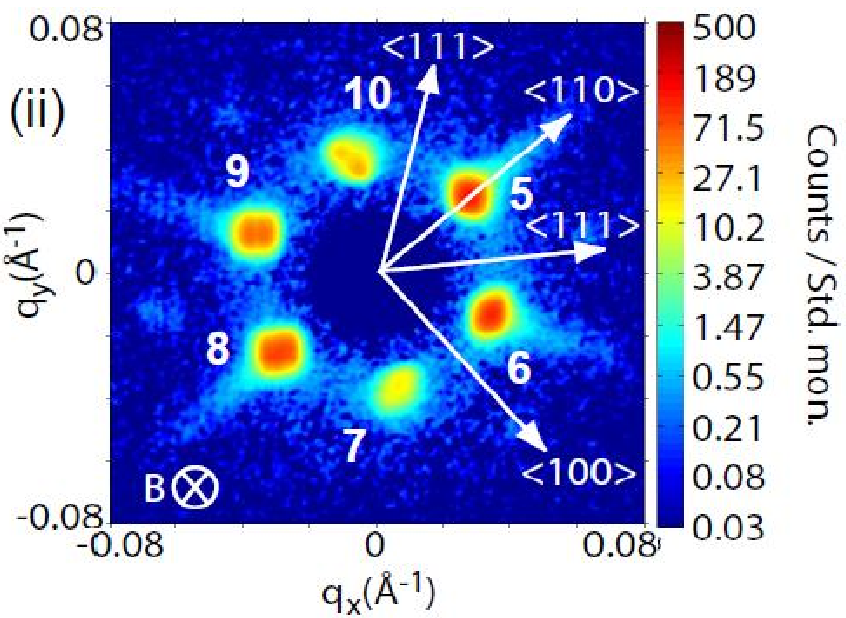
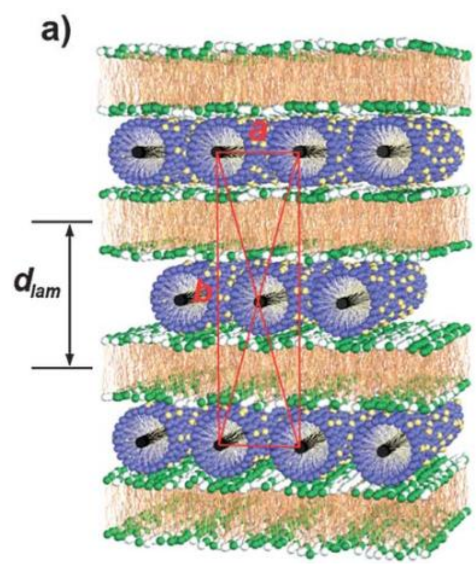
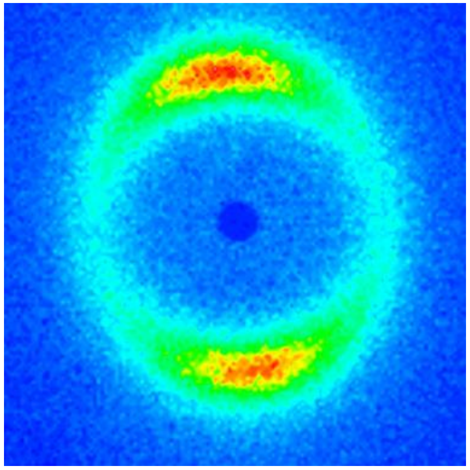
FRM II
Forschungs-Neutronenquelle
Heinz Maier-Leibnitz

MLZ
Heinz Maier-Leibnitz Zentrum

Physics with Neutrons II, SS 2016







Lecture 6, 6.6.2016

Plan for SS 2016

- VL1: Repetition of winter term, basic neutron scattering theory
- VL2: SANS, theory and applications
- VL3: Neutron optics
- VL4: Reflectometry and dynamical scattering theory
- VL5: Diffuse neutron scattering
- VL6: Magnetic scattering cross section
- VL7: Magnetic structures and structure analysis
- VL8: Polarized neutrons and 3d-polarimetry
- VL9: Inelastic scattering on magnetism
- VL10: 4.7.2016 (8:30!!) Phase transitions and critical phenomena as seen by neutrons
- VL11: Spin echo spectrocopy

Exam: Please register until 30.6.2016

Reminder:
Diffuse Neutron Scattering:
Looking between the Bragg spots

Diffuse Neutron Scattering

Master formula for elastic coherent scattering on crystals

$$\frac{d\sigma}{d\Omega}_{coh.el} = N_0 \frac{(2\pi)^3}{v_0} e^{-2W(\vec{\kappa})} \sum_{\vec{\tau}} |S_{\vec{\tau}}|^2 \, \delta(\vec{\kappa} - \vec{\tau})$$

Normalization   Debye-Waller   Structure factor   Bragg positions

Definition: Diffuse scattering is scattering that arises from departures of any kind from a perfect regular crystal lattice

- Phonons: Thermal diffuse scattering (TDS)
- Distortions due to defects: Huang scattering
- Static of dynamic displacements
- Stubstitution or interstitial defects
- Stacking faults
- (Micro-)Domains
- Magnetic effects
- ......

Diffuse neutron scattering can be static or dynamic

Diffuse Neutron Scattering

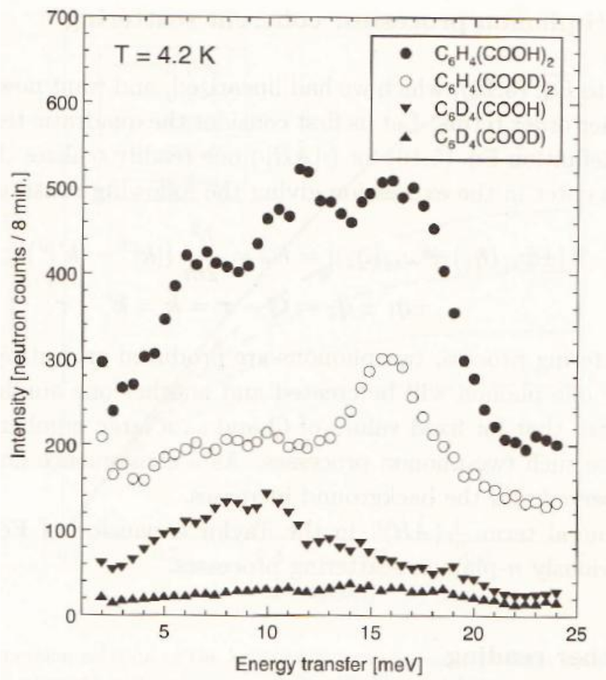
What we already know: Incoherent inelastic scattering

- Flat in Q (apart from DW)
- Gives Phonon DOS

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{1}{4M}\frac{k'}{k}\left(\langle b^2\rangle - \langle b\rangle^2\right) e^{-W(\boldsymbol{Q})} \times \langle(\boldsymbol{Q}\cdot e_s(\boldsymbol{q}))^2\rangle \cdot \frac{g(\omega)}{\omega} \cdot \left[\coth\frac{\hbar\omega}{2k_B T} \pm 1\right]$$

With phonon DOS g(ω)

$$\int_0^\infty g(\omega)d\omega = 3N$$



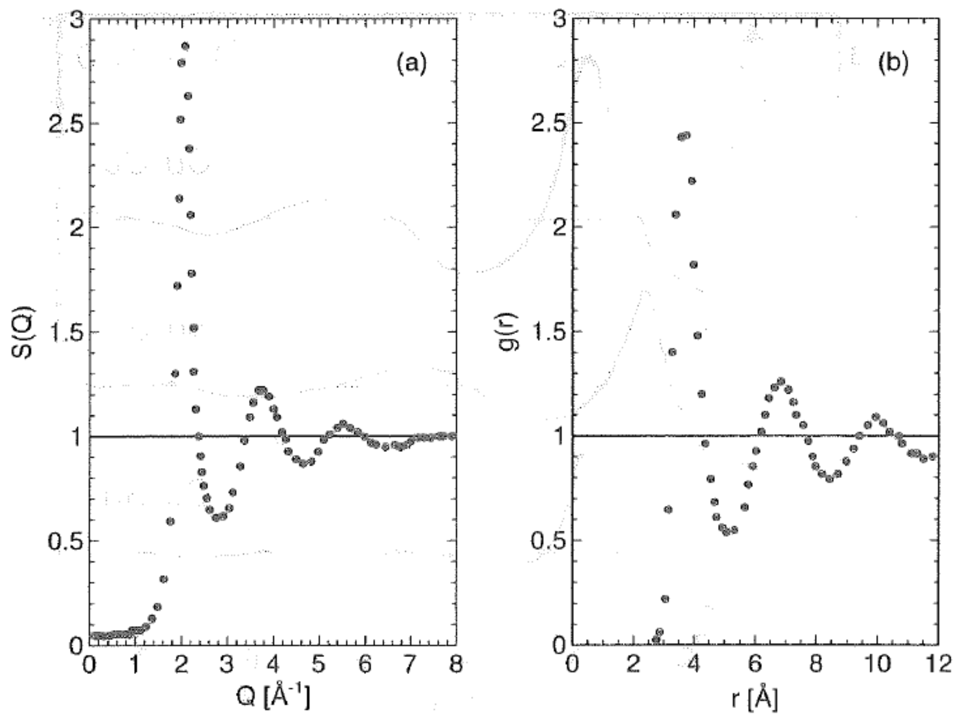
Diffuse Neutron Scattering

What we already know: <u>Scattering on liquids and amorphous materials</u>

- Measure of pair correlation function g(r)
- Quasielastic scattering: Diffusion

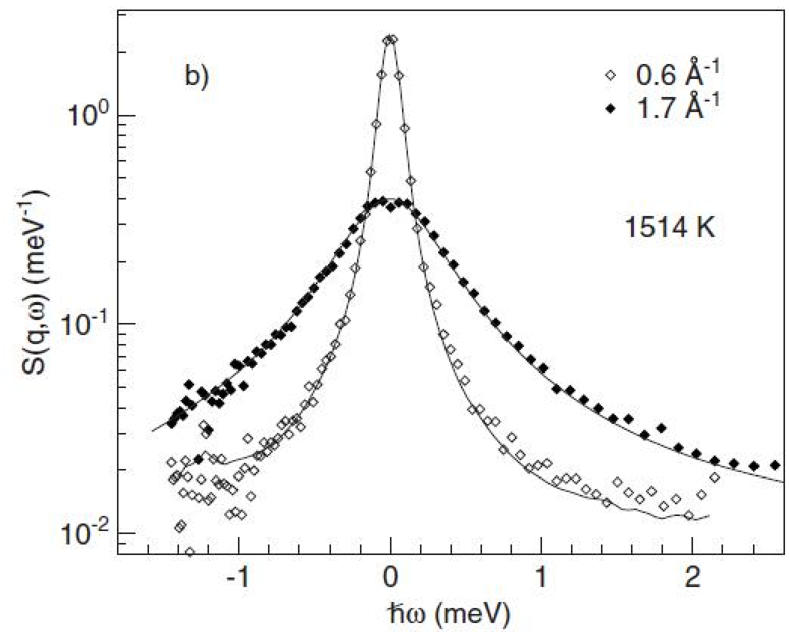
Static structure factor:
Deviations of the mean density n(r)

Dynamic structure factor:
Diffusive processes



Static structure factor:
Scattering function

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

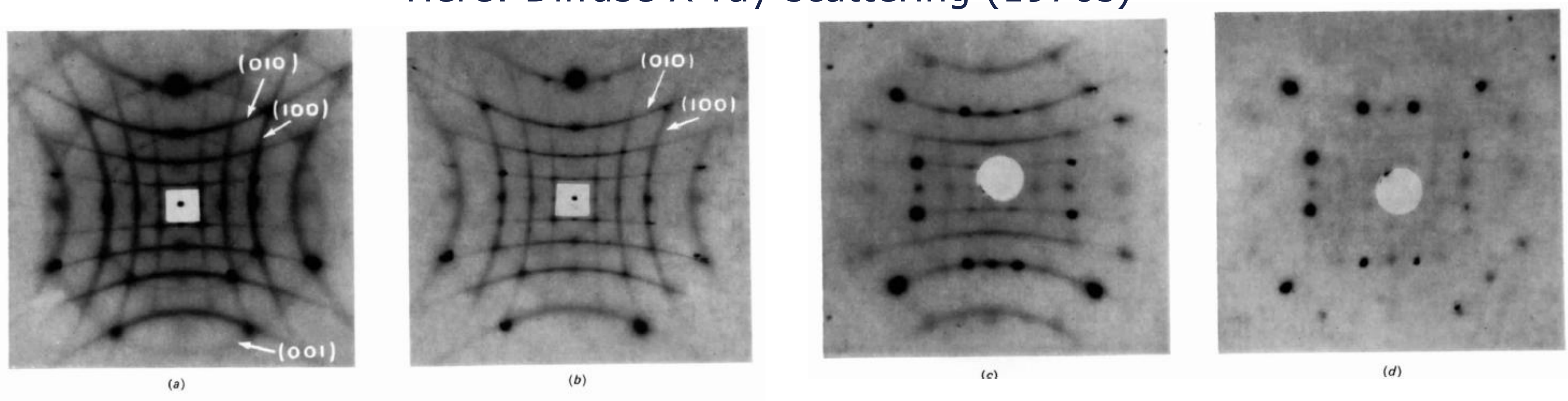


S(Q,w) ⟷ FT ⟷ G(r,t)

QENS, peaked at w=0

Example 1: Ferroelectric Perovskite $KNbO_3$

Here: Diffuse X-ray scattering (1970s)



(a) (b) (c) (d)

Two alternative models for diffuse sheets:

→ Formation of linear chains of correlated vibrational displacements due to a specific soft mode? Thermal diffuse scattering?

→ Formation of linear chains of correlated local displacements?

Diffuse Neutron Scattering

Ferroelectric Perovskite $KNbO_3$

Compare to $PbTiO_3$ and $BaTiO_3$

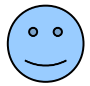
??

Formation of linear chains of correlated vibrational displacements due to a specific soft mode? Thermal diffuse scattering?

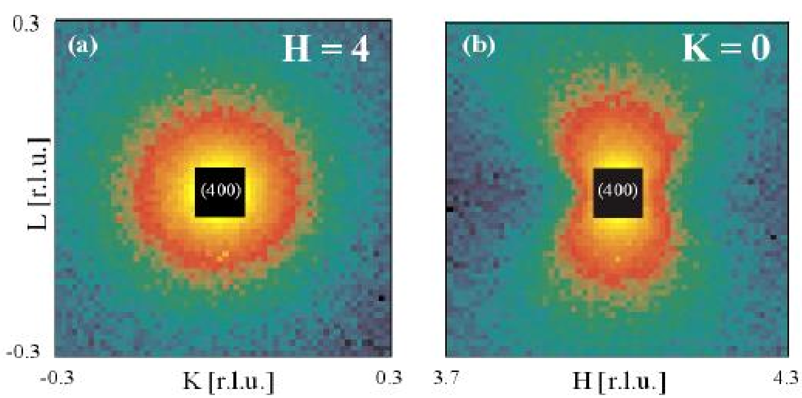
- Soft mode similar for $PbTiO_3$, $BaTiO_3$ and $KnbO_3$
- Expect similar results

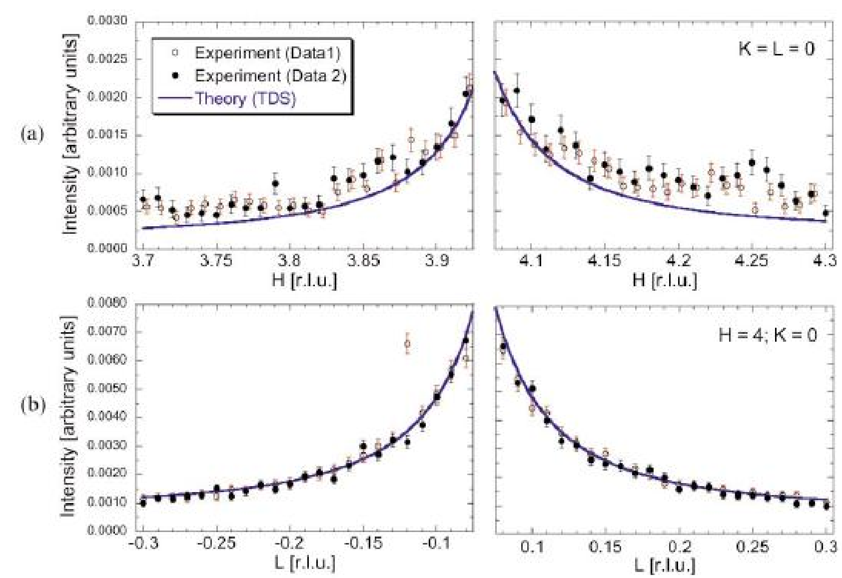
Formation of linear chains of correlated local displacements?

- Different local symmetry
- No diffuse sheets for $PbTiO_3$
- Expect different result



Thermal diffuse scattering $PbTiO_3$





Diffuse intensity around Bragg positions

Diffuse Neutron Scattering

Ferroelectric Perovskite $KNbO_3$

Formation of linear chains of correlated local displacements

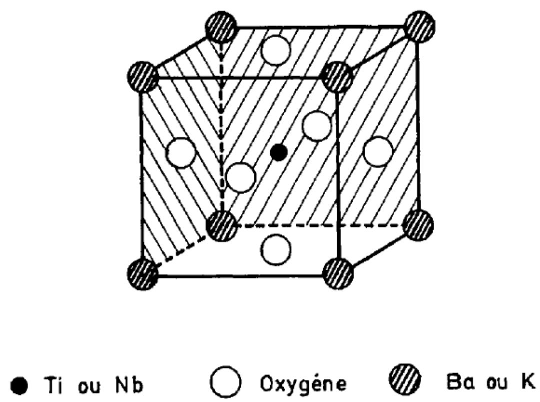
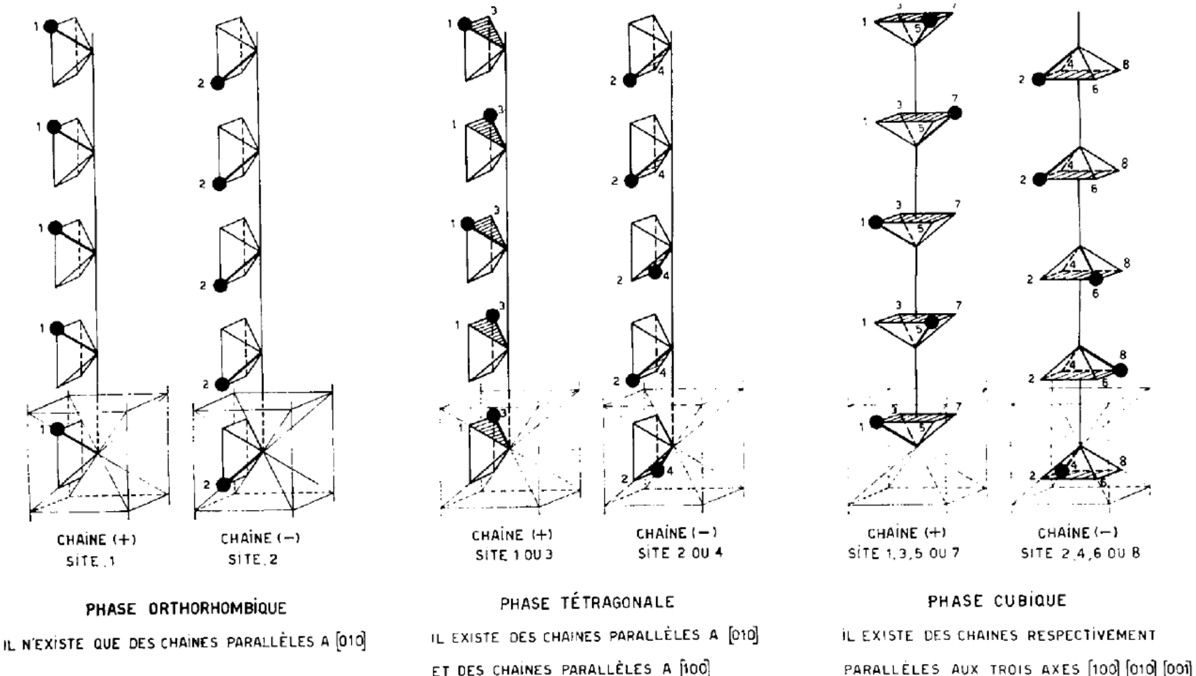


Fig. 5. La structure pérovskite idéale.



(a)

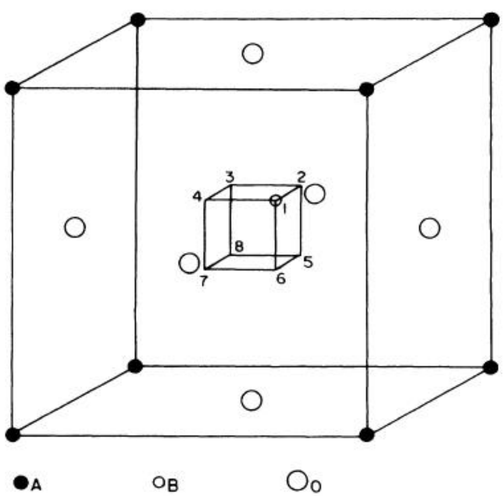
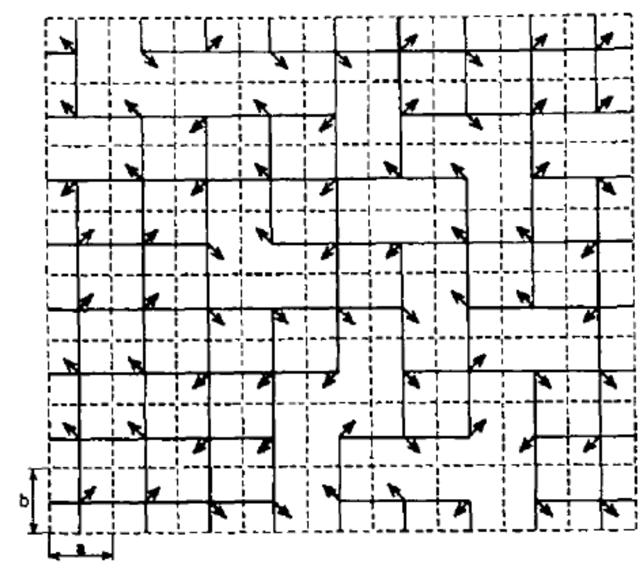


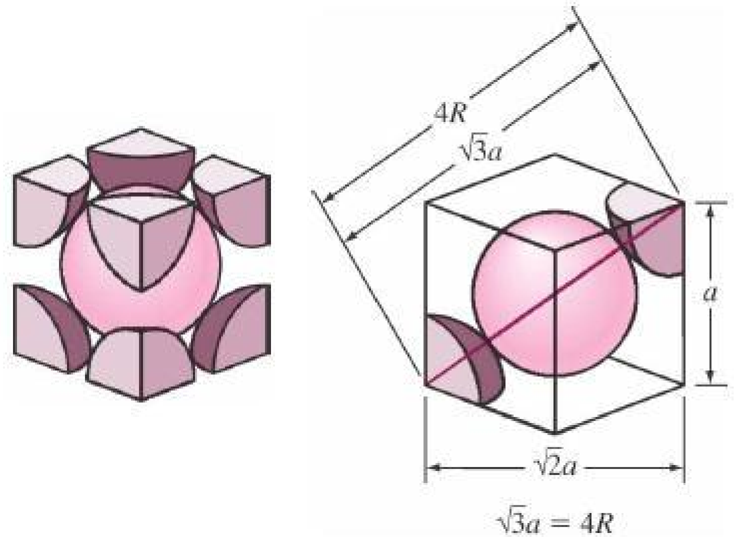
Fig. 10. Le croisement des chaines de corrélation en projection sur un plan (001) dans la phase tétragonale. Les flèches schématisent le déplacement de l'atome central projeté sur le plan (001); les mailles appartenant à une même chaine sont reliées en trait plein. En phase tétragonale la composante du déplacement perpendiculairement au plan de figure est constante; en phase cubique au contraire il existerait un système de chaines analogue suivant [001].



Diffuse Neutron Scattering

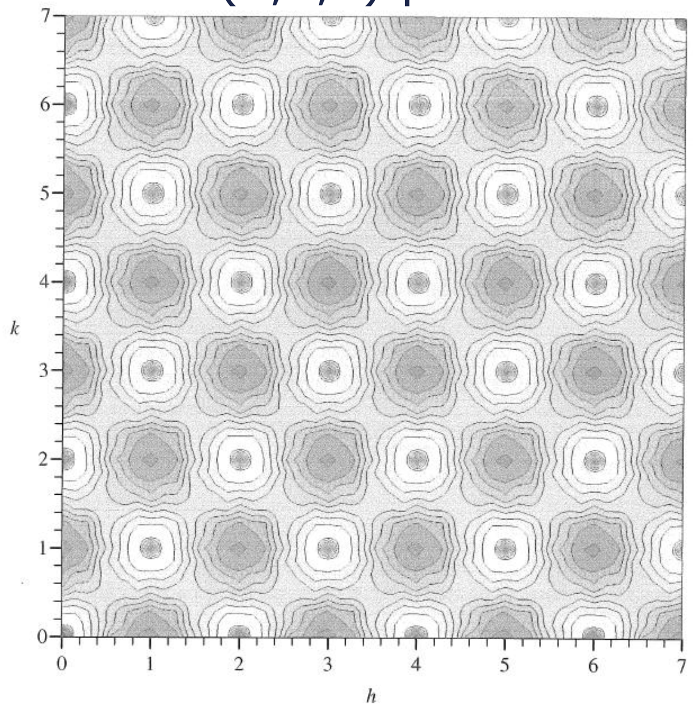
Example 2: Simple binary alloy on a bcc lattice

Crystallogaphic sice can be occuied by
- two specific atoms
- half occupacy
- random occupany



Simple rule:
- Similar atoms with scattering length b
- No imaginary part
- No variation of b
- Exactly one atom per unit call occupied, however, randomly distributed

(h,k,o) plane



„Random" distribution, flat in Q?

Diffuse scattering

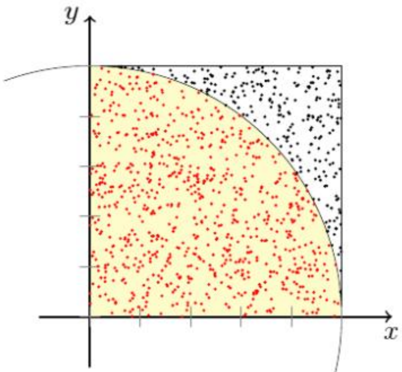
$$D^2 = \left| \sum_u s_u i b \sin(\mathbf{Q} \cdot \mathbf{d}) \exp(i\mathbf{Q} \cdot \mathbf{R}_u) \right|^2$$

\+ Bragg scattering

$$\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{coh}} = b^2 \sum_{j,j'} \exp(i\mathbf{Q} \cdot (\mathbf{R}_j - \mathbf{R}_{j'}))$$

Diffuse Neutron Scattering

Powerful tool for modeling diffuse scattering



Monte Carlo simulations (MC)

1) Choose appropriate descriptions of potentials

2) Generate arrangement of N atoms including boundary conditions

3) Calculate system energy

4) Randomly move atom(s)

5) Recalculate energy

6) Refine energy of the system with Monte Carlo method

→ Working from the potential to the structure

Reverse Monte Carlo simulations (RMC)

1) Generate arrangement of N atoms including boundary conditions and hard core potential to avoid overlap

2) Calculate F(Q) from arrangements

3) Randomly move atom(s)

4) Recalculate scattering signal F(Q)

5) Refine F(Q) of the system with Monte Carlo method

→ Working from the scattering signal to the structure

Diffuse Neutron Scattering

Diffuse neutron scattering: Lets try a classification:

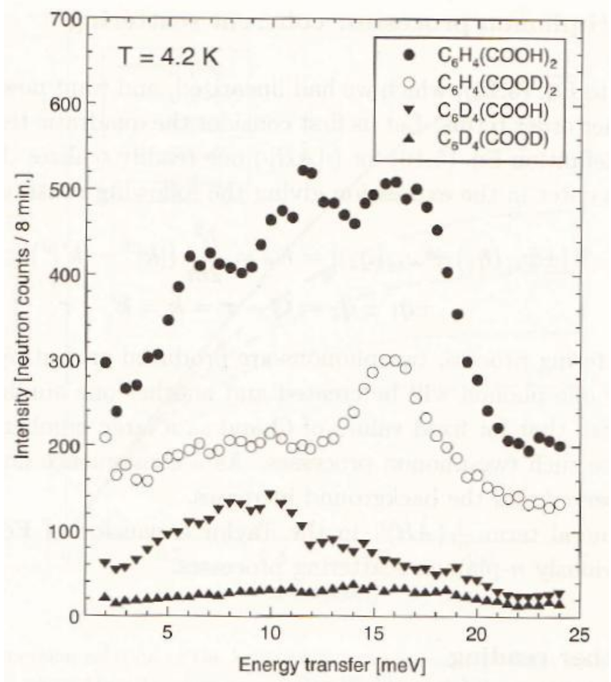
Diffuse background (flat in Q)

- Incoherent elastic scattering, radom disorder (isotope, spin, voids....)

$$\frac{d\sigma}{d\Omega_{inc.}} = (\langle b^2\rangle - \langle b\rangle^2) \sum_{j=j'} e^{-i\kappa(R_{j'}-R_j} = N(\langle b^2\rangle - \langle b\rangle^2)$$

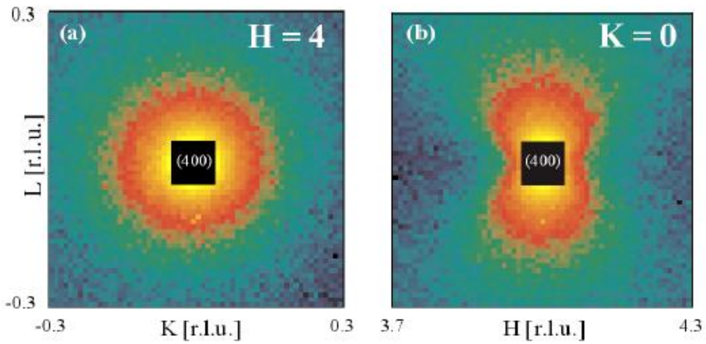
- Incoherent inelastic scattering, phonon DOS (Debye Waller, not really flat)

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{1}{4M}\frac{k'}{k}\left(\langle b^2\rangle - \langle b\rangle^2\right) e^{-W(\boldsymbol{Q})} \times \langle(\boldsymbol{Q}\cdot e_s(\boldsymbol{q}))^2\rangle \cdot \frac{g(\omega)}{\omega} \cdot \left[\coth\frac{\hbar\omega}{2k_BT} \pm 1\right]$$



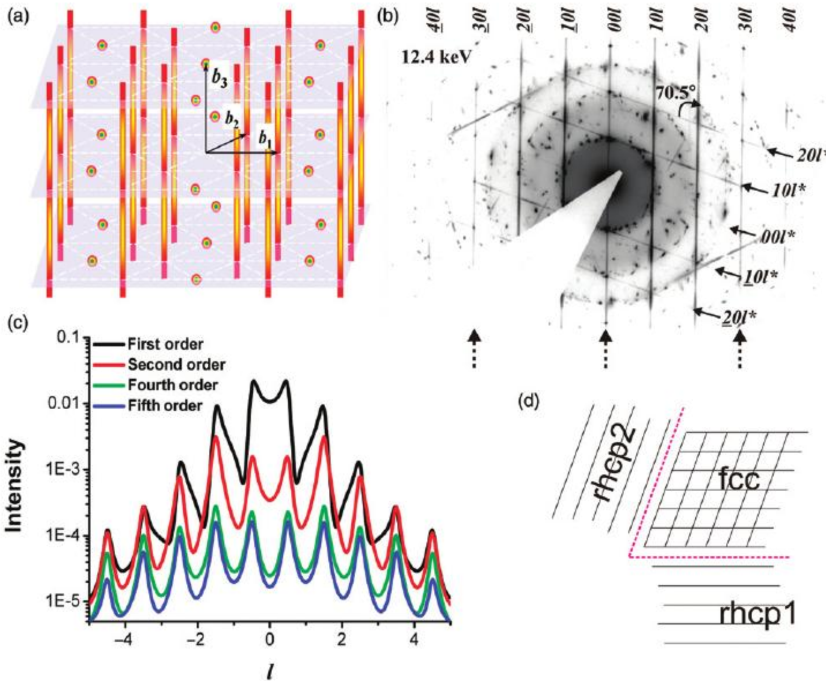
Odd Bragg peak shape (Butterflies, ellipoids..)

- Typically thermal diffuse scattering, Einstein model for optical phonons, measurement of lattice elasticity



Diffuse neutron scattering: Lets try a classification:

Bragg rods and planes



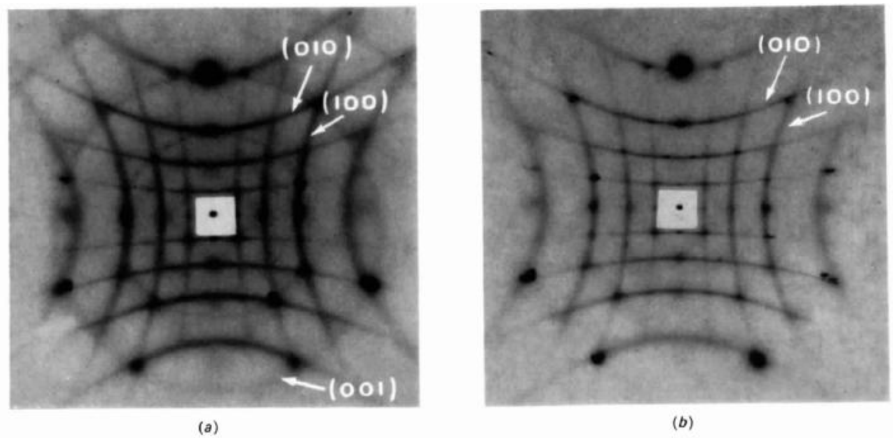
- Other than 3D order

  Line order (1D)
  - Bragg planes

  Planar correlations (2D oder)
  - Bragg rods

Diffuse but with structure in Q

- Partial order /suborder of systems
  See examples 1 and 2,
  Liquid and amorphous samples



Diffuse Neutron Scattering

In general: Size of distortion ~L

Spread in $\Delta q \sim 2\varpi/L$

Try to minimize parameter space

Short range features, spread over large Q

Work in real space (correlation appoach, microdomain approach)

Long range features, spread over small Q

Work in reciprocal space (Modulation wave approach)

Magnetic neutron scattering: Basic cross section

Magnetic neutron scattering

Starting point (similar to Winter term, now including the spin state of the neutron)

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}E'}\right) = \underbrace{\frac{k'}{k}}_{\text{Wavevectors}} \underbrace{\left(\frac{m}{2\pi\hbar}\right)^2}_{\text{Normalization}} \underbrace{\sum_{k',\sigma'}\sum_{k,\sigma}}_{\text{Sum over k and neutron spin states}} \underbrace{p_\lambda p_\sigma}_{\text{Probability of sample state and neutron spin state before scattering}} \underbrace{\left|\langle k'\sigma'\lambda' \,|U|\, k\sigma\lambda\rangle\right|^2}_{\text{Matrix element, Interaction}} \underbrace{\delta(\hbar\omega + E_\lambda - E_{\lambda'})}_{\text{Energy conservation}}$$

Until now: Only nuclear scattering

Interaction: Fermi pseudopotential

$$V(r) = \frac{2\pi\hbar^2}{m} b\delta(r)$$

Magnetic neutron scattering

Nuclear neutron scattering

Interaction: Fermi pseudopotential $$V(r) = \frac{2\pi\hbar^2}{m} b\delta(r)$$

- Scalar funtion
- Point like (delta function)
- For an incoming plane wave: s-wave scattering
- Spherical symmetry
- FT of delta function is constant

Magnetic neutron scattering:

Interaction: Magnetic moment of neutron interacts with local magnetic field

$$\hat{U} = \hat{\mu} \cdot \vec{H} \qquad \hat{U} = \hat{\mu} \cdot \vec{H} = \gamma \mu_N \hat{\sigma} \cdot \vec{H}$$

$$\vec{H} = \underbrace{\nabla \times \left(\frac{\vec{\mu}_e \times \vec{R}}{|\vec{R}|^3} \right)}_{\text{Spin of electrons}} \underbrace{- \frac{e}{c} \frac{\vec{v}_e \times \vec{R}}{|\vec{R}|^3}}_{\text{Orbital momentum}}$$

- Vector (dipole-dipole) interaction
- Extended range, not point like
- No s-wave scattering
- No spherical symmetry
- Magnetic moments: Unpaired electrons

Magnetic neutron scattering

Leaving away the maths

- For spin only scattering (neglect orbital momentum)
- For unpolarized neutrons (average over polarization states)
- For identical magnetic ions with localized moments

Master formula

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}E'}\right) = (\gamma r_0)^2 \frac{k'}{k} F^2(\vec{Q}) e^{-2W(\vec{Q})} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}) S^{\alpha\beta}(Q,\omega)$$

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \sum_{\lambda,\lambda'} p_\lambda \langle\lambda|S_{j'}^{\alpha}|\lambda'\rangle\langle\lambda|S_j^{\beta}|\lambda'\rangle\delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Magnetic neutron scattering

Master formula

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right) = (\gamma r_0)^2 \frac{k'}{k} F^2(\vec{Q}) e^{-2W(\vec{Q})} \sum_{\alpha,\beta} \underline{(\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2})} S^{\alpha\beta}(Q,\omega)$$

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \sum_{\lambda,\lambda'} p_\lambda \langle\lambda|S^\alpha_{j'}|\lambda'\rangle\langle\lambda|S^\beta_j|\lambda'\rangle \delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Dipole-dipole interaction: Angular dependence

Neutron polarization factor $(\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2})$

Only moments perpendicular to Q contribute to magnetic scattering

Don't confuse with polarized neutrons!

Magnetic neutron scattering

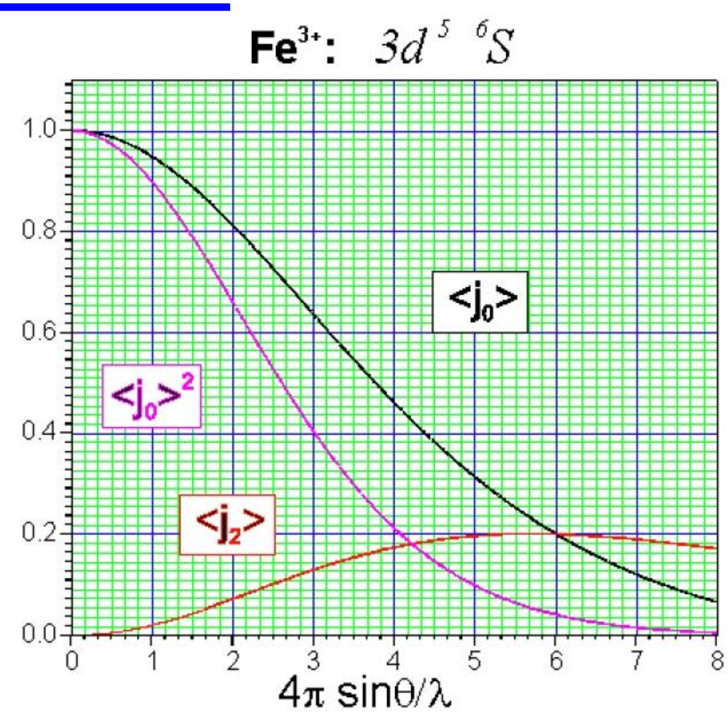
Master formula

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'}\right) = (\gamma r_0)^2 \frac{k'}{k} \underline{F^2(\vec{Q})} e^{-2W(\vec{Q})} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}) S^{\alpha\beta}(Q,\omega)$$

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \sum_{\lambda,\lambda'} p_\lambda \langle\lambda|S^\alpha_{j'}|\lambda'\rangle\langle\lambda|S^\beta_j|\lambda'\rangle \delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Dipole-dipole interaction: Magnetic form factor

- Fourier transform of electron cloud
- Useful to discriminate magnetic/nuclear scattering
- Check the tables for each ion!



Magnetic neutron scattering

For the case of orbital momentum + spin

$$\mu = -\mu_b(L + 2S)$$

Effective angular momentum operator

$$\hat{S}_j^\alpha = \frac{1}{2} g \hat{J}_j^\alpha$$

Landé splitting factor

$$g = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}$$

⇨ Approximation for small Q, spin+orbital momentum

Magnetic scattering function

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \underline{\langle S_{j'}^\alpha(0)\ S_{j'}^\beta(t) \rangle} e^{-i\omega t} \mathrm{d}t$$

<u>Spin correlation funtion:</u>

Correlation of magnetic moment at site j, time t=0
and site j', time t=t

⇨ <u>Fourier transform measured with neutrons!</u>

Magnetic neutron scattering

Why neutrons are so unique:

Fluctuation dissipation theorem

$$\boxed{S^{\alpha\beta}(Q,\omega) = \frac{N\hbar}{\pi}(1 - e^{-\frac{\hbar\omega}{k_b T}})^{-1} \mathrm{Im}\chi^{\alpha\beta}(\vec{Q},\omega)}$$

Neutrons directly measure generalized susceptibility tensor

$$\boxed{M^{\alpha}(\vec{Q},\omega) = \chi^{\alpha\beta}(\vec{Q},\omega) H^{\beta}(\vec{Q},\omega)}$$