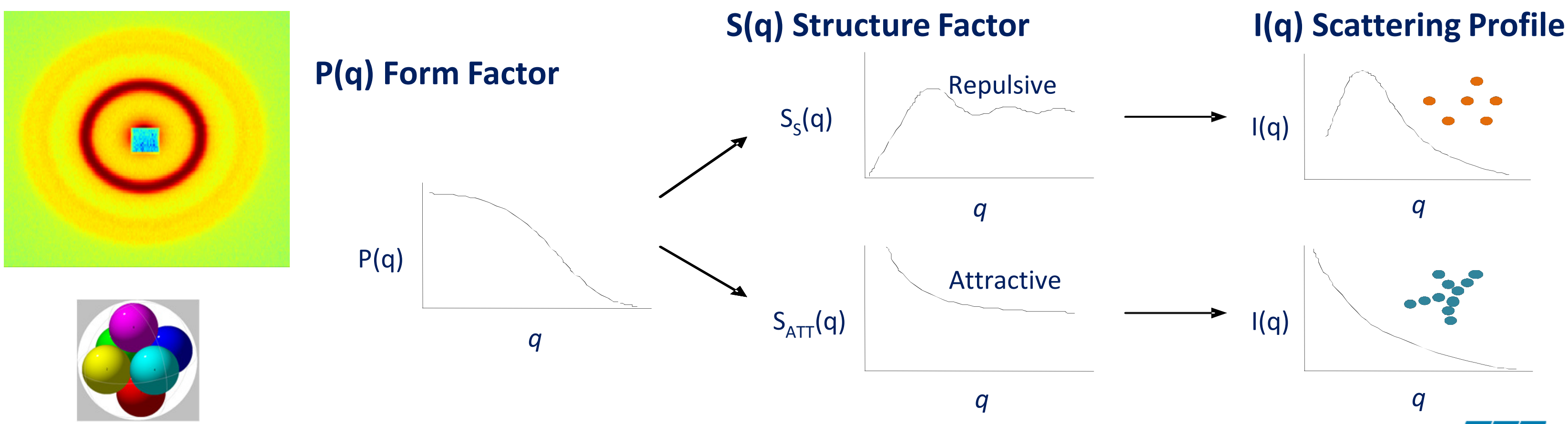
Form factor

Structure factor



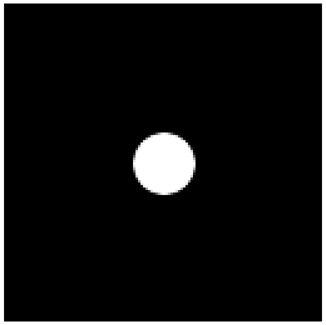
Small Angle Neutron Scattering is scattering from extended, nm-scale objects.

- Scattering from extended objects. Maybe diffraction if there is a periodic arrangement.
- Form factor: information about scattering objects.
- Structure factor: information about arrangement of particles.

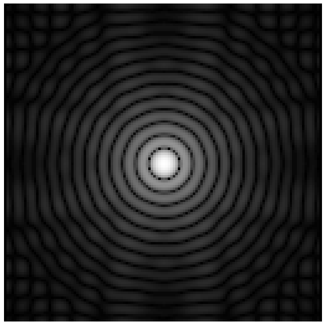


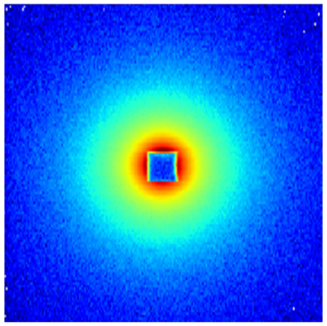
In dilute systems detector shows the fourier transform of your scattering objects (form factor).

Real space



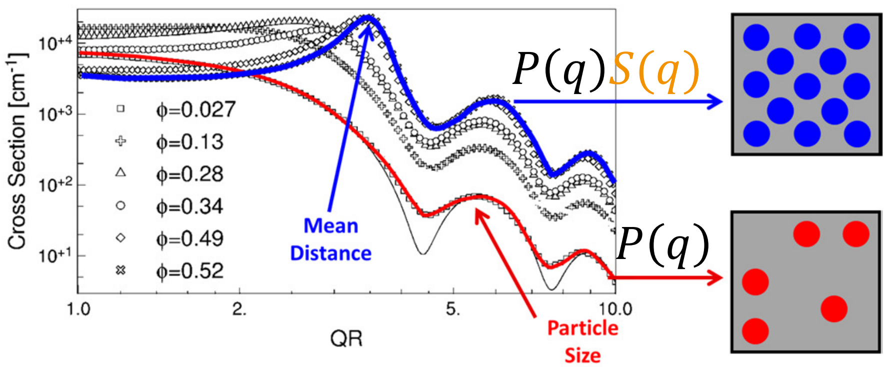
Reciprocal space



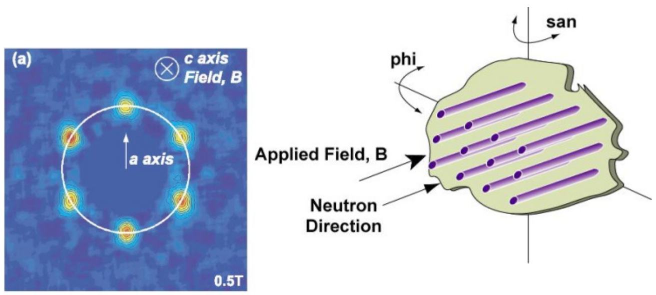


This gives information about the shape and size of the scattering objects, or at least their rotational averages.

In dense or ordered systems a structure factor appears.



Ordered systems → P(q)S(q) → Diffraction pattern

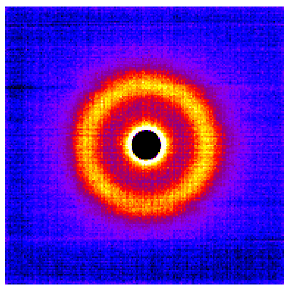
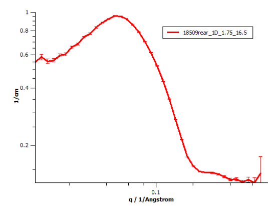


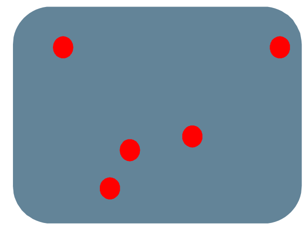
$$I(q) = \frac{d\Sigma}{d\Omega}(q) = \frac{X}{V}(\Delta\rho V_p)^2 P(q)S(q)$$

*Cubitt et al., PRL **91**, 047002 (2003);Cubitt et al., PRL **91**, 157002 (2003)*

*L.B. Lurio et al. PRL 84, **785**, (2000)*

The scattered intensity is determined by the scattering cross section.



The scattering intensity is determined by: How many particles there are, how visible they are, the FT of the shape of the particle (form factor) and the FT of the arrangement of particles (structure factor).

X/V: Number density of particles in the medium (aka N)

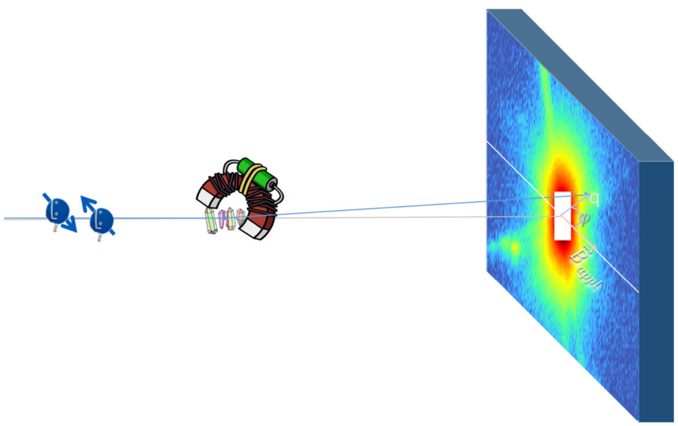
Difference between Particle (SLD – medium SLD)x particle Vol.: effective scattering power = cross section of the particles

$$I(q) = \frac{d\Sigma}{d\Omega}(q) = \frac{X}{V}(\Delta\rho V_p)^2 P(q)S(q)$$

Form factor   Structure factor

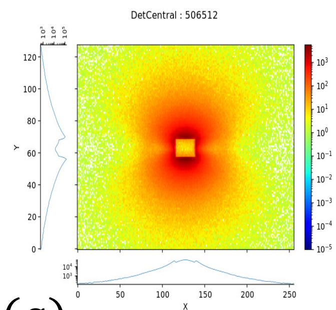
Magnetic scattering depends on the relative orientation of **M**, **B** and **q**.

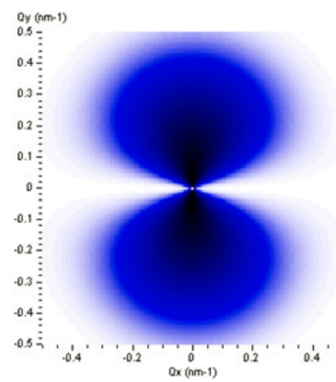
Magnetic scattering for $\boldsymbol{M} \perp \boldsymbol{q}$



SANS cross section for unpolarised neutron beam:

$$\frac{d\Sigma}{d\Omega}(q) = \frac{d\Sigma_{nuc}}{d\Omega}(q) + \frac{d\Sigma_{mag}}{d\Omega} = \frac{d\Sigma_{nuc}}{d\Omega}(q) + \frac{8\pi^3}{V} \; b_{m1}^2 \left|\tilde{Q}_{HJ}\right|^2$$





$$\frac{d\Sigma_{mag}}{d\Omega}(q, Bex_t) = \frac{X}{V}(\Delta\rho_m V_p)^2 \langle \sin^2\varphi \rangle P(q)$$

For $B_0 \perp$ neutron beam, M || B_0

Big assumption

$\sin^2\varphi$ modulation

Polarised SANS – incoming beam polarised.

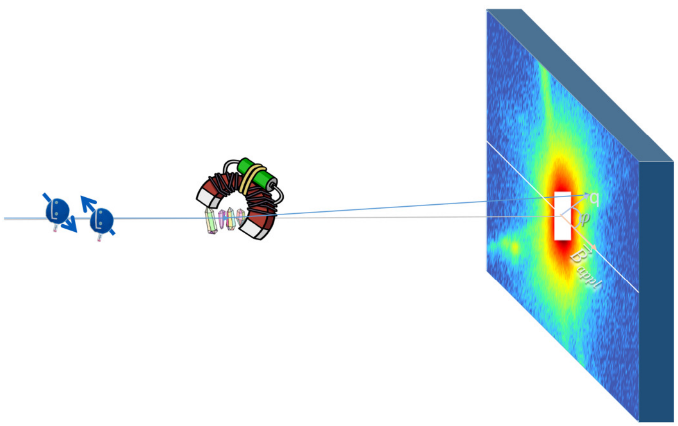
If magnetisation is saturated along external field axis and B perpendicular to neutron beam ($B_0 \perp$ neutron beam, M || B_0):

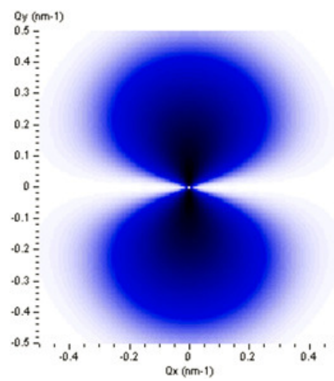
→ Horizontal cut on the detector gives pure nuclear scattering:

$$I^+ + I^- = \frac{d\Sigma_{nuc}}{d\Omega}(q) + \frac{d\Sigma_{mag}}{d\Omega} = 2\mathrm{FN}^2 + 2\mathrm{FM}^2 \langle \sin\varphi \rangle^2$$

→ Nuclear magnetic interference term $I^+ - I^- = -2\, FN\, FM \langle \sin\varphi \rangle^2$

Enhances magnetic contrast!



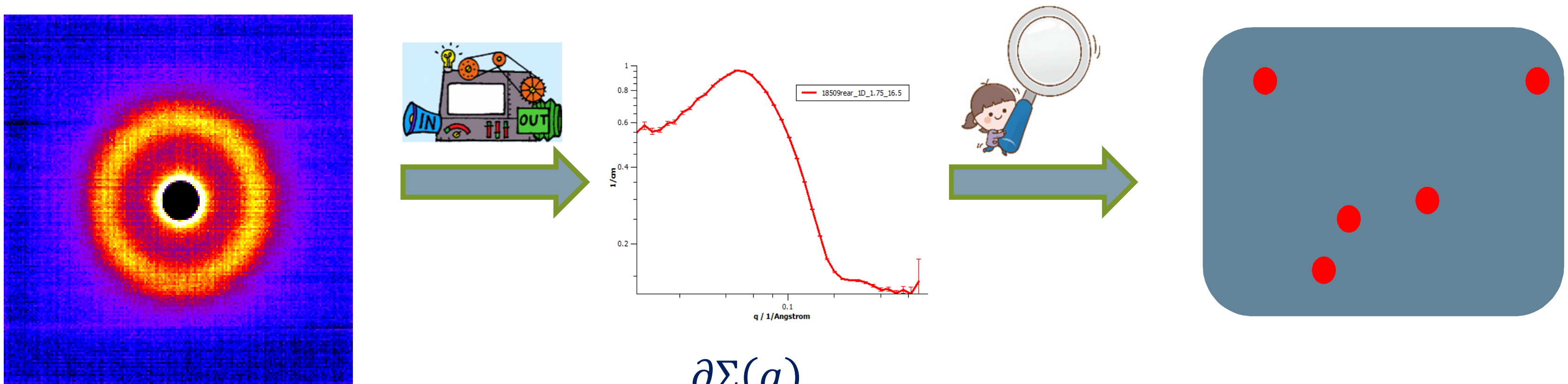


F_N, F_M – nuclear and magnetic scattering amplitude

In the real world polarisation efficiency corrections are necessary – see reference.

S Disch *et al* 2012 *New J. Phys.* **14** 013025

The intensity on the detector is determined by the scattered intensity and the reality of the experimental setup.



$$I(\theta,\lambda) = I_0(\lambda) \cdot \frac{\partial\Sigma(q)}{\partial\Omega} \cdot \Delta\Omega \cdot t \cdot T(\lambda) \cdot \eta(\lambda)$$

Solid angle $\Delta\Omega$, *t* = sample thickness, *T* = sample transmission η = detector efficiency …

In reality you will be interested only in the signal from your sample and therefore need to subtract the signal from the container (cell and solvent, nonmagnetic background etc…)

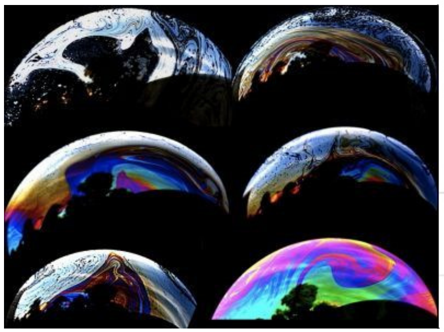
$$I_{sample}(\theta,\lambda) = I(\theta,\lambda) - Ico_{ntainer}$$

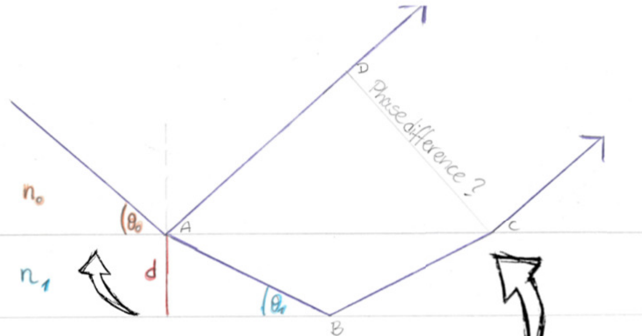
Two possible scattering geometries to capture small angles:

→ **Transmission:** particle like structures, like nanoparticles, polymers, superconducting flux line lattices, precipitates

→ **Reflection:** planar structures, like thin films, interfaces, multilayers, gratings, neutron guides, supermirrors

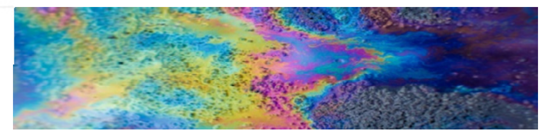
Reflectometry probes planar structures and buried interfaces through neutron interference.

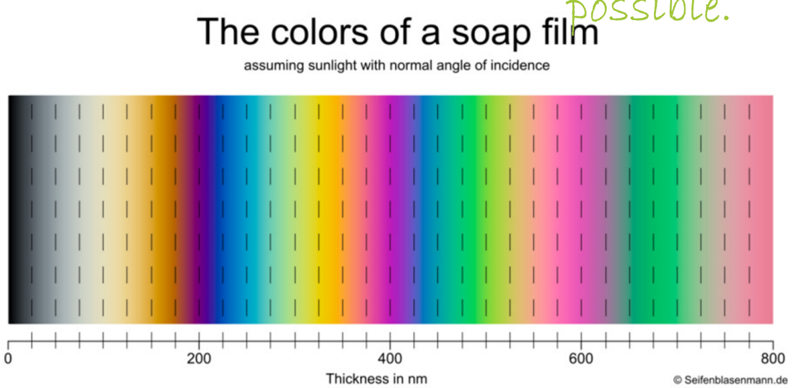




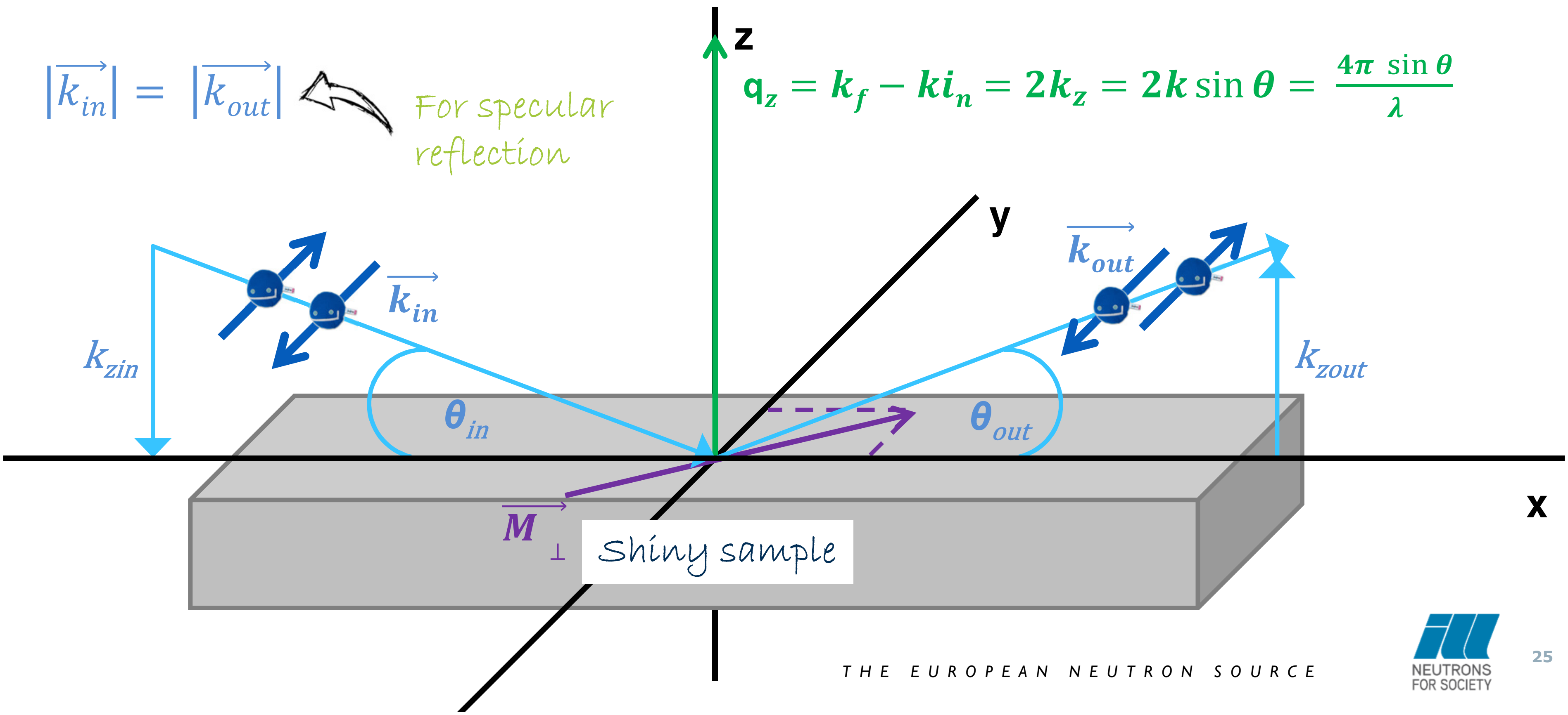
Phase shift at IF possible.

Constructive interference for $\overline{AD} = m\lambda$.





The neutron beam is reflected from the surfaces and interfaces at shallow angles.

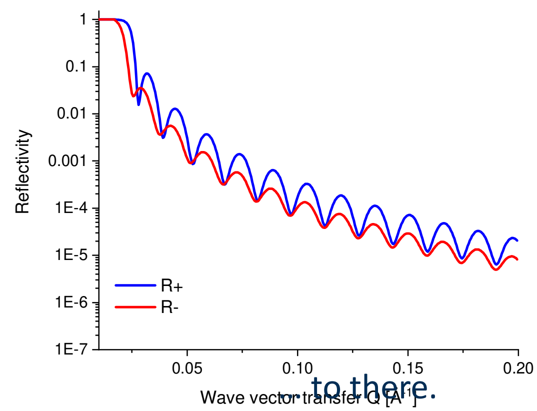


The problem is that in reflection the neutron-material interaction is not weak.

Neutron scatteri... perturbation
use **Born approxima**... work out scattering

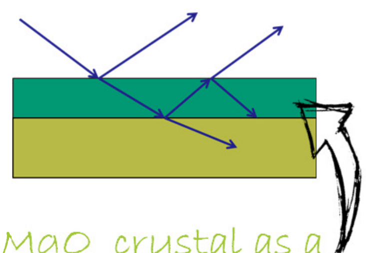
Sorry

Near total reflection region scattering is not weak! Still need to work out solutions to time independent Schrödinger equation .



to there.

Reflectivity is neutron interference from thin layers.



MgO crystal as a reflectometrist sees it

$$\frac{\hbar^2}{2m}\frac{d^2\psi(z)}{dz^2} + (\mathrm{E} - V)\psi(z) = 0$$

small angles → small q → length scale probed is large!

$$d = \frac{2\pi}{\Delta q} \quad \Rightarrow \quad \rho = \sum_i N_i\, b_i$$

density approach

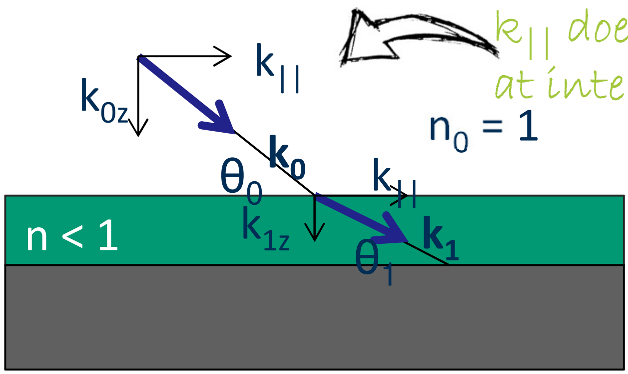
nucleus-neutron interaction (Born approximation):

$$V_n = \frac{2\pi\hbar^2}{m_n} b\delta(\boldsymbol{r})$$

$V_n(z) = \frac{2\pi\hbar^2}{m_n}\rho$ (assume V independent of x and y)

neutron mass

Doing optics with neutrons…. **Total reflection**



$k_{||}$ does not change at interface!

Homogeneous medium: **refractive index n**
For neutrons (and X-rays) typcially < 1

$$n = 1 - \frac{\lambda^2}{2\pi}\rho - i\beta$$

Snell's law

$$\cos\theta_1 = \frac{k_{||}}{k_1}$$

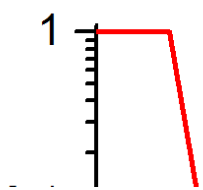
$$\frac{\cos\theta_0}{\cos\theta_1} = \frac{k_1}{k_0} = \frac{n}{n_0} = n < 1$$

$$\cos\theta_c = n$$

$$\theta_c = \lambda\sqrt{\frac{\rho}{\pi}}$$

$$Q_c = \frac{4\pi}{\lambda}\sin\theta_c = 4\sqrt{\pi\rho}$$

used small angle approximation and Taylor expansion



Total external reflection: $\theta_1 = 0$

$$\cos\theta_0 \leq \cos\theta_c = n$$

Reflection and transmission of a neutron from a single interface is treated in like the quantum mechanical potential step/well problem (should be familiar to those who took undergraduate physics lectures*).

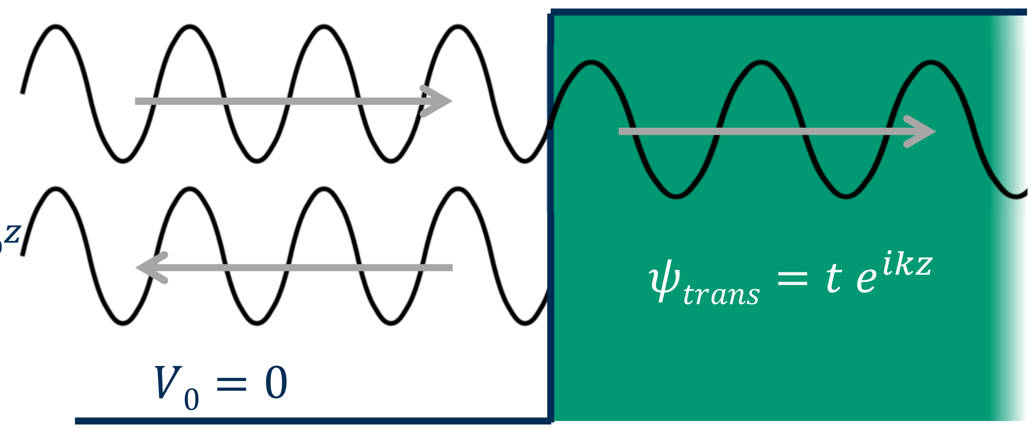
$$\frac{\hbar^2}{2m}\frac{d^2\psi(z)}{dz^2} + (\mathrm{E} - V)\psi(z) = 0$$

Ignoring the parallel component to the surface because it does not change.

Solution for SE is a wave: $\psi_z = A_{right}\, e^{ikz} + B_{left}\, e^{-ikz}$

$\psi_{in} = e^{ik_0 z}$

$\psi_{out} = r\, e^{-ik_0 z}$



* You can have evernescent waves, resonances, wave guide effects etc. …

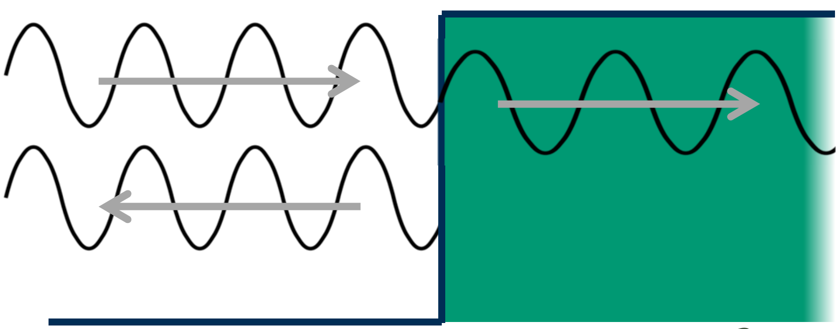
Amplitude of incoming wave is 1

$\psi^{vac}{}_z = e^{ik_0 z} + re^{-ik_0 z}$

$\psi^{med}{}_z = te^{ikz}$

There is no other interface to reflect from so there is no reflected wave

Solutions for the Schrödinger and Helmholtz equations give expression for r, t and refractive index.



$$V_0 = 0;\ k_0 = \frac{2\pi}{\lambda} \qquad V_{med} = \frac{2\pi\hbar^2}{m_n}\rho;$$

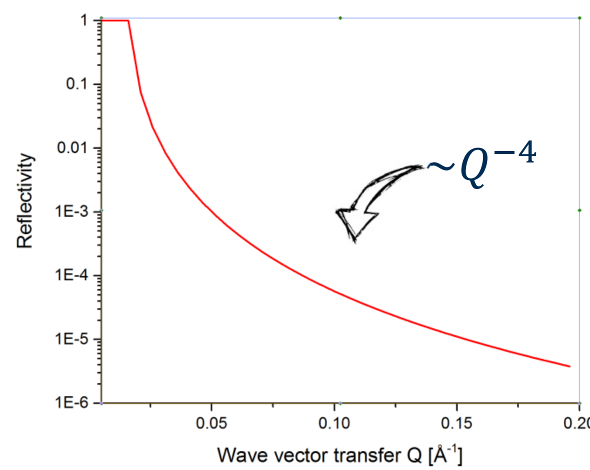
$$k_{med}{}^2 = k_{0z}{}^2 - 4\pi\rho$$

Boundary conditions: ψ_z and $\nabla\psi_z$ continuous at interface → work out r and t (appendix)

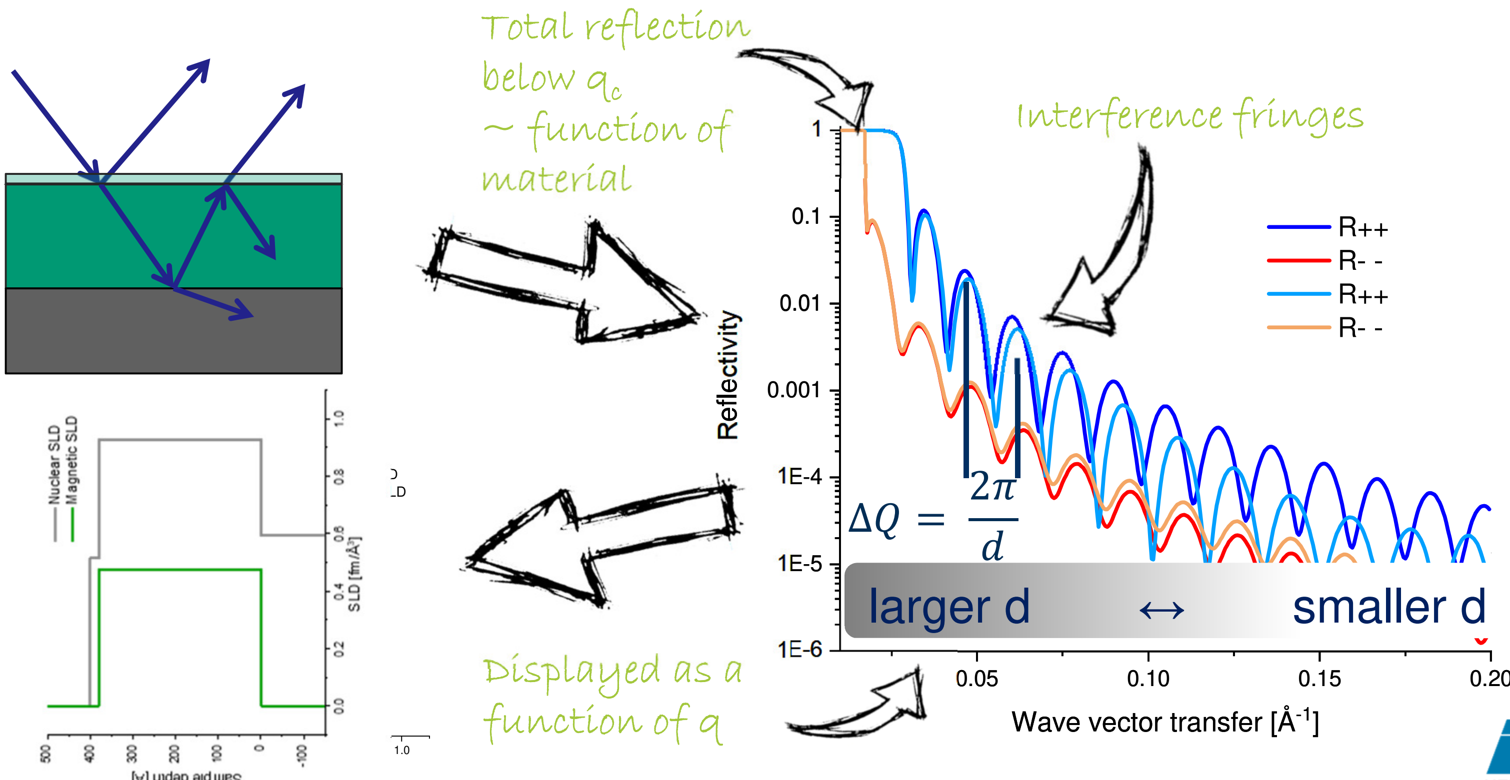
Conservation of energy means k has to change in medium

For large k_z:

$$R = r^2 \approx \frac{k_c{}^4}{16 k_{0z}{}^4} \propto Q^{-4}$$



(Polarised) neutron travelling waves reflect from the various interfaces in a planar structure and the resulting interference pattern gives information of the (magnetic and) chemical depth profile in a planar structure (through the refractive index changes).



The magnetic potential is determined by dipolar interaction between neutron magnetic moment and moments in the magnetic layer. Can be described by a magnetic scattering length density.

$$V_{mag} = -\vec{\mu_n}\,\vec{B}$$

Magnetic interaction potential™

$\boldsymbol{B} = \boldsymbol{B}_0 + \mu_0 \boldsymbol{M}_\perp$

Zeeman, neglected here.

neutron magnetic moment:
$\mu_n \approx 0.001\,\mu_B$

anti-parallel to neutron spin!

$$b_{m1} = \frac{m_n \mu_n \mu_0 \mu_B}{2\,\pi\,\hbar^2}\, m_{layer} = 2.699\text{ fm}\, m_{layer}$$

In fractions of μ_B/atom, i.e. $m_{Fe} = 2.2$

$$V_{layer} = V_{nuc} + V_{mag} = \frac{2\pi\hbar^2}{m_n} bN - \vec{\mu_n}\,\vec{B}$$
$$= \frac{2\pi\hbar^2}{m}(b_{nuc} \pm b_{mag})\,N$$

We need to talk about directions…
But this slide works without modification when the magnetisation and neutron spin are parallel and in the film plane

The amplitude and type (SF or NSF) of magnetic scattering is determined by the relative directions of the neutron magnetic moment, magnetisation and momentum transfer q.

Vectorial quantities to deal with:
- → Neutron magnetic moment $\overrightarrow{\mu_n}$
- → Magnetisation $\vec{M}$
- → Wave vector transfer $\vec{q}$

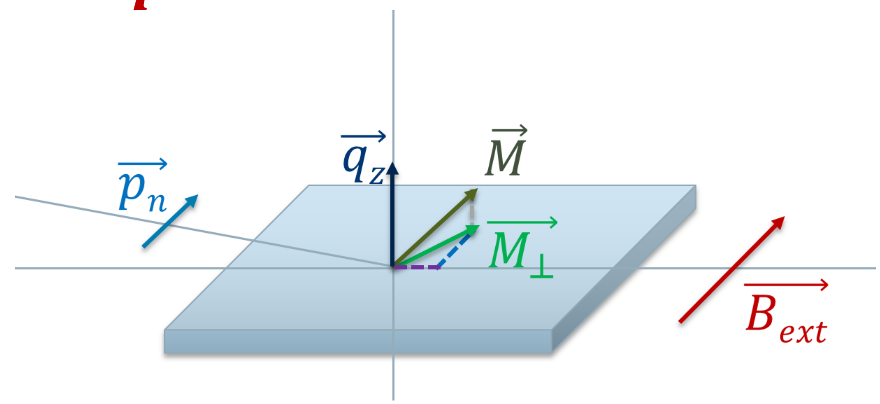
$$V_{mag} = -\overrightarrow{\mu_n}\,\vec{B}$$

Only $\vec{M} \perp \vec{q}$ visible!

$$\boldsymbol{B} = \boldsymbol{B_0} + \mu_0 \boldsymbol{M_\perp}$$

Magnetic interaction vector for neutron scattering (Halpern, Johnson):

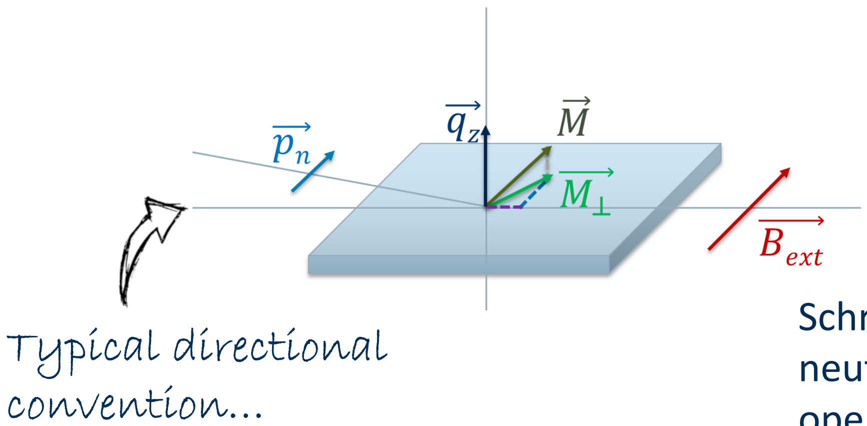
$$\boldsymbol{M_\perp} \propto \boldsymbol{Q_{HJ}} = \boldsymbol{q} \times (\boldsymbol{M} \times \boldsymbol{q}) = \boldsymbol{M} - \boldsymbol{q}(\boldsymbol{Mq})$$



Reflectivity measures as a function of q_z **→ can NOT measure out of plane magnetisation!**

Probably biggest disadvantage of Neutron reflectometry...

Magnetic scattering can be spin-flip or non spin-flip.



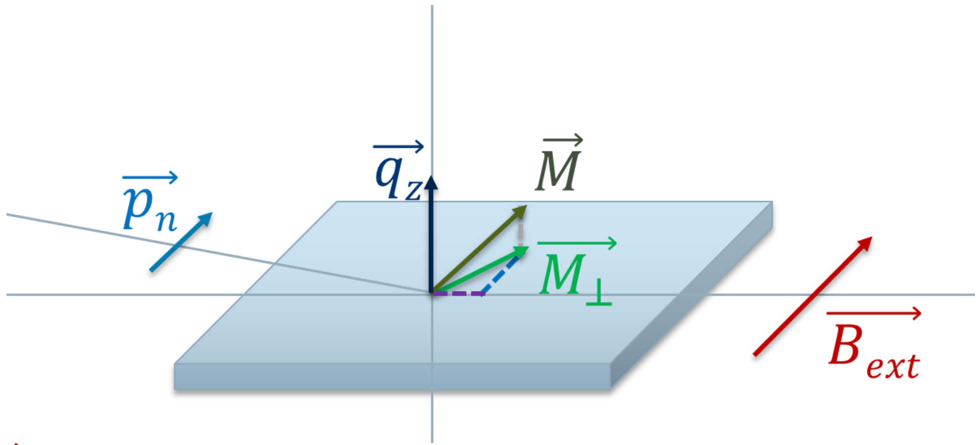
$$V_{mag} = -\vec{\mu_n}\,\vec{B}$$

Schrödinger equation now includes wave functions for neutron spin up and down and interaction includes spin operator

Szenarios:

- M collinear with neutron spin → two independent equations, no spin flip scattering (i.e. no spin-mixing terms), introduce effective magnetic scattering length (slide 38).
- M perpendicular to neutron spin → two coupled equations for the two spin states containing spin mixing, spin flip scattering
- Measure four reflectivity components (spin in, spin out): (++), (+-), (-+), (--)

Most common polarised neutron reflectometry setup, but the vectorial relations are valid also for other geometries.



$\overrightarrow{p_n}, \overrightarrow{\mu_n} \parallel \overrightarrow{B_{ext}}$

$\overrightarrow{q_z} \perp \overrightarrow{M_{visible}} = \overrightarrow{M_\perp}$

$\vec{M} \parallel \overrightarrow{p_n}$ - no spin flip scattering

$\vec{M} \perp \overrightarrow{p_n}$ - spin flip scattering

Magnetic interaction - summary.

→ y and x components of magnetisation are separated out into non-spin flip and spin flip components.

→ Non-spin flip: Nuclear and magnetic scattering $V_{noflip} \sim (b_n \pm b_m \cos\gamma)$

→ Spin flip: Magnetic scattering only

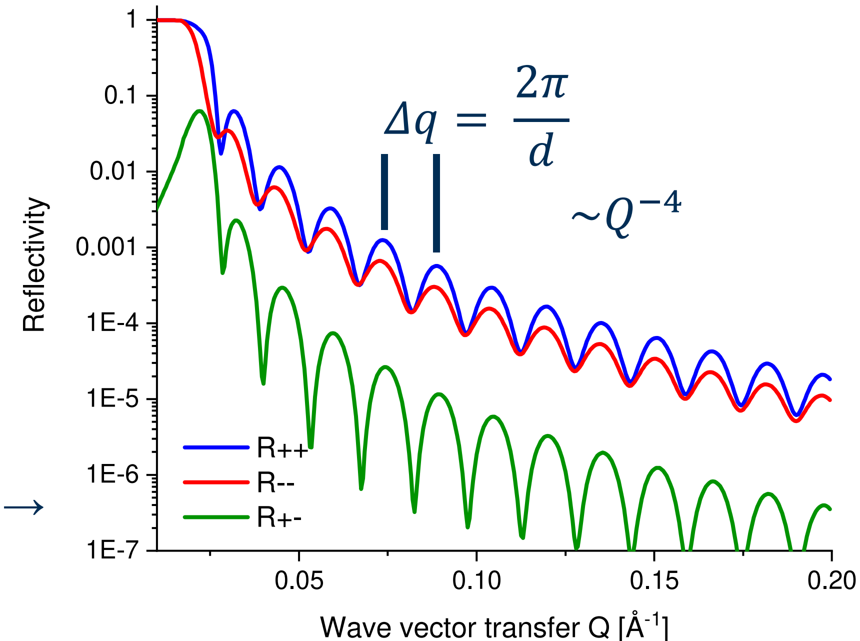
$$V_{flip} \sim b_m \sin\gamma$$

→ Specular reflectivity: $R^{+-} = R^{-+}$

Only $\vec{M} \perp \vec{q}$ visible!

Reflectivity measures as a function of q_z → can NOT measure out of plane magnetisation!

$$q_c = 4\sqrt{\pi(\rho_{nuc} \pm \varrho_m)}$$



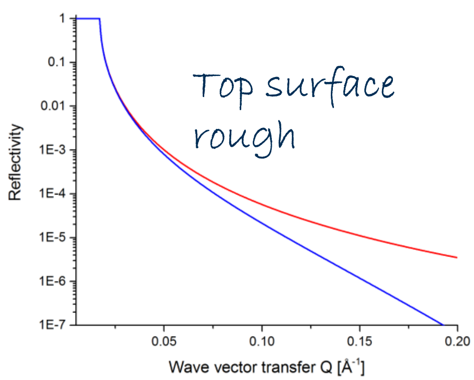
Real samples are complicated!

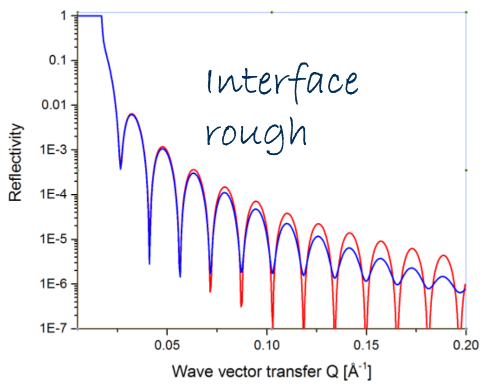
Roughness is hard to describe!

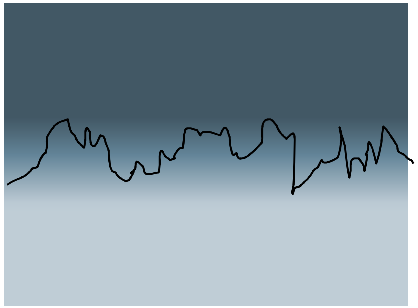
- Roughness is complicated, approximations are heuristic.
- Depends on length scale.
- Specular reflectometry does not distinguish between correlated and uncorrelated roughness. (but there may be a diffuse background present in the data).
- Thickness gradient: blurs Kiessig fringes, looks like a loss of (λ-)resolution

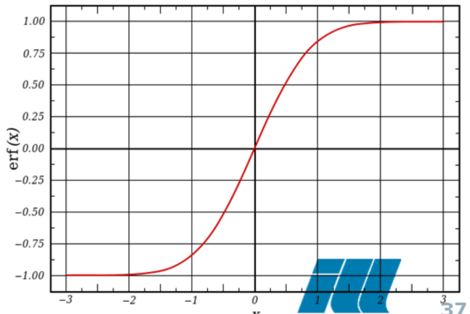
Croce-Nevot factor
Multiply each layer transfer matrix with
$e^{-k_{zj}k_{zj+1}<z>2}$
assumes small, uncorrelated fluctuations

Graded interface:
Replace random z fluctuations with smoothly varying scattering length density profile. (Describe ρ variation with a function and split into many layers)

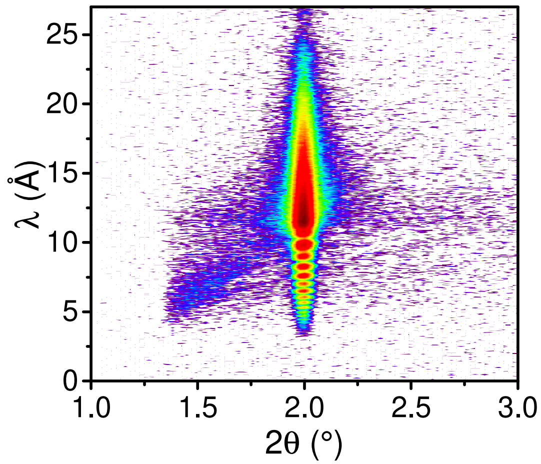


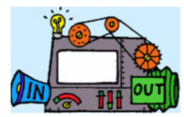


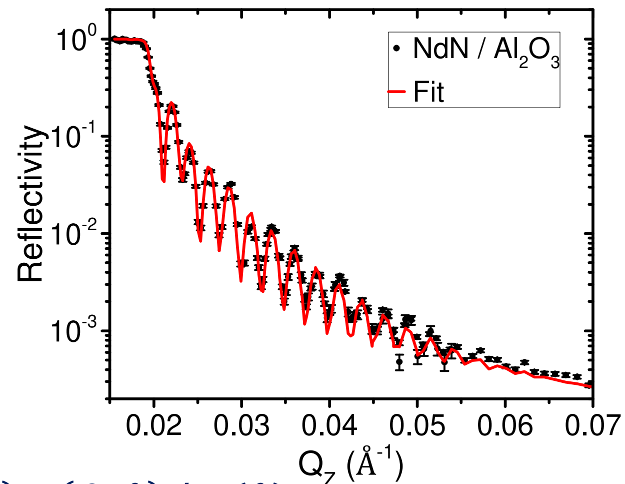




Data reduction transforms the detector image into a reflectivity curve.



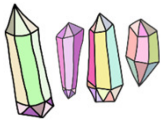




$$R(\theta,\lambda) = (I_{meas}(\theta,\lambda) - Ibc_{kg})\,\eta(\theta,\lambda)/I_0(\lambda)$$

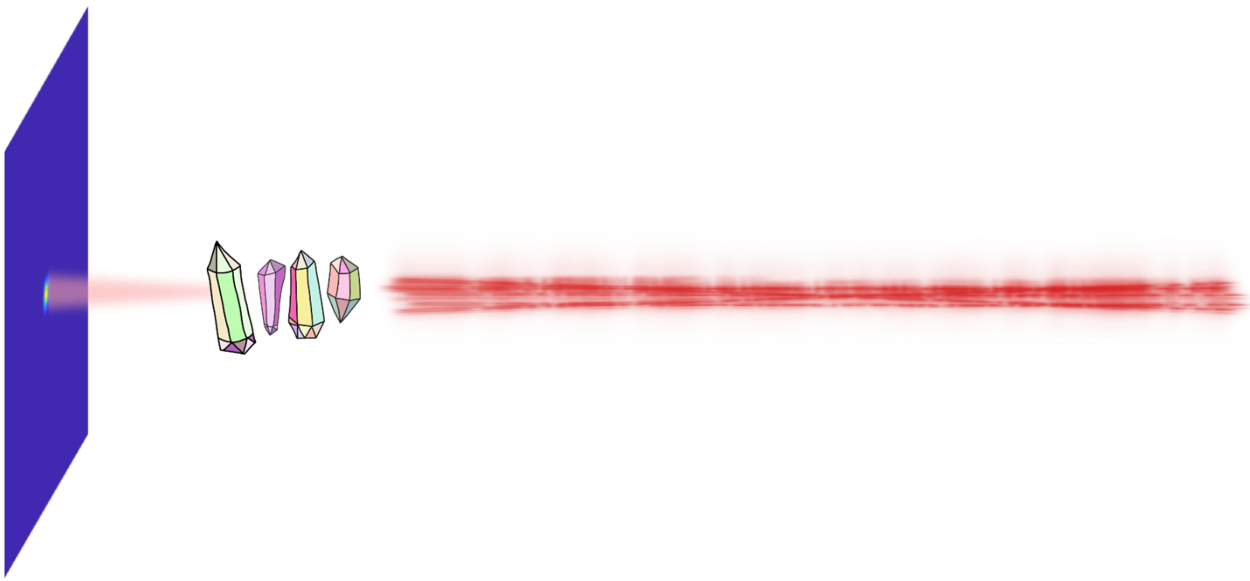
- Background is usually measured simultaneously on the same detector
- Need to find suitable integration range for θ (can be tricky).
- For polarised data polarisation efficency corrections also need to be applied.

Let’s build a SANS machine



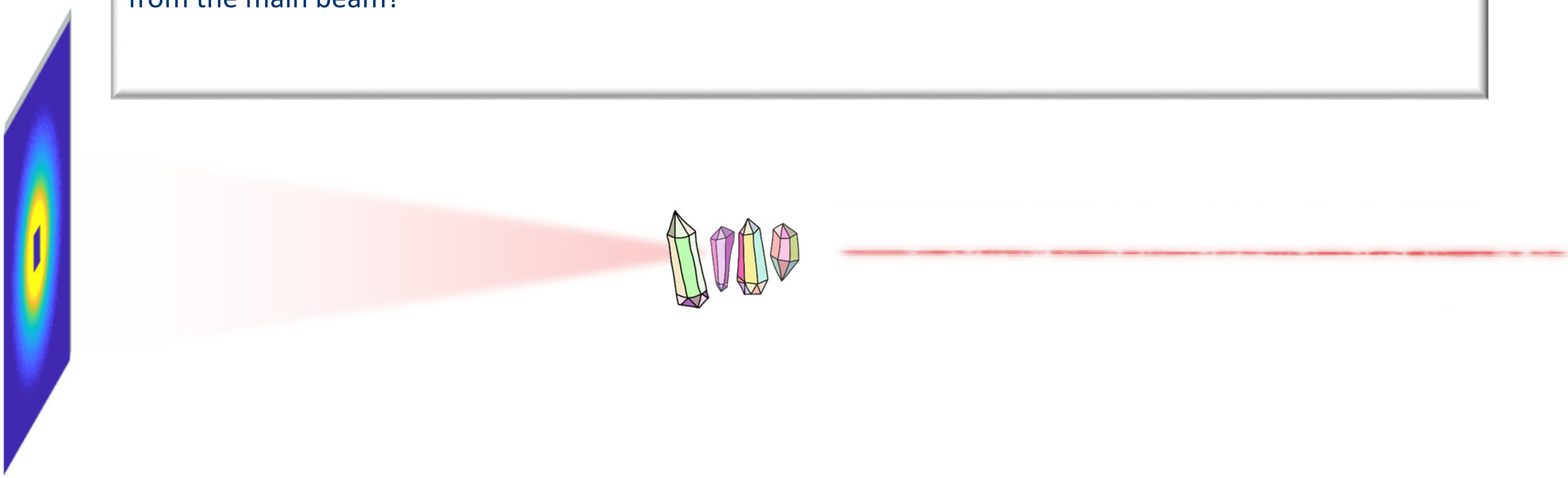
Let’s build a SANS machine

Let’s build a SANS machine



Let’s build a SANS machine

Question: How far do I need to move the detector from the sample to separate a 0.1deg scattering angle from the main beam?

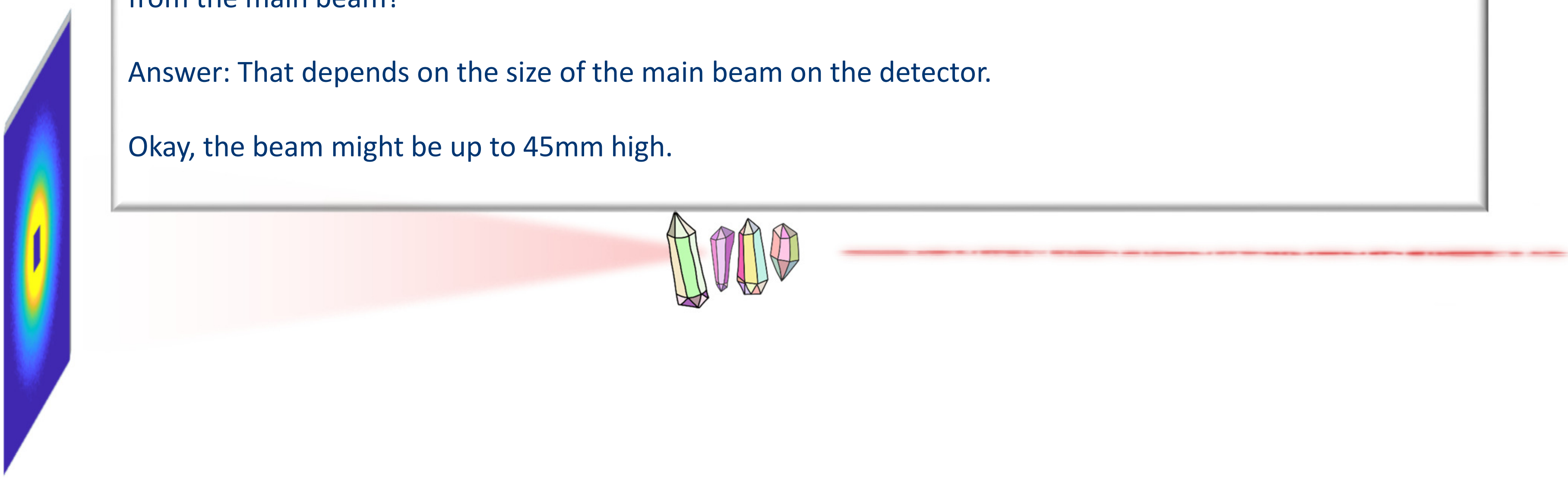


Let’s build a SANS machine

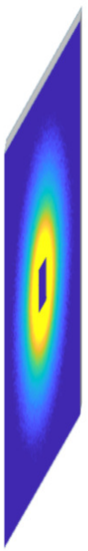
Question: How far do I need to move the detector from the sample to separate a 0.1deg scattering angle from the main beam?

Answer: That depends on the size of the main beam on the detector.

Okay, the beam might be up to 45mm high.



Let’s build a SANS machine



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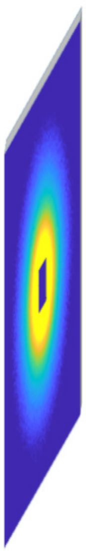
Then 22.5mm/distance = tan(0.1deg), the bare minimum distance is ~12.9m

How can I resolve smaller angles?

Let’s build a SANS machine



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Okay, the beam might be up to 45mm high.

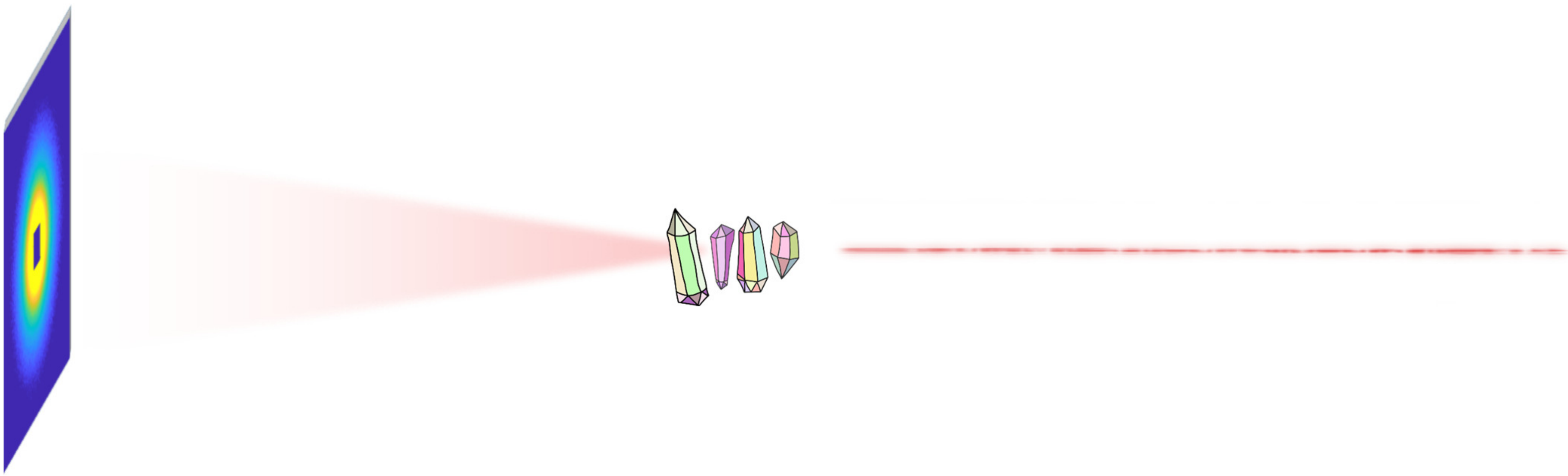
Then 22.5mm/distance = tan(0.1deg), the bare minimum distance is ~12.9m

How can I resolve smaller angles?

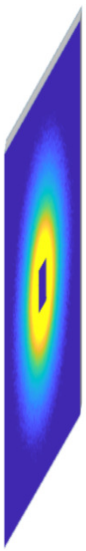
With larger detector distances

Yes, but only if the angular spread (aka angular divergence) of your beam is smaller than the scattering angle. -> You need smaller angular divergence.

Let’s build a SANS machine



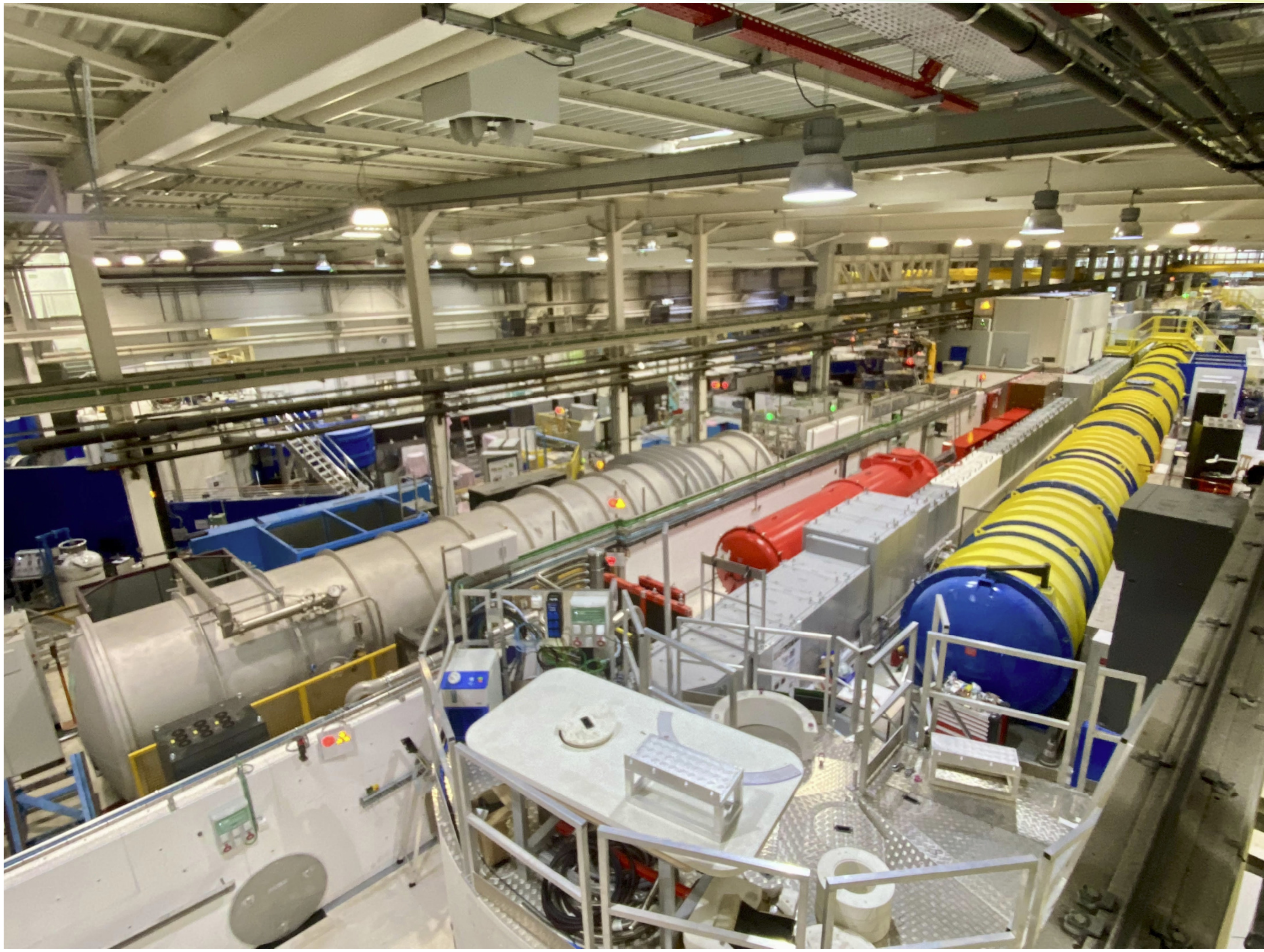
Let’s build a SANS machine



Yes, but only if the angular spread (aka angular divergence) of your beam is smaller than the scattering angle. -> You need smaller angular divergence.

How do we limit the angular divergence?

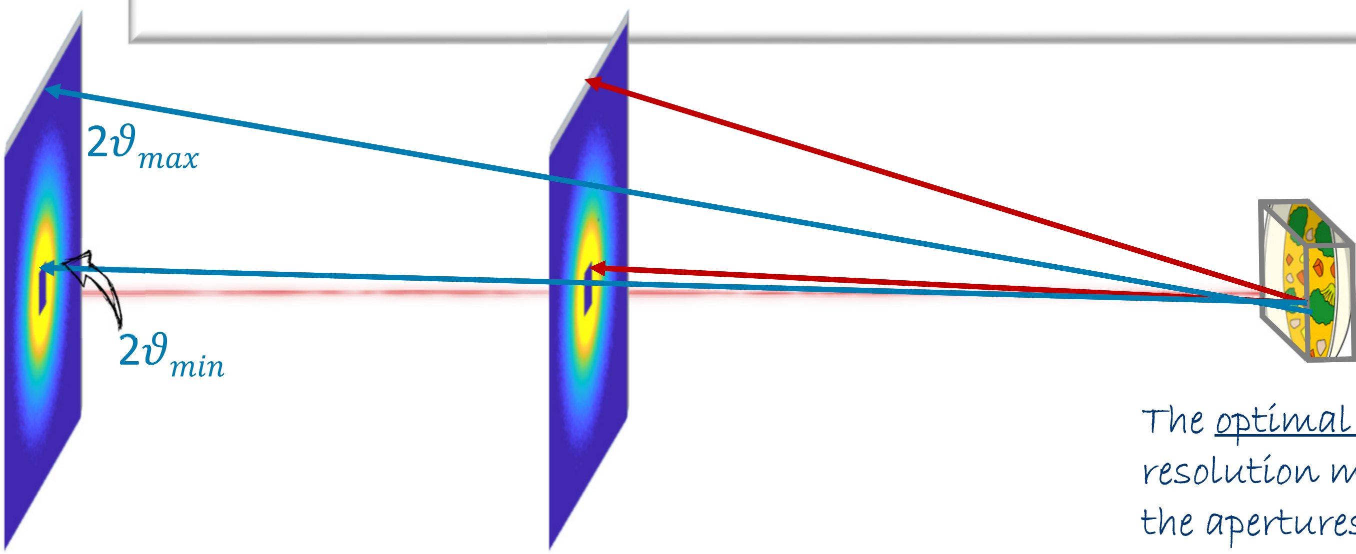
Smaller apertures, move apertures further apart…



NEUTRONS FOR SOCIETY

Let's build a SANS machine

Question: What determines the q-range I can access with my specific setup?



$$q = \frac{4\pi \sin\theta}{\lambda}$$

$$\tan 2\vartheta \approx 2\vartheta = \frac{distance\ pixel\ to\ beam\ centre}{detector\ distance}$$

The optimal setup considering flux and resolution matches the distances between the apertures defining the incoming beam with the detector distance. Sometimes we need to deviate from this though.

What if we need a larger q-range?

increase θ range

$$q = \frac{4\pi \sin\theta}{\lambda}$$

more detector distances (standard monochromatic SANS):

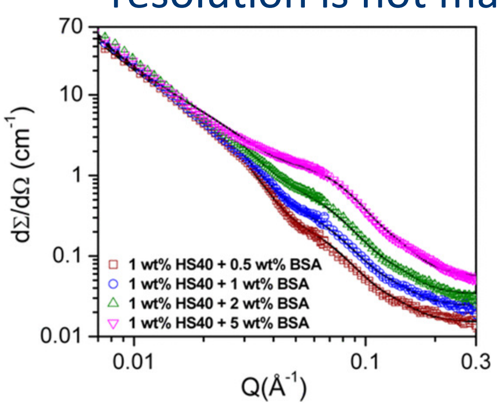
→ advantage: well established, works for asymmetric data, great for Bragg peaks

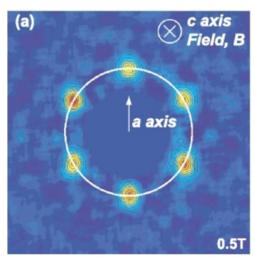
→ disadvantage: slow to change configuration, not good for kinetics, resolution is not matched

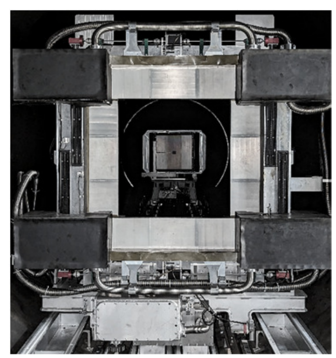
more detectors:

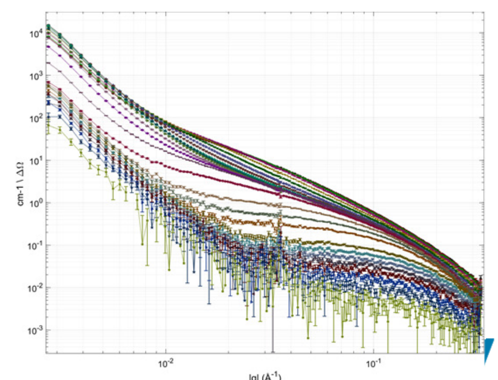
→ advantage: large q-range, single λ (good for polarisation and polarisation analysis), kinetics, quite fast

→ disadvantage: resolution is not matched, not good for asymmetric data, instrument cal more complicated









What if we need a larger q-range?

increase λ range

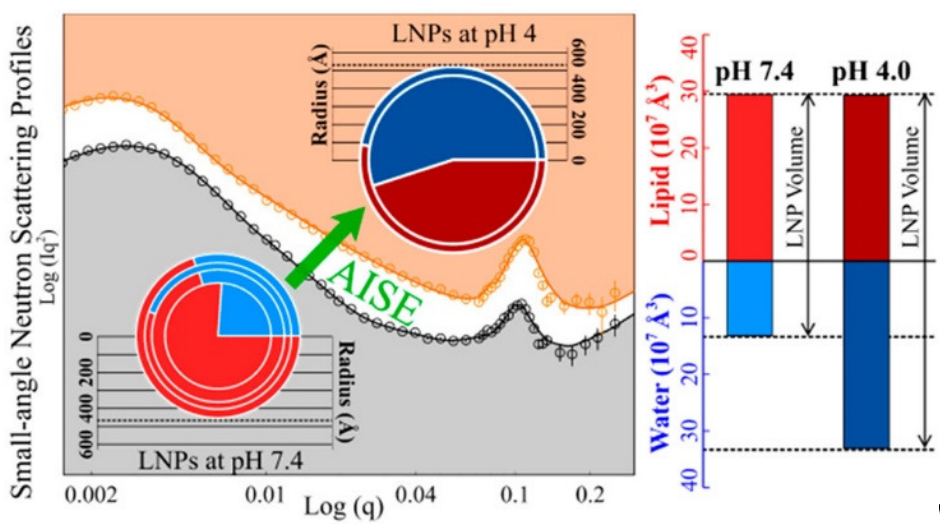
$$q = \frac{4\pi \sin\theta}{\lambda}$$

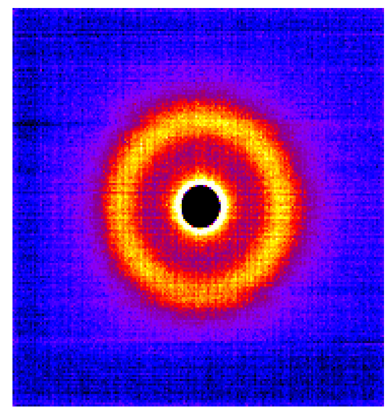
TOF mode

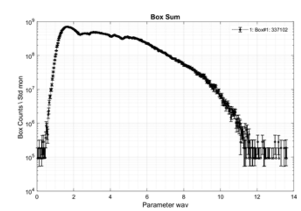
→ advantage: very large simoultaneous q-range, kinetics

→ disadvantage: can be slow, data treatment complicated, q-range edges, inefficient for bragg peaks, H can be an issue

Only possible mode on spallation sources!

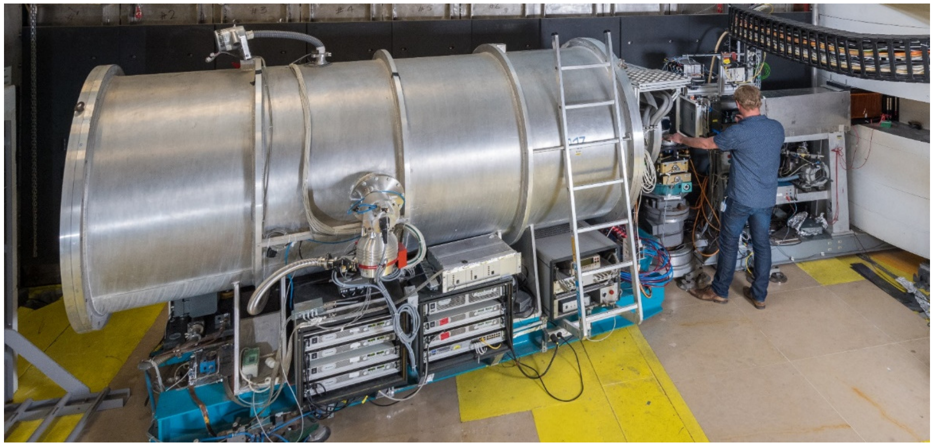


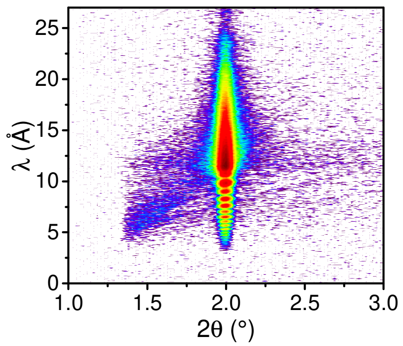




ACS Nano 2023, 17, 2, 979–990

And how does a reflectometer look different?

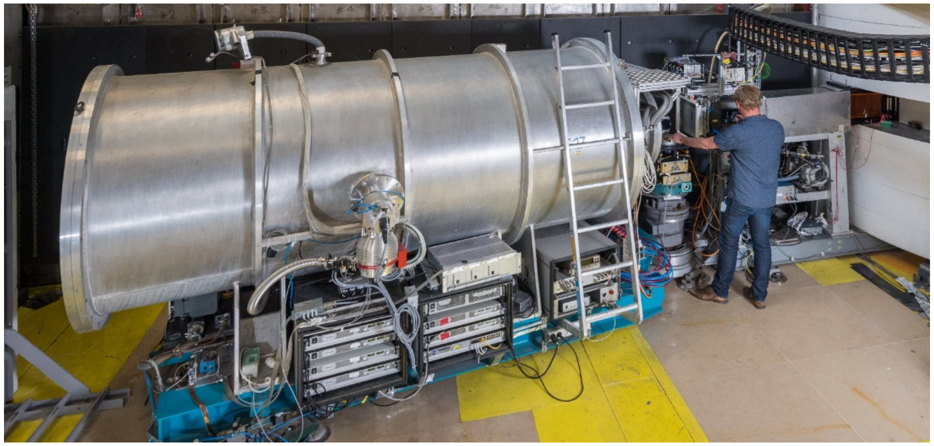


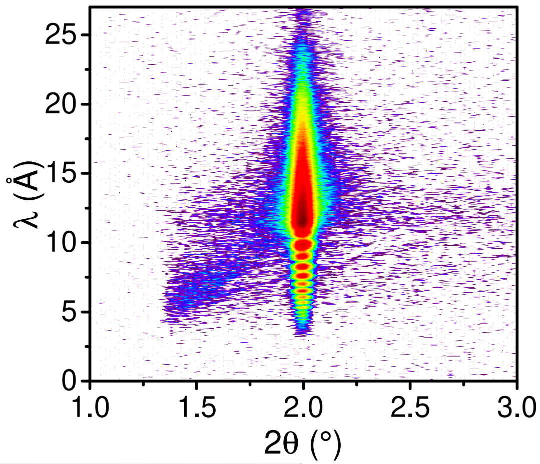


Question: What beam shape would you use for a reflectometer?

Very collimated perpendicular to the sample surface, but can be large parallel to the sample → letterbox

And how does a reflectometer look different?

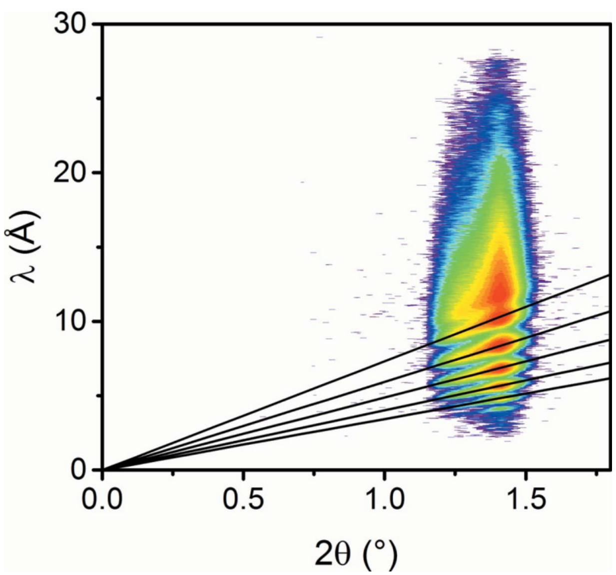




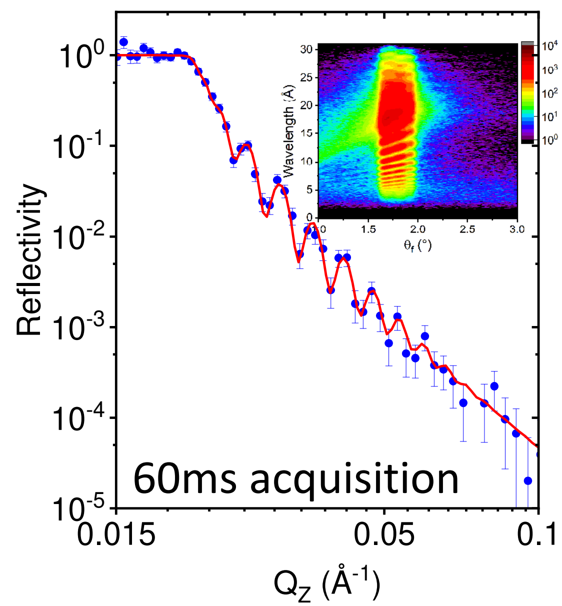
Question: TOF or Monochromatic?
Both work, but the majority of reflectometers are now TOF (large simultaneous q-range often wins, less of a q-range edge problem, less H in the beam as samples thin)

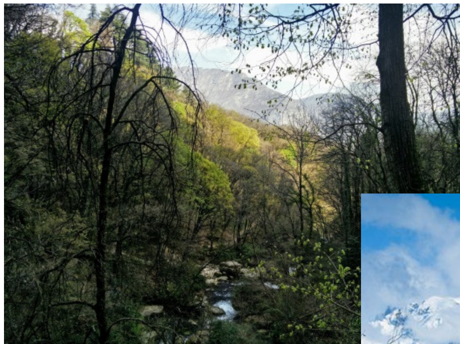
New ways do deal with real samples make use of modern area detectors to reconstruct reflectivity from wavy surfaces or large beams.

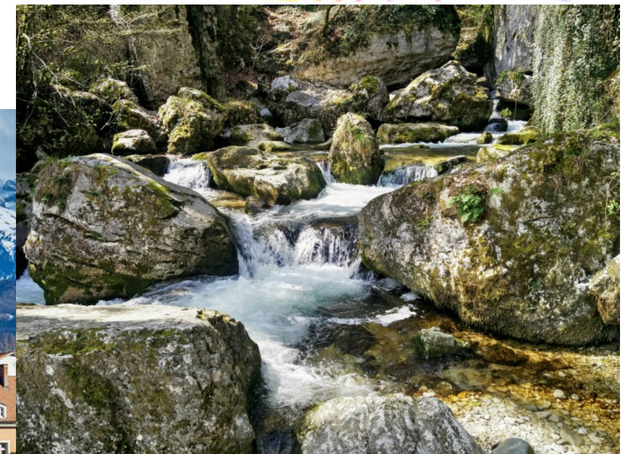
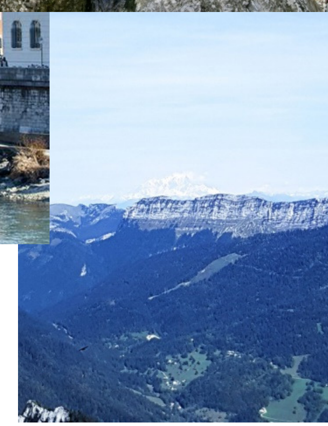
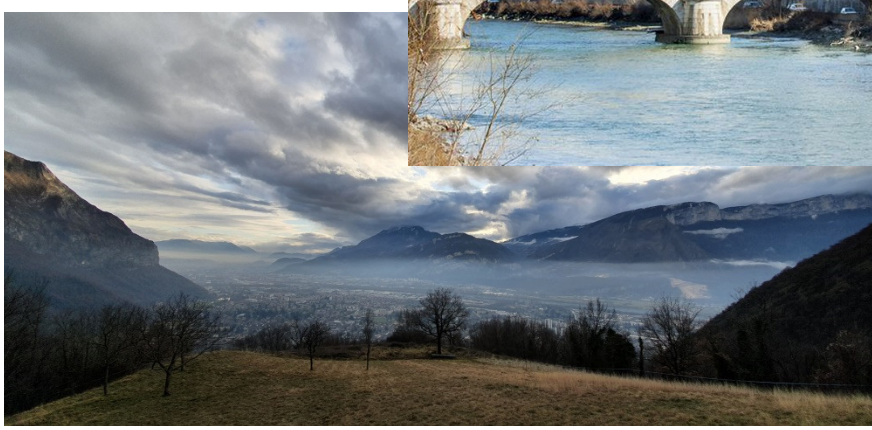
- → Roughness for NR is short scale
- → Long scale roughness (wavy, bent surfaces or facets) does not lose coherent information. (Provided thickness is constant).
- → Information can be retrieved with position sensitive detector and the right data treatment.



Cubitt et al., J. Appl. Cryst. ***48****, 2006 (2015)*





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Appendix

d – d-spacing in sample
θ – ½ scattering angle of neutron (total scattering angle = 2θ)
k – neutron wavevector, $\overrightarrow{k_{in}}$ incoming wave vector, in vacuum $k_0 = \frac{2\pi}{\lambda}$
q – wave vector transfer, qc wave vector transfer at critical edge
Ψ – neutron wavefunction
m, m_n – neutron mass
μ_n – neutron magnetic moment
ρ, ρ_{nuc}, ρ_m – scattering length density (general, nuclear, magnetic)
R_g – radius of gyration (rotational average)
t – sample thickness, T – sample transmission
Q_{HJ} – Halpern-Johnson vector of magnetic interaction
b, b_c – bound coherent nuclear scattering length
b_m – magnetic scattering length, b_{m1} magnetic scattering length for magnetic moment of 1 μ_B
N – atomic density (number of atoms per volume), X - number of particles
E – energy of neutron
V, V_n – potential energy of neutron(V_{nuc}, V_{mag} nuclear and magnetic potential)
$\delta(\boldsymbol{r})$ – delta function
$V_{unit\ cell}$ – unit cell volume
η – detector efficiency
λ – neutron wavelength
φ – angle between external field axis and q (in the detector plane) relevant for magnetic SANS
γ – angle between magnetisation in sample and q-vector, determines spin flip or non spin flip scattering
All vectors are written either in bold **v** or like this $\vec{v}$.

Back to Schrödinger: determine the refractive index n…

→ In vacuum potential V = 0, energy of neutron is: $\mathrm{E} = \frac{\hbar^2 {k_0}^2}{2m}$

→ In a medium: $V(z) = \frac{2\pi\hbar^2}{m} bN$

Time independent Schrödinger equation ↔ Helmholtz propagation equation

$$\frac{\hbar^2}{2m}\frac{d^2\psi(z)}{dz^2} + (\mathrm{E} - V)\psi(z) = 0 \qquad \frac{d^2\psi}{dz^2} + \mathrm{k}^2\psi(z) = 0$$

compare: $k^2 = \frac{2m}{\hbar^2}(\mathrm{E} - V)$

use $n^2 = \frac{k^2}{{k_0}^2}$, $k_0 = \frac{2\pi}{\lambda}$

$$n^2 = 1 - \frac{\lambda^2}{\pi}\mathrm{Nb} \rightarrow n \approx 1 - \frac{\lambda^2}{2\pi}\rho$$

Work out reflection from a single interface:

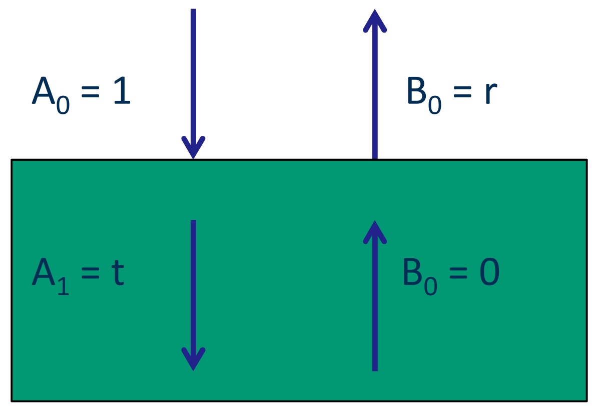
General solution for SE: $\psi_z = A\,e^{ikz} + B\,e^{-ikz}$

$$\frac{\hbar^2}{2m}\frac{d^2\psi(z)}{dz^2} + (\mathrm{E} - V)\psi(z) = 0$$

From previous slides: $k_z^{\,2} = \frac{2m}{\hbar^2}(\mathrm{E} - V) = k_{0z}^{\,2} - 4\pi\rho$ (remember $k_{||}$ does not change)

z < 0: $\psi_z = e^{ik_{0zz}} + re^{-ik_{0zz}}$

z > 0: $\psi_z = te^{ik_z z}$



Boundary conditions: ψ_z and $\nabla\psi_z$ continuous at interface

reflection amplitude:

$$r = \frac{k_{0z} - kz}{k_{0z} + kz}$$

transmission amplitude:

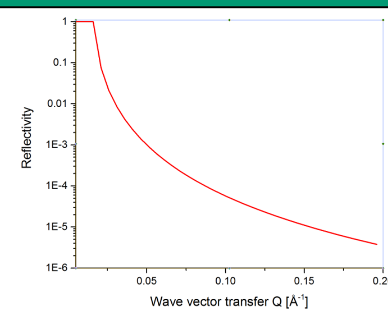
$$t = \frac{2k_{0z}}{k_{0z} + kz}$$

For large k_z:

$$\sqrt{a^2 - b^2} \approx a - {b^2}/{2a},\ a \to \infty$$

$$R = r^2 = \left(\frac{k_{0z} - \sqrt{k_{0z}^{\ 2} - kc^2}}{k_{0z} + \sqrt{k_{0z}^{\ 2} - kc^2}}\right)^2 \approx \frac{k_c^{\ 4}}{16k_{0z}^{\ 4}} \propto Q^{-4}$$

$Q = 2k_{0z}$, $Q_c = 2k_c$



For many interfaces the transmitted wave of the n^th interface becomes the incoming wave of the n+1^th interface but the formalism remains the same.

→ Work out reflection and transmission amplitudes in successive layers using boundary conditions as before: at each interface ψ_z and $\nabla\psi_z$ continuous.

→ Also remember phase change of travelling wave in each layer.

→ Write all this in matrix formalism:

$$\begin{bmatrix}1\\r\end{bmatrix} = \prod_j M_j \begin{bmatrix}t\\0\end{bmatrix} = \begin{bmatrix}M_{11} & M_{12}\\M_{21} & M_{22}\end{bmatrix}\begin{bmatrix}t\\0\end{bmatrix}$$

Travelling waves will interfere.

Looks horrible but is basically the same as before, easy for a computer.

Blundell & Bland, PRB 46(6), 3391 (1992)