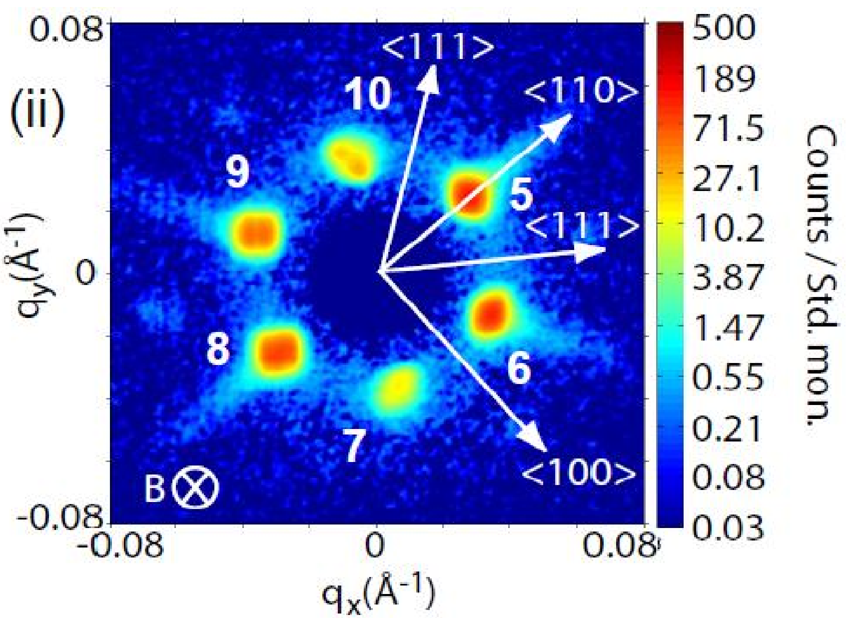
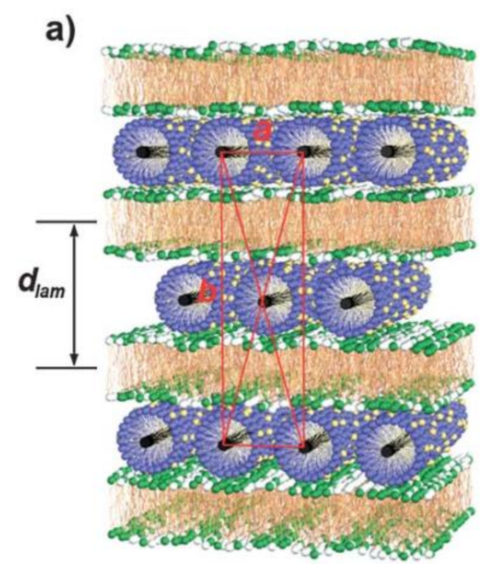
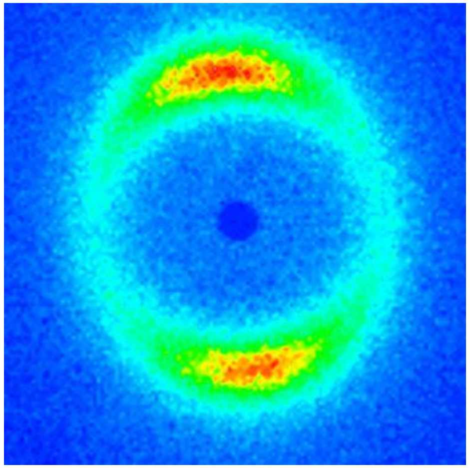
FRM II
Forschungs-Neutronenquelle
Heinz Maier-Leibnitz

MLZ
Heinz Maier-Leibnitz Zentrum

Physics with Neutrons II, SS 2016







Lecture 1, 11.4.2016

Organization

Lecture: Monday 12:00 – 13:30, PH227
Sebastian Mühlbauer (MLZ/FRM II)
Sebastian.muehlbauer@frm2.tum.de
Tel:089/289 10784

Tuturials: Friday 12:00 – 13:30, 2224 (E21)
(first tutorial 24.4.2016)
Lukas Karge
Lukas.karge@frm2.tum.de
Tel:089/289 11774

http://wiki.mlz-garching.de/n-lecture02:index

Organization

Suggested:

Seminar ***Methoden und Experimente in der Neutronenstreuung***
(PH-E21-1), Wednesday 9:00-10:30, PH2224, (Start 20.4.2016)
P. Böni, S. Mühlbauer, C. Hugenschmidt

Lecture ***Grundlagen zur Instrumentierung mit Neutronen***
Thursday, 8:30-9:00, PH2224, (Start 21.4.2016)
P.Böni

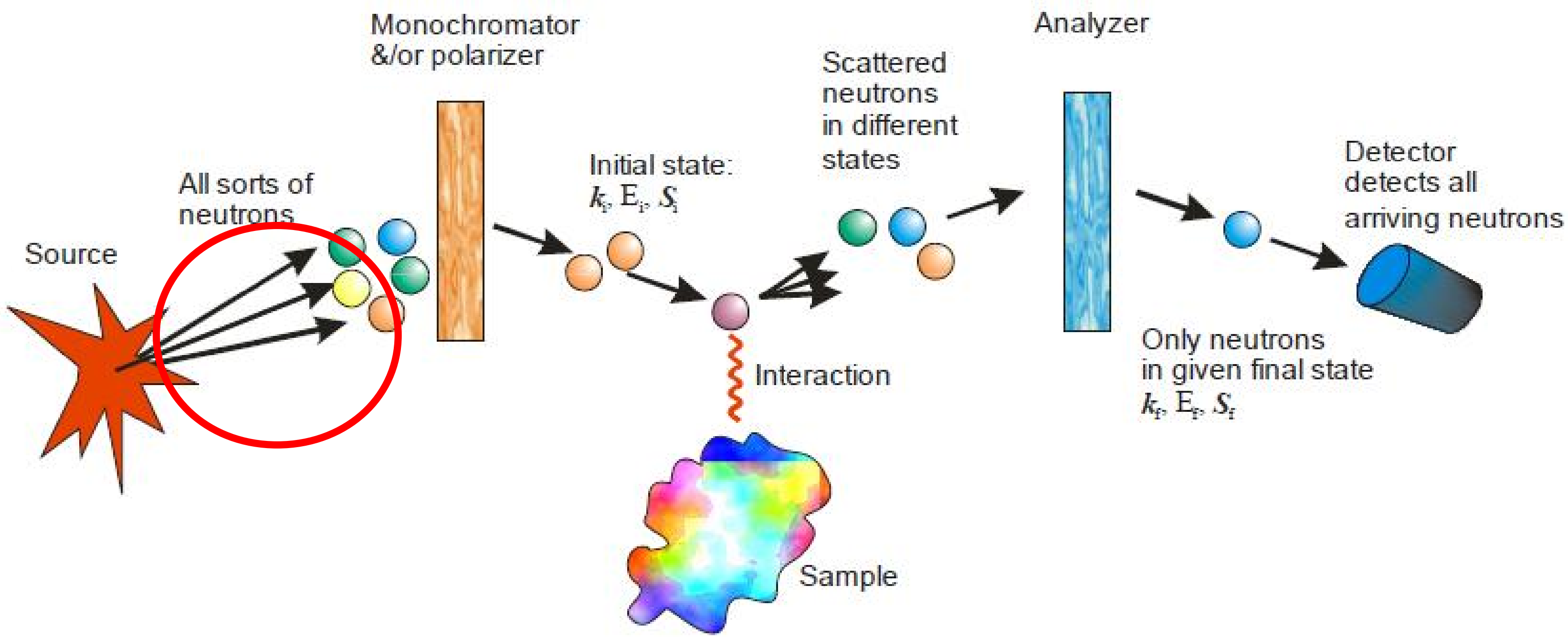
Seminar ***Neutrons in Science and Industry***
(PH-21-4), Monday 14:30-15:45, HS3, (Start 11.4.2016)
Organization: P.Böni, W.Petry, T. Schöder, T. Schrader

Plan for SS 2016

- VL1: Repetition of winter term, basic neutron scattering theory
- VL2: SANS, GISANS and soft matter
- VL3: Neutron optics, reflectometry and dynamical scattering theory
- VL4: Diffuse neutron scattering
- VL5: Cross sections for magnetic neutron scattering
- VL6: Magnetic elastic scattering (diffraction)
- VL7: Magnetic structures and structure analysis
- VL8: Polarized neutrons and 3d-polarimetry
- VL9: Inelastic scattering on magnetism
- VL10: Magnetic excitations Magnons, spinons
- VL11: Phase transitions and critical phenomena as seen by neutrons
- VL12: Spin echo spectrocopy

Essence of a neutron scattering experiment

Fundamental principle of a neutron scattering experiment



Basic neutron scattering theory

Fermis Golden Rule

Fermis Golden Rule for scattering process: Born approximation

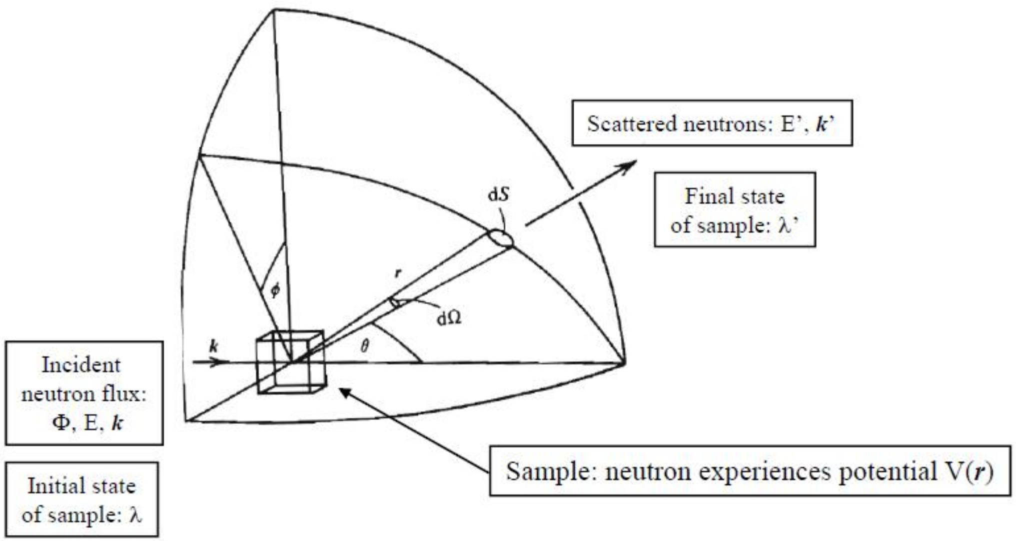
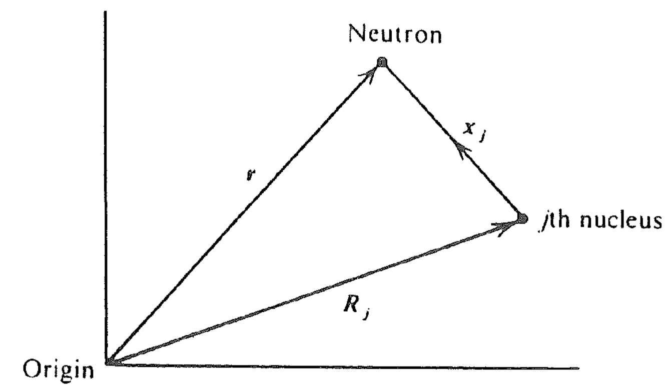


Fig. 2.1 Coordinates of nucleus and neutron.



$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\lambda\to\lambda'} = \frac{1}{\Phi}\frac{1}{\Omega}\sum_{k'\ in\ \mathrm{d}\Omega} W_{k,\lambda\to k',\lambda'}$$

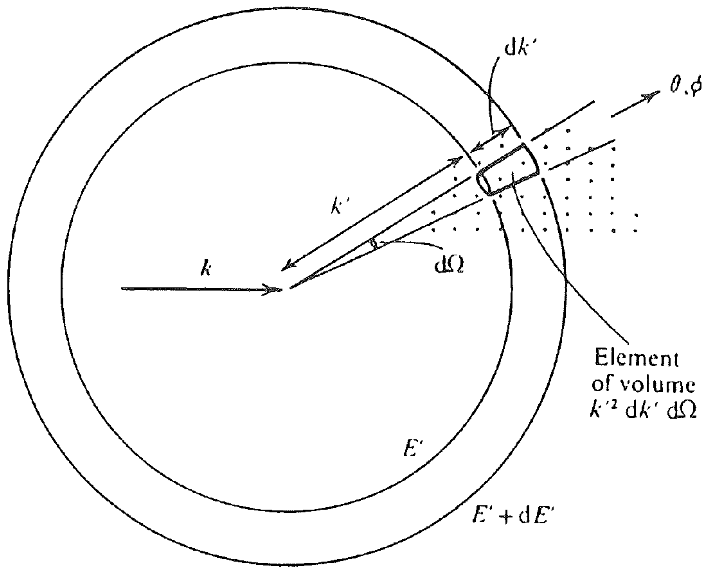
$$\sum_{k'\ in\ \mathrm{d}\Omega} W_{k,\lambda\to k',\lambda'} = \frac{2\pi}{\hbar}\varrho_k \left|\langle k'\lambda' |V| k\lambda\rangle\right|^2$$

$$\langle k'\lambda' |V| k\lambda\rangle = \int \Psi_{k'}^* \chi_{\lambda'}^* V \Psi_k^* \chi_\lambda^* \mathrm{drdR}$$

Challenge: Rewrite this mixed expression in terms of sample properties

Box Integration

1st step: Box Normalization to calculate ρ_k



$$\left(\frac{d\sigma}{d\Omega}\right)_{\lambda\to\lambda'} = \underline{\frac{k'}{k}\left(\frac{m}{2\pi\hbar}\right)^2} \left|\langle k'\lambda' | V | k\lambda\rangle\right|^2$$

Energy Conservation, Integration

2nd step: Energy conservation

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}E'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar}\right)^2 \left|\langle k'\lambda'|V|k\lambda\rangle\right|^2 \underline{\delta(E_\lambda - E_{\lambda'} + E - E')}$$

$$\int \delta(E_\lambda - E_{\lambda'} + E - E') = 1$$

3rd step: Integration with respect to neutron coordinate r

$$\langle k'\lambda'|V|k\lambda\rangle = \sum_j V_j(\kappa)\,\langle\lambda'|e^{i\kappa R_j}|\lambda\rangle \qquad \underline{V_j(\kappa) = \int V_j(x_j)e^{i\kappa x_j}\mathrm{d}x_j}$$

$$\kappa = k - k'$$

Interaction = Fourier transform of the potential function

Fermi Pseudopotential

4th step: Ansatz: Delta function potential for single nucleus

$$V(r) = a\delta(r)$$

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

- b can be bound or free scattering length!
- Fermi pseudopotential does NOT represent physical reality

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\left|\sum_j b_j \left\langle k'\lambda' \left| e^{i\kappa R_j} \right| k\lambda \right\rangle\right|^2 \delta(E_\lambda - E_{\lambda'} + E - E')$$

Integral Representation of Delta Function

5th step: Integral representation of the delta function for energy.
Idea: Stick all the time dependence into the matrix element

$$\delta(E_\lambda - E_{\lambda'} + E - E') = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} e^{i(E_\lambda - E_{\lambda'})t/\hbar} e^{-i\omega t} \mathrm{d}t$$

H is the Hamiltonian of the scattering system with Eigenfunctions λ and Eigenvalues E_λ

$$H|\lambda\rangle = E_\lambda|\lambda\rangle$$

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}E'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'} b_j b_{j'} \int_{-\infty}^{\infty} \langle\lambda| e^{-i\kappa R_{j'}} |\lambda'\rangle \langle\lambda'| e^{iHt/\hbar} e^{i\kappa R_j} e^{-iHt/\hbar} |\lambda\rangle\, e^{-i\omega t} \mathrm{d}t$$

No terms of λ and λ' outside the matrix element anymore!

Sum Over Final States

6th step: Sum over final states, average over initial states

- We use:

$$\sum_{\lambda'} \langle\lambda|A|\lambda'\rangle\langle\lambda'|B|\lambda\rangle = \langle\lambda|AB|\lambda\rangle$$

$$p_\lambda = \frac{1}{Z} e^{\frac{-E_\lambda}{k_b T}}$$

$$\langle A\rangle = \sum_\lambda \langle\lambda|A|\lambda\rangle$$

- Stick the time evolution into the operator for the position R_j $\quad R_j(t) = e^{iHt/\hbar} R_j e^{-iHt/\hbar}$

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'} b_j b_{j'} \int_{-\infty}^{\infty} \underline{\left\langle e^{-i\kappa R_{j'}(0)} e^{-i\kappa R_j(t)}\right\rangle} e^{-i\omega t} dt$$

Correlation function

Coherent/Incoherent Scattering

Assume a large sample with statistically variations of b_j

No correlations among b: $\overline{b_{j'}b_j} = (\bar{b})^2, j' \neq j$

$$\overline{b_{j'}b_j} = (\overline{b^2}), j' = j$$

Double differential crosssection spilts up into two terms:

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{coherent} = \frac{\sigma_{coh.}}{4\pi}\frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'}\int_{-\infty}^{\infty}\underline{\left\langle e^{-i\kappa R_{j'}(0)}e^{-i\kappa R_j(t)}\right\rangle} e^{-i\omega t}dt$$

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{incoherent} = \frac{\sigma_{inc.}}{4\pi}\frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'}\int_{-\infty}^{\infty}\underline{\left\langle e^{-i\kappa R_{j}(0)}e^{-i\kappa R_j(t)}\right\rangle} e^{-i\omega t}dt$$

$$\sigma_{coh.} = 4\pi\bar{b}^2 \qquad \sigma_{inc.} = 4\pi(\overline{b^2} - \bar{b}^2)$$

Coherent/Incoherent Scattering

Coherent

Spatial and temporal correlations between <u>different</u> atoms

- Interference effects:
- Given by average of b
  Bragg scattering

Incoherent

Spatial and temporal correlations between <u>the same</u> atom

- Constant in *Q*
- Given by variations in b due to spin, disorder, random atomic motion....

Neutron diffraction on crystals

Elastic Scattering, Diffraction on Crystals

Starting point: Coherent elastic cross-section

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'} b_j b_{j'} \int_{-\infty}^{\infty} \left\langle e^{-i\kappa R_{j'}(0)} e^{-i\kappa R_j(t)} \right\rangle e^{-i\omega t}\mathrm{d}t$$

Sample: Single X-tal in thermal equilibrium, consider static correlations!

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

Drop the operator formalism for R_j as we look at static correlations

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

Information on the position of the atoms

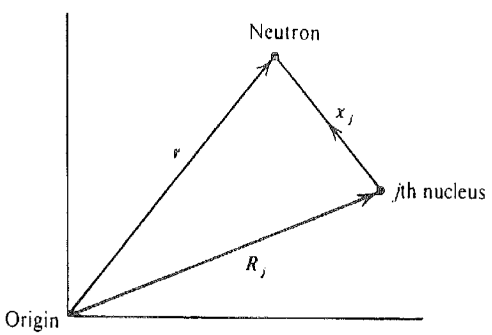
$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega_{inc.}} = (\langle b^2 \rangle - \langle b \rangle^2) \sum_{j=j'} e^{-i\kappa(R_j - R_j)} = N(\langle b^2 \rangle - \langle b \rangle^2)$$

Isotropic, constant elastic background

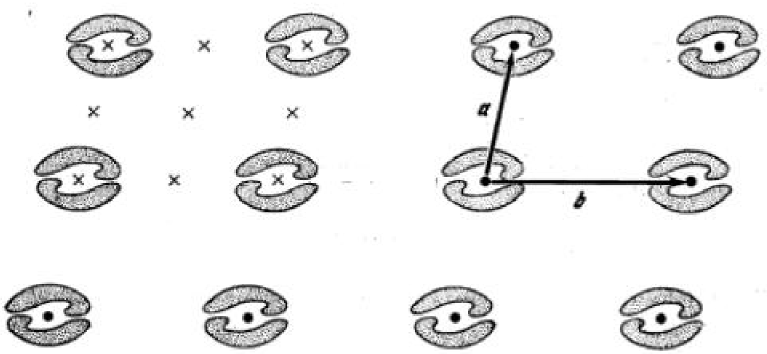
Elastic Scattering, Diffraction on Crystals: Lattice Sums

What we used:

Fig. 2.1 Coordinates of nucleus and neutron.



Our sample is a periodic crystal!



Crystal= Basis+lattice

$$\frac{d\sigma}{d\Omega}_{coh.el} = \langle b\rangle^2 N_0 \sum_T e^{i\vec{\kappa}\vec{T}} \quad \text{with} \quad \vec{R}_{j'} - \vec{R}_j = \vec{T}$$

Lattice sum: $$\sum_{\vec{l}} e^{i\vec{\kappa}\vec{l}} = \frac{(2\pi)^3}{v_0} \sum_{\vec{\tau}} \delta(\vec{\kappa} - \vec{\tau})$$

Real space lattice

Reciprocal space lattice

Lattice Sums & Reciprocal Lattice

Lattice sum: $$\sum_{\vec{l}} e^{i\vec{\kappa}\vec{l}} = \frac{(2\pi)^3}{v_0} \sum_{\vec{\tau}} \delta(\vec{\kappa} - \vec{\tau})$$

Real space lattice

Reciprocal space lattice

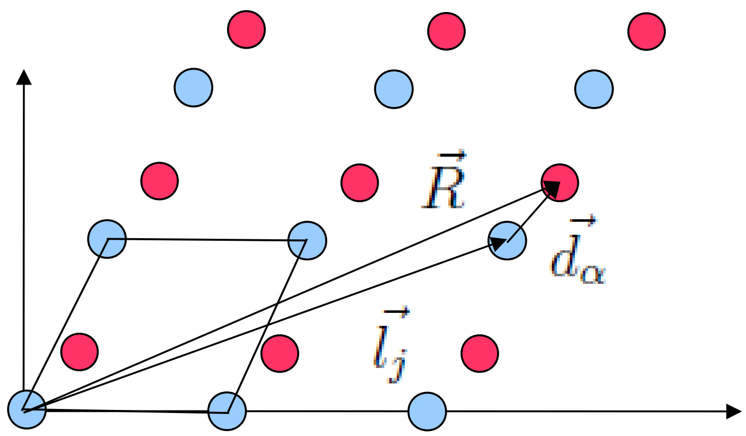
Braggs law: $$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}_{coh.el} = N_0 \frac{(2\pi)^3}{v_0} \langle b \rangle^2 \sum_{\vec{\tau}} \delta(\vec{\kappa} - \vec{\tau})$$

- Scattering occurs when κ meets a vector of the reciprocal lattice τ
- Reciprocal lattice vectors τ are perpendicular to a corresponding lattice plane indexed by the Miller index (h,k,l) and

$$d_{(h,k,l)} = \frac{2\pi}{|\vec{\tau}_{h,k,l}|}$$

Structure Factor

More than one atom in the unit cell: $\vec{R} = \vec{l_j} + \vec{d_\alpha}$



$\vec{l_j}$ Position of the j-th unit cell

$\vec{d_\alpha}$ Position of the α-th atom in the unit cell

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

$$S_{\vec{\tau}} = \sum_{\vec{d}} b_d e^{i\vec{\kappa}\vec{d}}$$

Structure factor (sum over one unit cell)

Master Formula for Neutron Diffraction

Going from operator to standard vector notation of R_j:

Neglected thermal vibration of atoms around their equilibrium position!

$$2W(\vec{\kappa}) = \frac{1}{3}\kappa^2 \langle u^2 \rangle$$

Debye-Waller factor describes mean dispalcement $\langle u^2 \rangle$

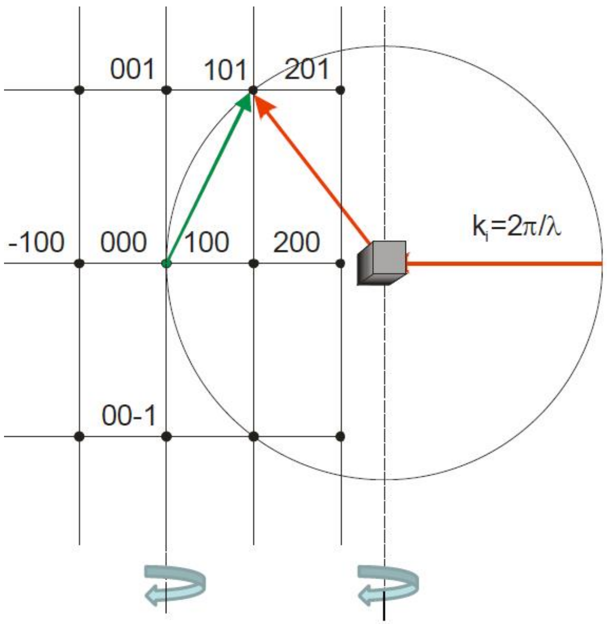
Master formula for elastic coherent scattering on crystals

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

Normalization   Debye-Waller   Structure factor   Bragg positions

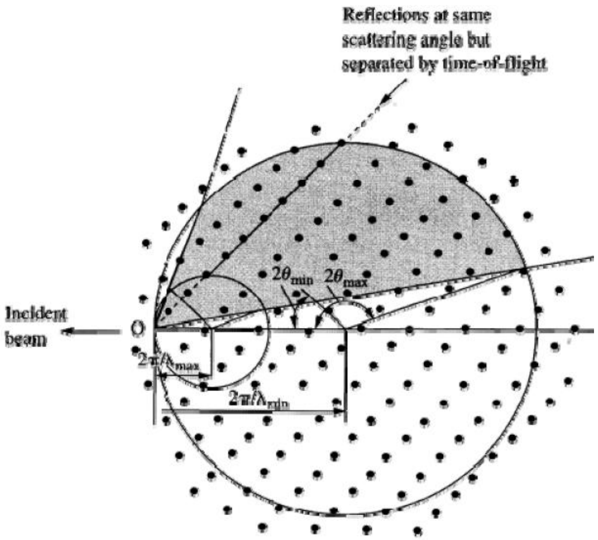
Monochromatic vs. TOF vs. Laue

Monochromatic beam



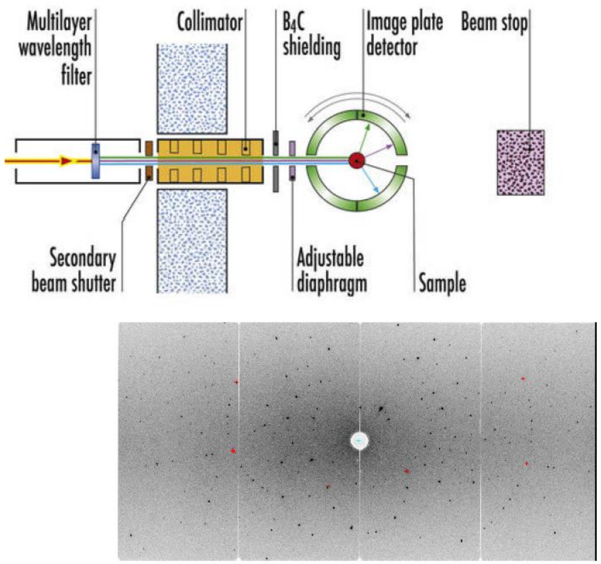
- Ewald construction
- Less intensity
- Rocking curve gives intensity of Bragg peak
- Clean data

Time-of-flight (TOF)



- Ewald construction for each wavelength in the beam
- Rocking curve distributed in time and detector
- Waste less neutrons

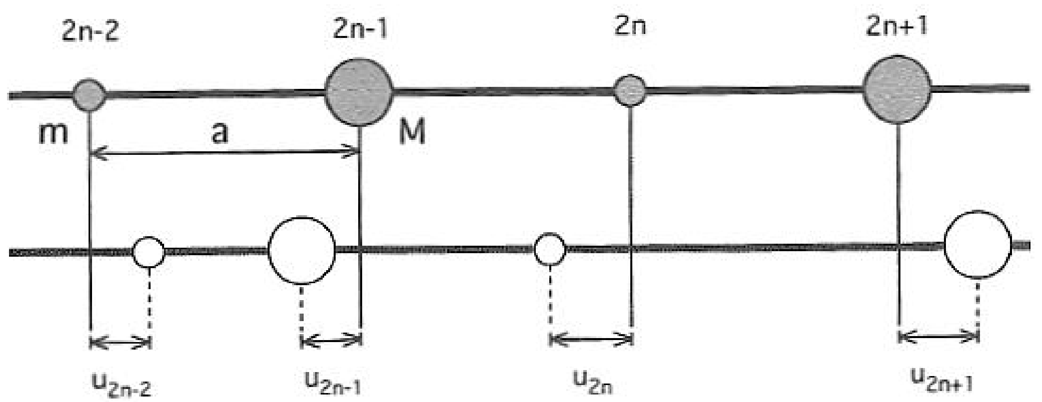
Laue (polychromatic beam)

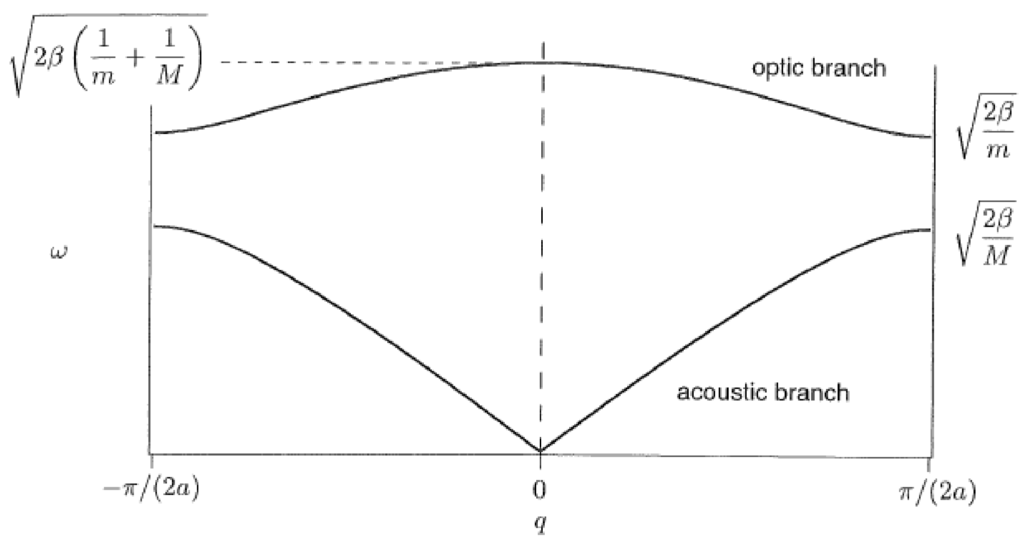


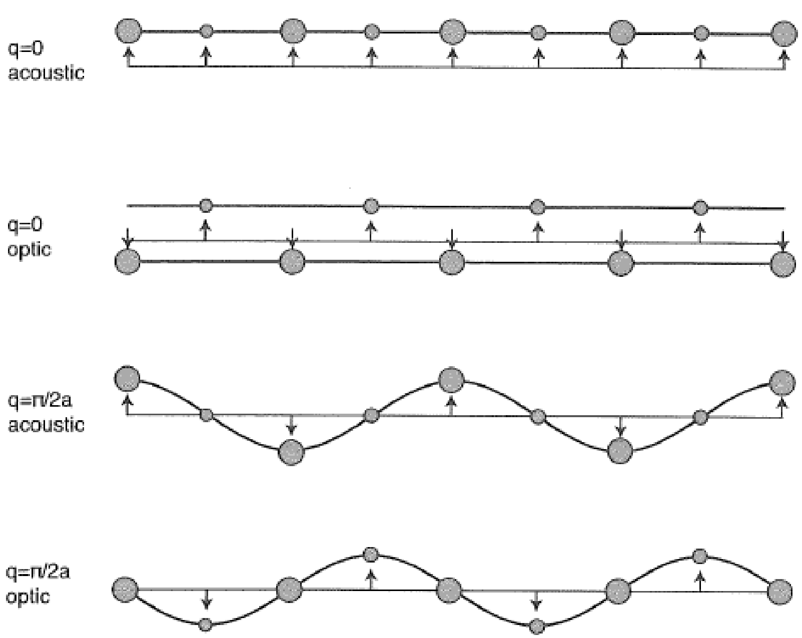
- Essentially white beam
- More Bragg peaks (not stronger)
- Hard to get intensities
- Large background

Inelastic neutron scattering: Coherent excitations - Phonons

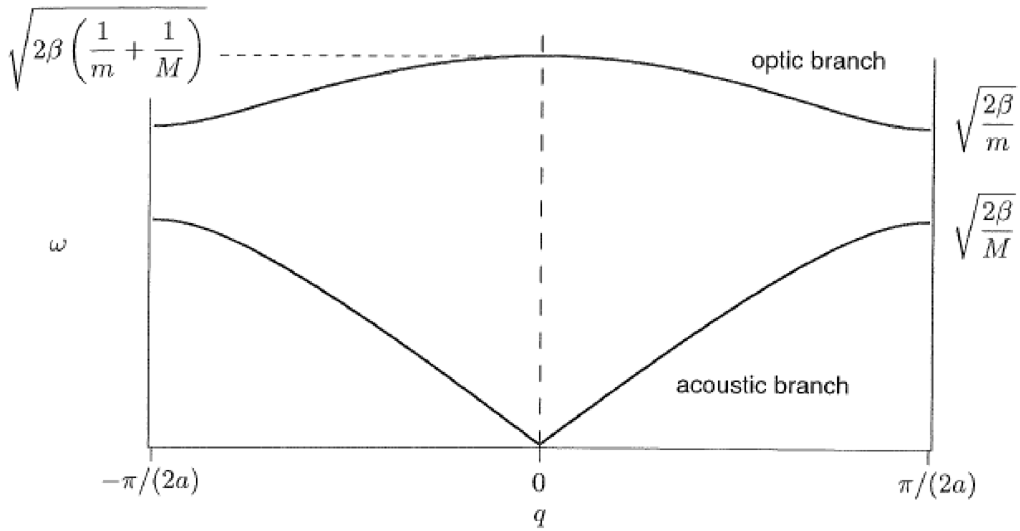
Basic idea: Phonons in a linear chain

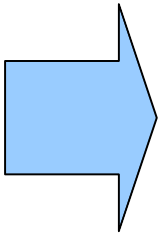






Properties of Phonons

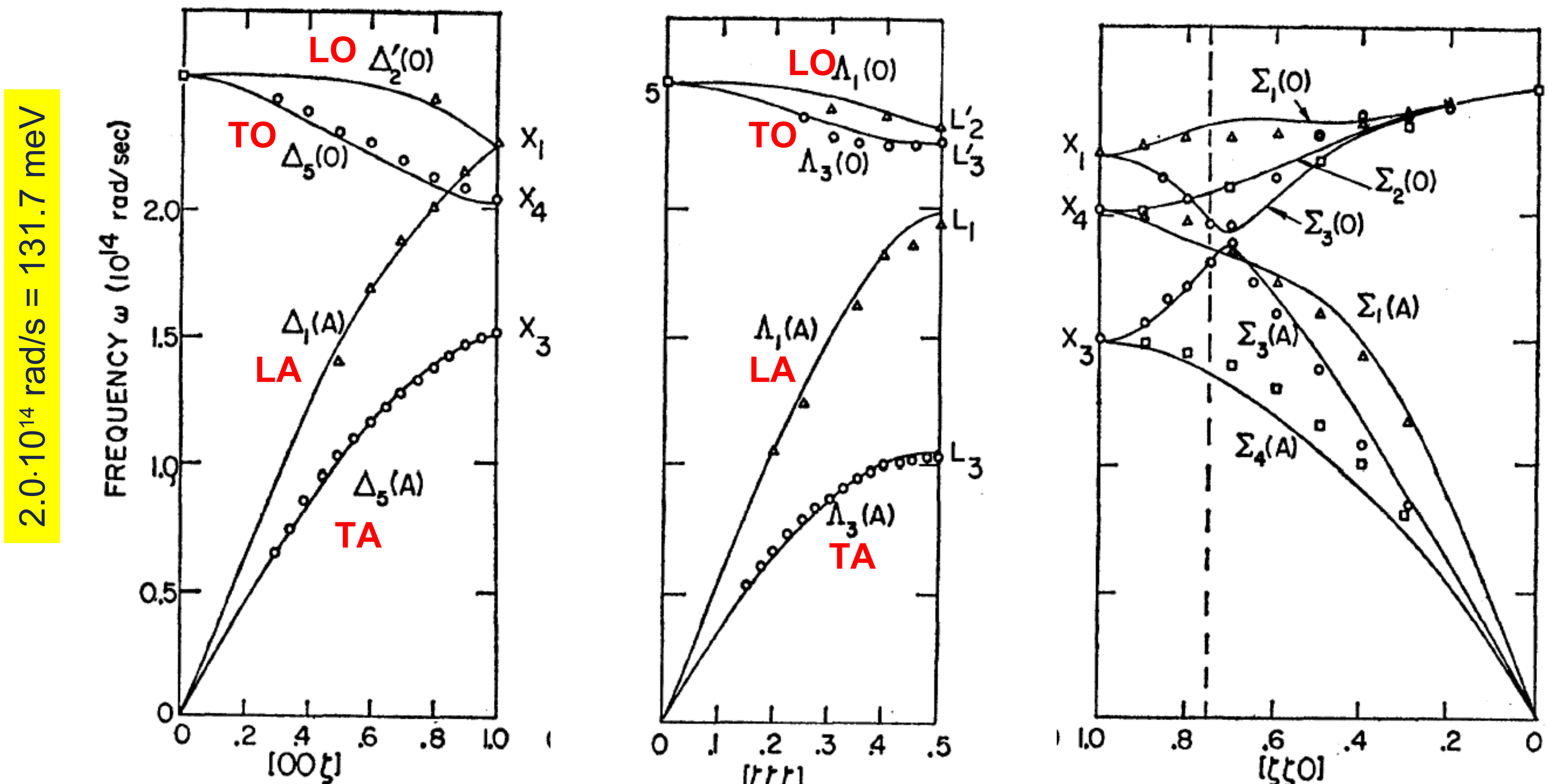




Collective excitation of atoms
“Live” in the first Brillouin zone
Description as dilute, non-interacting phonon gas”
Quasiparticles (3N phonons for N atoms)
Follow quasi-continuous dispersion relation
Obey Bose-Einstein statistics (specific heat!)
3P phonon branches (3p-3 optical, 2 transverse acoustic, 1 longitudinal acoustic for P-atomic basis)
QM picture (raising and lowering operator)

Inelastic scattering: Phonon dispersion

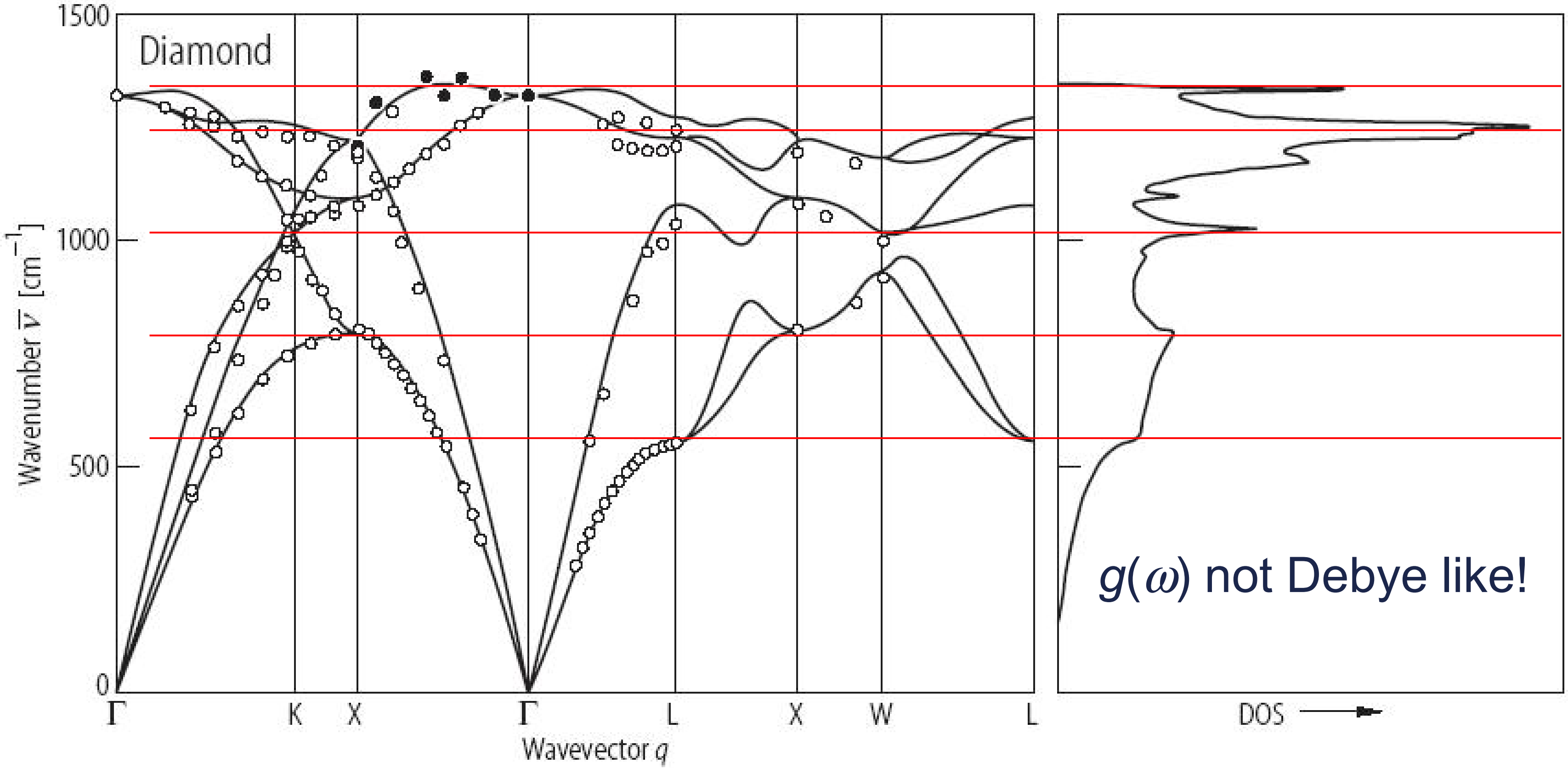
diamond: fcc, basis: (000), (¼ ¼ ¼) , T = 296 K, shell model fits



After: J. L. Warren et al., Phys. Rev. **158**, 805 (1967).

For X-ray data see: E. Burkel, Inelastic Scattering of X-Rays with Very High Energy Resolution, Springer Berlin (1991)

Reminder: Phonon DOS, specific specific heat, Debye approximation



Van Hove singularities are kinks or discontinuities in the density of states due to flat portions of the dispersion curves.

Inelastic scattering: Cross-section for phonon emission/absporption

Time dependendent position operator $\hat{\boldsymbol{R}}_j(t) = \boldsymbol{l}_j + \hat{\boldsymbol{u}}_j(t)$

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'} b_j b_{j'} e^{\imath\boldsymbol{Q}\cdot(\boldsymbol{l}_j-\boldsymbol{l}_{j'})}\int_{-\infty}^{\infty}\langle e^{-\imath\boldsymbol{Q}\cdot\hat{\boldsymbol{u}}_{j'}(0)} e^{\imath\boldsymbol{Q}\cdot\hat{\boldsymbol{u}}_j(t)}\rangle e^{-\imath\omega t}\mathrm{d}t.$$

Displacement expressed in terms of normal mode (QM harmonic oscillator)

$$\hat{\boldsymbol{u}}_j(t) = \sqrt{\frac{\hbar}{2MN}}\sum_{s,\boldsymbol{q}}\frac{\boldsymbol{e}_s(\boldsymbol{q})}{\sqrt{\omega_s(\boldsymbol{q})}}\left(\hat{a}_s(\boldsymbol{q})e^{\imath[\boldsymbol{q}\cdot\boldsymbol{l}_j-\omega_s(\boldsymbol{q})t]} + \hat{a}_s^+(\boldsymbol{q})e^{-\imath[\boldsymbol{q}\cdot\boldsymbol{l}_j-\omega_s(\boldsymbol{q})t]}\right)$$

Ladder operators of QM oscillator

$$\hat{a}_s^+(\boldsymbol{q})|\lambda_n\rangle = \sqrt{n+1}|\lambda_{n+1}\rangle,$$
$$\hat{a}_s(\boldsymbol{q})|\lambda_n\rangle = \sqrt{n}|\lambda_{n-1}\rangle,$$
$$\langle\lambda_n|\hat{a}_s(\boldsymbol{q})\hat{a}_s^+(\boldsymbol{q})|\lambda_n\rangle = n_s(\boldsymbol{q})+1$$
$$\langle\lambda_n|\hat{a}_s^+(\boldsymbol{q})\hat{a}_s(\boldsymbol{q})|\lambda_n\rangle = n_s(\boldsymbol{q}),$$

Bose-Einstein statistics of phonons

$$n_s(\boldsymbol{q}) = \left(\exp\left(\frac{\hbar\omega_s(\boldsymbol{q})}{k_B T}\right)-1\right)^{-1}$$

Inelastic scattering: Cross-section for phonon emission/absportion

Abbreviaton of the exponents

$$\hat{A} = -\imath \boldsymbol{Q} \cdot \hat{\boldsymbol{u}}_{j'}(0) = -\imath \sum_{s,\boldsymbol{q}} \left(\alpha_s(\boldsymbol{q})\hat{a}_s(\boldsymbol{q}) + \alpha_s^*(\boldsymbol{q})\hat{a}_s^+(\boldsymbol{q})\right) \qquad \alpha_s(\boldsymbol{q}) = \sqrt{\frac{\hbar}{2MN}} \sum_{s,\boldsymbol{q}} \frac{\boldsymbol{Q}\cdot \boldsymbol{e}_s(\boldsymbol{q})}{\sqrt{\omega_s(\boldsymbol{q})}} e^{\imath \boldsymbol{q}\cdot \boldsymbol{l}_{j'}},$$

$$\hat{B} = \imath \boldsymbol{Q} \cdot \hat{\boldsymbol{u}}_j(t) = \imath \sum_{s,\boldsymbol{q}} \left(\beta_s(\boldsymbol{q})\hat{a}_s(\boldsymbol{q}) + \beta_s^*(\boldsymbol{q})\hat{a}_s^+(\boldsymbol{q})\right), \qquad \beta_s(\boldsymbol{q}) = \sqrt{\frac{\hbar}{2MN}} \sum_{s,\boldsymbol{q}} \frac{\boldsymbol{Q}\cdot \boldsymbol{e}_s(\boldsymbol{q})}{\sqrt{\omega_s(\boldsymbol{q})}} e^{\imath [\boldsymbol{q}\cdot \boldsymbol{l}_j - \omega_s(\boldsymbol{q})t]}$$

Taylor expansion of the time evolution

$$e^{\langle \hat{A}\hat{B}\rangle} = \underline{1} + \underline{\langle \hat{A}\hat{B}\rangle} + \frac{1}{2}\langle \hat{A}\hat{B}\rangle^2 + \cdots + \frac{1}{n!}\langle \hat{A}\hat{B}\rangle^n + \ldots$$

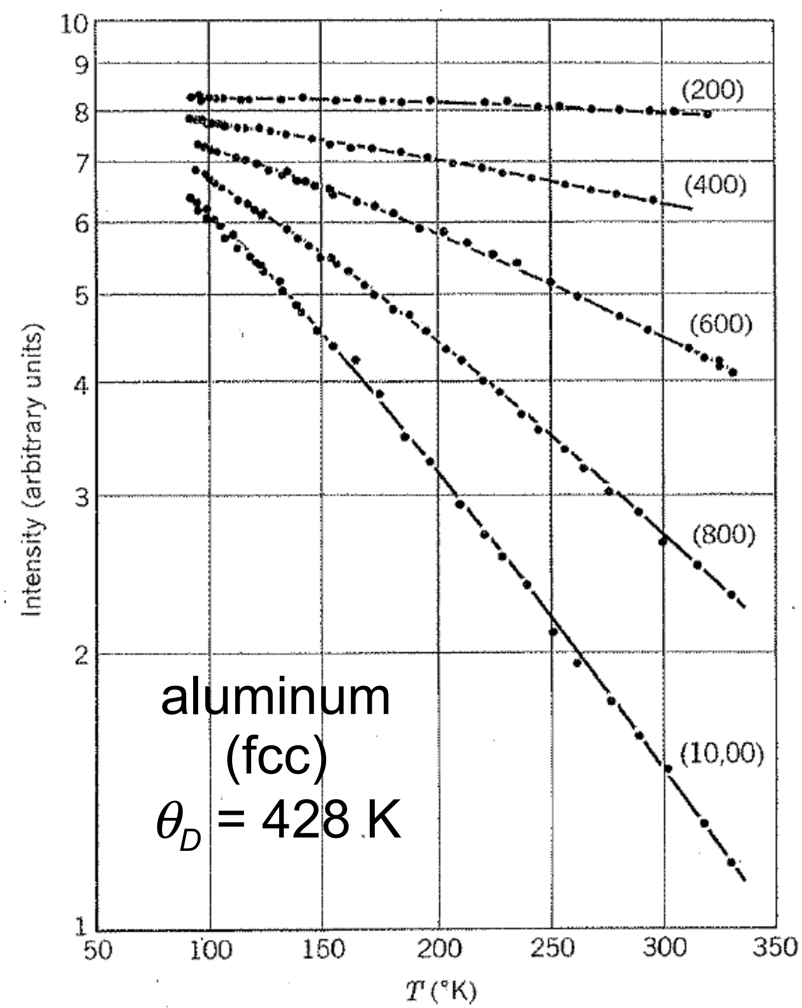
Elastic scattering (+ Debye Waller factor)

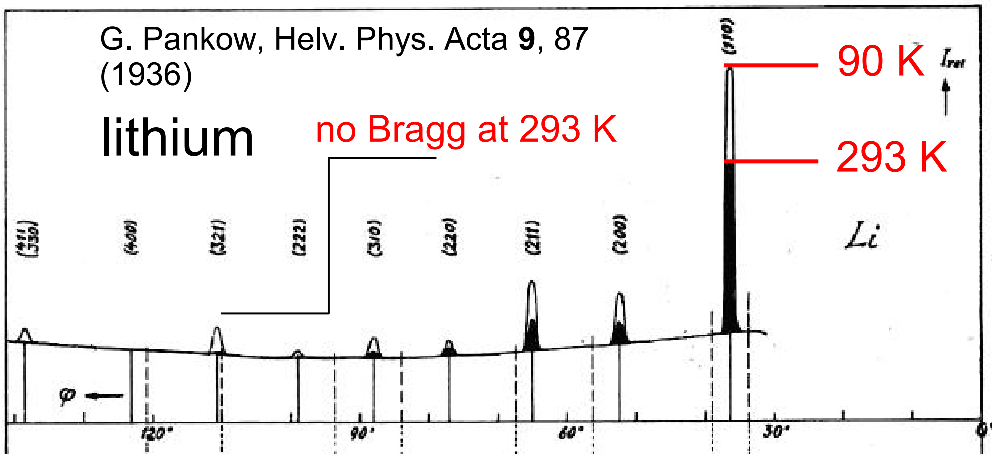
Single phonon processes

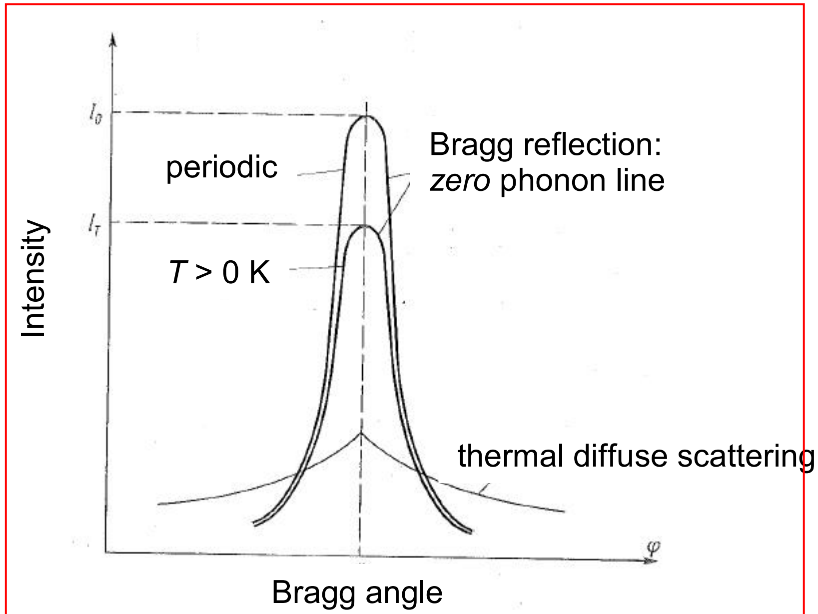
Write linear term in term of sample properties (QM harmonic oscillator!)

$$\langle \lambda_n | \hat{A}\hat{B} | \lambda_n \rangle = \langle \lambda_n | \sum_{s,\boldsymbol{q}} [\alpha_s(\boldsymbol{q})\beta_s^*(\boldsymbol{q})\hat{a}_s(\boldsymbol{q})\hat{a}_s^+(\boldsymbol{q}) + \alpha_s^*(\boldsymbol{q})\beta_s(\boldsymbol{q})\hat{a}_s^+(\boldsymbol{q})\hat{a}_s(\boldsymbol{q})] | \lambda_n \rangle$$

Inelastic scattering: Debye Waller factor: Aluminium/Lithium







Master formula for coherent inelastic scattering

Consider only coherent scattering (j≠j')

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{1}{4\pi M} \cdot \frac{k'}{k} \langle b \rangle^2 e^{-2W(\boldsymbol{Q})} \sum_{s,\boldsymbol{q}} \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s(\boldsymbol{q}))^2}{\omega_s(\boldsymbol{q})}$$

$$\times \left[(n_s(\boldsymbol{q})+1) \sum_{\boldsymbol{l}} e^{\imath(\boldsymbol{Q}-\boldsymbol{q})\cdot\boldsymbol{l}} \int_{-\infty}^{\infty} \mathrm{d}t e^{\imath(\omega_s(\boldsymbol{q})-\omega)t} \right.$$

$$\left. + n_s(\boldsymbol{q}) \sum_{\boldsymbol{l}} e^{\imath(\boldsymbol{Q}+\boldsymbol{q})\cdot\boldsymbol{l}} \int_{-\infty}^{\infty} \mathrm{d}t e^{-\imath(\omega_s(\boldsymbol{q})+\omega)t} \right].$$

Convert integrals to delta functions using lattice sums (as for diffraction)

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{4\pi^3}{v_0 M} \cdot \frac{k'}{k} \langle b \rangle^2 e^{-2W(\boldsymbol{Q})} \sum_{s,\boldsymbol{q}} \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s(\boldsymbol{q}))^2}{\omega_s(\boldsymbol{q})}$$

$$\times \left[\underline{(n_s(\boldsymbol{q})+1)\delta(\omega-\omega_s(\boldsymbol{q})) \sum_{\boldsymbol{\tau}} \delta(\boldsymbol{Q}-\boldsymbol{q}-\boldsymbol{\tau})} \right.$$

Phonon emission

$$\left. \underline{+ n_s(\boldsymbol{q})\delta(\omega+\omega_s(\boldsymbol{q})) \sum_{\boldsymbol{\tau}} \delta(\boldsymbol{Q}+\boldsymbol{q}-\boldsymbol{\tau})} \right].$$

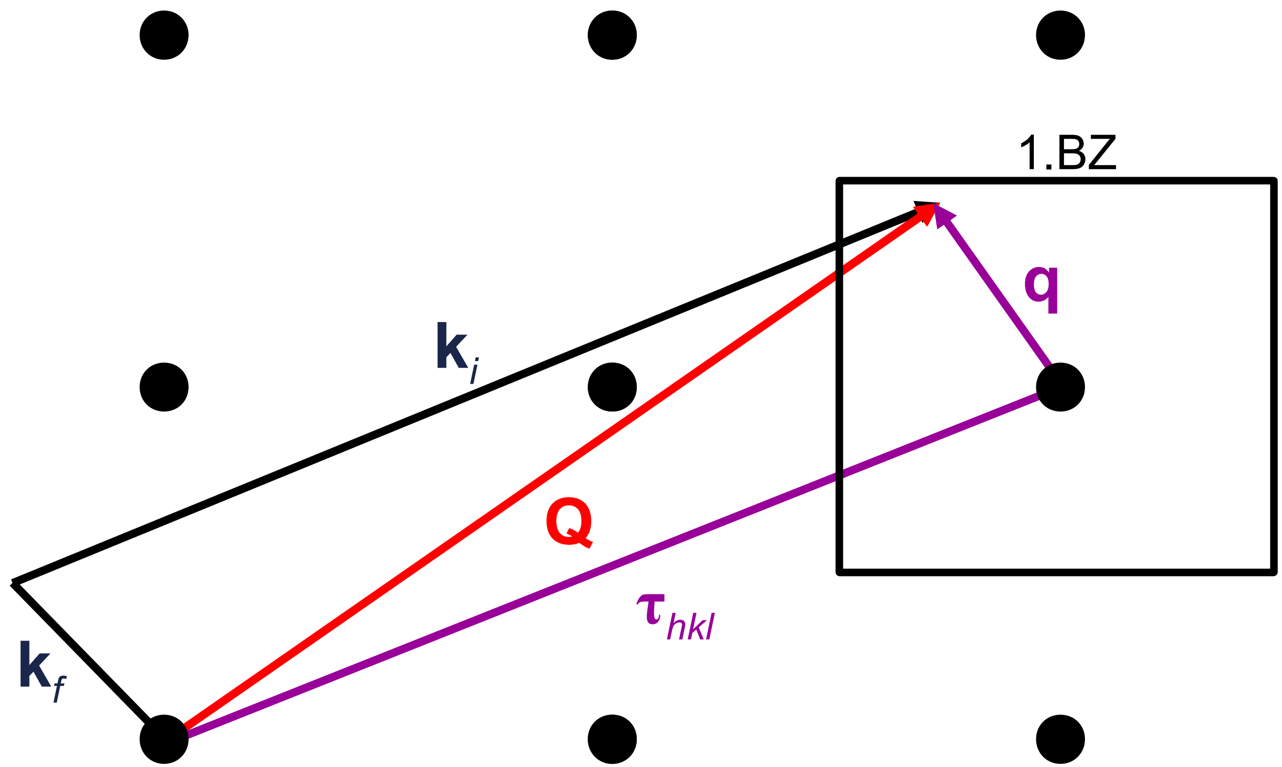
Phonon absorption

Scattering „triangle“ for inelastic scattering

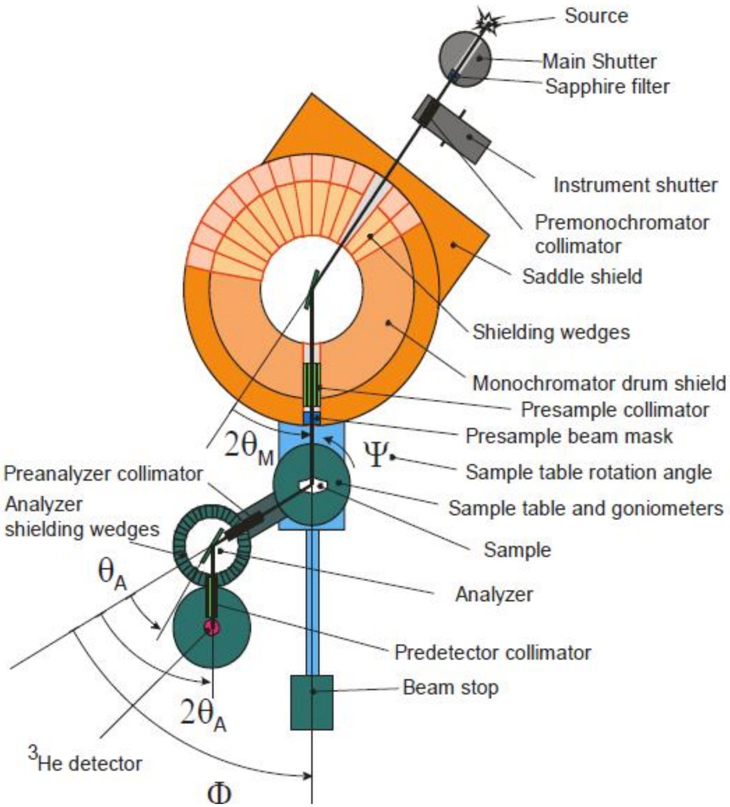
Example:
phonon creation

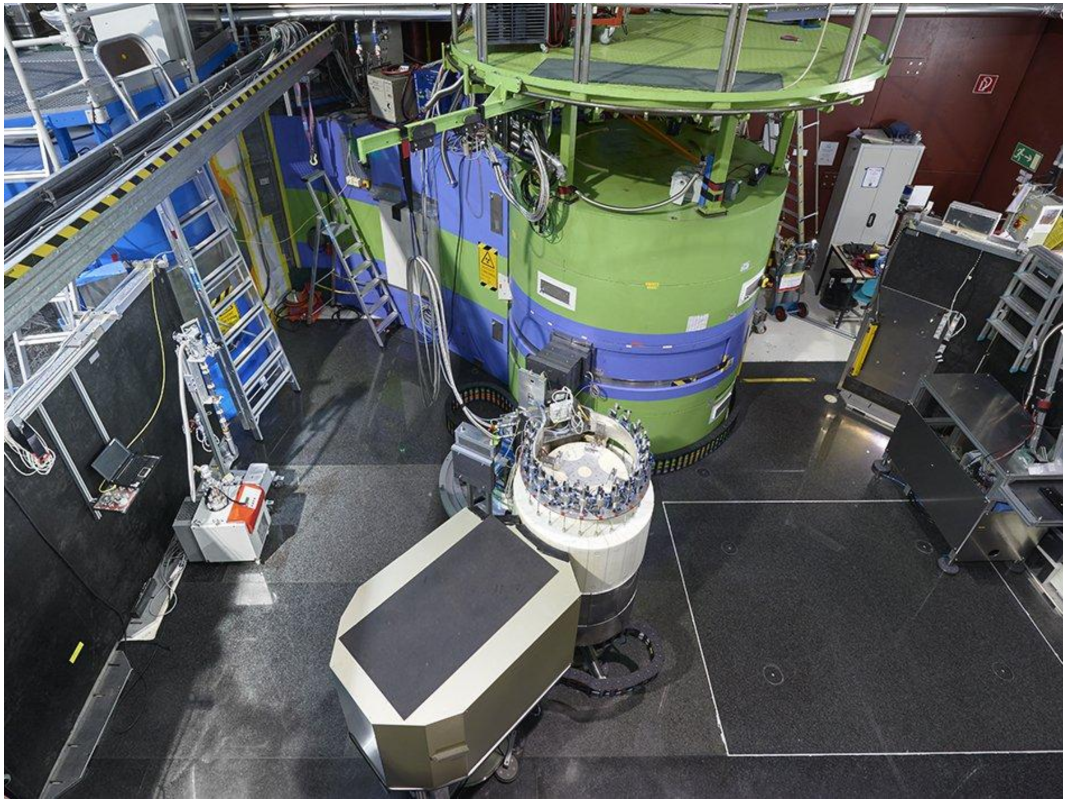
$$\omega = \omega_s(\mathbf{q})$$

$$\Rightarrow \; k_i^2 > k_f^2$$



Triple axis spectrometer: Workinghorse for phonons and magnons





„Working horse" for phonons/magnons in magnetism/superconductivity
Clean data at a fixed point in momentum/energy space
Slow, wasting a lot of neutrons

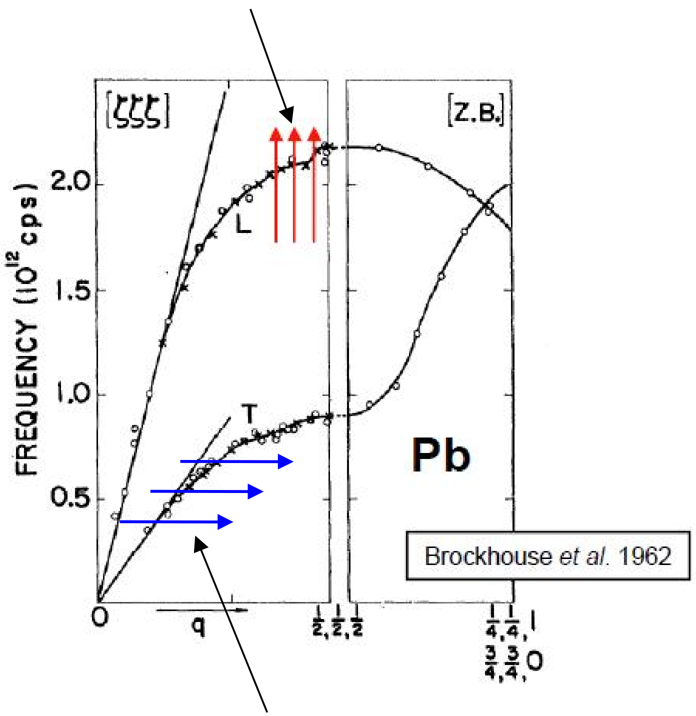
Cold TAS (PANDA)
Best energy resolution: 20µeV
Energy transfer <20meV
Momentum transfer <6Å^{-1}

Thermal TAS (PUMA)
Best energy resolution: 600µeV
Energy transfer <100meV
Momentum transfer <12Å^{-1}

Triple axis spectrometer: Scattering „triangle" S(q,w)

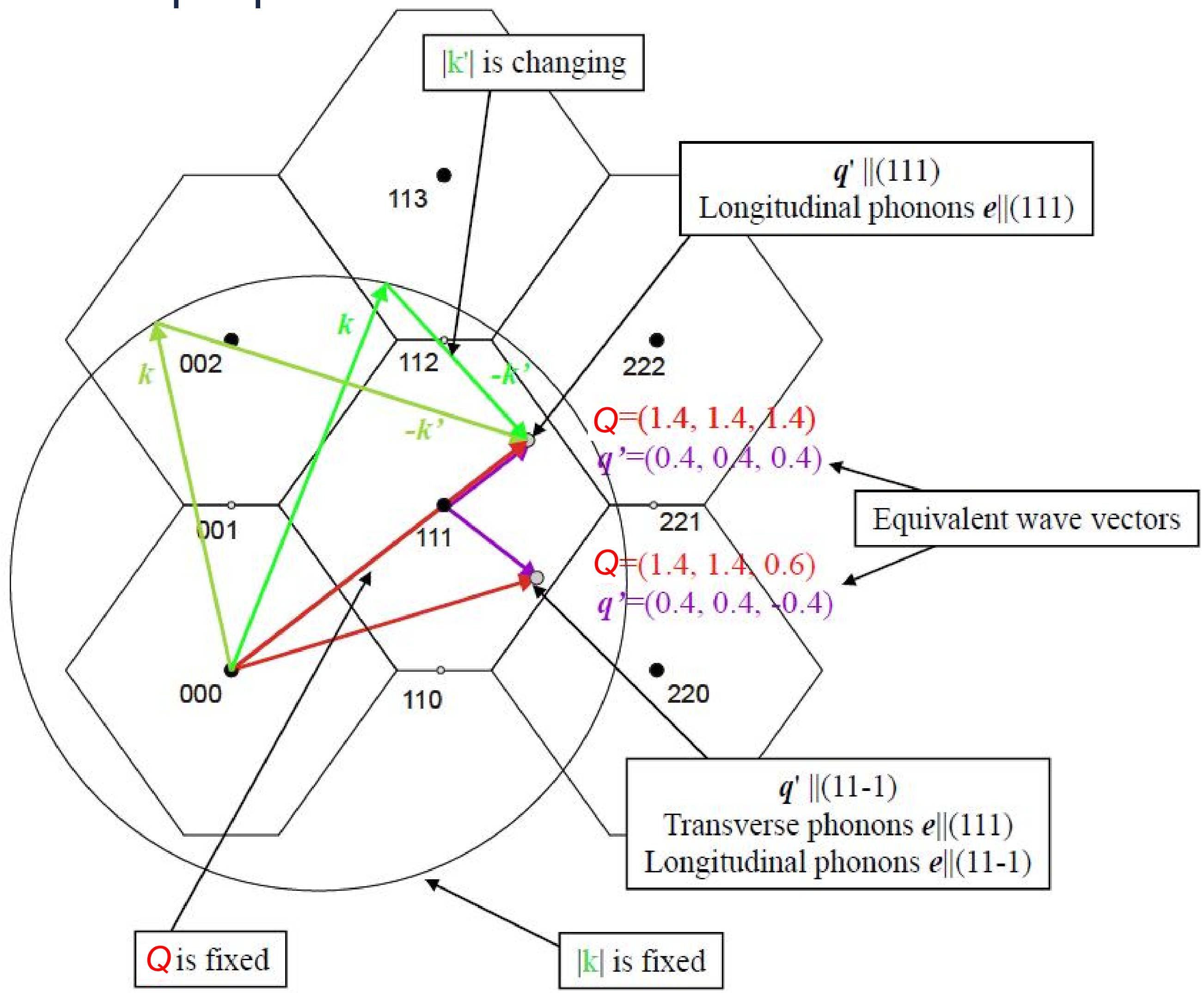
Scanning S(Q,w):

constant *q* scan



constant *E* scan

Fixed k_i constant *q* scan: Measure the properties of normal vibrational modes



Incoherent inelastic: Phonon DOS

Inelastic incoherent scattering: Phonon DOS

Now consider incoherent scattering (j=j')

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{k'}{k}\frac{1}{2\pi\hbar}e^{-2W(\boldsymbol{Q})}\sum_j b_j^2 \int_{-\infty}^{\infty} e^{\langle\hat{A}\hat{B}\rangle} e^{-\imath\omega t}\mathrm{d}t.$$

Similar to the coherent part, only consider the linear term in the Taylor expansion

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{1}{2M}\frac{k'}{k}\left(\langle b^2\rangle - \langle b\rangle^2\right)e^{-2W(\boldsymbol{Q})}\sum_{s,\boldsymbol{q}}\frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s(\boldsymbol{q}))^2}{\omega_s(\boldsymbol{q})}$$
$$\times\left[(n_s(\boldsymbol{q})+1)\delta\left(\omega-\omega_s(\boldsymbol{q})\right)+n_s(\boldsymbol{q})\delta\left(\omega+\omega_s(\boldsymbol{q})\right)\right]$$

Phonon emission

Phonon absorption

Inelastic incoherent scattering: Phonon DOS

Compare to coherent part:

Energy conservation is fulfilled
No momentum conservation is fulfilled

All phonons with energy ω_s contribute!

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

With phonon DOS $g(\omega)$

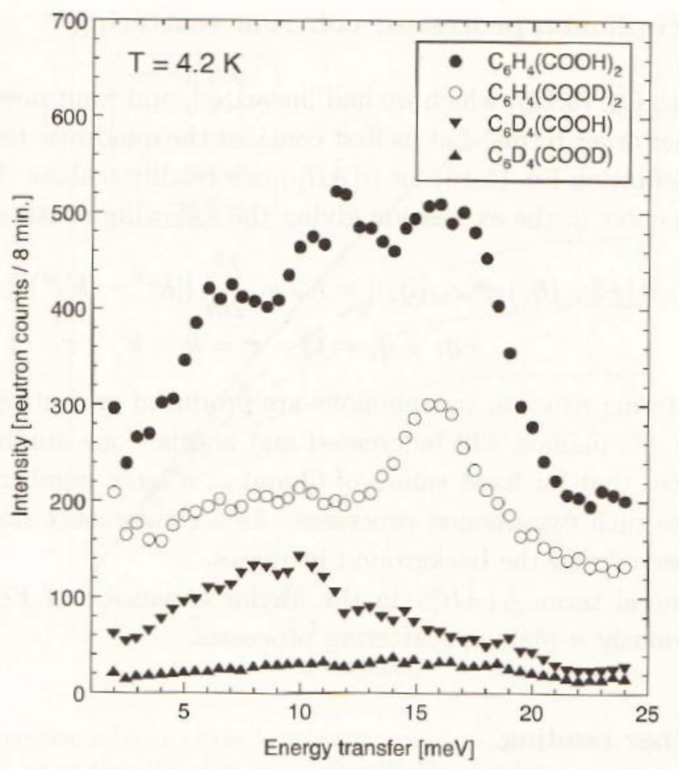
$$\int_0^\infty g(\omega)\mathrm{d}\omega = 3N$$

Inelastic incoherent scattering: Phonon DOS

For a (cubic) Bravais lattice only

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{1}{12M}\frac{k'}{k}\left(\langle b^2\rangle - \langle b\rangle^2\right) e^{-W(\boldsymbol{Q})} Q^2 \times \frac{g(\omega)}{\omega}\cdot\left[\coth\frac{\hbar\omega}{2k_B T} \pm 1\right].$$

Inelastic incoherent scattering directly measures phonon DOS g(ω)



Correlation functions of neutron scattering

Starting point: General cross-section

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'} b_j b_{j'} \int_{-\infty}^{\infty} \langle e^{-i\boldsymbol{Q}\hat{\boldsymbol{R}}_j\,(0)} e^{-\boldsymbol{Q}\hat{\boldsymbol{R}}_{j'}(t)}\rangle e^{-i\omega t} \mathrm{dt}$$

$$I(\boldsymbol{Q},t) = \frac{1}{N} \qquad \int_{-\infty}^{\infty} \langle e^{-i\boldsymbol{Q}\hat{\boldsymbol{R}}_j\,(0)} e^{-\boldsymbol{Q}\hat{\boldsymbol{R}}_{j'}(t)}\rangle$$

⇨ Intermediate scattering function

Fourier transform (space) $G(\boldsymbol{r},t) = \frac{1}{(2\pi)^3}\int I(\boldsymbol{Q},t) e^{-i\boldsymbol{Q}\boldsymbol{r}} \mathrm{dQ}$

⇨ Pair correlation function

Fourier transform (time) $S(\boldsymbol{Q},\omega) = \frac{1}{(2\pi\hbar)}\int I(\boldsymbol{Q},t) e^{-i\omega t} \mathrm{dt}$

⇨ Scattering function, directly connected to cross-section

Physical meaning of pair correlation function G(r,t)

$$G(\boldsymbol{r},t) = \frac{1}{N}\sum_{j,j'}\int \langle \delta(\boldsymbol{r}' - \hat{\boldsymbol{R}}_{j'}(0))\delta(\boldsymbol{r}' + \boldsymbol{r} - \hat{\boldsymbol{R}}_j(t))\rangle \mathrm{d}r'$$

Correlation between atom j' at time t=0 at position r' and atom j at time t=t and position r'+r

Splits up in

$$G_s(\boldsymbol{r},t) = \frac{1}{N}\sum_{j}\int \langle \delta(\boldsymbol{r}' - \hat{\boldsymbol{R}}_{j}(0))\delta(\boldsymbol{r}' + \boldsymbol{r} - \hat{\boldsymbol{R}}_j(t))\rangle \mathrm{d}r'$$

Self correlation function

$$G_d(\boldsymbol{r},t) = \frac{1}{N}\sum_{j\neq j'}\int \langle \delta(\boldsymbol{r}' - \hat{\boldsymbol{R}}_{j'}(0))\delta(\boldsymbol{r}' + \boldsymbol{r} - \hat{\boldsymbol{R}}_j(t))\rangle \mathrm{d}r'$$

Correlation function

Coherent and incoherent part

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega}\right)_{coh} = N\frac{k'}{k}\langle b\rangle^2 S_{coh}(\boldsymbol{Q},\omega)$$

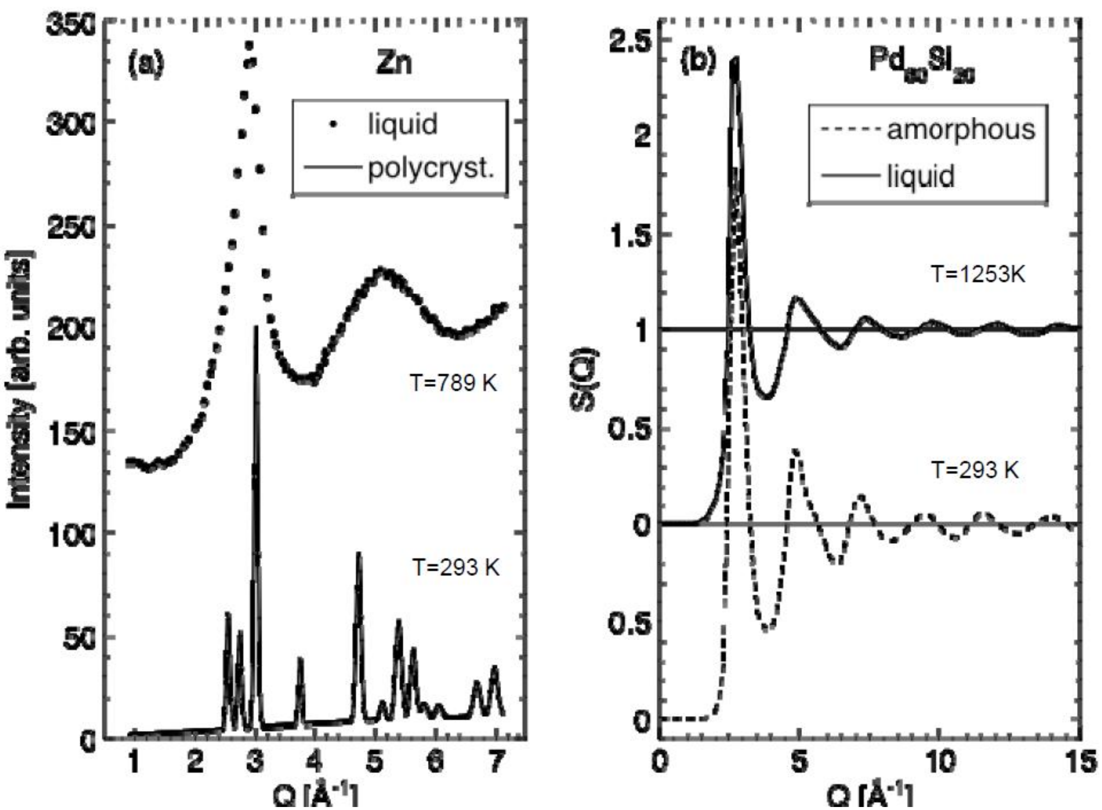
$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega}\right)_{inc} = N\frac{k'}{k}(\langle b^2\rangle - \langle b\rangle^2) S_{inc}(\boldsymbol{Q},\omega)$$

Neutron scattering on liquids and amorphous materials

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

Liquid (amorphhous) sample ⟺ Crystalline sample

⇨ Similar density, no LRO, only short range correlations

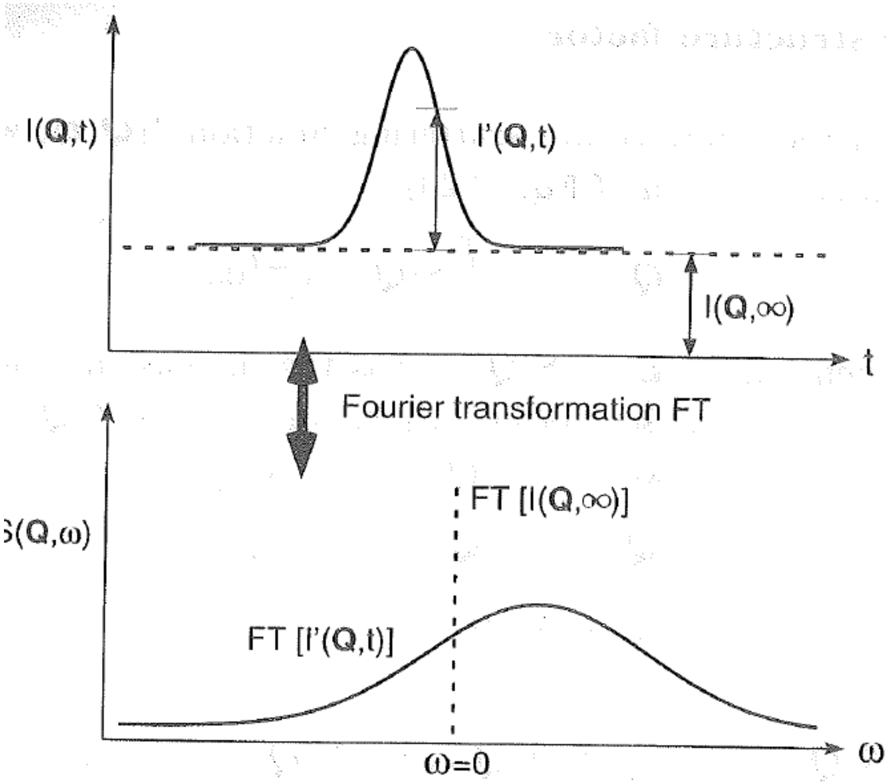


Static structure factor

Start with I(Q,t) and split into into two parts:

$$I(\boldsymbol{Q},t) = \hbar \int S(\boldsymbol{Q},\omega) e^{\omega t} \mathrm{d}\omega$$

$$I(\boldsymbol{Q},t) = I(\boldsymbol{Q},\infty) + I'(\boldsymbol{Q},t)$$



$$S(\boldsymbol{Q},\omega) = \underbrace{\frac{1}{\hbar}\delta(\omega) I(\boldsymbol{Q},\infty)}_{\text{Elastic part}} + \underbrace{\frac{1}{2\pi\hbar}\int I'(\boldsymbol{Q},t) e^{-i\omega t}\mathrm{d}t}_{\text{Inelastic part}}$$

Elastic part　　Inelastic part

➡ Infinite time correlations

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

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

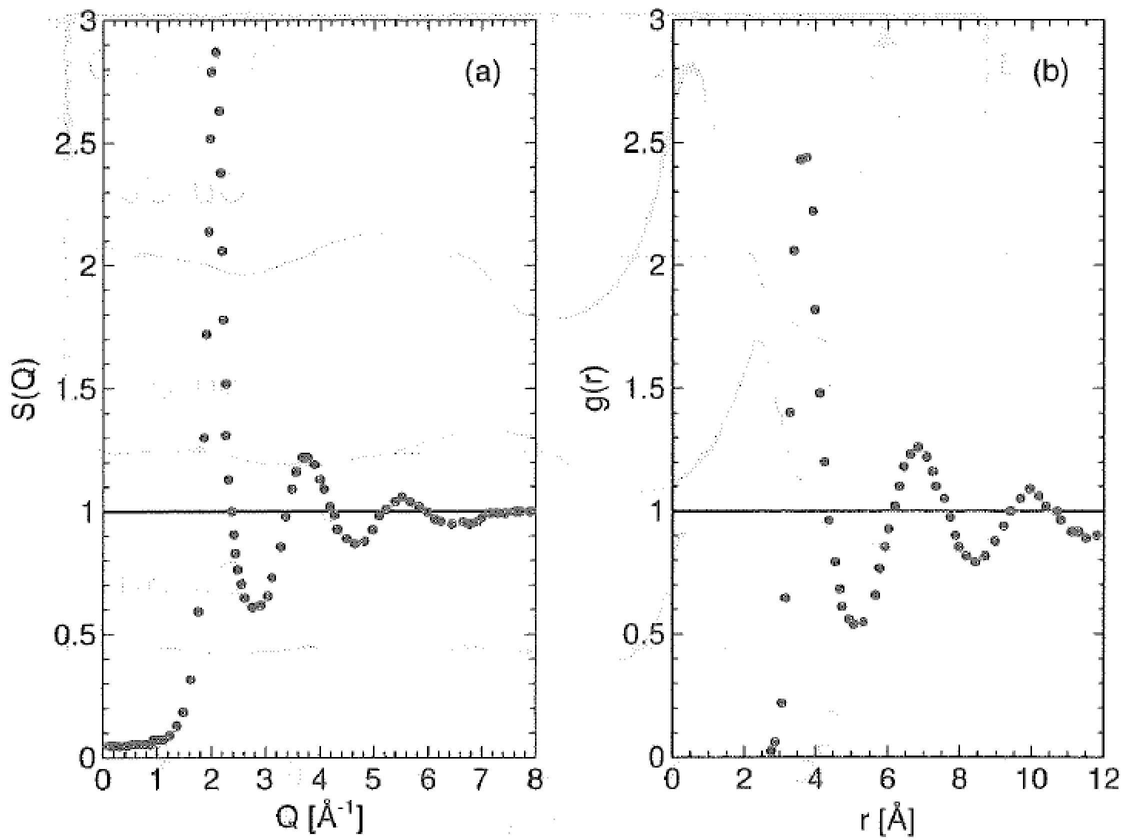
Elastic scattering from liquids

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

g(r): pair correlation function

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

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

Dynamic structure factor: Looking at diffusive processes

S(Q,w) ⇔ FT ⇔ G(r,t)

Liquid state

Large values of r,t
small values of Q,w

⇒ Long time behaviour

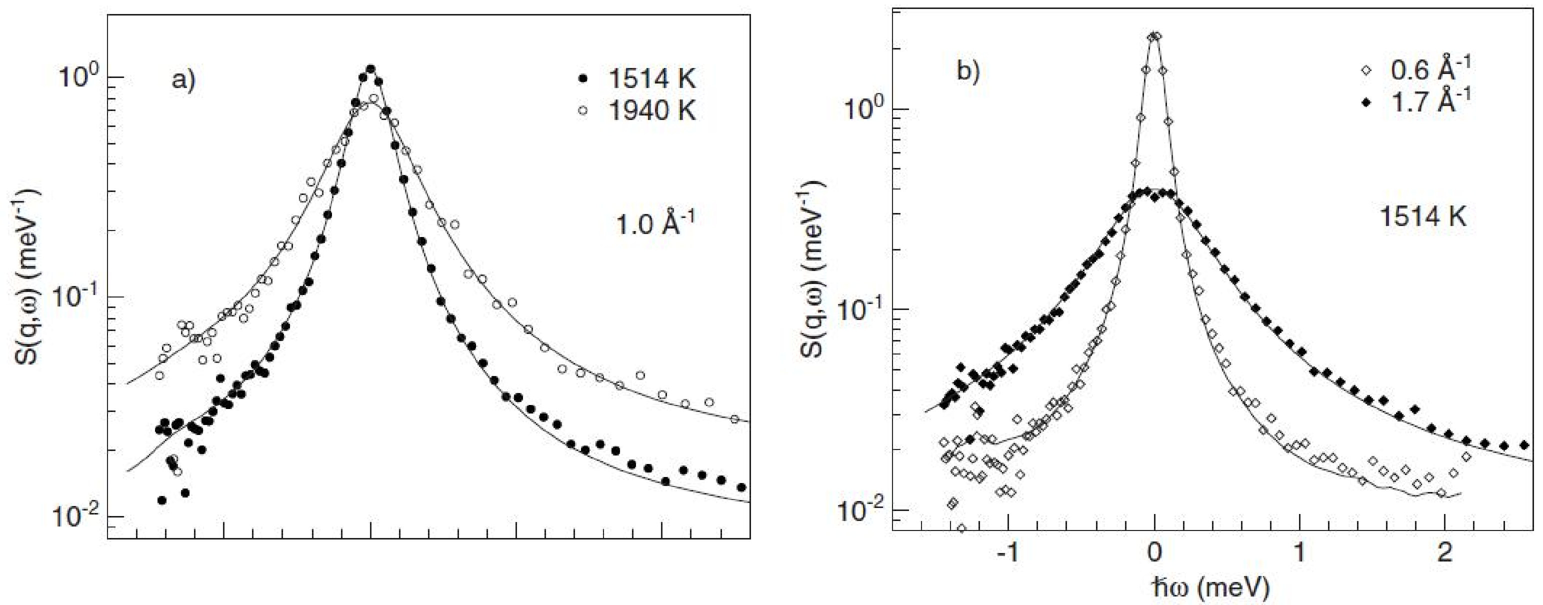
⇒ Diffusion

Small values of r,t
large values of Q,w
$G_s(r,t)$ peaked at t=0 for small r

⇒ Short time behaviour

⇒ Ideal gas

$t \geq 10^{-12}$s ⇒ QENS, peaked at w=0 $t \leq 10^{-13}$s



Diffusive behaviour (low Q):

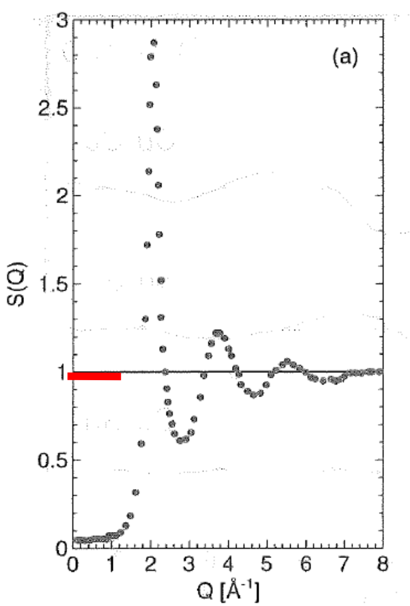
Fick´s law: $$\frac{\partial n(\boldsymbol{r},t)}{\partial t} = D\nabla^2 n(\boldsymbol{r},t)$$ Diffusion constant D

Incoherent scattering: $$S_{\mathrm{inc}}(Q,\omega) = \frac{1}{2\pi\hbar}\int I_s(Q,t)e^{-\imath\omega t}\mathrm{d}t = \frac{1}{\pi\hbar}\frac{DQ^2}{\omega^2+(DQ^2)^2}$$

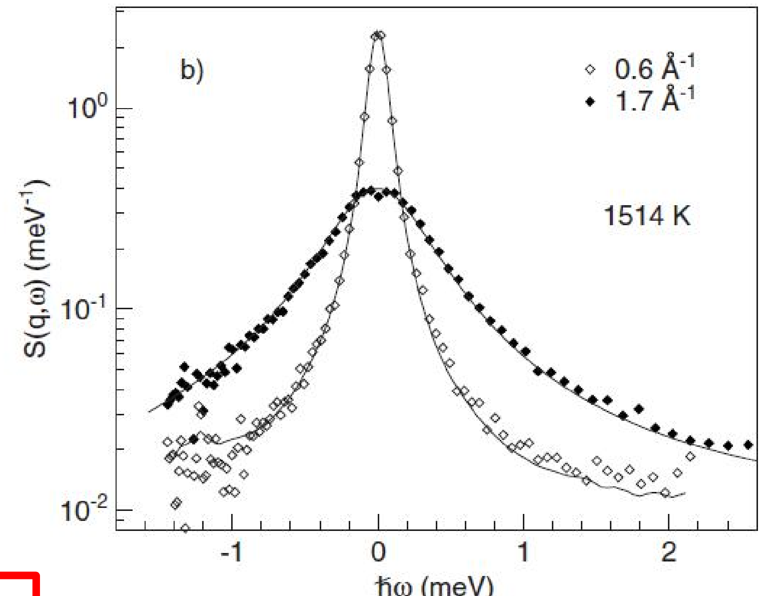
Lorentzian function centered at w=0

$$\Gamma^{\mathrm{fwhm}} = 2\hbar DQ^2$$

Valid only for q^{-1}» mean distance



Otherwise: microscopic details!



QENS ⟺ Diffusion constant

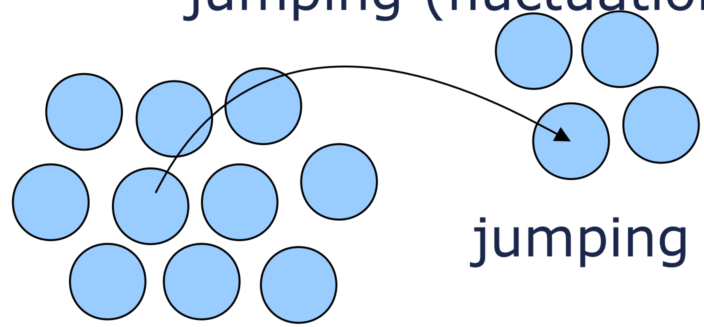
Width increases with Q

Diffusive behaviour (higher Q):

⇨ Macroscopic model fails for $q^{-1} \sim$ mean distance

jumping (fluctuation) time t_1

Miccroscopic model:
Jump diffusion
$t_0 \gg t_1$



jumping vector l_0

equilibrium pos. r_0
relaxation time t_0

$$S_{\mathrm{inc}}(Q,\omega) = \frac{1}{2\pi\hbar}\int I_s(Q,t)e^{-\imath\omega t}\mathrm{d}t$$

$$= \frac{1}{2\pi\hbar}\int e^{-f(Q)t}\cos\omega t = \frac{1}{\pi\hbar}\frac{f(Q)}{\omega^2 + f^2(Q)}$$

Again:Lorentzian function centered at w=0

$$\Gamma^{\mathrm{fwhm}} = 2\hbar f(Q) \qquad f(Q) = \frac{1}{\tau_0}\left(1 - \frac{1}{\left(1+(Ql_0)^2\right)^2}\right)$$