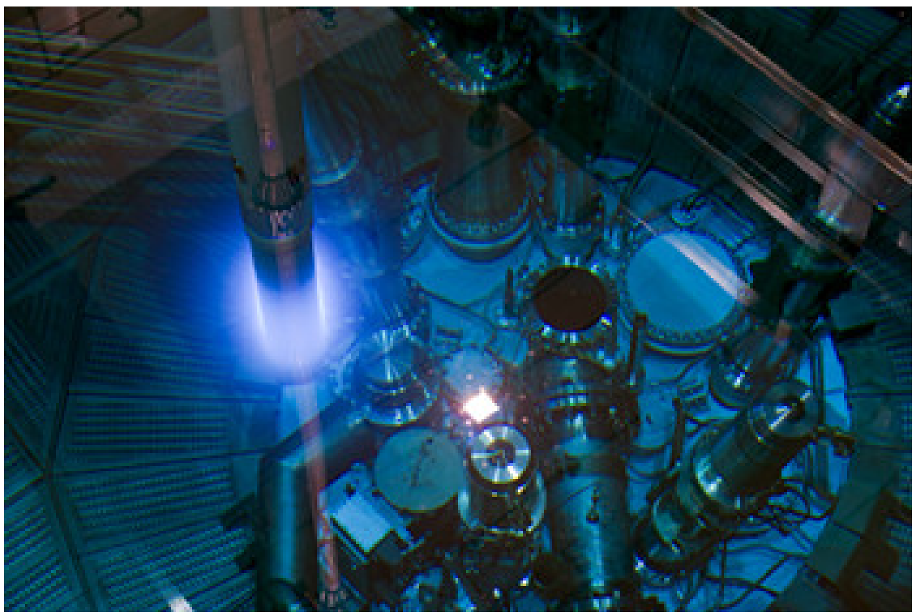
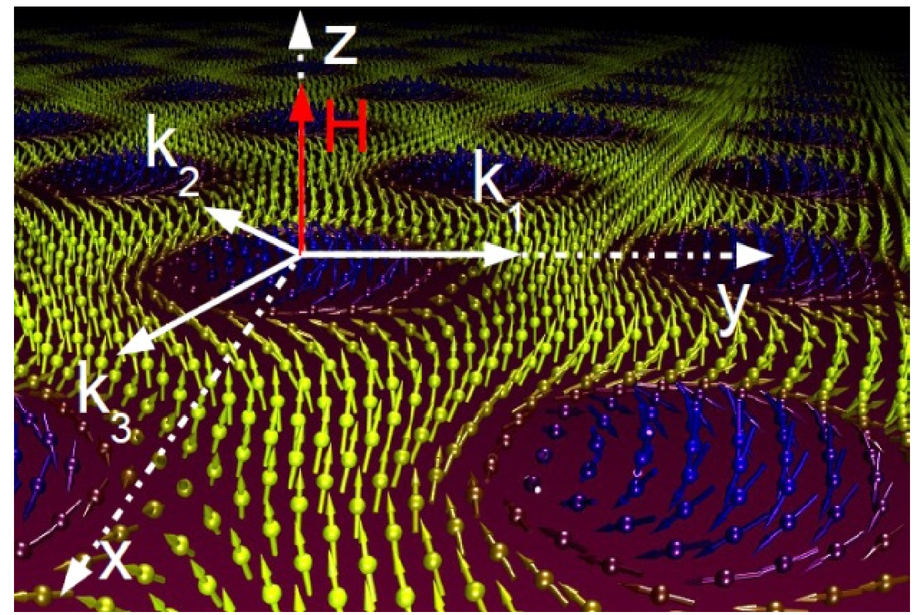
FRM II
Forschungs-Neutronenquelle
Heinz Maier-Leibnitz

MLZ
Heinz Maier-Leibnitz Zentrum

Physics with Neutrons I, WS 2015/2016





Lecture 14, 1.2.2016

MLZ is a cooperation between:

JÜLICH
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Helmholtz-Zentrum
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Zentrum für Material- und Küstenforschung

TUM
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Reminder: Correlation functions

Starting point: General cross-section

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

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

➡ 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

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

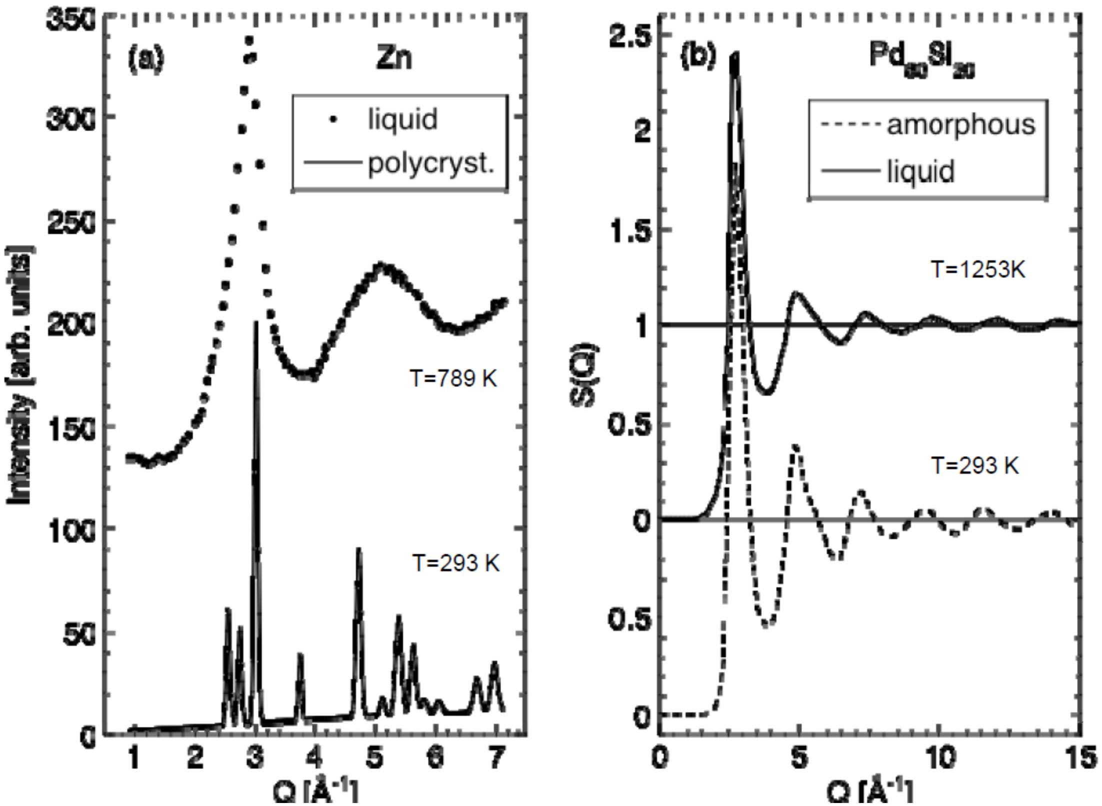
$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega}\right)_{inc} = N\frac{k'}{k}(\langle b^2\rangle - \langle b\rangle^2) S_{inc}(\boldsymbol{Q},\omega)$$

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

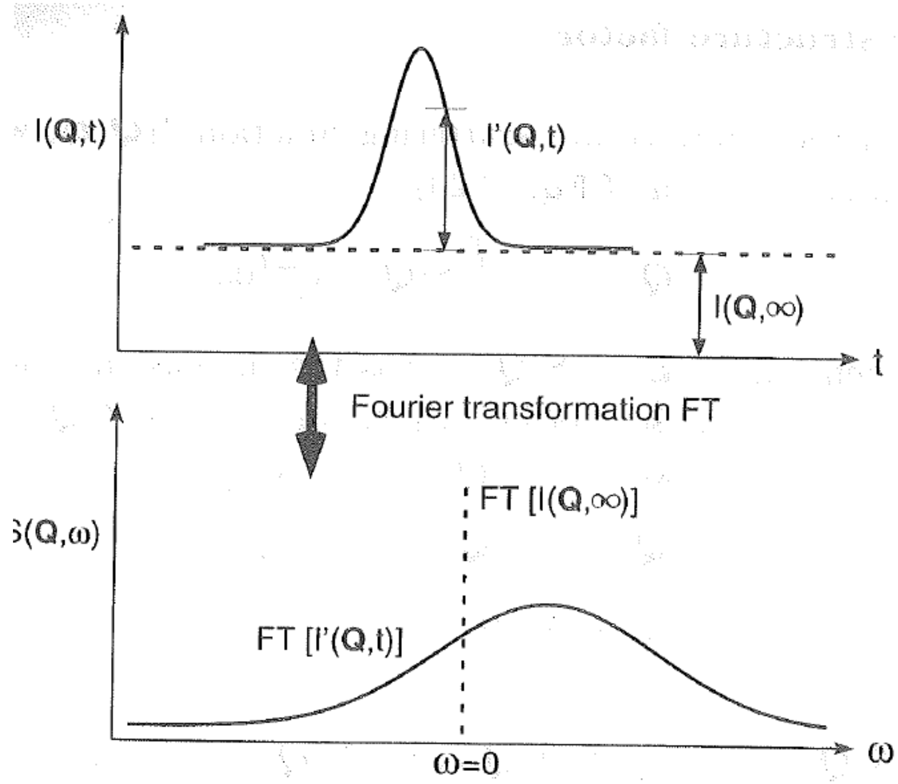


Static structure factor

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

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

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



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

Elastic part   Inelastic part

⇨ Infinite time correlations

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

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

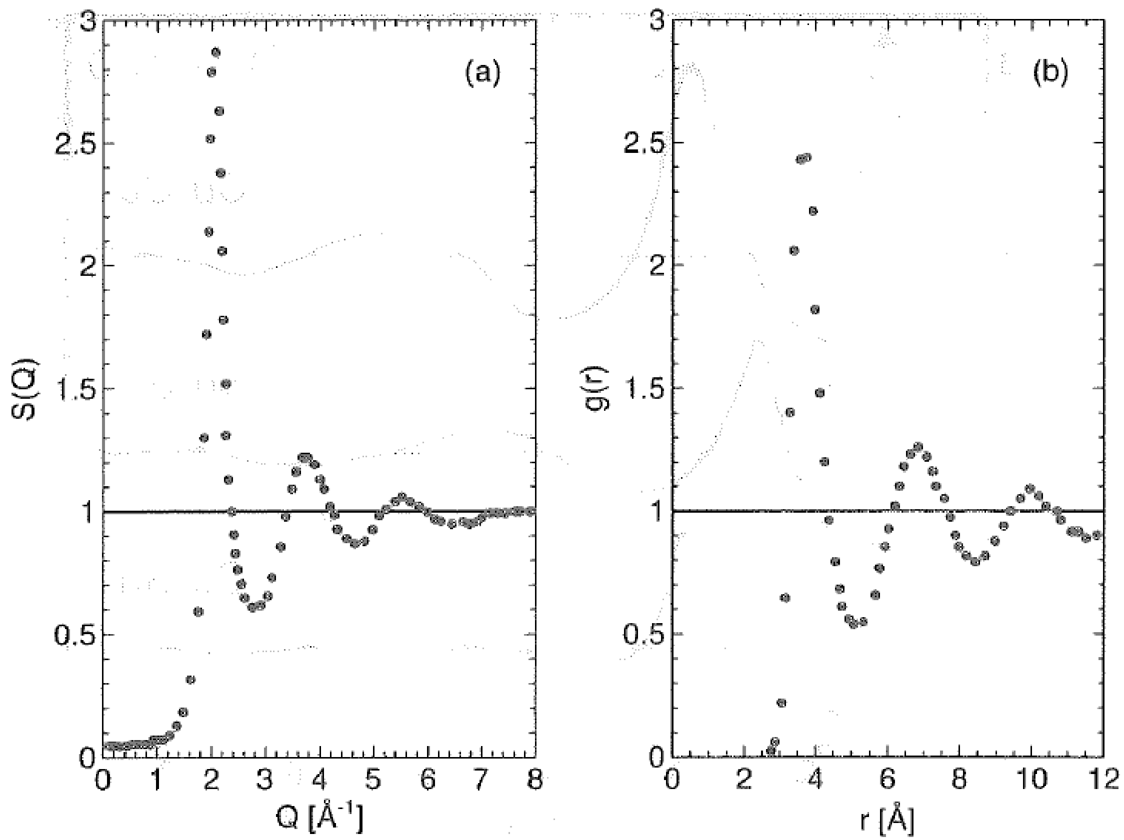
Elastic scattering from liquids

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

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->0 $S(Q=0)=1$

Limit Q->∞ $S(Q\to\infty)=n_0\kappa_T k_B T$ 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

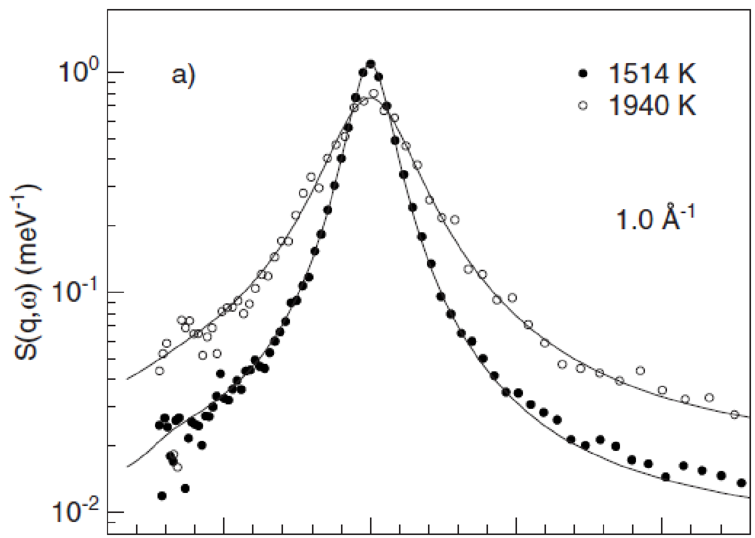
⇨ Diffusion

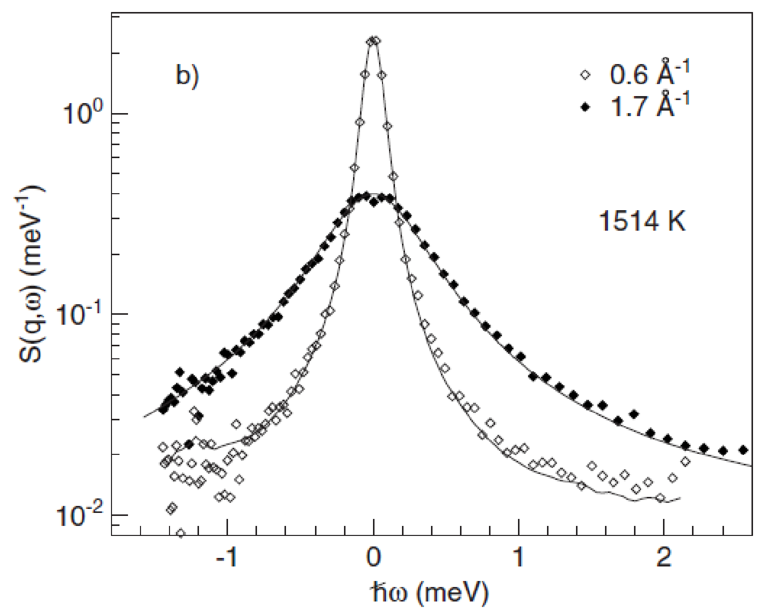
Small values of r,t
large values of Q,w
$G_s(r,t)$ peaked at t=0 for small r

⇨ Short time behaviour

⇨ Ideal gas

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





Diffusive behaviour (low Q):

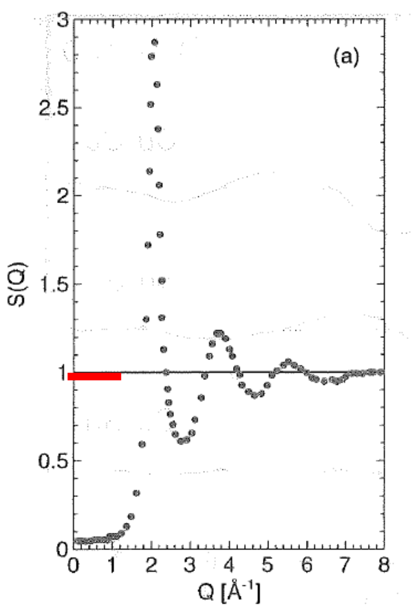
Fick´s law: $\frac{\partial n(\boldsymbol{r},t)}{\partial t} = D\nabla^2 n(\boldsymbol{r},t)$ Diffusion constant D

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

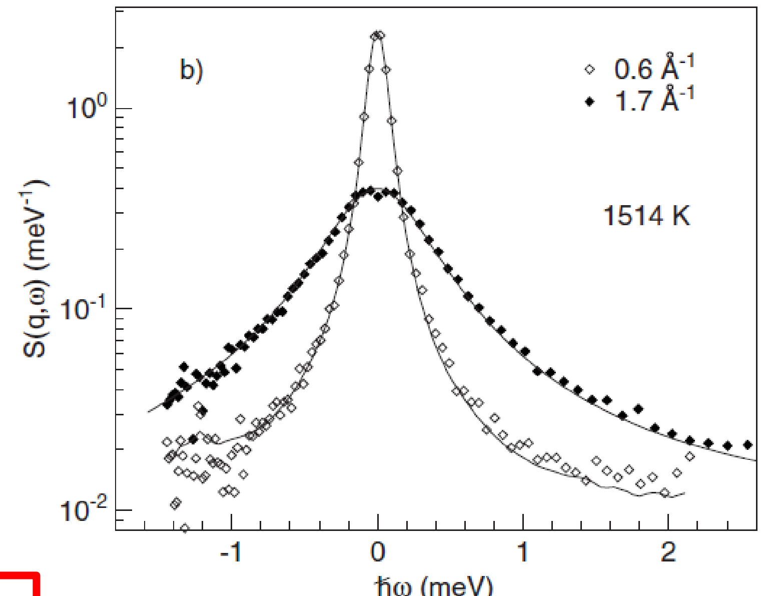
Lorentzian function centered at w=0

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

Valid only for q^{-1}» mean distance



Otherwise: microscopic details!



Width increases with Q

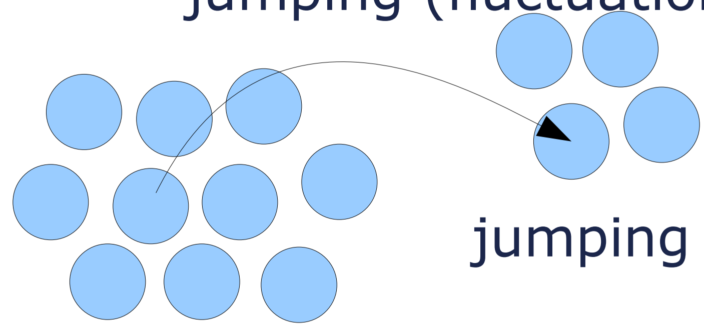
QENS ⟺ Diffusion constant

Diffusive behaviour (higher Q):

➡ Macroscopic model fails for q^{-1}~ 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

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

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

Again:Lorentzian function centered at w=0

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

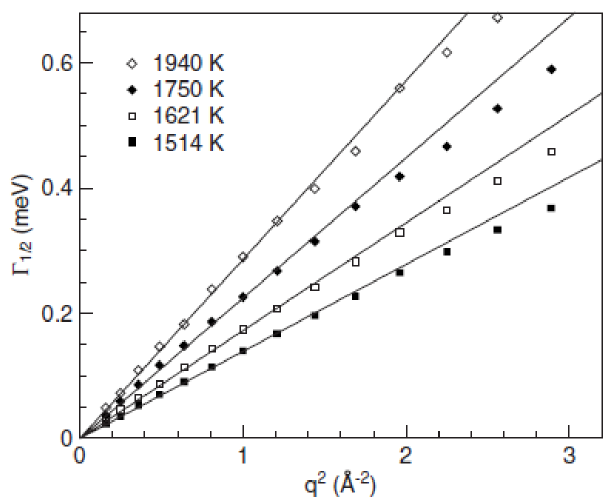
Applications of QENS on liquids:

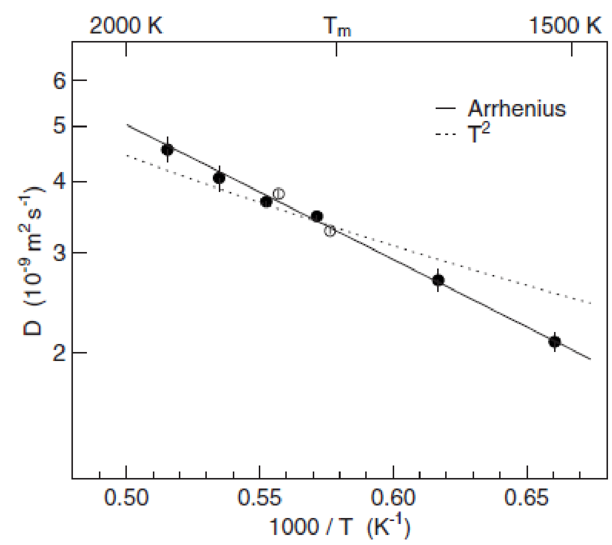
Diffusion constants important for crystallization and solidification of (metallic) melts

QENS not altered by convection (microscopic!!)
Containerless levitation: Udercool molten Ni by 200°C

PHYSICAL REVIEW B 77, 092201 (2008)

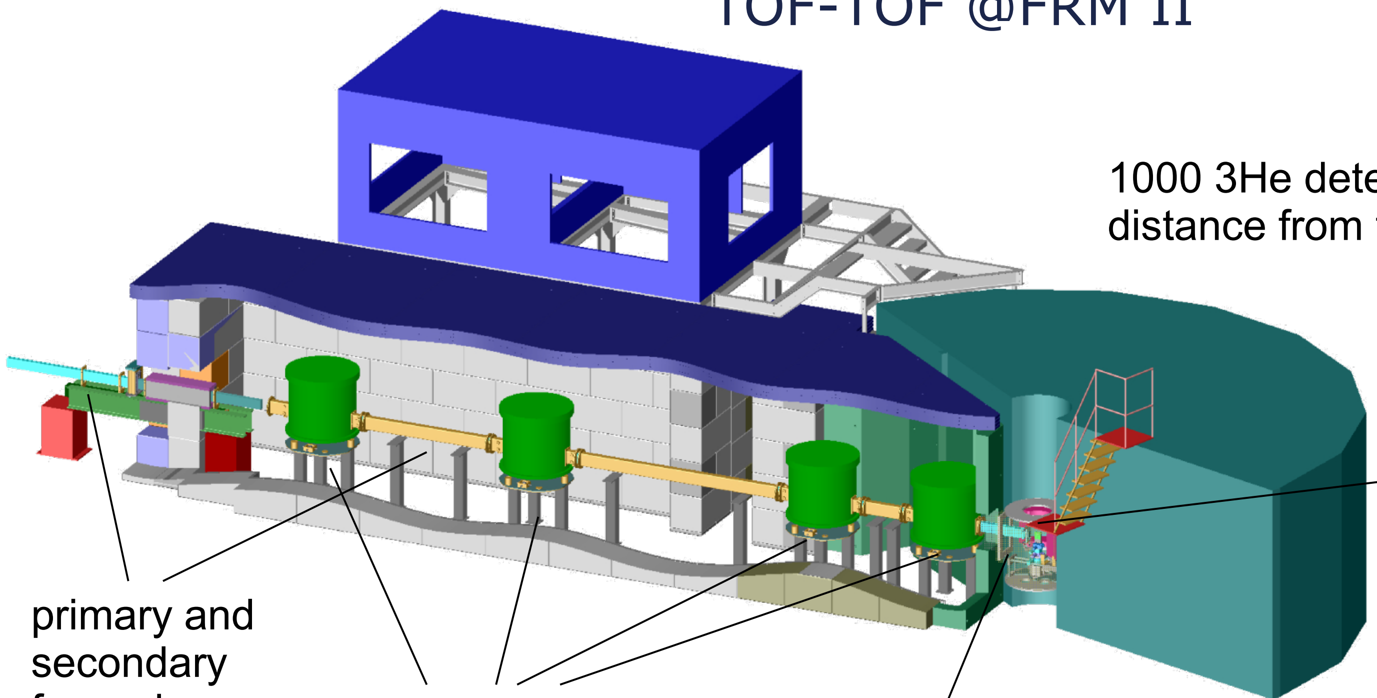
Determination of self-diffusion coefficients by quasielastic neutron scattering measurements of levitated Ni droplets

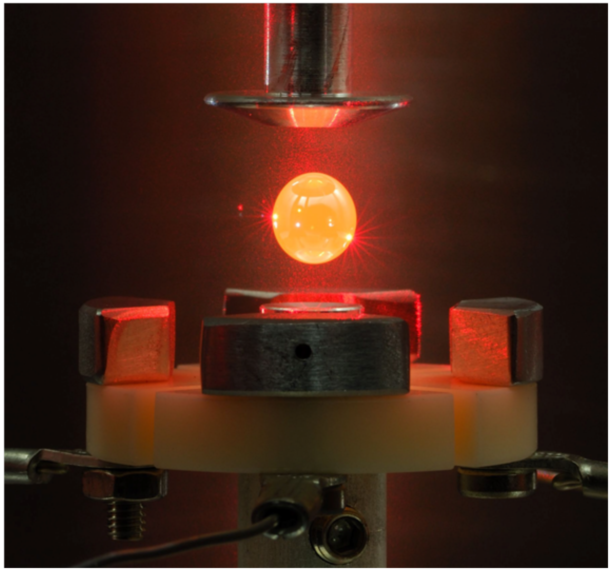




Scattering on liquids

TOF-TOF @FRM II





Outlook: Summer term 2016

VL1: Repetition of winter term, basic neutron scattering theory
VL2: Neutron optics, reflectometry and dynamical scattering theory
VL3: GISANS and soft matter
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
VL13: Spin echo spectrocopy
VL12: 1D Magnetism

TISANE: Kinetic small angle neutron scattering