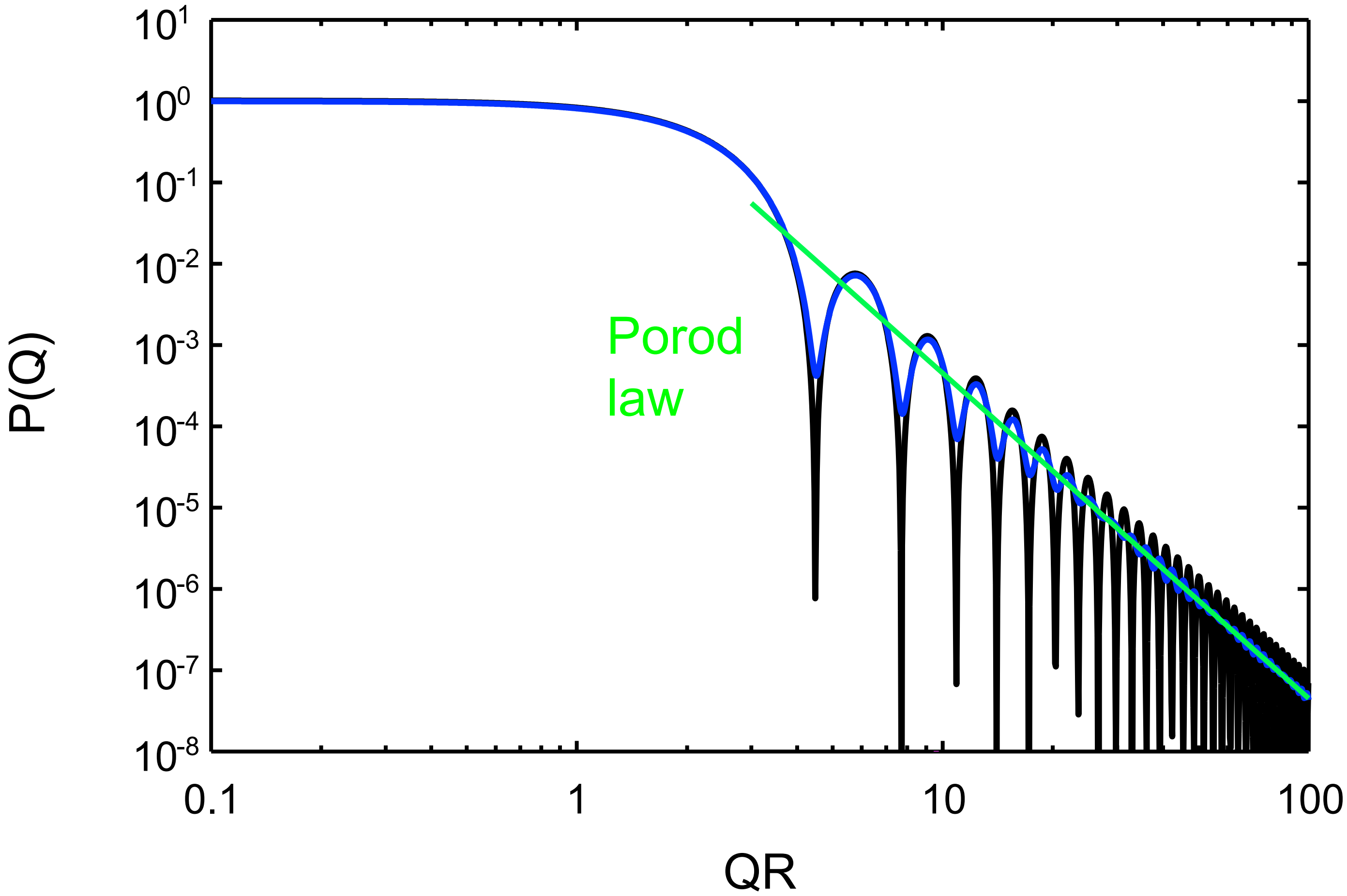
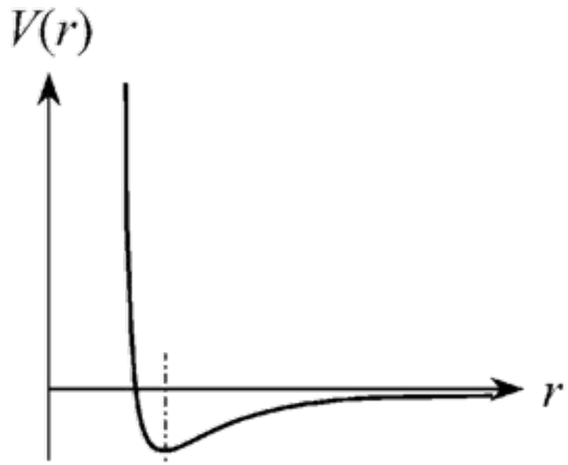
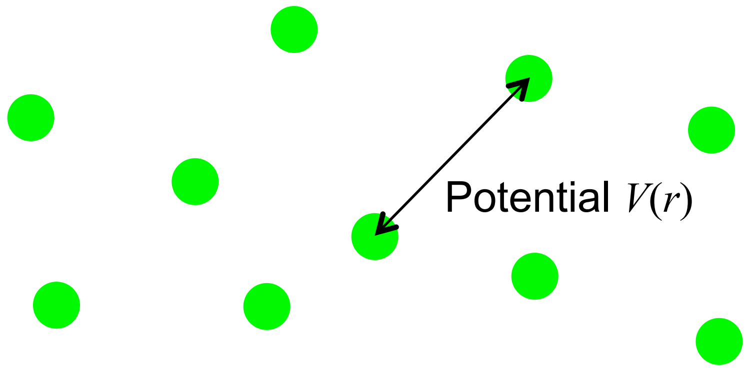
Form factor of a sphere



Example: liquid, pair correlation function





Difficult calculation: approximations, computer simulation

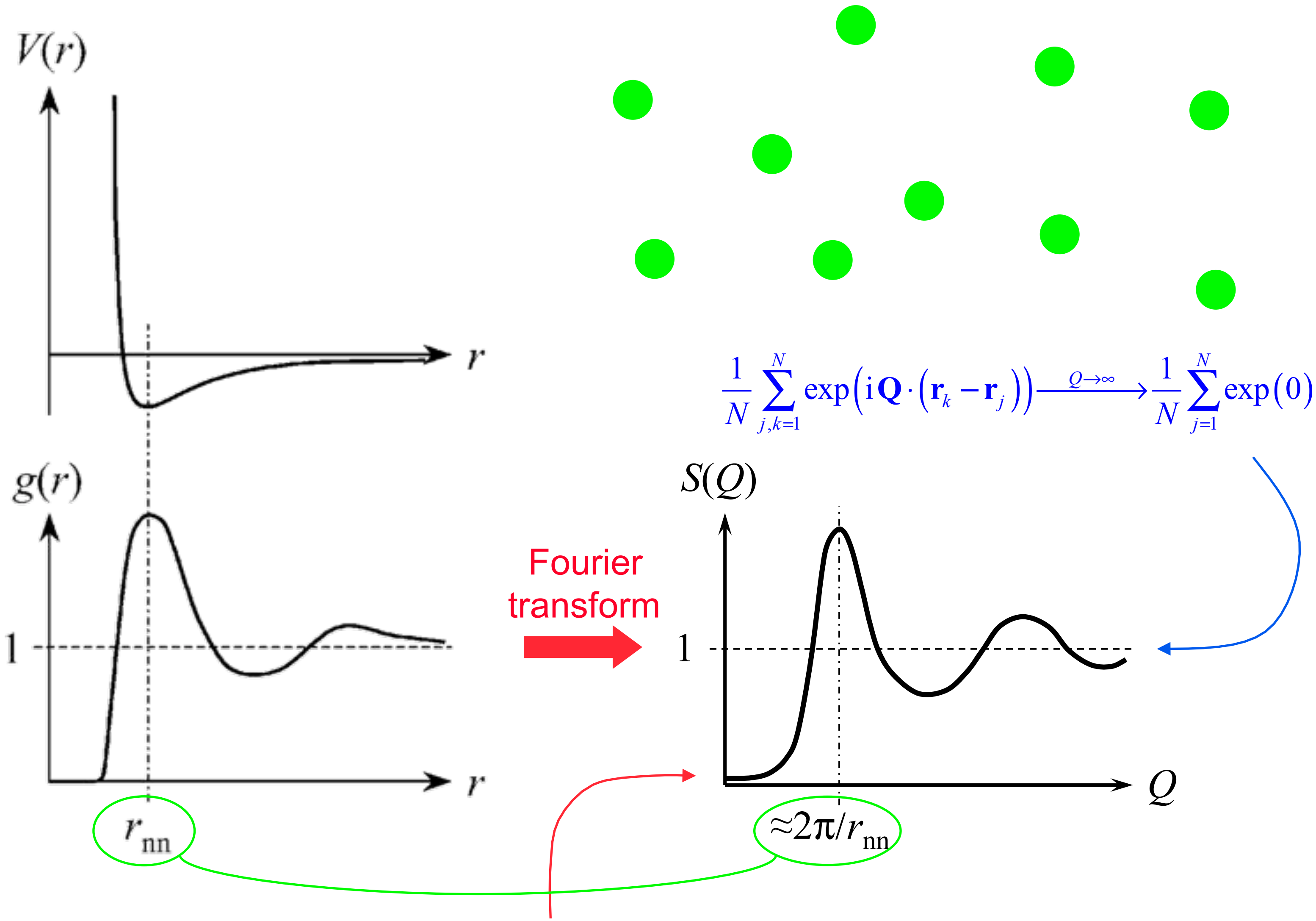
Correlation fades for long distances:

$$g(\mathbf{r}) = \frac{\langle \rho(\mathbf{0})\rho(\mathbf{r}) \rangle}{\rho_0^{\ 2}} \rightarrow \frac{\rho_0^{\ 2}}{\rho_0^{\ 2}} = 1$$

Distances smaller 2x particle radius not allowed

Preferred nearest-neighbour distance

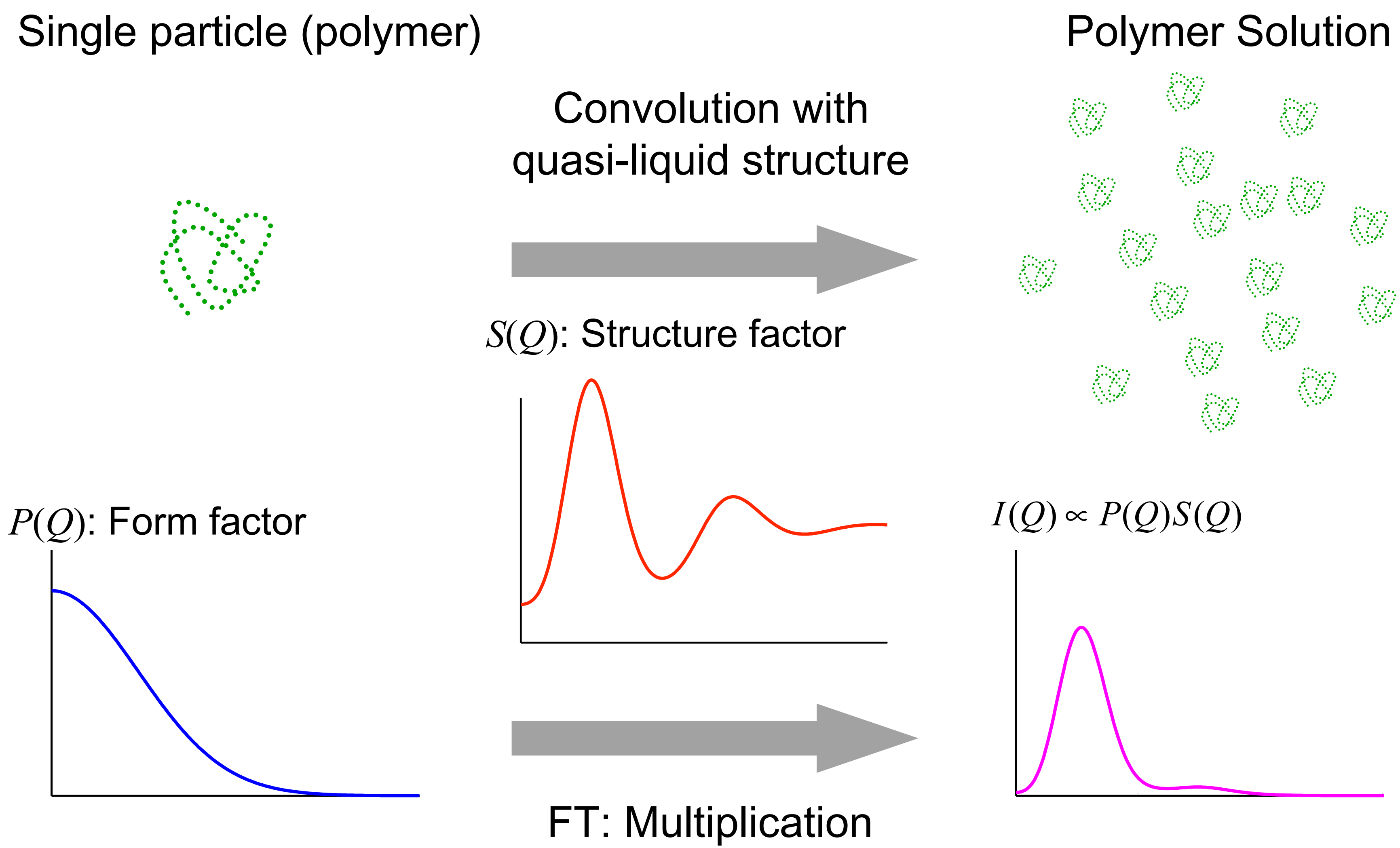
Example: liquid, structure factor



$$\frac{1}{N}\sum_{j,k=1}^{N}\exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_k-\mathbf{r}_j\right)\right)\xrightarrow{Q\to\infty}\frac{1}{N}\sum_{j=1}^{N}\exp(0)=1$$

$$S(Q)\xrightarrow{Q\to 0}\rho_0 k_\mathrm{B} T \kappa_T$$

Form factor and structure factor



Summary elastic scattering

- The scattering law is the absolute square of the Fourier transform of the density.
- The scattering law is the Fourier transform of the density correlation function.
- The scattering vector **Q** is the vectorial difference of the incident and the scattered wave vector (scattering triangle):

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

- Fourier transform ⇒ Q roughly corresponds to a length $2\pi/Q$.
- Repetitive structure: product of form- and structure factor.

Scattering from dynamic systems (inelastic scattering)

„Neutrons tell us where the atoms are *and how they move*.“

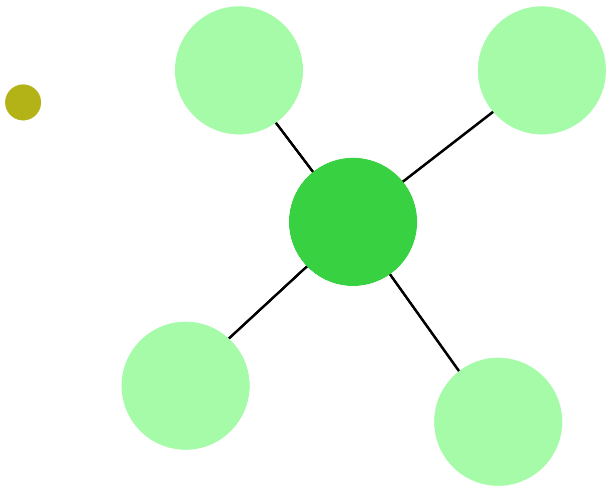
[C. G. Shull ?]

Elastic and inelastic scattering

What is the difference between elastic and inelastic scattering?

Elastic scattering

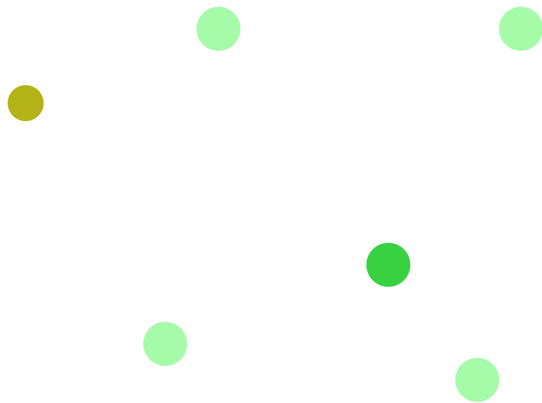
- scatterers are (assumed) fixed in space
- no energy transfer



- Information about
  - Structure

Inelastic scattering

- scatterers are moving or moved by neutron
- energy transfer

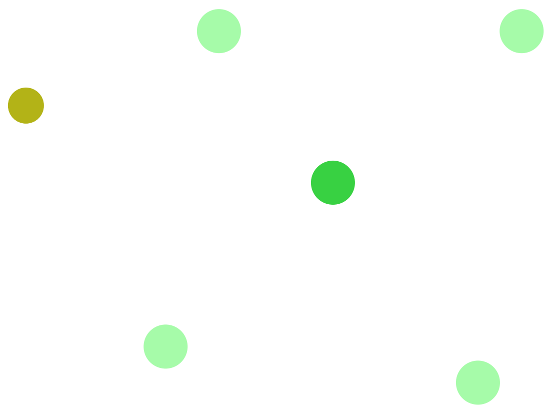


- Information about
  - Structure
  - Dynamics

Inelastic scattering

Energy loss

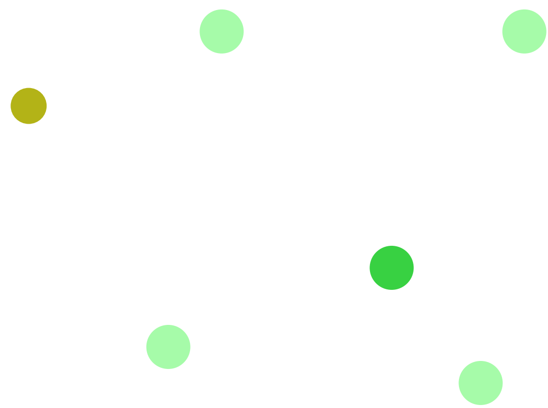
- Energy is transferred from the neutron to the system:



- $\hbar\omega = E' - E < 0$
- $\lambda' > \lambda$
- $k' < k$

Energy gain

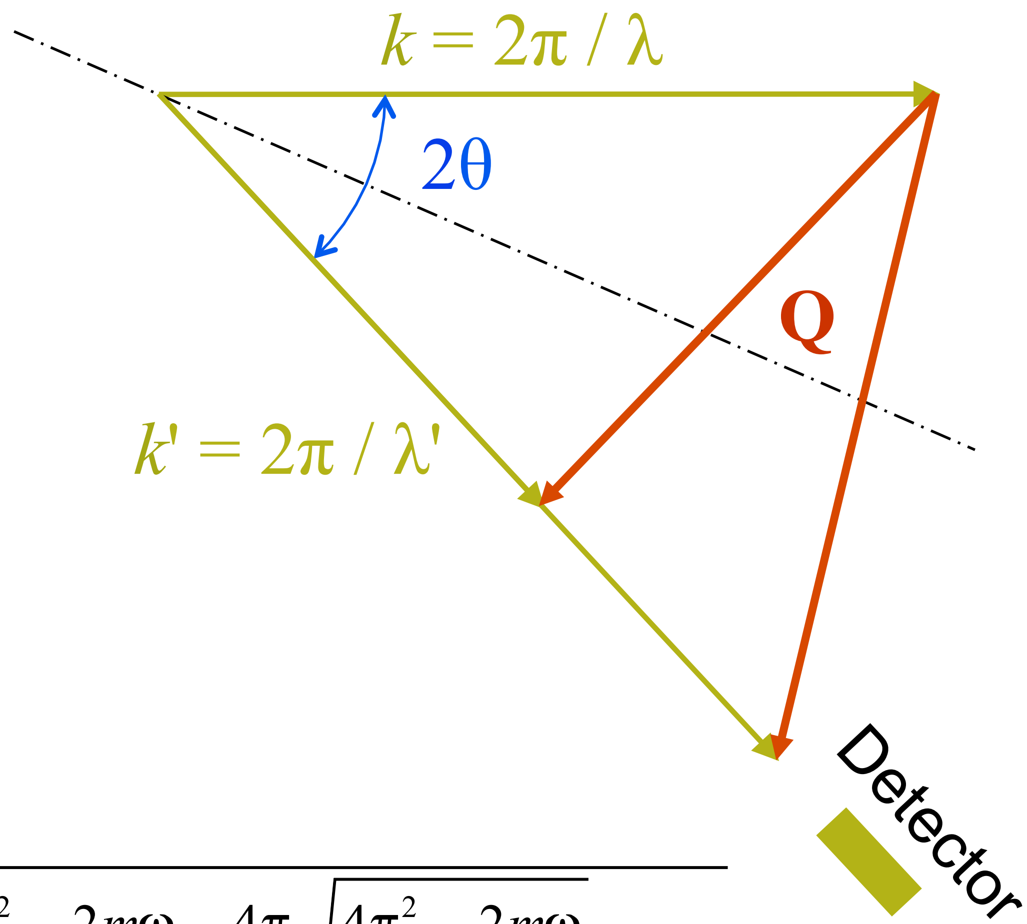
- Energy is transferred from the system to the neutron:



- $\hbar\omega = E' - E > 0$
- $\lambda' < \lambda$
- $k' > k$

The scattering triangle (inelastic scattering)

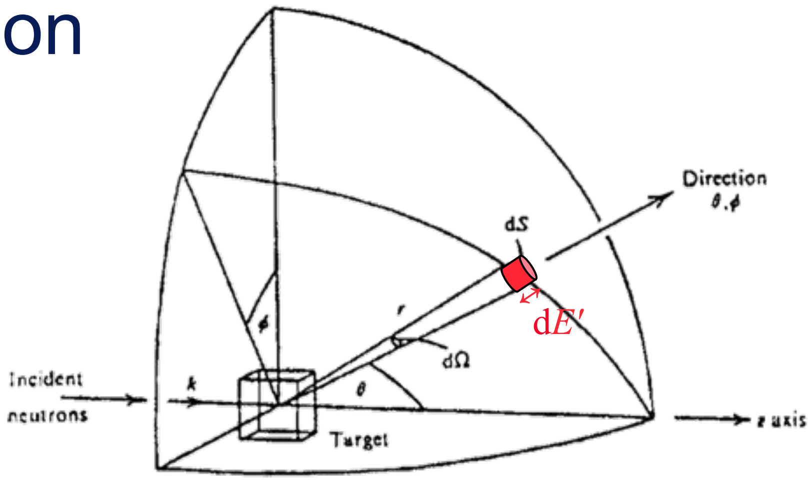
Source



The scattering triangle is not isosceles, Q is not determined by the position of source and detector only, Q is a function of θ and $\hbar\omega$:

$$Q = \sqrt{k^2 + k'^2 - 2kk'\cos 2\theta} = \sqrt{\frac{8\pi^2}{\lambda^2} + \frac{2m\omega}{\hbar} - \frac{4\pi}{\lambda}\sqrt{\frac{4\pi^2}{\lambda^2} + \frac{2m\omega}{\hbar}}\cos 2\theta}$$

Double differential cross section



Inelastic scattering:

- Experiment resolves final energy of the neutron E'.
- Scattering depends on energy transfer $\hbar\omega = E' - E$.
- ⇒ Scattering cross section becomes ‚double differential':

$$\frac{d\sigma}{d\Omega} = |b|^2 N\, S(Q)$$

$$\Downarrow$$

$$\frac{d\sigma}{d\Omega\, d\omega} = |b|^2 N\, S(Q,\omega)$$

- Structure Factor $S(Q)$ ⇒ Scattering Function $S(Q,\omega)$

Inelastic neutron scattering law recipe

… you gotta believe (or hear another one hour lecture)

1. Start with a static formula:

$$S(\mathbf{Q}) = \left\langle \sum_{j,k=1}^{N} \exp\left(\mathrm{i}\,\mathbf{Q}\cdot\left(\mathbf{r}_k - \mathbf{r}_j\right)\right)\right\rangle$$

2. Introduce time difference:
   intermediate scattering function

$$I(\mathbf{Q},t) = \left\langle \sum_{j,k=1}^{N} \exp\left(\mathrm{i}\,\mathbf{Q}\cdot\left(\mathbf{r}_k(t) - \mathbf{r}_j(0)\right)\right)\right\rangle$$

3. Take the Fourier transform from
   time to frequency:

$$S(\mathbf{Q},\omega) = \frac{1}{2\pi N}\int_{-\infty}^{\infty} \mathrm{d}t\, \exp(-\mathrm{i}\,\omega t)\left\langle \sum_{j,k=1}^{N} \exp\left(\mathrm{i}\,\mathbf{Q}\cdot\left(\mathbf{r}_k(t) - \mathbf{r}_j(0)\right)\right)\right\rangle$$

Coherent and incoherent scattering (neutrons)

$$\sum_{j,k=1}^{N} b_j^* b_k \exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_k(t)-\mathbf{r}_j(0)\right)\right)=$$

$$=\left|\bar{b}\right|^2 \sum_{j\neq k=1}^{N} \exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_k(t)-\mathbf{r}_j(0)\right)\right)+\overline{\left|b\right|^2}\sum_{j=1}^{N}\exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_j(t)-\mathbf{r}_j(0)\right)\right)=$$

$$=\left|\bar{b}\right|^2 \sum_{j,k=1}^{N} \exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_k(t)-\mathbf{r}_j(0)\right)\right)+\left(\overline{\left|b\right|^2}-\left|\bar{b}\right|^2\right)\sum_{j=1}^{N}\exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_j(t)-\mathbf{r}_j(0)\right)\right)$$

$S_{\mathrm{coh}}(\mathbf{Q},\omega)$ $S_{\mathrm{inc}}(\mathbf{Q},\omega)$

Wrapping it all up: $$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega\,\mathrm{d}\omega}=N\frac{k'}{k}\left(\underbrace{\left|\bar{b}\right|^2}_{\sigma_{\mathrm{coh}}/4\pi} S_{\mathrm{coh}}(\mathbf{Q},\omega)+\underbrace{\left(\overline{\left|b\right|^2}-\left|\bar{b}\right|^2\right)}_{\sigma_{\mathrm{inc}}/4\pi} S_{\mathrm{inc}}(\mathbf{Q},\omega)\right)$$

with the scattering functions

$$S_{\mathrm{coh}}(\mathbf{Q},\omega)=\frac{1}{2\pi N}\int_{-\infty}^{\infty}\mathrm{d}t\exp(-\mathrm{i}\omega t)\left\langle\sum_{j,k=1}^{N}\exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_k(t)-\mathbf{r}_j(0)\right)\right)\right\rangle$$

$$S_{\mathrm{inc}}(\mathbf{Q},\omega)=\frac{1}{2\pi N}\int_{-\infty}^{\infty}\mathrm{d}t\exp(-\mathrm{i}\omega t)\left\langle\sum_{j=1}^{N}\exp\left(\mathrm{i}\mathbf{Q}\cdot\left(\mathbf{r}_j(t)-\mathbf{r}_j(0)\right)\right)\right\rangle$$

Intermediate scattering functions

$$S_{\text{coh}}(\mathbf{Q},\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(-\mathrm{i}\,\omega t) I_{\text{coh}}(\mathbf{Q},t)\,\mathrm{d}t \quad \text{with} \quad I_{\text{coh}}(\mathbf{Q},t) = \left\langle \frac{1}{N}\sum_{j,k=1}^{N} \exp\left(\mathrm{i}\,\mathbf{Q}\cdot\left(\mathbf{r}_k(t)-\mathbf{r}_j(0)\right)\right)\right\rangle$$

$$S_{\text{inc}}(\mathbf{Q},\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(-\mathrm{i}\,\omega t) I_{\text{inc}}(\mathbf{Q},t)\,\mathrm{d}t \quad \text{with} \quad I_{\text{inc}}(\mathbf{Q},t) = \left\langle \frac{1}{N}\sum_{j=1}^{N} \exp\left(\mathrm{i}\,\mathbf{Q}\cdot\left(\mathbf{r}_j(t)-\mathbf{r}_j(0)\right)\right)\right\rangle$$

Relation to diffraction:

$$I_{\text{coh}}(\mathbf{Q},0) = \left\langle \frac{1}{N}\sum_{j,k=1}^{N} \exp\left(\mathrm{i}\,\mathbf{Q}\cdot\left(\mathbf{r}_k(0)-\mathbf{r}_j(0)\right)\right)\right\rangle = S(\mathbf{Q})$$

Integral theorem of Fourier transform:

$$I_{\text{coh}}(\mathbf{Q},0) = \int_{-\infty}^{\infty} S_{\text{coh}}(\mathbf{Q},\omega)\,\mathrm{d}\omega$$

This is mathematically what the diffraction experiment does: integration over all energy transfers.

Van-Hove correlation functions

One piece missing: Is there something like $g(r)$ for inelastic scattering?

Yes, similar to the static case one can define the van-Hove correlation functions

$$G(\mathbf{r},t) = \frac{1}{N}\left\langle \sum_{j,k=1}^{N} \delta\left(\mathbf{r} - \mathbf{r}_k(t) + \mathbf{r}_j(0)\right) \right\rangle$$

$$G_\mathrm{s}(\mathbf{r},t) = \frac{1}{N}\left\langle \sum_{j=1}^{N} \delta\left(\mathbf{r} - \mathbf{r}_j(t) + \mathbf{r}_j(0)\right) \right\rangle$$

which have the intermediate scattering functions as Fourier transform:

$$I_\mathrm{coh}(\mathbf{Q},t) = \int_{V_d} G(\mathbf{r},t)\exp(\mathrm{i}\mathbf{Q}\cdot\mathbf{r})d^3r$$

$$I_\mathrm{inc}(\mathbf{Q},t) = \int_{V_d} G_\mathrm{s}(\mathbf{r},t)\exp(\mathrm{i}\mathbf{Q}\cdot\mathbf{r})d^3r$$

Relation to static case ($t = 0$ as before): $G(\mathbf{r},0) = \dfrac{\langle \rho(\mathbf{0})\rho(\mathbf{r})\rangle}{\rho_0} = \delta(\mathbf{r}) + \rho_0 g(\mathbf{r})$

Fourier transform relations

Experiment level:

Scattering functions

$S_{coh}(\mathbf{Q},\omega)$ $S_{inc}(\mathbf{Q},\omega)$

↓ temporal inverse Fourier transform | ↑ temporal Fourier transform

Intermediate scattering functions

$I_{coh}(\mathbf{Q},t)$ $I_{inc}(\mathbf{Q},t)$

↓ spatial inverse Fourier transform | ↑ spatial Fourier transform

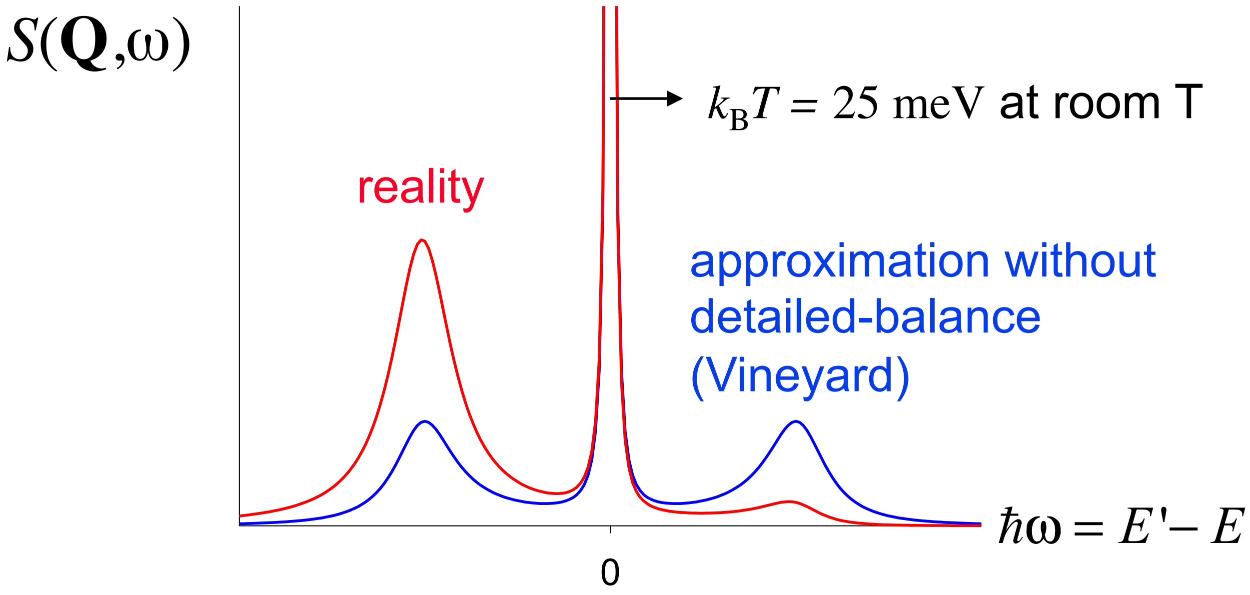
Microscopic level:

Van Hove correlation functions

$G(\mathbf{r},t)$ $G_s(\mathbf{r},t)$

Detailed balance factor

Asymmetry:



Detailed balance condition:

$$S(\mathbf{Q},-\omega) = \exp(\hbar\omega / k_B T)\, S(\mathbf{Q},\omega)$$

Energy loss is more probable than energy gain.

Asymmetry of S(Q,ω) ⇒ $I(Q,t)$ and $G(r,t)$ are complex!

Can be neglected for not too fast processes:

$t \gg \hbar / k_B T \approx 0.03$ ps at room temperature

Example: Diffusion

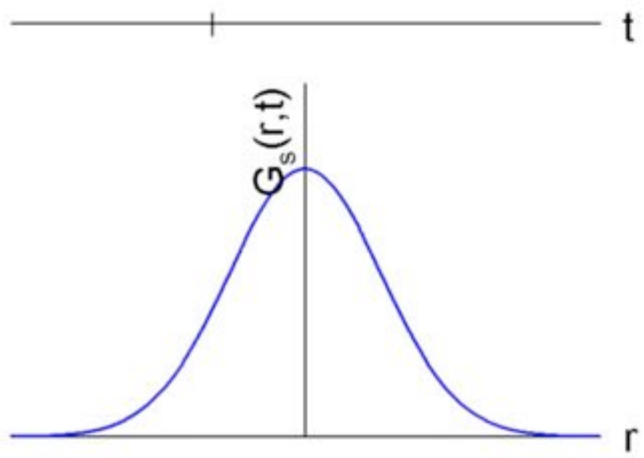
Example for Gaussian dynamics:

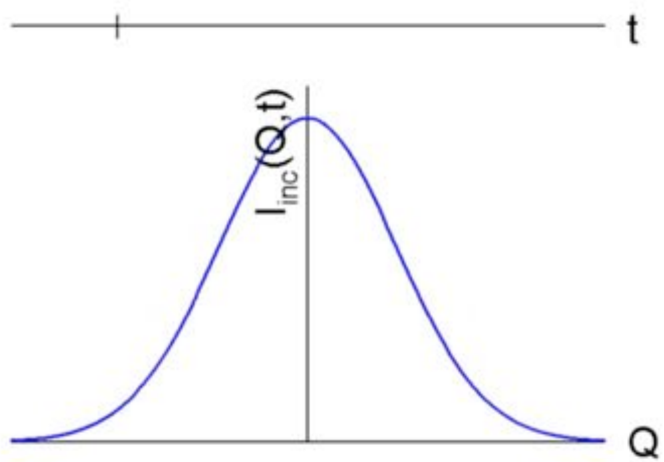
Diffusion, $\langle r^2 \rangle = 6Dt$

$$G_s(r,t) = \frac{1}{(4\pi D t)^{3/2}} \exp\left(-\frac{r^2}{4Dt}\right)$$

Fourier transform of a Gaussian is a Gaussian with reciprocal width.

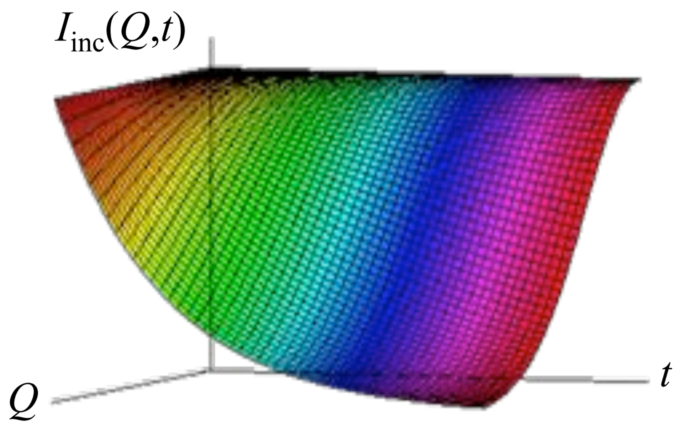
$$I_{inc}(Q,t) = \exp(-DQ^2 t)$$





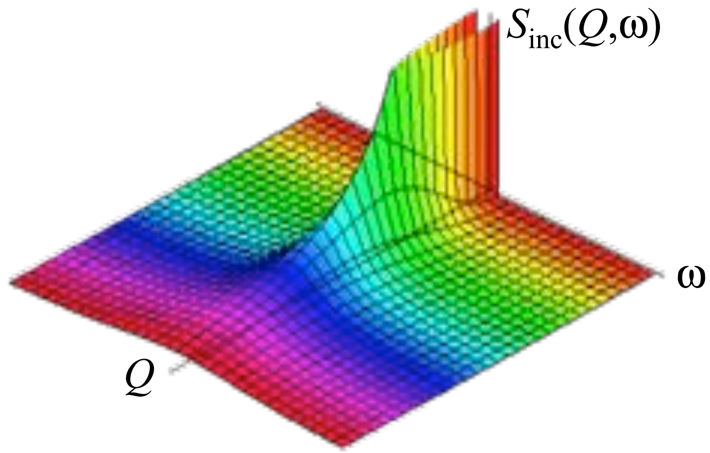
Diffusion 2

$$I_{\mathrm{inc}}(Q,t) = \exp(-DQ^2 t)$$



Fourier transform of a an exponential decay is a Lorentzian.

$$S_{\mathrm{inc}}(Q,\omega) = \frac{1}{\pi}\frac{DQ^2}{\omega^2 + (DQ^2)^2}$$



Summary inelastic scattering

- The scattering cross section depends on the scattering vector $\mathbf{Q}$ and the energy transfer $\hbar\omega$.
- The scattering vector $\mathbf{Q}$ is (as in elastic scattering) the vectorial difference of the incident and the scattered wave vector (scattering triangle), but depends now on ω too.
- Q roughly corresponds to a length $2\pi/Q$.
- ω roughly corresponds to a time $2\pi/\omega$.
- The exact relation between spatial correlation and scattering is a Fourier transform.
- The exact relation between temporal correlation and scattering is a Fourier transform.
- Inelastic scattering contains two components:
  - coherent: from average scattering lengths, measures pair correlations
  - incoherent: from variance of scattering length, measures self correlation