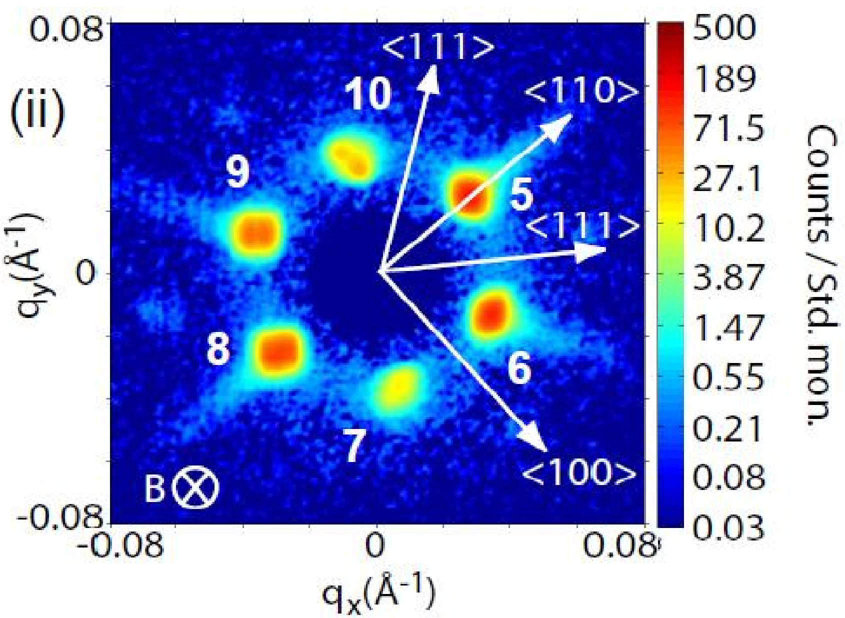
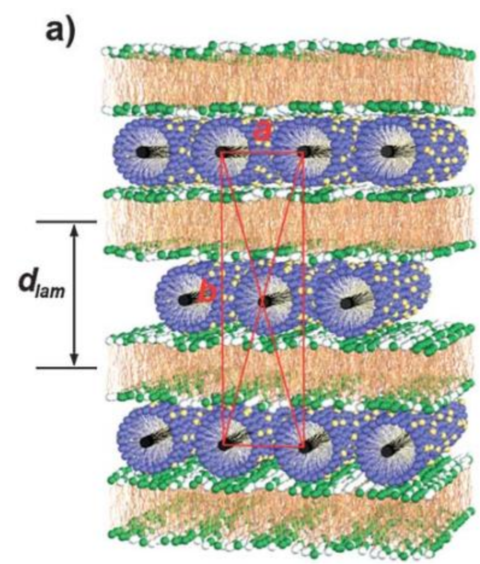
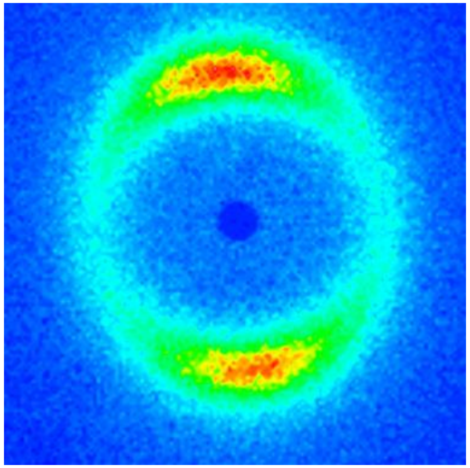
# Physics with Neutrons II, SS 2016



## Lecture 2, 25.4.2016

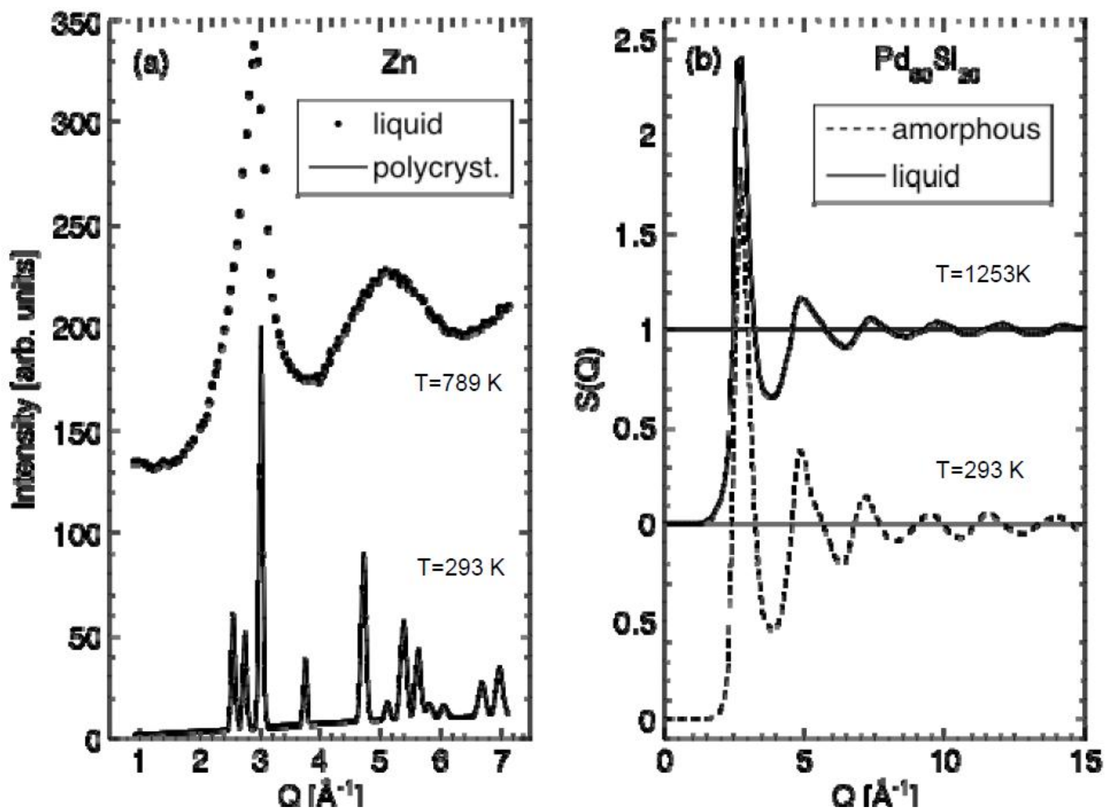
# Reminder: Scattering on liquids



Pair correlation function G(r,t) useful for description of liquids

Liquid (amorphhous) sample ⟺ Crystalline sample

⇨ Similar density, no LRO, only short range correlations



Static structure factor: Looking at deviations of the mean density n(r)

$$G'(\boldsymbol{r}) = \frac{1}{N}\int \langle n(\boldsymbol{r}' - \boldsymbol{r}) - \langle n(\boldsymbol{r}' - \boldsymbol{r})\rangle)(n(\boldsymbol{r}') - \langle n(\boldsymbol{r}')\rangle)\rangle \mathrm{d}\mathrm{r}'$$

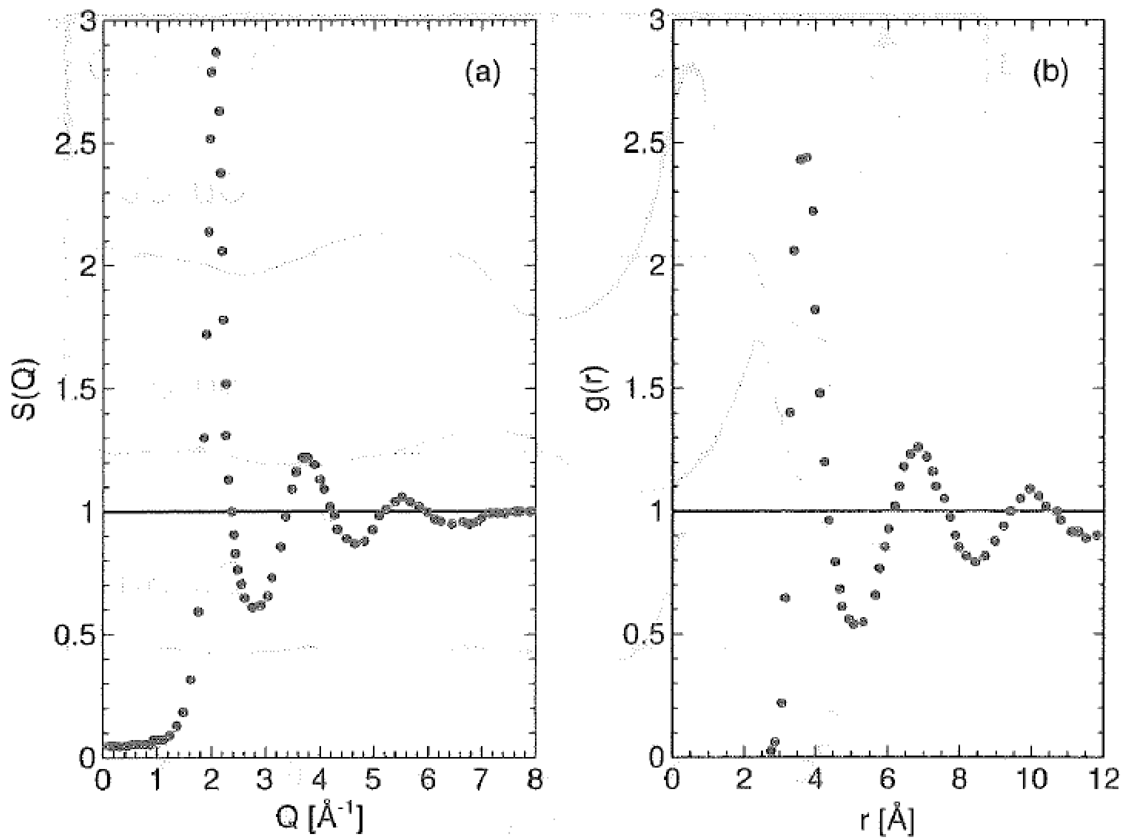
Elastic scattering from liquids

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = N\langle b\rangle^2(1 + \int (g(\boldsymbol{r}) - n_0)e^{i\boldsymbol{Q}\boldsymbol{r}}\mathrm{d}\mathrm{r}$$

g(r): pair correlation function

$$S(Q) = 1 + 4\pi\int_0^\infty (g(r) - n_0)\frac{\sin Qr}{Qr}r^2\mathrm{d}\mathrm{r}$$

Static structure factor: Looking at deviations of the mean density n(r)




Static structure factor:
Scattering function

g(r) pair correlation function:
Deviations from mean density n(r).

Limit Q->∞ $S(Q=\infty)=1$

Limit Q->0 $S(Q\to 0)=n_0\kappa_T k_B T$ Isothemal compressibility

# SANS – applications and extensions

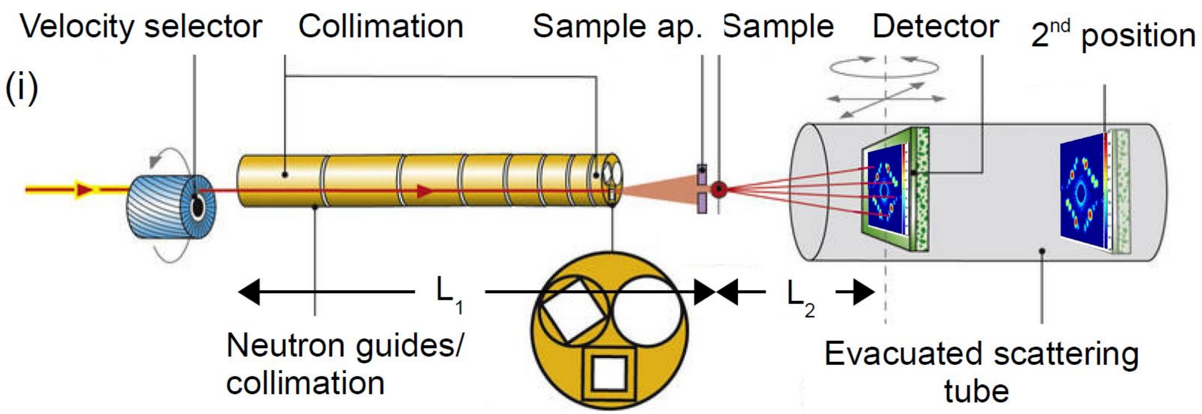
# SANS – Basic theory

# SANS



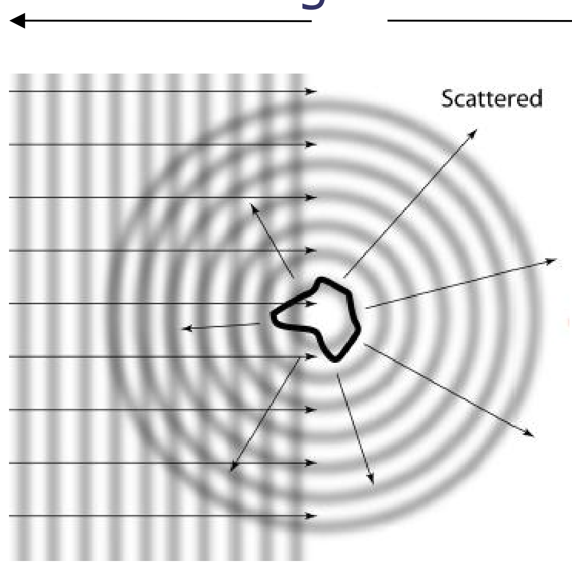
Large scales in real space 10-40000Å ⟺ Low Q, small scattering angles 0.54 $Å^{-1}$ - $6 \cdot 10^{-4}$ $Å^{-1}$

Diffractometer specialized for small scattering angles

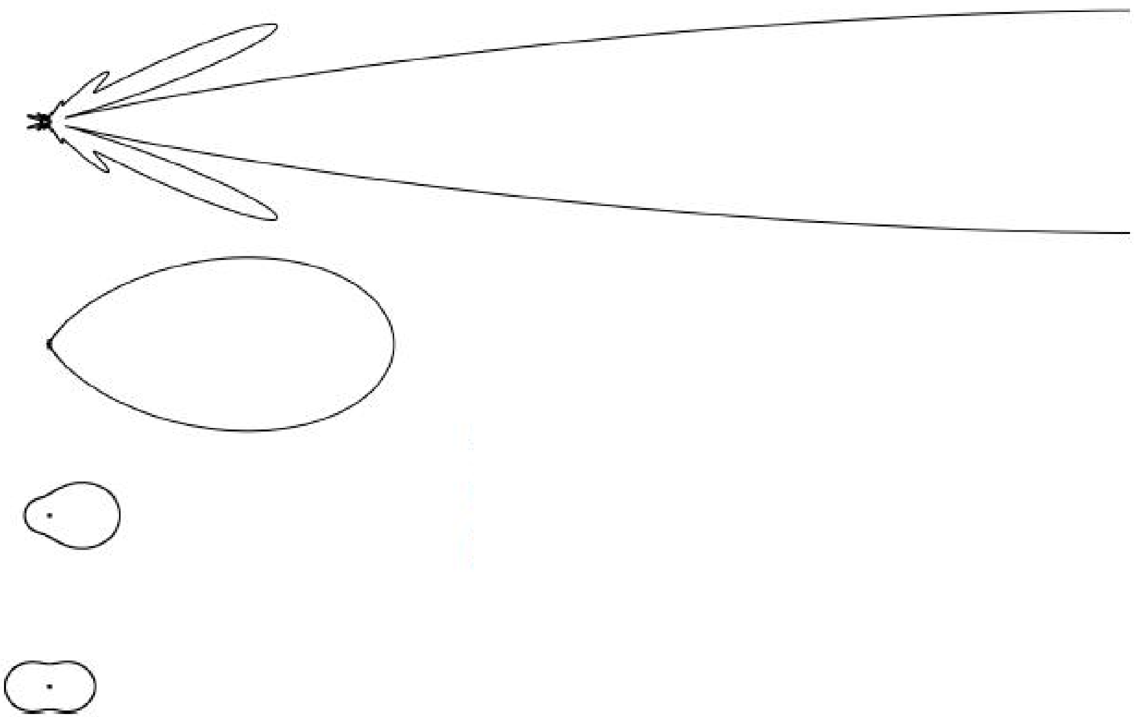



# Forward scattering-Why is it useful?

Backscattering Forward scattering




Phase function for different x




$$x = \frac{2\pi r}{\lambda}$$

Lots of intensity scattered in forward direction

# Scattering length density



Starting point: coherent elastic cross section

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \int_{\infty}^{\infty} \frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'} \mathrm{d}(\hbar\omega) = \sum_{j,j'} b_j b_{j'} \left\langle e^{-iqr_{j'}} e^{-iqr_j} \right\rangle$$

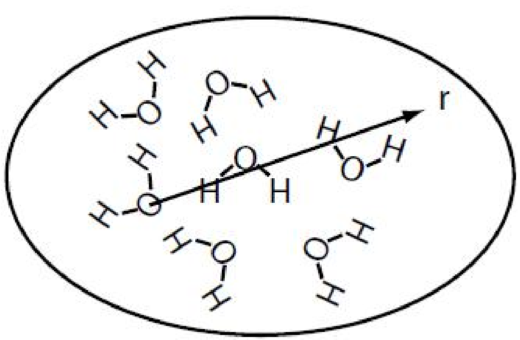
Sum over identical atoms $\frac{d\sigma}{d\Omega}(\mathbf{q}) = \frac{1}{N} \left| \sum_i^N b_i e^{i\mathbf{q}\cdot\mathbf{r}} \right|^2$

➡ SANS: low q averages over large r

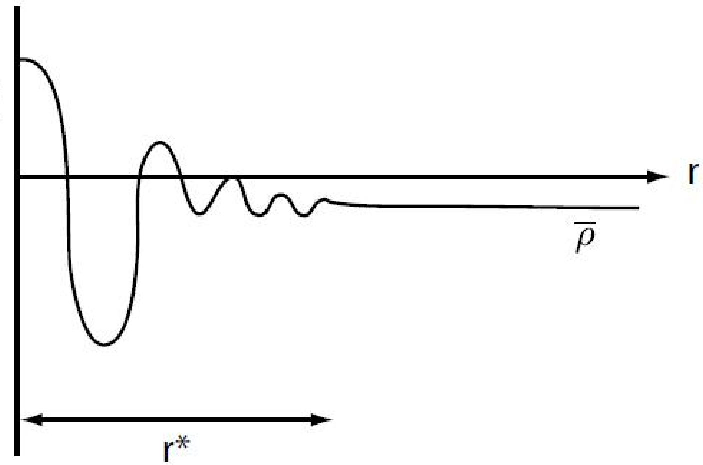
Scattering length b ➡ Scattering length density $\rho = \frac{\sum_i^n b_i}{V}$

$$\rho(\mathbf{r}) = b_i \delta(\mathbf{r} - \mathbf{r_i})$$

Example: water




$$\rho = \frac{\sum_i b_i}{\overline{v}}$$



# Principle of Babinet



Insert scattering length density into coherent elastic cross section:

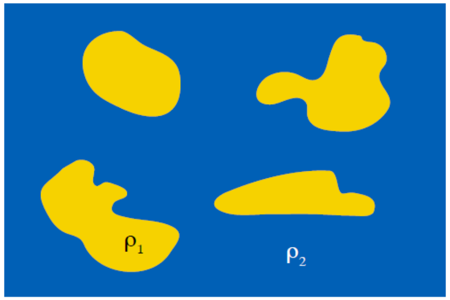
Rayleigh-Gans Equation $$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{N}{V}\frac{d\sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}\left|\int_V \rho(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r}\right|^2$$

⇨ SANS measures inhomogeneities of scattering length density

Assume a general two phase system

$$V = V_1 + V_2$$

$$\rho(r) = \begin{cases} \rho_1 & \text{in } V_1 \\ \rho_2 & \text{in } V_2 \end{cases}$$




Split up the integral over the sample, break up into the two subvolumes

$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}\left|\int_{V_1} \rho_1 e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1} + \int_{V_2} \rho_2 e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_2}\right|^2$$

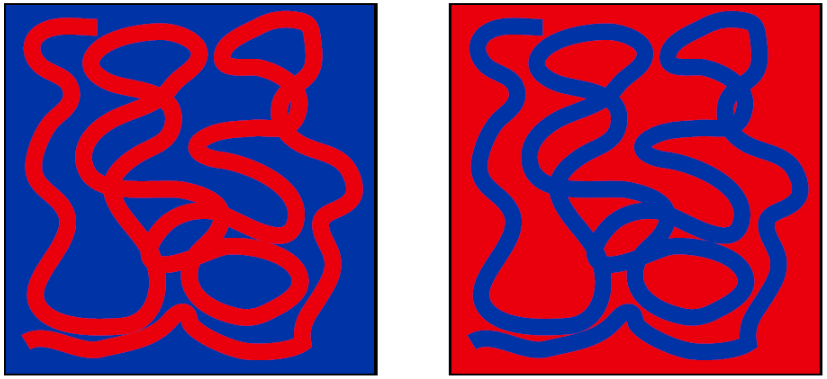
$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}\left|\int_{V_1} \rho_1 e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1} + \rho_2\left\{\int_V e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} - \int_{V_1} e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1}\right\}\right|^2$$

⇨ $$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}(\rho_1 - \rho_2)^2\left|\int_{V_1} e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1}\right|^2$$

# Principle of Babinet

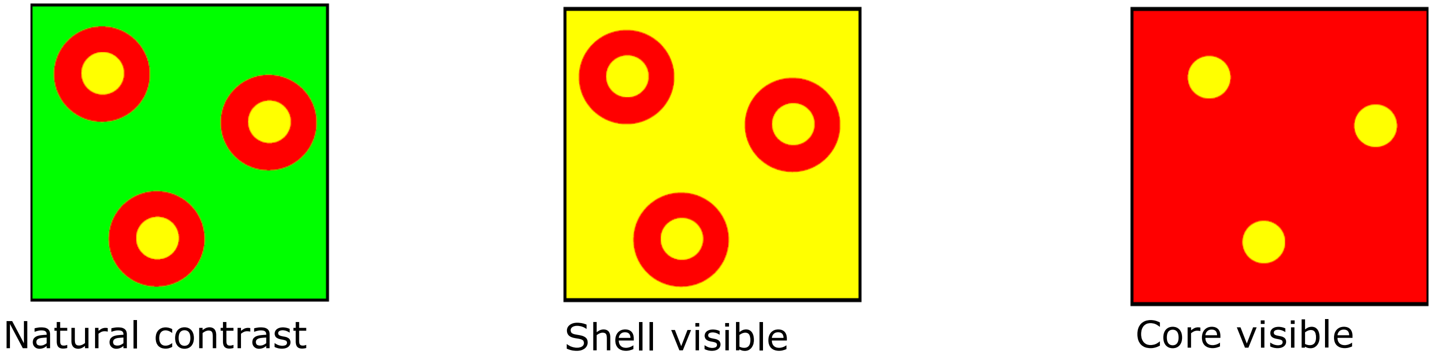


$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}(\rho_1 - \rho_2)^2 \left| \int_{V_1} e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1} \right|^2$$



Principle of Babinet: Same coherent scattering

Contrast variation and contrast matching



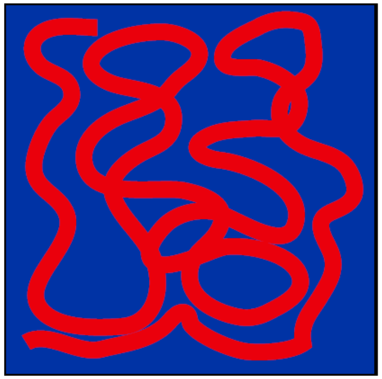

Often: Mixture of H2O/D2O, isotope variation

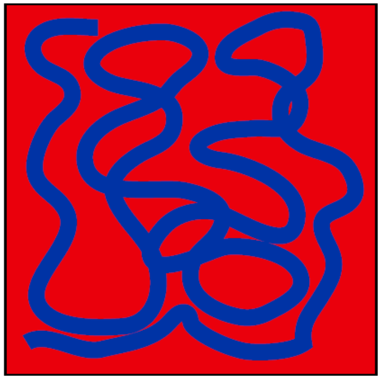
FRM II Forschungs-Neutronenquelle Heinz Maier-Leibnitz

# Structure & Form Factor

MLZ Heinz Maier-Leibnitz Zentrum

$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}(\rho_1 - \rho_2)^2 \left| \int_{V_1} e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1} \right|^2$$





Split up the integral over the sample

$$\frac{d\Sigma}{d\Omega}(q) = \frac{N}{V}(\rho_1 - \rho_2)^2 V_p^2 P(q) S(q)$$

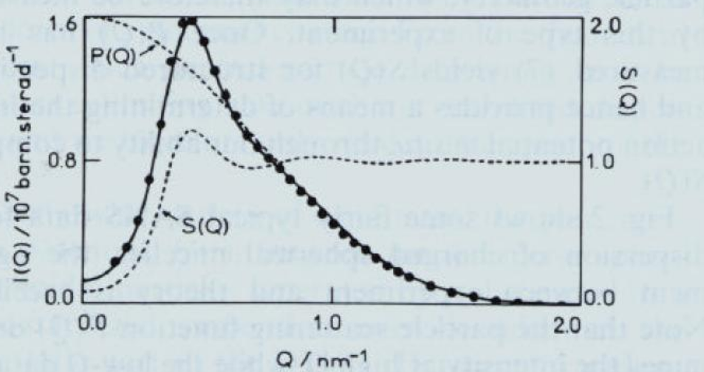



Fig. 2. Observed (●) and calculated (——) scattered intensity $I(Q)$ as a function of momentum transfer $Q$ for a charged micellar dispersion: 0·03 mol dm$^{-3}$ hexadecyltrimethylammonium chloride in $D_2O$ at 313 K. The functions $P(Q)$ and $S(Q)$ are discussed in the text. (1 barn sterad$^{-1}$ = $10^{-28}$ $M^2$ sterad$^{-1}$).

**Form factor P(Q)**

Interference of neutrons scattered at the same object

Shape, surface and densitiy distribution of objects

**Structure factor S(Q)**

Interference of neutrons scattered from different objects

Arrangement or superstructure of objects

Remember: Bragg scattering and Debye Waller

# Form Factor



Form factor P(Q)

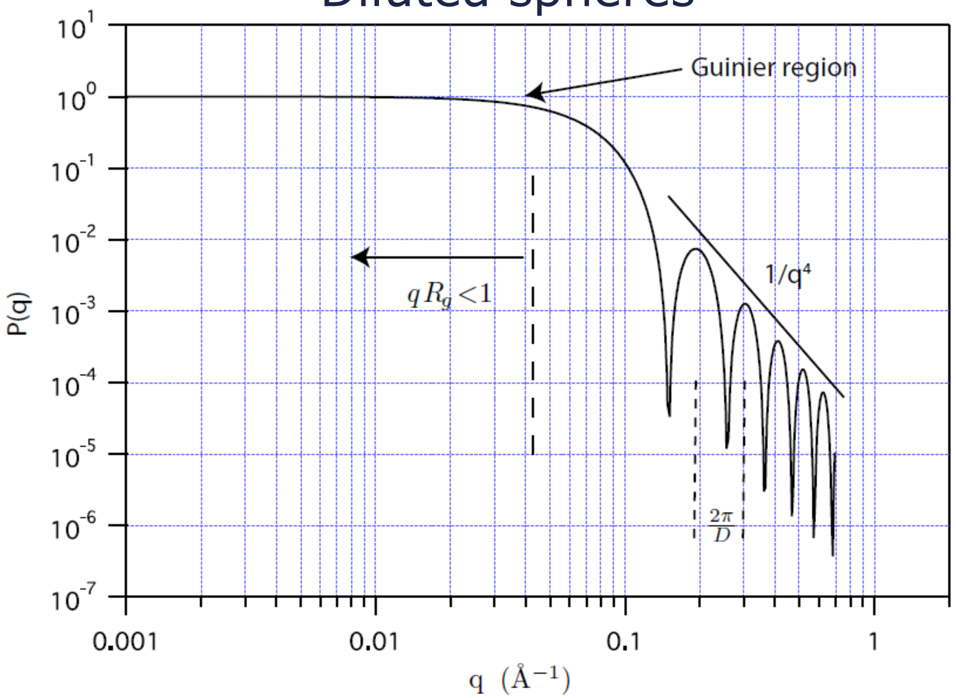
Interference of neutrons scattered at the same object

For isotropic solutions:

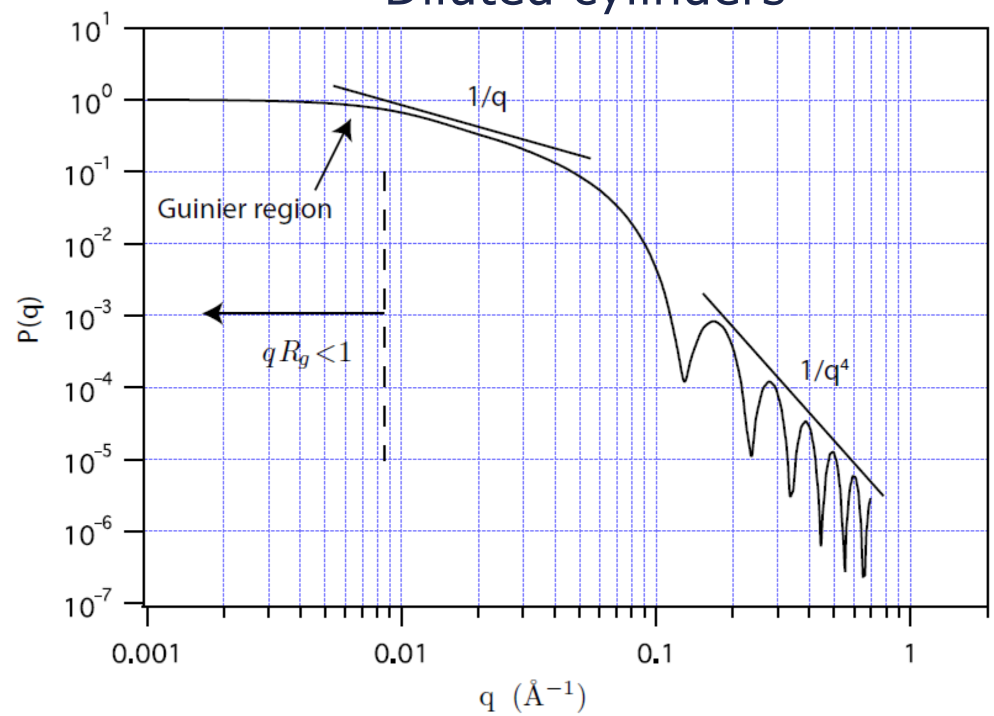
$$S(q) = 1 + 4\pi N_p \int_0^\infty [g(r) - 1] \frac{\sin(qr)}{qr} r^2 dr$$

g(r) pair correlation function

Diluted spheres




Diluted cylinders

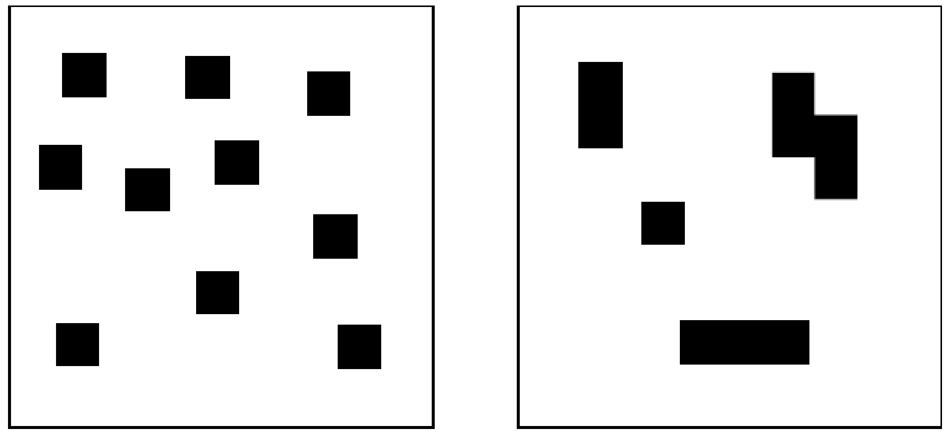



More about that in the tutorial!

# Scattering invariants



Two samples with 10% white and 90% black



Integrate with respect to Q

$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{1}{V}(\rho_1 - \rho_2)^2 \left| \int_{V_1} e^{i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r_1} \right|^2$$

For a two phase system

$$\frac{Q}{4\pi} = Q^* = 2\pi^2 \phi_1 (1 - \phi_1)(\rho_2 - \rho_1)^2$$

Scattering invariant, total small angle scattering is constant!

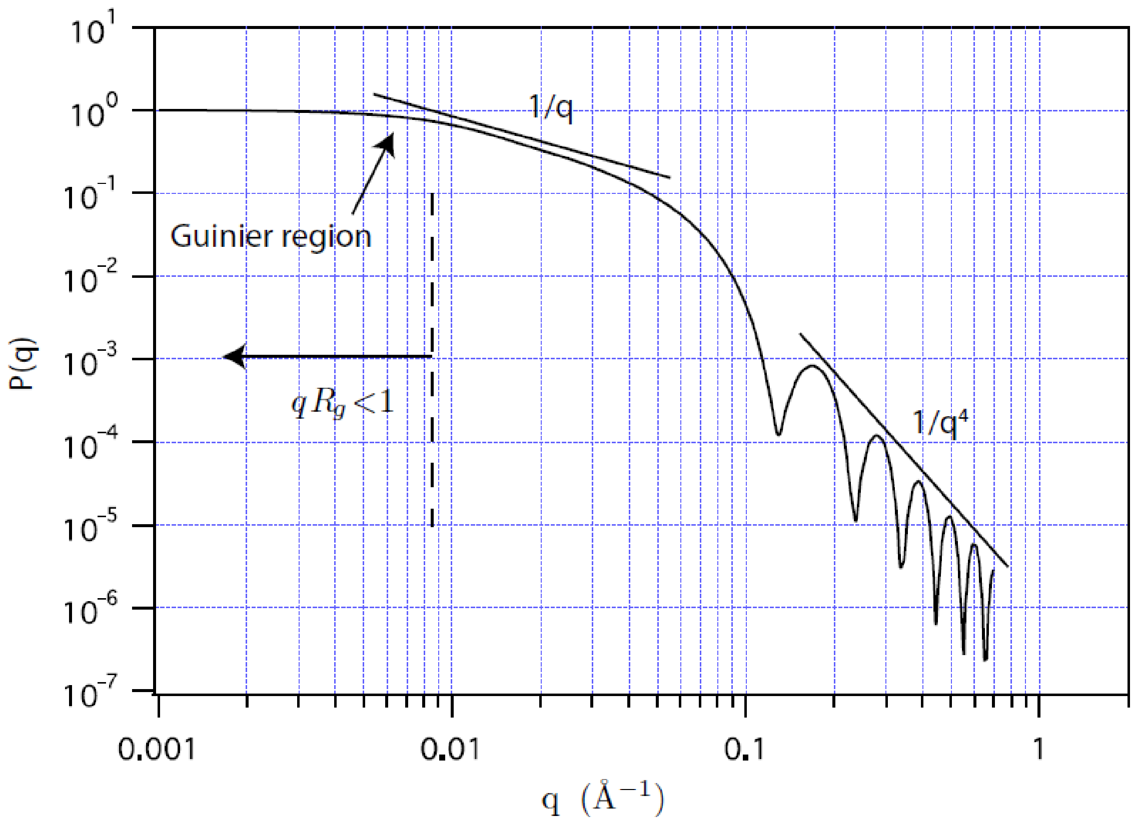
# Porod and Guinier regime

Porod scattering for smooth surfaces and
$Q \gg 1/D$

$$I(q) \propto (q)^{-4}$$

$$\frac{\pi}{Q^{\star}} \cdot \lim_{q \to \infty} (I(q) \cdot q^4) = \frac{S}{V}$$

Form factor for diluted cylinders radius 30Å, length 400Å
No structure factor!

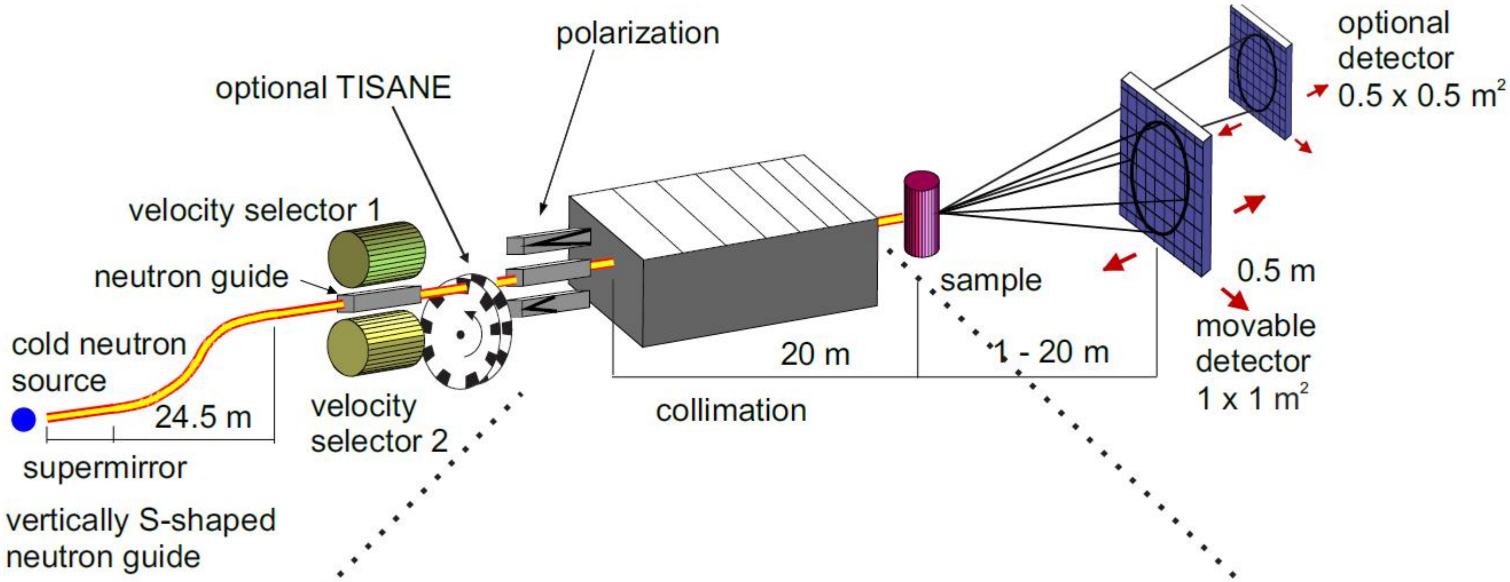


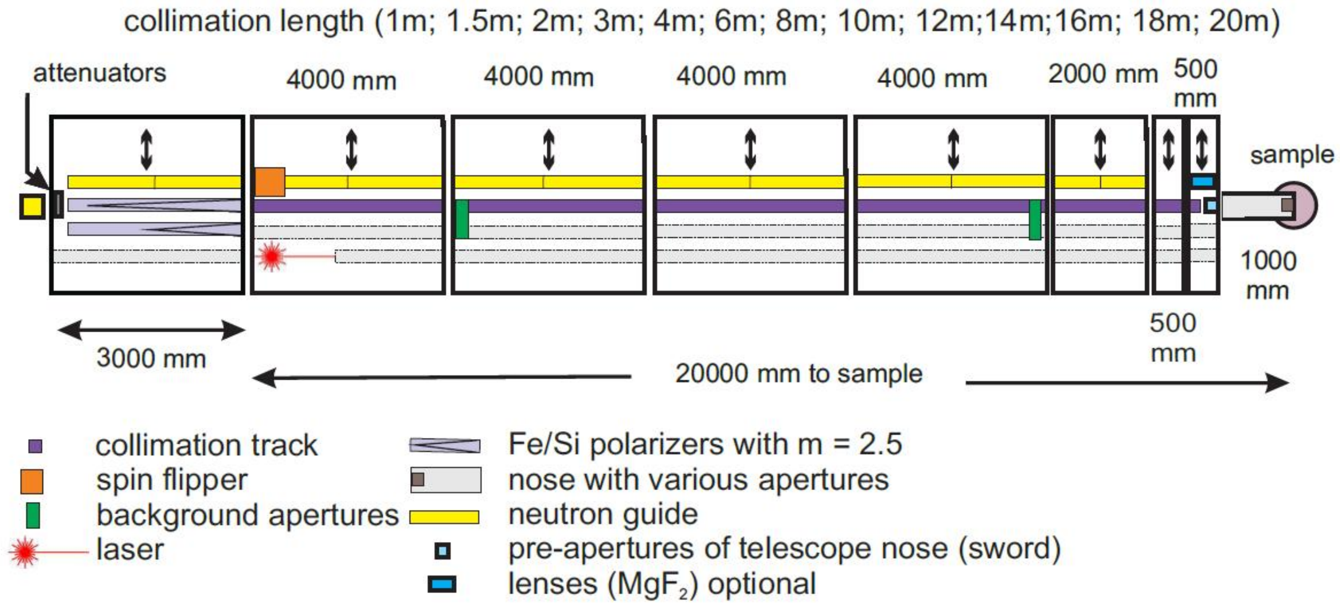

Guinier scattering for dilute, monodisperse and isotropic solutions of particles:
$QR_G \ll 1$

$$I(q) = I(0) e^{\frac{-(qR_g)^2}{3}}$$

# SANS – Instrumentation

# Elements of a SANS Instrument

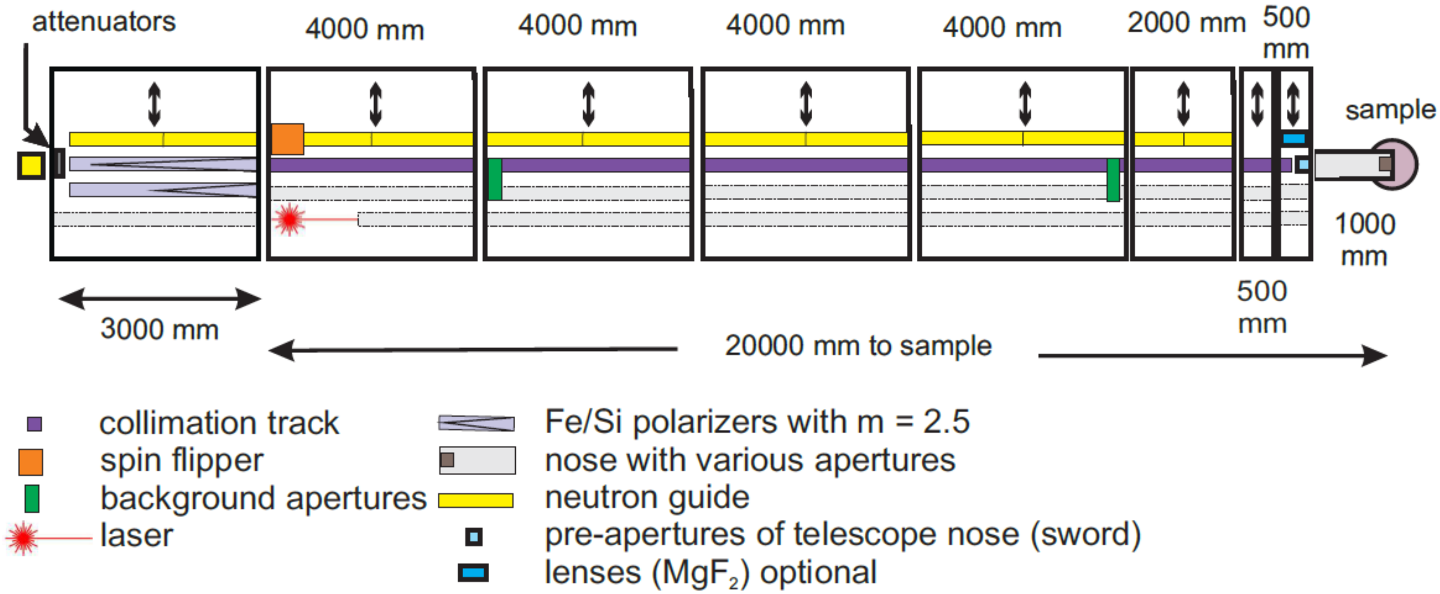


# Collimation section



Velocity selector




Collimation: Define resolution and intensity

Aperture system/neutron guides (supermirror)

Alignment extremely critical

Well-defined and homogenous wavelength /divergence profiles

Transmission polarizer for the use of polarized neutrons

Parasitic background scattering has to be avoided (whole collimation system is evacuated, edge scattering, incoherent background)

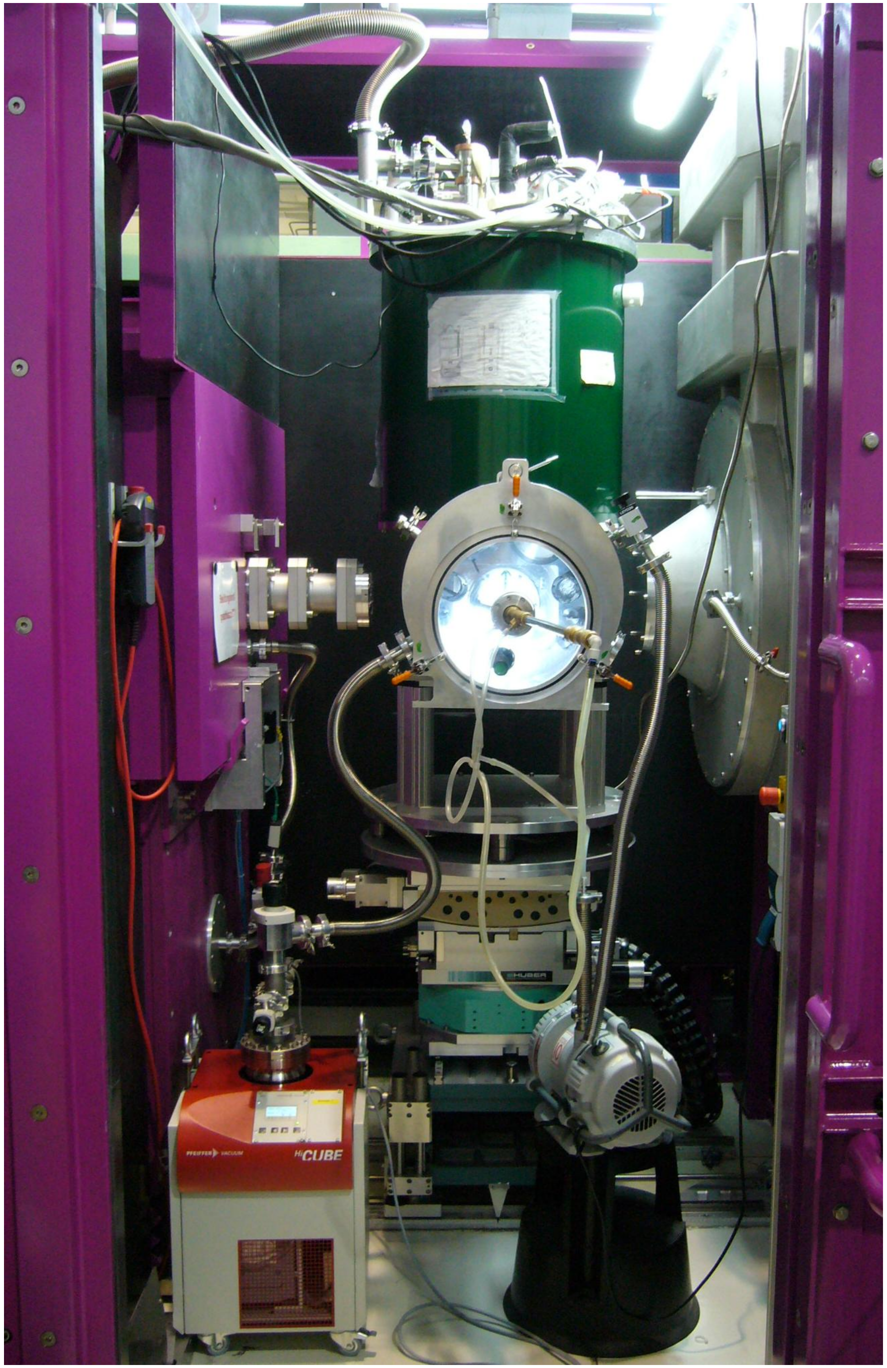
# Sample stage



Provide necessary sample environment (similar to any other neutron diffractometer or spectrometer)

Parasitic background scattering has to be avoided (extremely critical!)

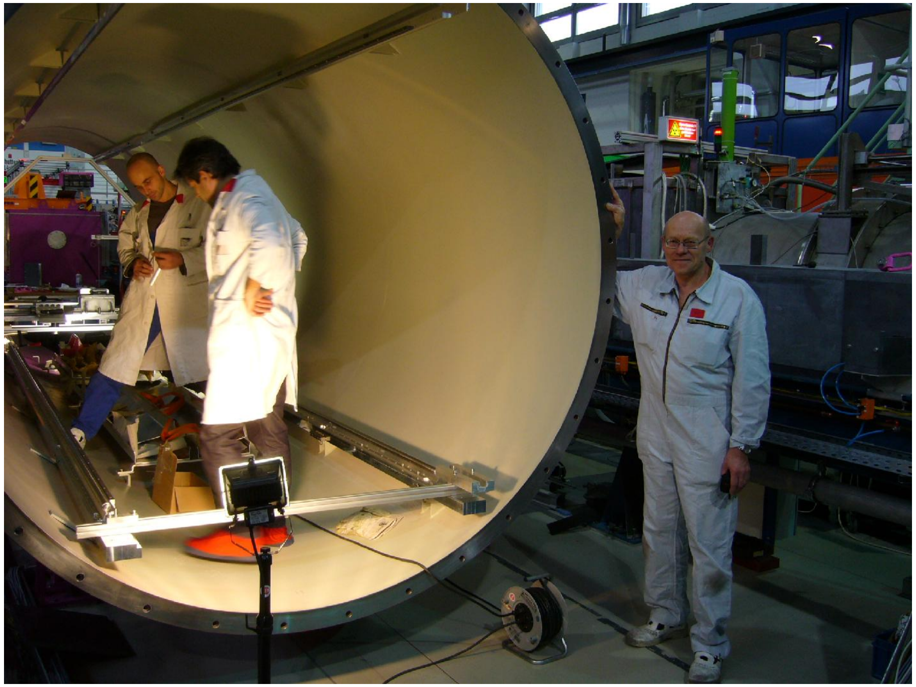
- Minimize neutrons travelling in air (few cm can be too much)
- Avoid Aluminum neutron windows (single crystalline sapphire is better)
- Get rid of scattering at edges (use conical slits)

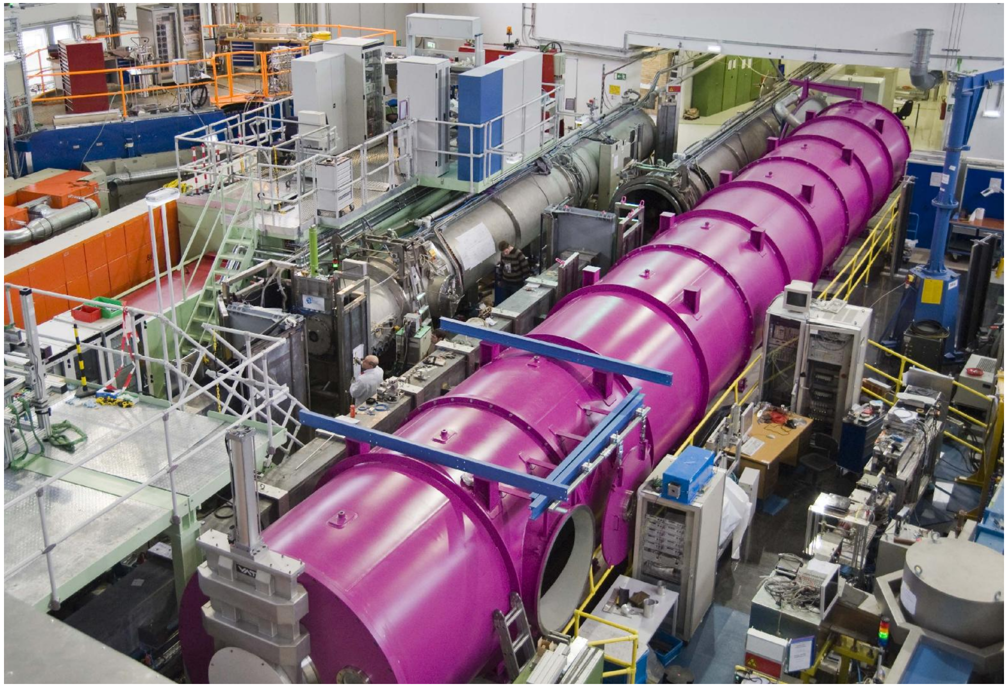


# Detector tube



Vacuum vessel for detector to provide lowest possible background





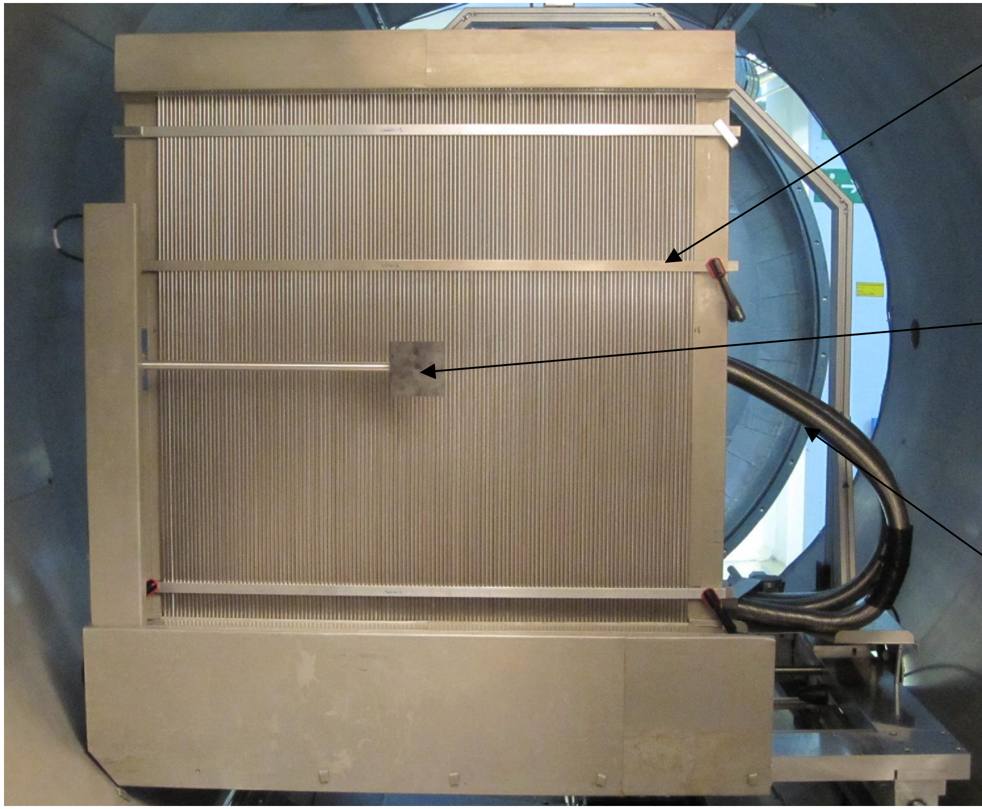
Sample detector length adjustable (select Q-range)

One (or several) $He^3$ position sensitive detectors (typical $1m^2$ with 5mm resolution)

Typical length 10-40m

Interior completely covered with neutron absorbing Cadmium

# Detector

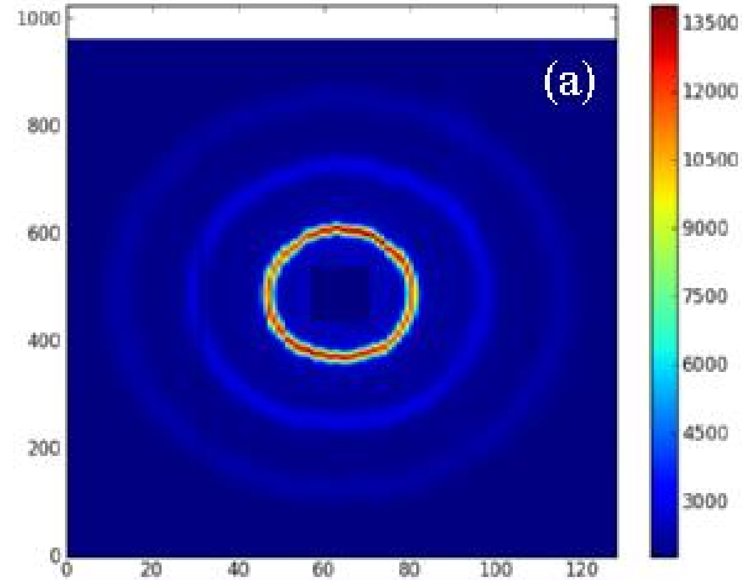


Cd strips for calibration

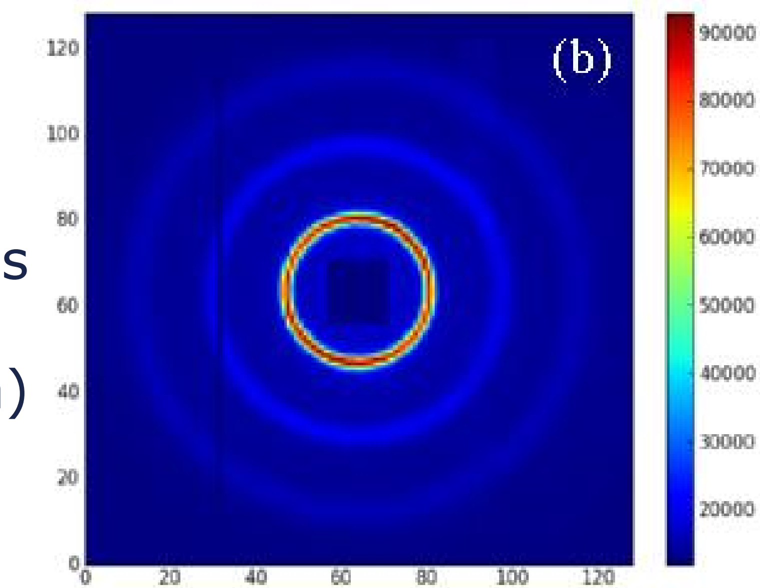
Beam stop

Air cooling for detector electronics

Raw data




Position calibration

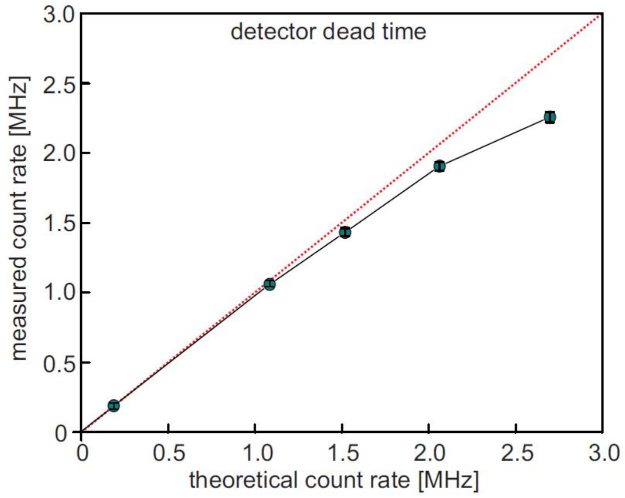



Array of 128 position sensitive $^3$He Reuter Stokes tube detectors
8mm x 8mm position resolution (charge division)
Detector distance from 1-20m, moves on rails
Maximal count rate 4MHz

# Detector corrections

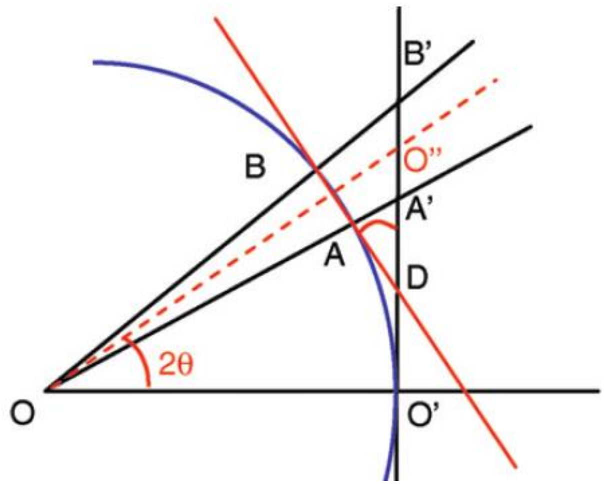


## Dead time corrections



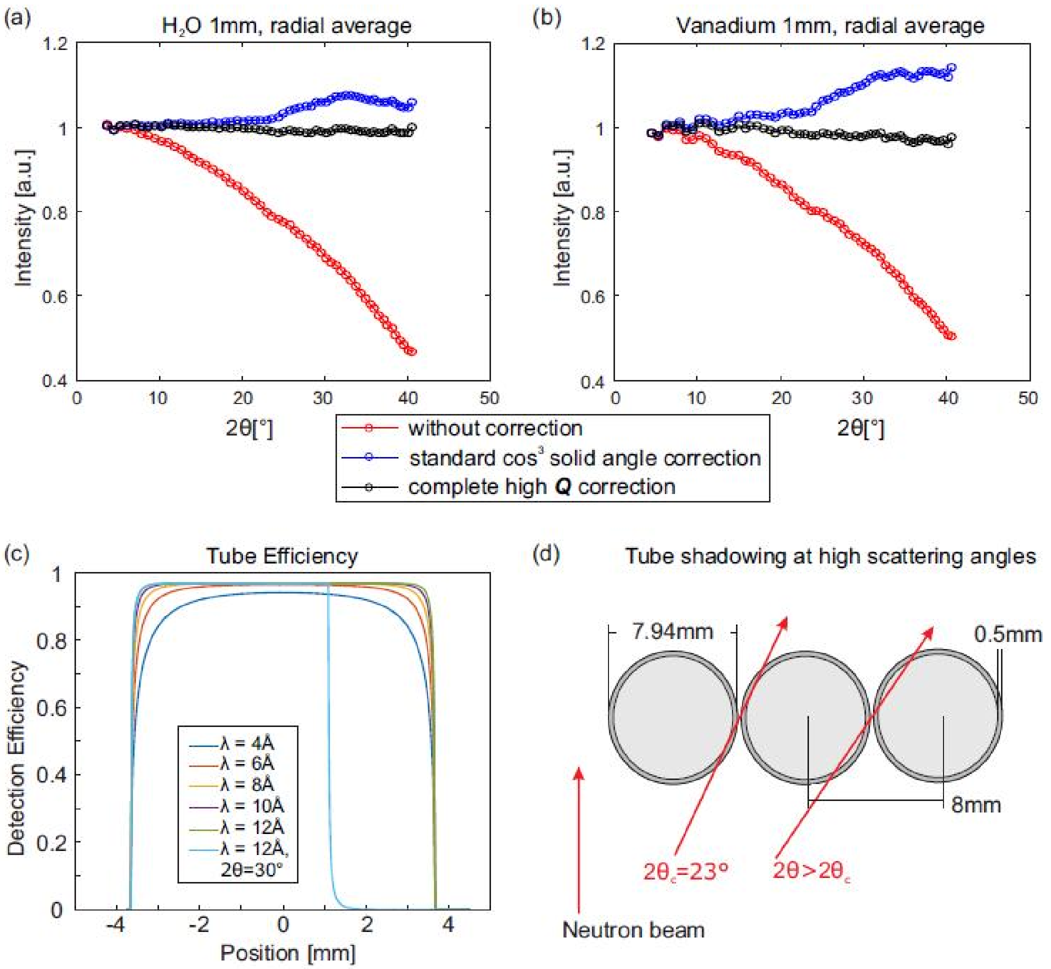

## Solid angle correction

$$\Delta\sigma(2\Theta) = \frac{p_x p_y \cos(2\Theta)^3}{D(2\Theta = 0)}$$



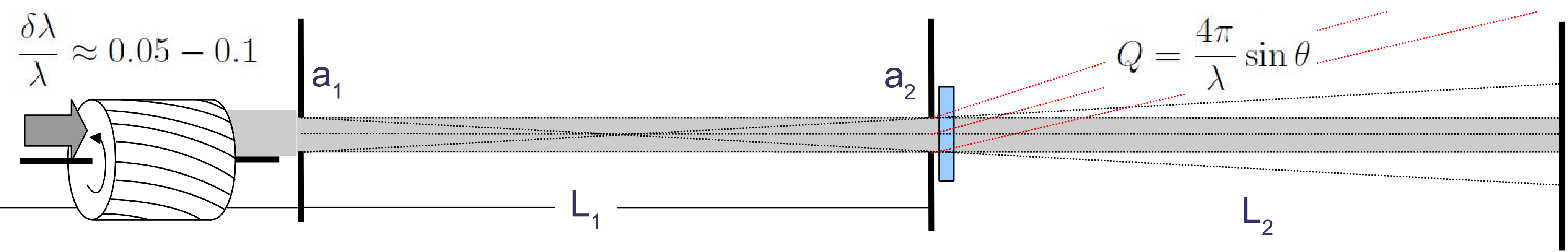

## Anisotropic solid angle correction for tube array detector

$$\Delta\sigma(2\Theta) = \frac{p_x p_y \cos(\Theta_x)\cos(2\Theta)^2}{D(2\Theta = 0)}$$



# SANS – Resolution & Intensity

# SANS – Resolution




- Angular resolution
- Monochromaticity
- Detector resolution
- Gravity

} Treat as Gaussian distributions: $\left\langle \frac{\delta Q^2}{Q^2} \right\rangle = \left\langle \frac{\delta\lambda^2}{\lambda^2} \right\rangle + \left\langle \frac{\cos^2\theta\,\delta\theta^2}{\sin^2\theta} \right\rangle$

$$\left\langle \frac{\delta Q^2}{Q^2} \right\rangle = 0.0025 + \left\langle \frac{\delta\theta^2}{\theta^2} \right\rangle$$

⇒ Angular resolution: $\delta\theta \approx \sqrt{\frac{5}{12}\frac{a}{L}}$

What is the largest object SANS can detect (limit small Q)?

For large scattering angles (large Q) wavelength resolution dominates.

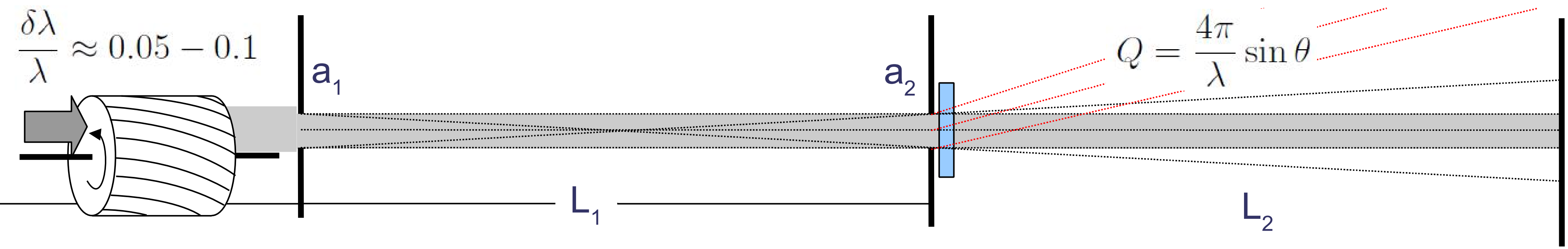
$$a_1 = a_2 = a \qquad L_1 = L_2 = L$$

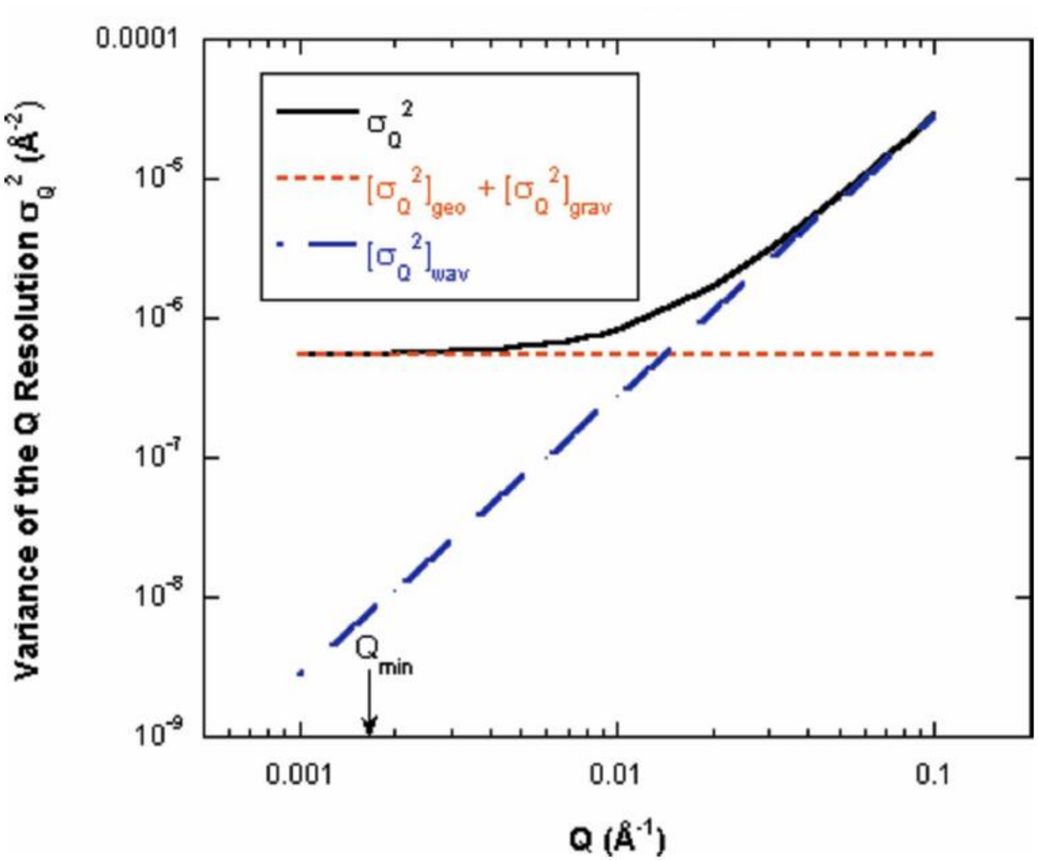
$$\Rightarrow \delta Q \approx \frac{\delta\theta}{\theta_{min}} Q_{min} \approx \delta\theta \frac{4\pi}{\lambda} \approx \frac{2\pi a}{\lambda L}$$

$$\frac{2\pi}{\delta Q} = \frac{\lambda L}{a}$$

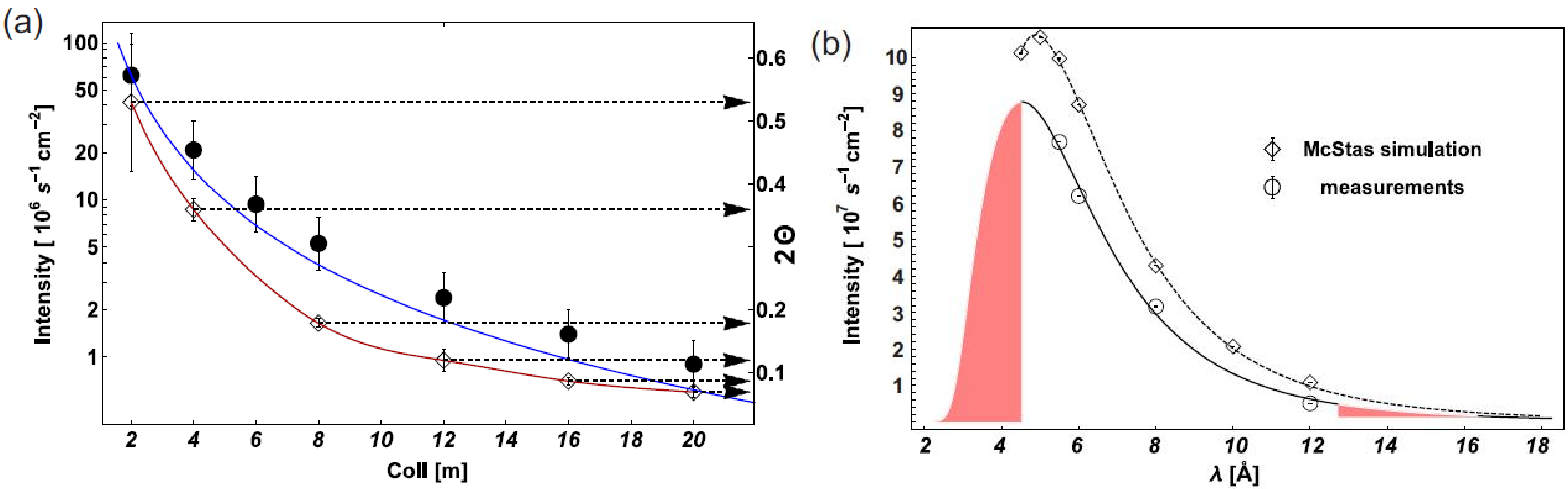
On D11, ILL: L=40m, λ=15Å ⇒ D≈ 5μm

# SANS – Resolution



# SANS – Intensity & Resolution

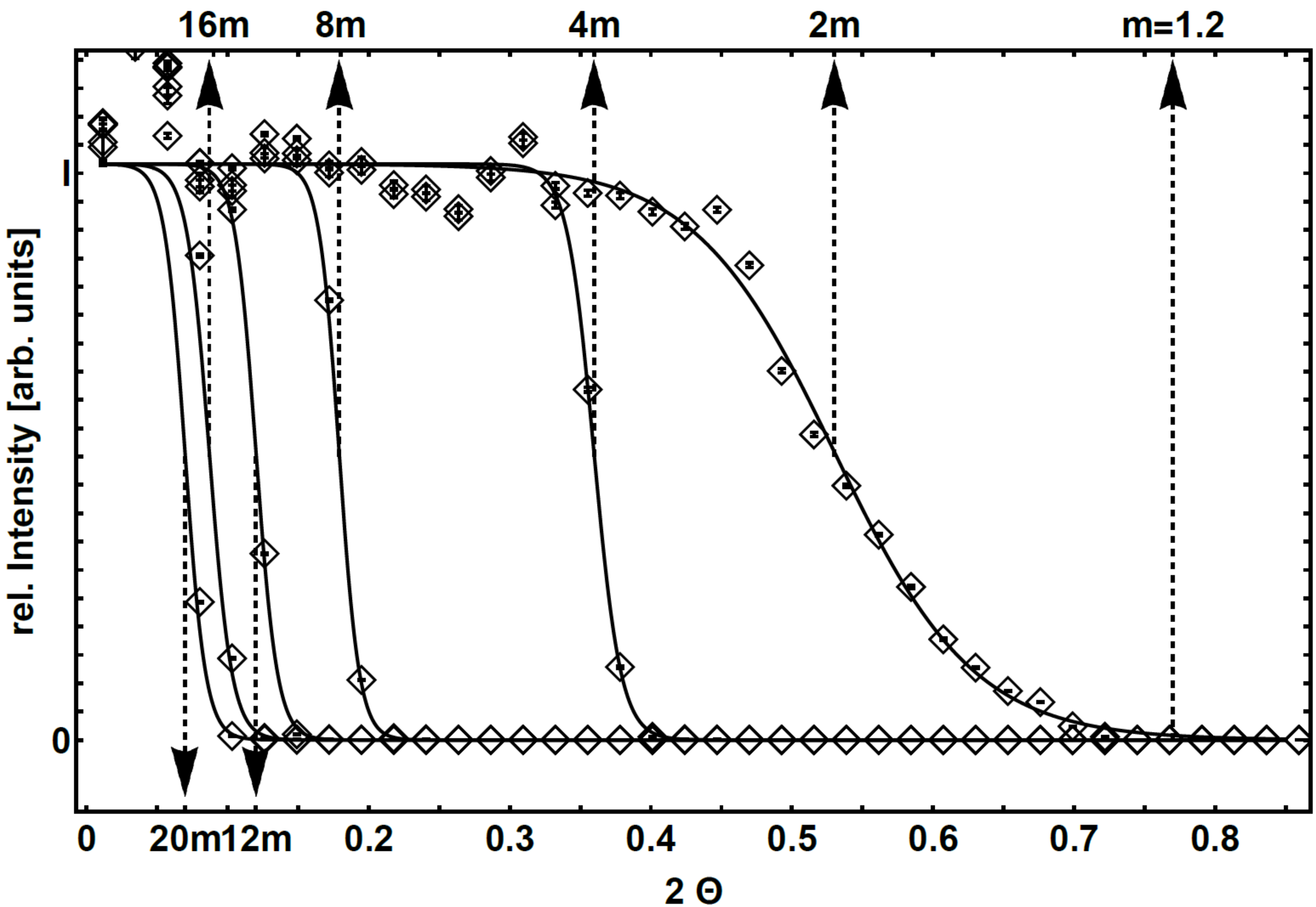



Intensity: Quadratic decrease with source to sample distance (collimation length)

Wavelength: Decrease of intensity with $\lambda^{-4}$

$$\delta Q \approx \frac{\delta\theta}{\theta_{min}} Q_{min} \approx \delta\theta \frac{4\pi}{\lambda} \approx \frac{2\pi a}{\lambda L}$$

# SANS – Intensity & Resolution

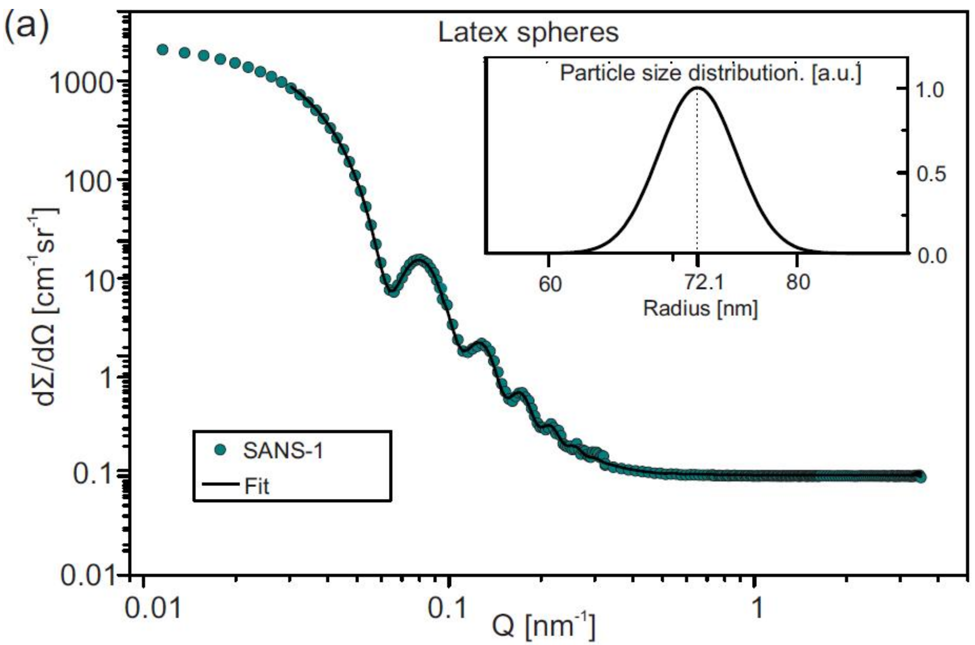


# Detector corrections



Typical SANS dataset:

- Sample (at different L)
- Water (absolute scale)
- Empty sample holder/cuvette
- Background




$$\left(\frac{d\Sigma}{d\Omega}\right)_{\text{sample}} = \frac{1}{F_{\text{sc}}}\left(\frac{d\Sigma}{d\Omega}\right)^{\text{real}}_{\text{H}_2\text{O}} \frac{\left[\frac{I_{\text{sample}} - I_{\text{B4C}}}{Tr_{\text{sample}}} - \frac{I_{\text{sample-EC}} - I_{\text{B4C}}}{Tr_{\text{sample-EC}}}\right]\frac{1}{e_{\text{sample}}}}{\left[\frac{I_{\text{H}_2\text{O}} - I_{\text{B4C}}}{Tr_{\text{H}_2\text{O}}} - \frac{I_{\text{H}_2\text{O-EC}} - I_{\text{B4C}}}{Tr_{\text{H}_2\text{O-EC}}}\right]\frac{1}{e_{\text{H}_2\text{O}}}}$$

➡ Fit model of the sample (conv. with resolution to the dataset)

➡ SANS tells you:

- Shape of scattering object
- Size(distribution) of scattering objects
- Surface of scattering objects
- Scattering length density distribution
- Arrangement (Superstructure?)

# SANS – applications

Soft matter
Hard matter
Magnetism

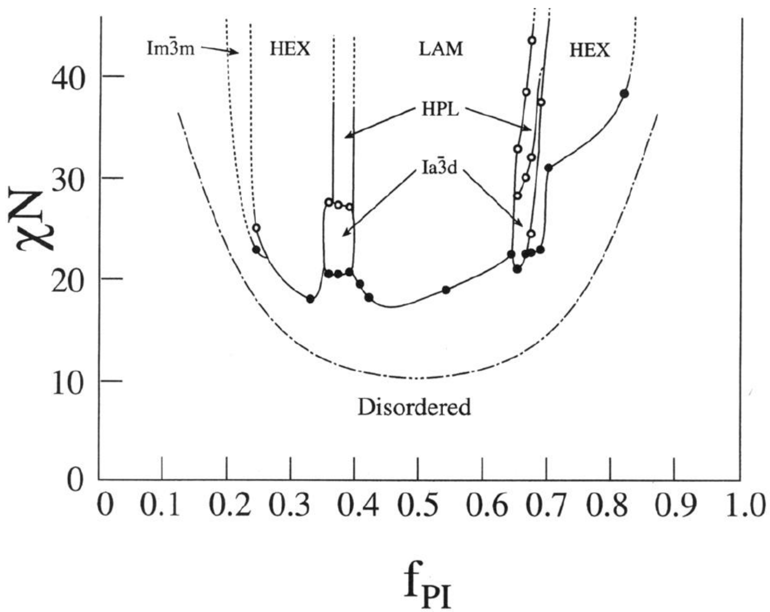
# Applications of SANS



## Polyisoprene – Polystyrene Diblock Copolymer Phase Diagram

Two (or more) homopolymers units linked by covalent bonds
Microphase separation: Complex nanostructures phases

- Flory-Huggins segment-segment interaction
- Degree of polymerization
- Volume fraction





SANS pattern:
Aligned by shear

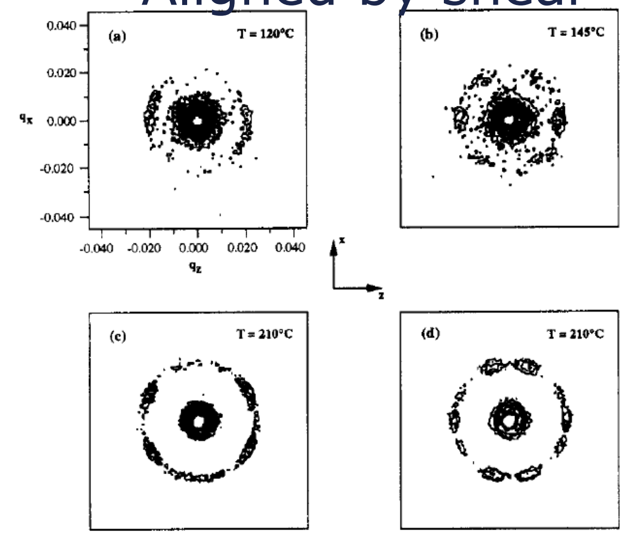



**Figure 10.** Contour plots of SANS patterns for the sample IS-68 in three different ordered states (A, D, B). A nearly featureless pattern (a) was observed in state A (120 °C), which is attributed to a parallel orientation of lamellae with respect to the shear plane. After heating the sample to 145 °C (state D) and application of dynamic shearing ($\dot{\gamma} = 0.1\ s^{-1}$ with $|\gamma| =$ 300%), a weak hexagonal scattering pattern was observed (b). This is consistent with a hexagonal in-plane arrangement of perforations in the minority (PS) layers. Further heating the sample to 210 °C, without shear (state B), produced a predominantly four-peak pattern (c), which transformed into result (d) when a shear rate of 2.2 $s^{-1}$ was applied. The azimuthal relationship between the combined 10 reflections in (c) and (d) is consistent with the SANS data reported for sample IS-39 (ref 22) and the $Ia\bar{3}d$ space group symmetry.

Macromolecules, Vol. 28, No. 26, (1995)
Physics Today, p. 32, Feb. (1999)

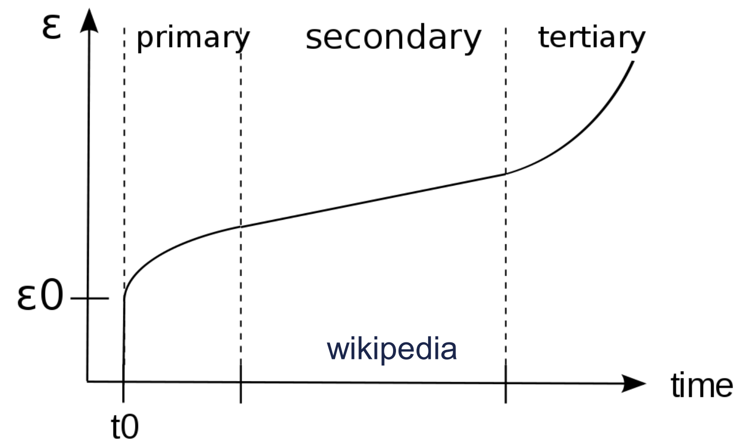
# Applications of SANS



Creep cavitation damage in steel at high T

Volume fraction and size distribution of cavities can be measured with SANS and USANS.

Grain boundary cavitation dominant failure mode.



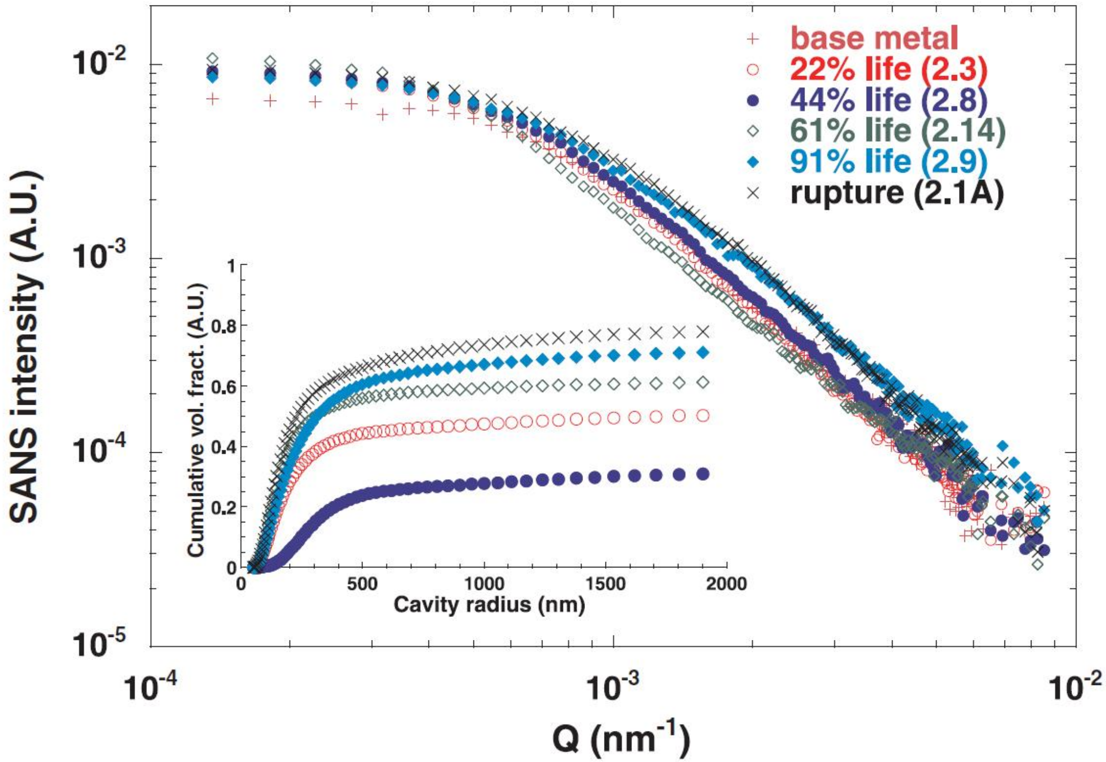
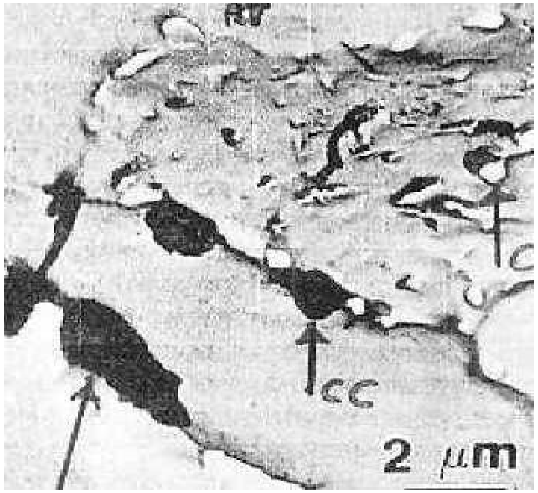
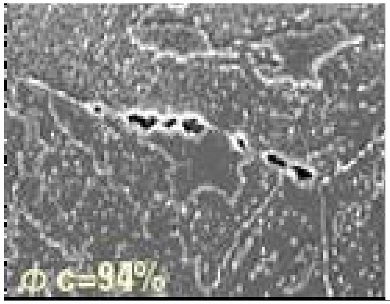
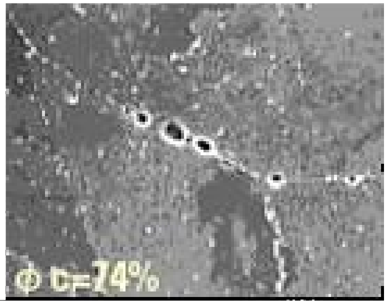




**Fig. 3.** SANS intensity for the investigated specimens, measured in the double-crystal experiment at HMI-BENSC (*Inset*: Cumulative cavity volume fraction as a function of cavity radius)



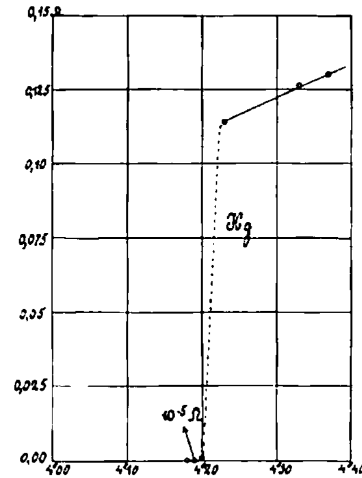



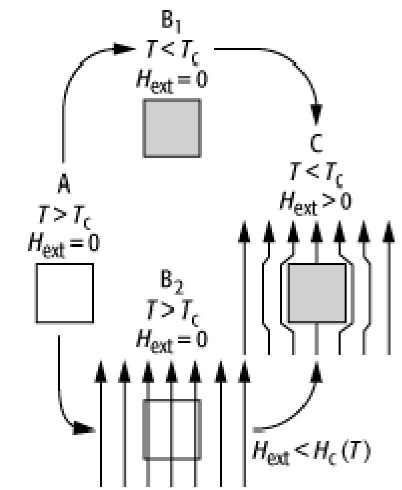



# Applications of SANS



## Superconductivity (H. Kamerlingh Onnes 1911)






- Total loss of resistivity
- Expulsion of magnetic fields (Meissner Ochsenfeld Effect)

Two length scales:

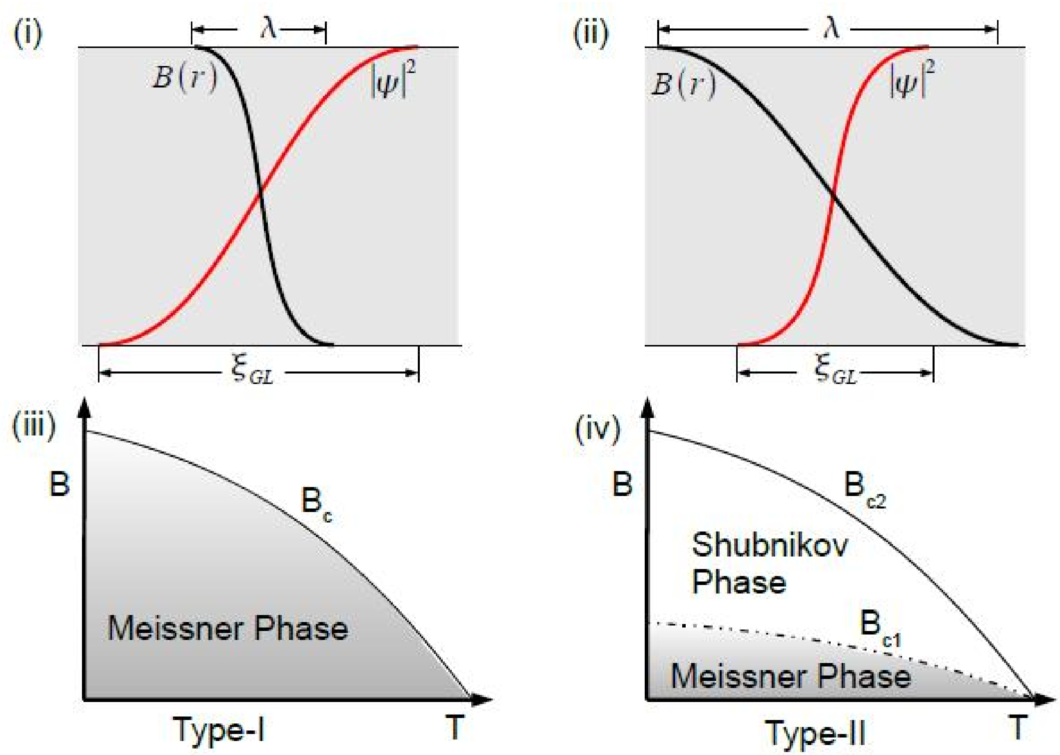
Penetration depth

$$\lambda^2(T) = \frac{m^* c^2}{4\pi n_s e^{*2}}$$

Coherence length

$$\xi^2(T) = \frac{\hbar^2}{2m^*|\alpha(T)|} \propto \frac{1}{1-\frac{T}{T_c}}$$

- Interface energy

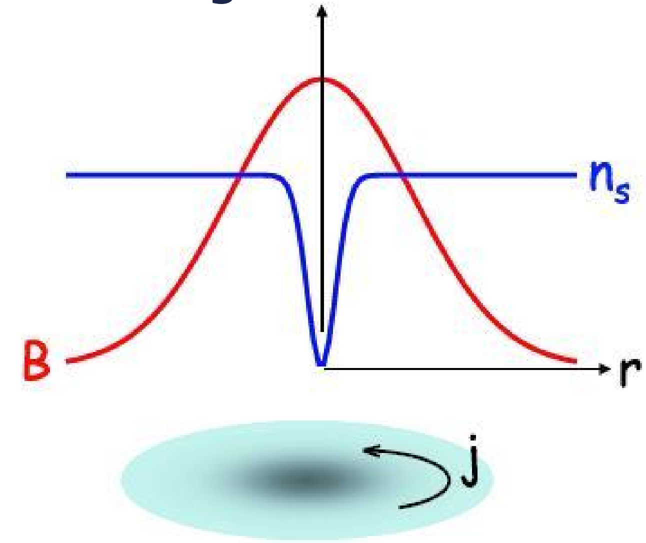


# Applications of SANS



Stabilized by negative energy of super-/normal conducting interface.

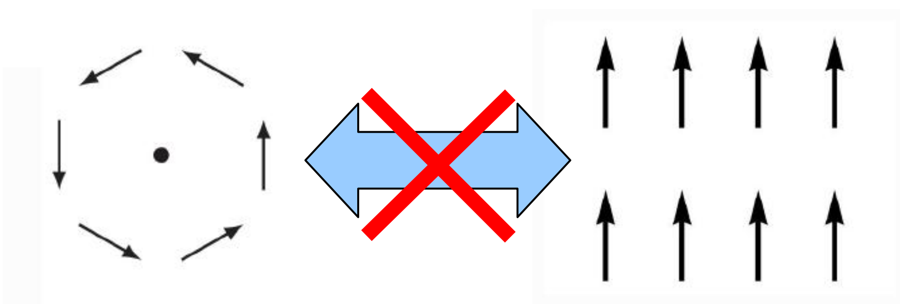
⇨ Quantization of magnetic flux $\phi_0 = \frac{h}{2e}$




Now consider the topology!

Superconducting vortex: Topological defect of the superconducting OP.

No continuous transformation from no vortex to a vortex state.




Protected by topology: Particle-like properties

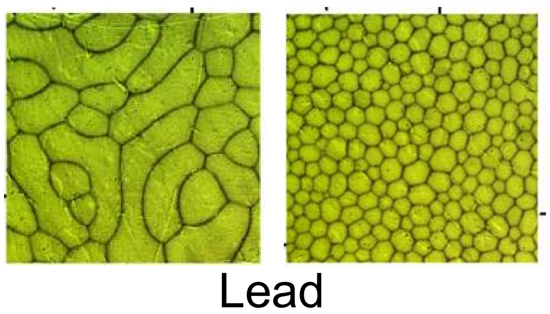
# Applications of SANS

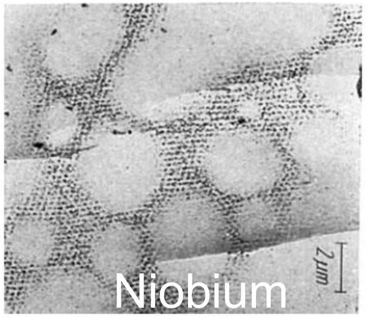


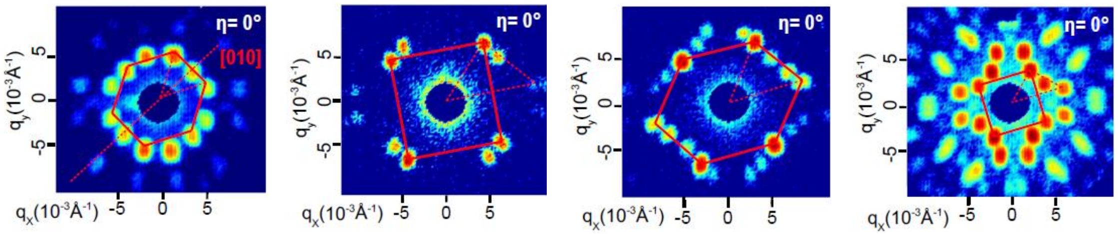
Condensed matter ⟺ superconducting vortex matter

Properties of superconducting VM reflect underlying physics
Model system for general questions

Domain structure

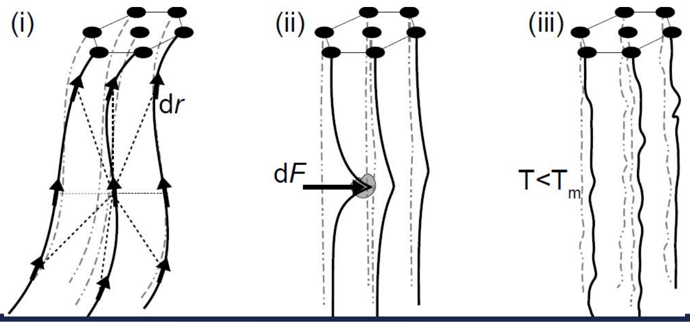



Symmetry and structure

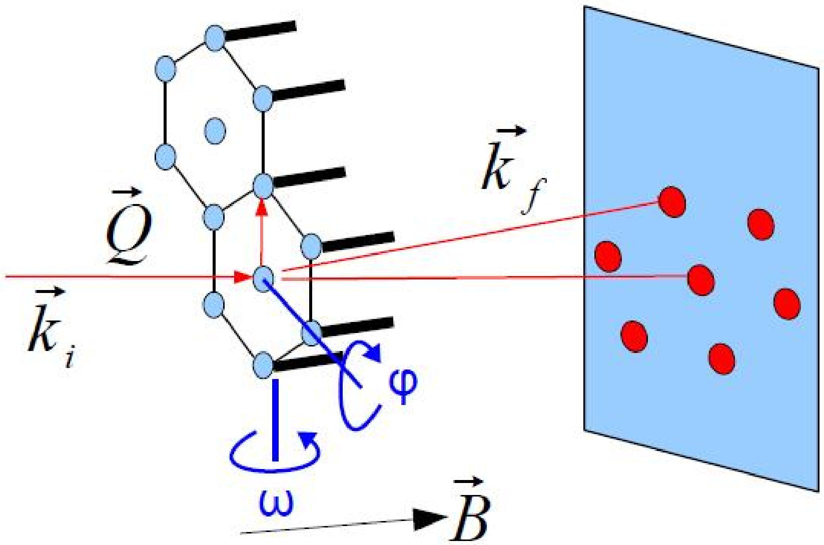
Elasticity & melting



# SANS & Vortex Lattices



Vortex lattice 2D magnetic Bravais lattice




- One flux quantum per unit cell

$$\phi_0 = \frac{h}{2e} \qquad |\vec{a}_i| = \left(\frac{2\phi_0}{\sqrt{3}B}\right)^{\frac{1}{2}} \qquad |\vec{a}_i| = \frac{2\pi}{|\vec{Q}|}$$

- Typical values:

$B$=1500 G

$A_0$=1260 Å

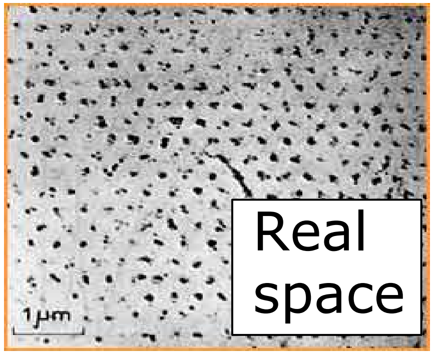
$|\vec{Q}|$=0.0057 Å$^{-1}$

Intensity Bragg peak

$$R = \frac{2\pi\gamma^2\lambda_n^2 t}{16\phi_0^2 Q}|h(Q)|^2$$

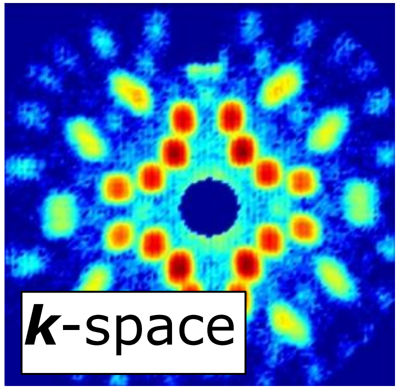
Form factor

$$h(Q) = \frac{\phi_0}{(2\pi\lambda)^2} e^{\frac{-\pi B}{B_{c2}}}$$




- Rocking gives all bragg peaks

90° rot. around the n-beam

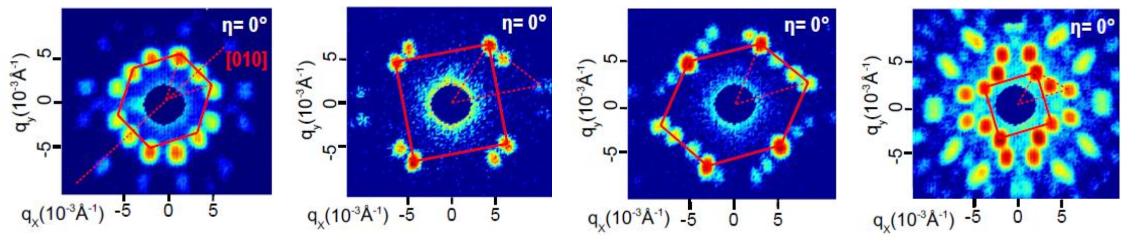


# SANS & Vortex Lattices



## Symmetry and structure

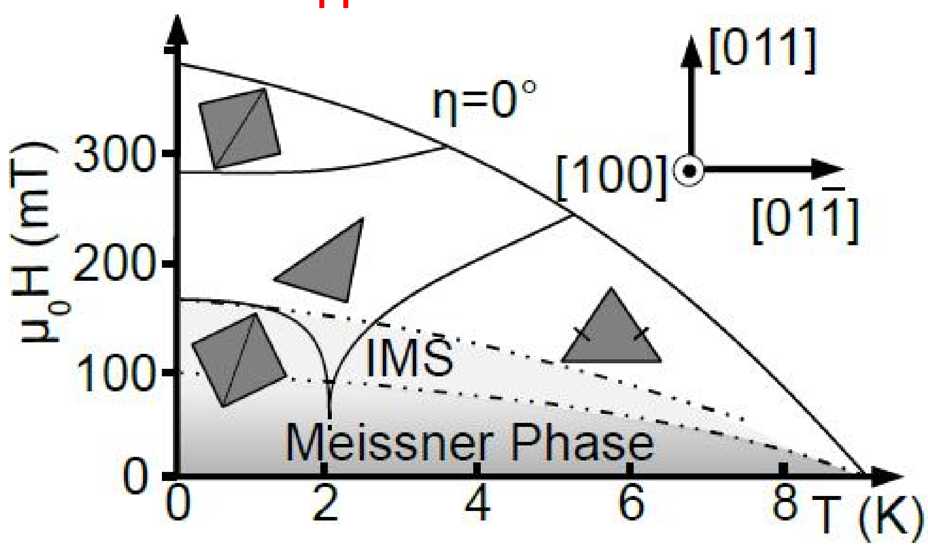
Nature of OP, symmetry of Fermi surface. Intricate to separate different influences.



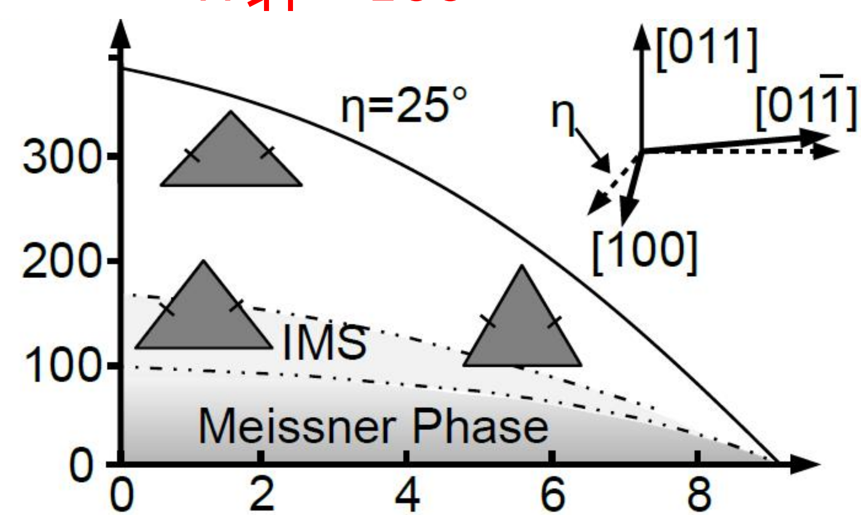

S. Mühlbauer et al. *Phys. Rev. Lett.* 102, 136408 (2009)

Six-fold symmetry of vortex lattice ⟷ **Frustration** ⟷ Four fold crystal symmetry ⇒ Four VL phases, break crystal symmetry!

H || <100>



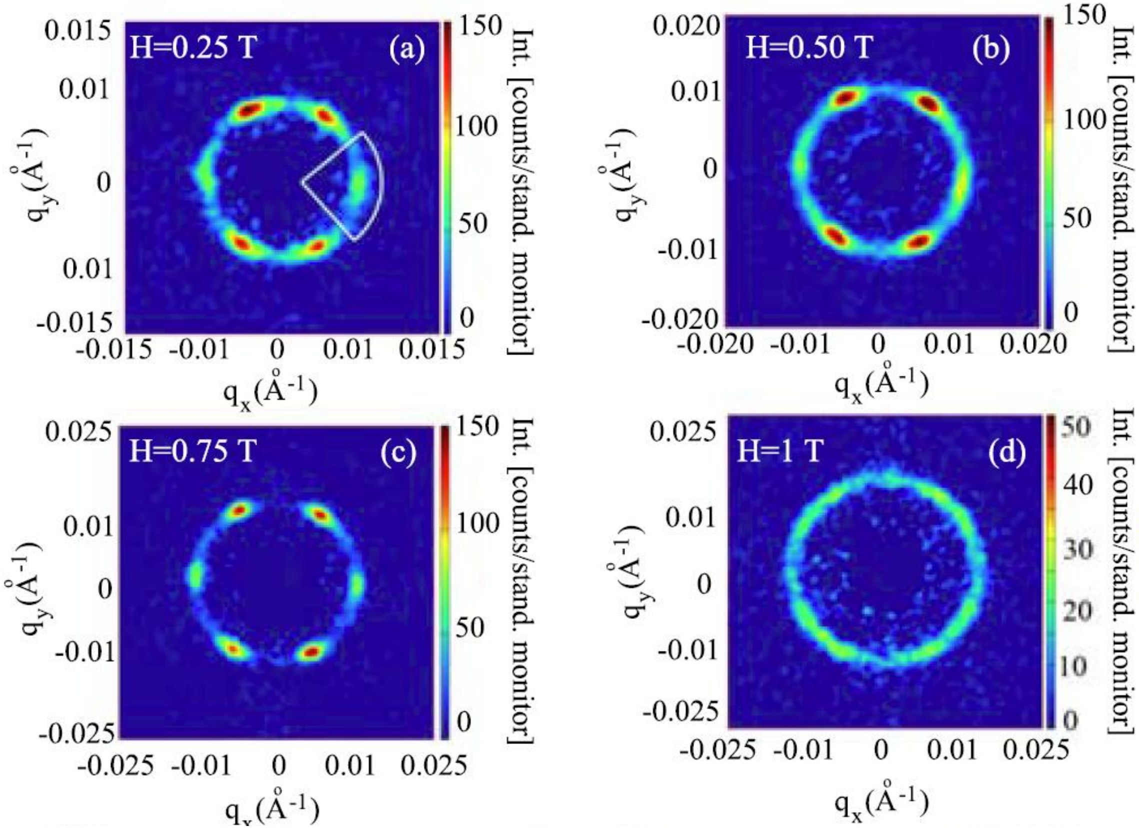

H ∦ <100>

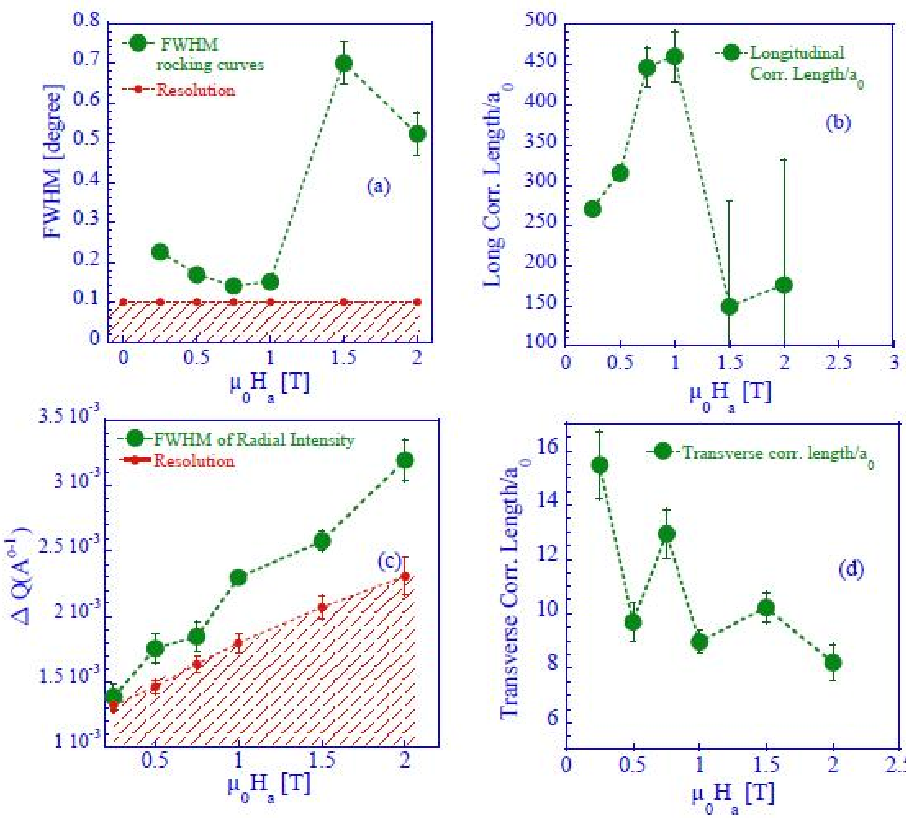


# Structure & form factor, correlation lengths

## Optimally doped $Ba_{(1-x)}K_xFe_2As_2$

Longitudinal and transverse correlation lengths of the vortex lattice




S. Demirdis et al. submitted to *PRB (2015)*

# SANS – extensions to lower Q

Focusing
VSANS
USANS (Bonse Hart Camera)
MSANS

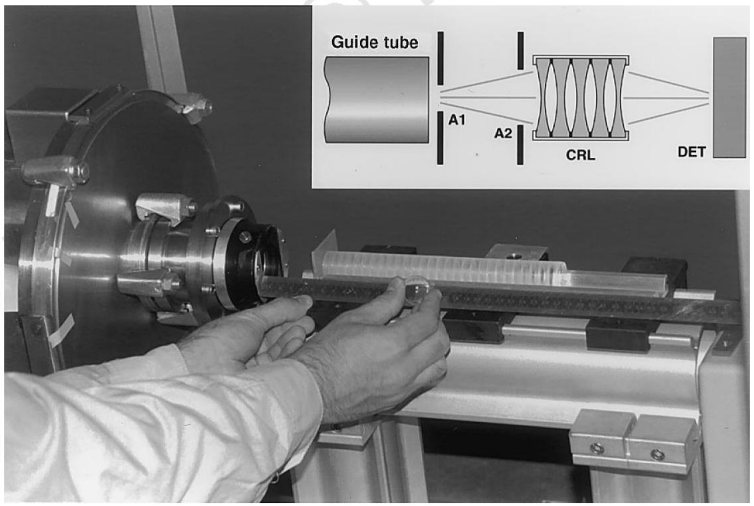
# Extensions to lower Q: Focusing



Refractive index <1 $n = 1 - \rho b_c \frac{\lambda^2}{2\pi}$

Focal length (single $MgF_2$ lens ) ≈200m

➡ Stack of lenses is used



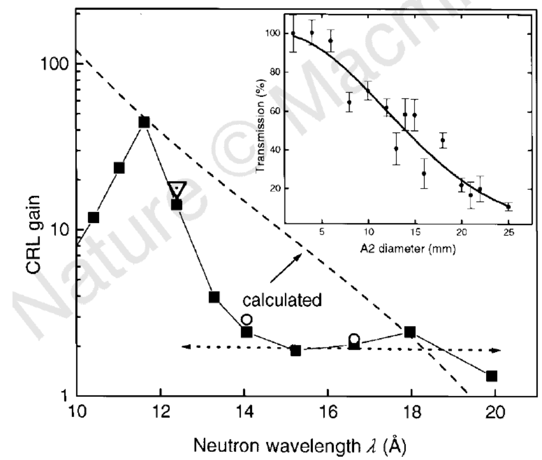

Nature **391**, p 563 (1998)

Ideal lens:

➡ No absorption

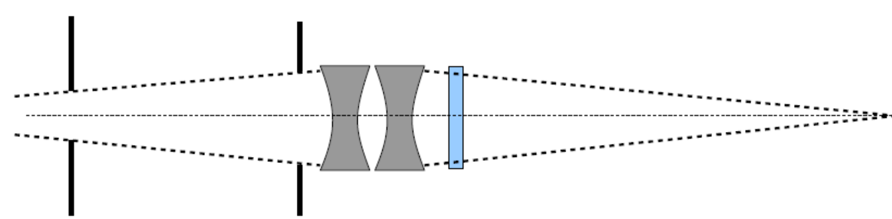
➡ No incoherent scattering

➡ Large refractive index



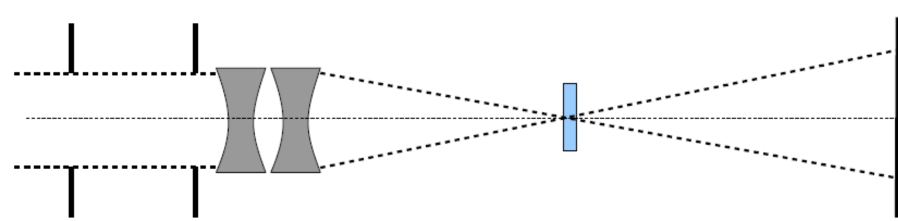

Boosting the resolution: Focus the neutron beam on the detector.

➡ Large sample needed.



Boosting the intensity: Focus the neutron beam on the sample.

➡ Sacrifice Q-resolution.



# Extensions to lower Q: VSANS

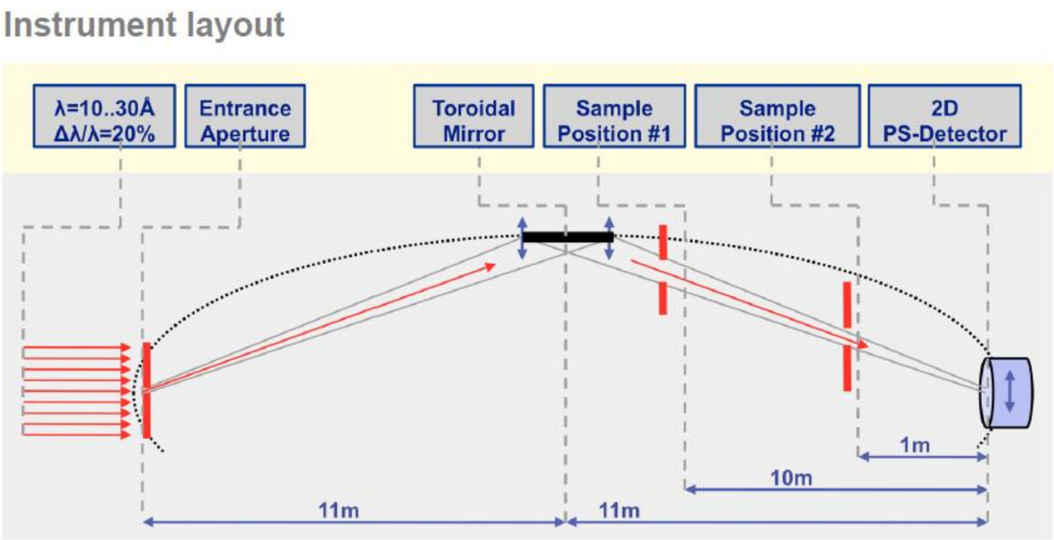


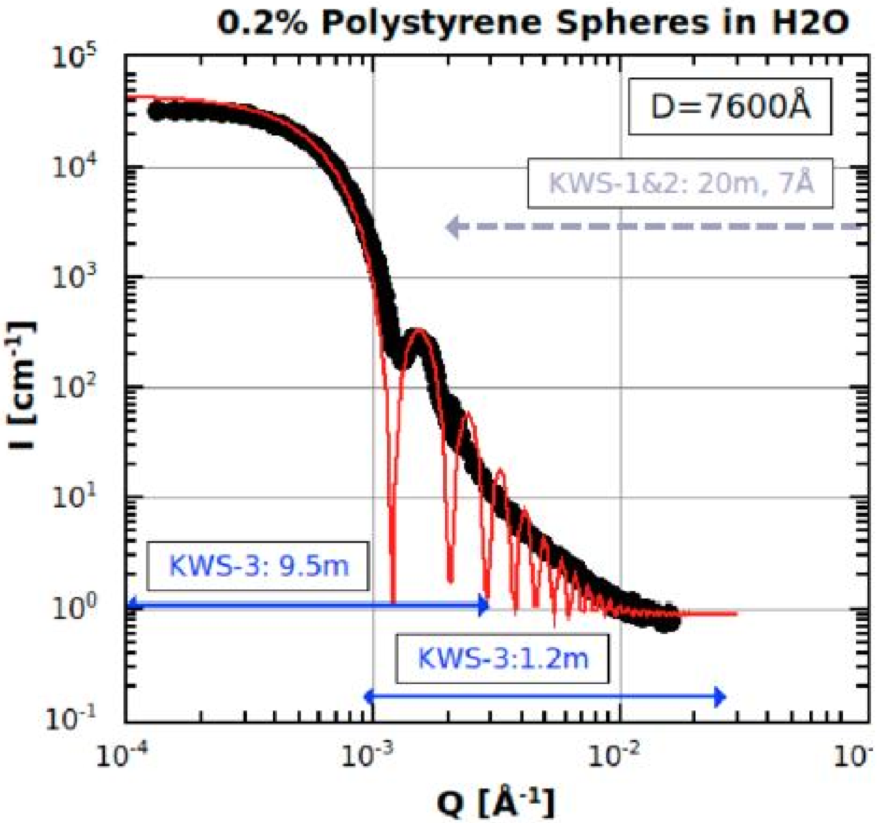
VSANS

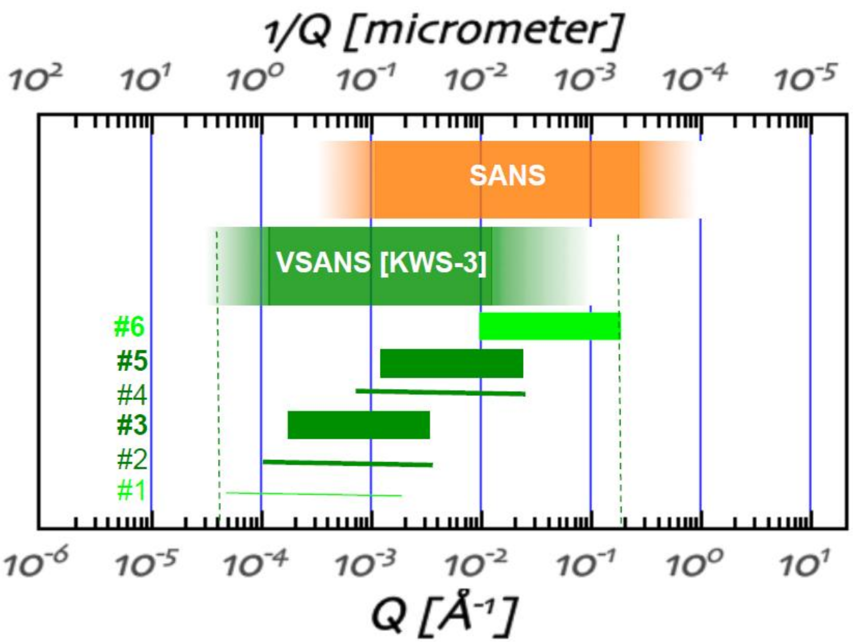
(Toroidal elliptical mirror)

$10^{-4}$ Å$^{-1}$ < Q < $3\cdot10^{-3}$ Å$^{-1}$

KWS 3 (FRM II)



# Extensions to lower Q: DCD

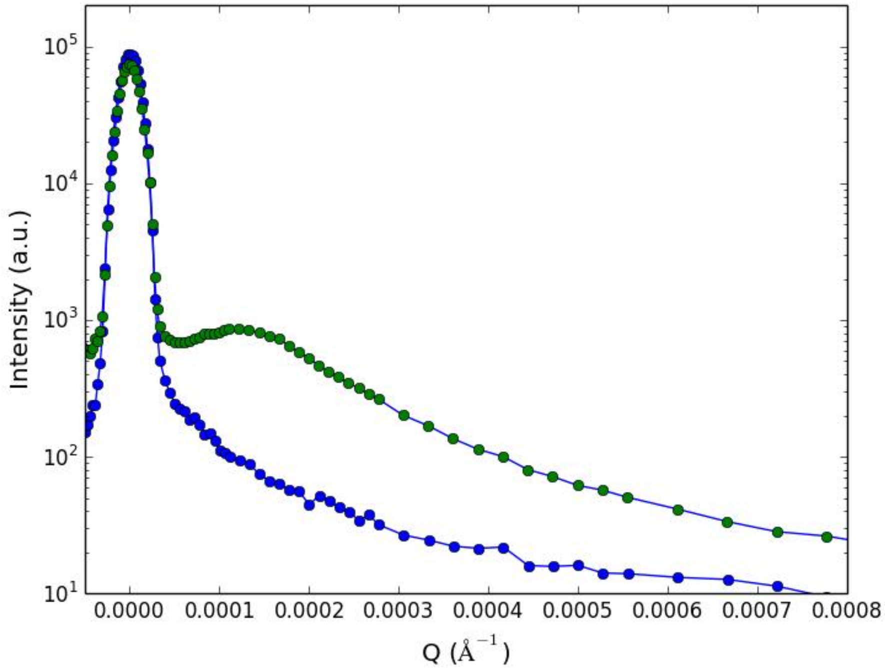


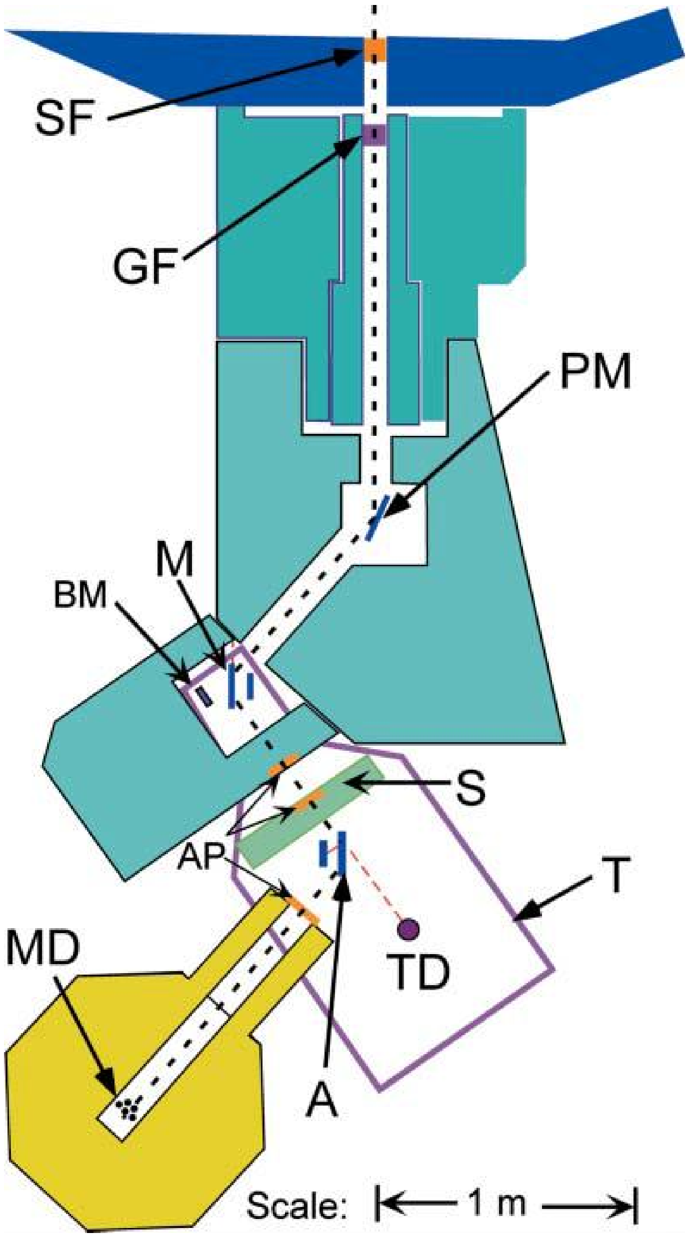
USANS (Bonse-Hart Camera)

$3 \cdot 10^{-5}$ Å$^{-1}$ < Q < $5 \cdot 10^{-3}$ Å$^{-1}$

S18 (ILL), BT5 (NIST), Kookaburra (ANSTO), V12 (HMI)

- Use multiple reflection at channel cut perfect single crystal monochromators
- Slit smeared data



# Extensions to lower Q: Multi SANS

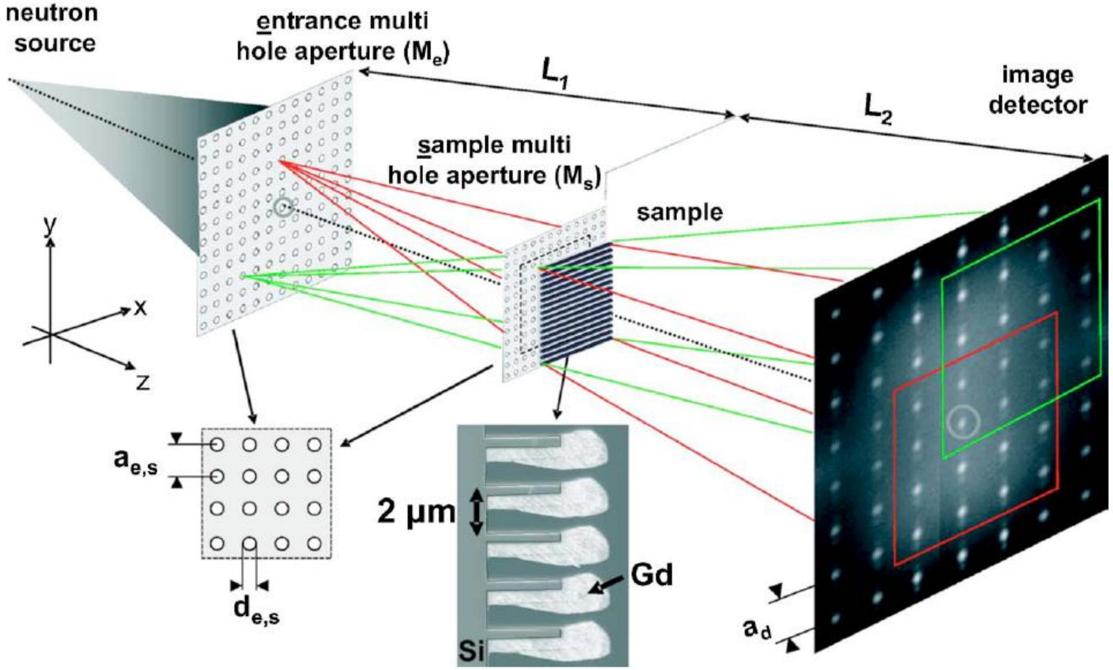


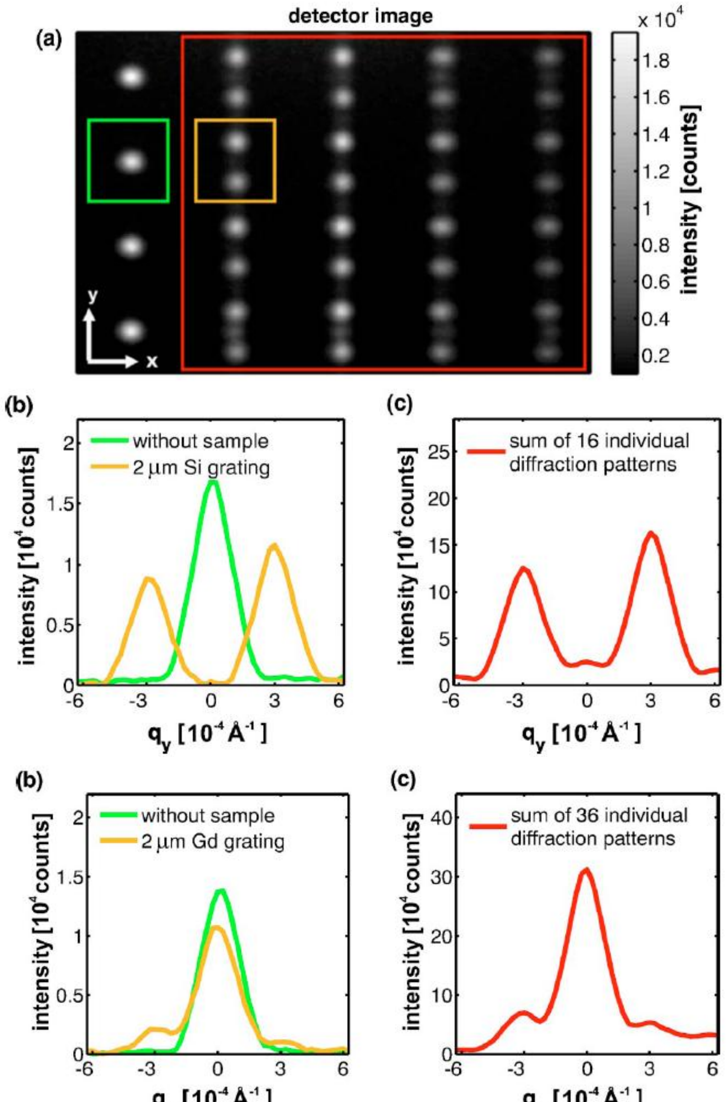
Multi-pinhole masks.

Theorem of intersecting lines.

$$\frac{a_d}{a_s} = \frac{L_1 + L_2}{L_1} \qquad \frac{a_d}{a_e} = \frac{L_2}{L_1}$$

Coherent summation of different scattering patterns at the detector




Resolution of $3 \cdot 10^{-4}$Å$^{-1}$ with 2.6m collimation

Useful for particular samples and Q-range

Problem: Edge scattering and background!