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National Neutron Scattering Facility
Outline QuasiElastic Neutron Scattering from an user point of view

Part I: why neutrons?

context and experimental probes

Role of the experimentalist: user, facility scientist

QENS spectrometers

Part II: what are the observables?

constraints on measurements, limitations

Models and theories

Sample environment: next challenges
good reasons to use neutrons

- Cover large scales of time and space simultaneously
- A unique probe for the magnetism
- Complementary to the other techniques; give access directly in observable relevant and defined well (S(Q), S(Q,w), F (Q, t), dispersion curves)
- Neutral, non-destructive and penetrating; precise, sensitive and selective
- Neutron scattering methods are pluridisciplinary
  - it provides final answers to fundamental questions and
  - a solid background to support any other techniques (X rays, NMR, Numerical Simulations..)

thanks to well controlled and well known observables providing absolute quantities.
Comparison with x rays and light

\[ Q \equiv k_1 - k_2 \]
\[ Q^2 = k_1^2 + k_2^2 - 2|k_1||k_2| \cos \theta \]
\[ \hbar \omega = E_1 - E_2 \]
\[ Q_{el} = 2|k_1| \sin (\theta/2) \]
\[ = (4\pi/\lambda) \sin (\theta/2) \]

Neutrons:
\[ E = \frac{\hbar^2}{2m_n} \left( \frac{1}{\lambda} \right)^2 \]
\[ E (\text{meV}) = \frac{81.81}{\lambda^2} \]

X Rays and Light scattering:
\[ E = \hbar c \left( \frac{1}{\lambda} \right) \]
\[ E (\text{keV}) = \frac{12.4}{\lambda} \]

(\(\lambda\) in Å)
\[ \vec{Q} = \vec{k}_1 - \vec{k}_0 \]
\[ h\omega = E_f - E_i \]

Measured Intensity

\[ \frac{\partial^2 \sigma}{\partial \Omega \partial \omega} \approx N^* \sigma_{\text{Scat}} * S(\vec{Q}, \omega) \]

Dynamical Structure Factor

Scattering cross-section

van Hove correlation function: \( G(r,t) \) is the probability to find a particle at a distance \( r \), at time \( t \), provided it was at \( r=0 \), at \( t=0 \).

\[ S(\vec{Q}, \omega) = \frac{1}{2\pi} \int G(\vec{r},t) e^{i(\vec{Q}\vec{r} - \omega t)} d\vec{r} dt \]
QUASI ELASTIC NEUTRON SCATTERING: QENS

Measure of scattering processes involving small amounts of energy exchange, classical approximation ( $|\hbar \omega| << \frac{1}{2} k_B T$ )
i.e. in the low energy region of inelastic spectra close to 0

Dynamical phenomena at $10^{-13}$ to $10^{-7}$ s
Motions explored in space on length scales comparable with $\lambda$ of the neutrons

Vibrational displacements, librations, jump distances, diffusion paths, correlation lengths (nano to micro)

Observables: Dynamical structure factor, $S(Q,w)$ or intermediate scattering function, mean square displacements, self diffusion coefficient, relaxation time, Reptation Rouse modes, friction coefficient, Rotation diffusion, EISF, VDOS, $C_p$
Sample where atoms are moving

Neutron Source
The probe

Neutrons have a spin
Scattering with or without spin flipping
Elastic, quasielastic and inelastic scattering of neutrons

Incoming Neutron

$\lambda : 2 \text{ to } 10 \ \text{Å}$

$E_0 : 1 \text{ to } 10 \ \text{meV}$

$Q_{\text{elastic}} = \frac{4\pi}{\lambda_0} \sin \theta$ and $|\vec{k}| = |\vec{k}_0|$  

Technically a TOF spectrum is accumulated at a fixed scattering angle for each detector

$E = E_0 + \hbar \omega$

$|\vec{k}| = \sqrt{\frac{2mE}{\hbar}}$

$2\theta$

$Q$

Detector

$E_0$

$E_0$

Number of neutrons

Energy

$\Delta \omega$

$\Delta \omega$

Energy

$E_0$

$E_0$

Energy

Number of neutrons

Elastic scattering

Elastic and inelastic scattering

$\text{diffusif (}10^{-9} - 10^{-12} \text{ s)}$

quasi elastic broadening
scenario of quasielastic scattering as T increases

Molecular liquids and polymers

\[
\ln S_{el}(Q, \Delta \omega, T)/S_{el}(Q, \Delta \omega, T=2K)
\]

harmonic and anharmonic behavior

\(T_{\text{dynamic}} \sim T_{\text{glass}}\)

To cover a wide range requires the combination of several instruments
**Speed and wave length of the neutron**

<table>
<thead>
<tr>
<th>$\lambda$ (Å)</th>
<th>$v$ (m/s)</th>
<th>$k$ (Å$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>7 912</td>
<td>12.6</td>
</tr>
<tr>
<td>1.0</td>
<td>3 956</td>
<td>6.3</td>
</tr>
<tr>
<td>1.5</td>
<td>2 637</td>
<td>4.2</td>
</tr>
<tr>
<td>2.0</td>
<td>1 978</td>
<td>3.1</td>
</tr>
<tr>
<td>2.5</td>
<td>1 582</td>
<td>2.5</td>
</tr>
<tr>
<td>5.0</td>
<td>791</td>
<td>1.3</td>
</tr>
<tr>
<td>10.0</td>
<td>396</td>
<td>0.6</td>
</tr>
<tr>
<td>20.0</td>
<td>198</td>
<td>0.3</td>
</tr>
</tbody>
</table>
Sequences of steps for an experiment

Before the measurement

- Facility scientists, engineers and technicians
- Thermalisation
- Monochromator
- Collimator
- Polarisation
- Detector efficiency

User interacting at the facility

- Sample holder and position
- Sample environment (T, p, B)
- Choice of the E and Q resolution
- Low background, parasites

Interpretation of the data

Models

Theories

Applications

Questions

After the measurement

- Speed, (particle character)
- Wavelength, (wave character)
- Direction,
- Polarisation

Preparation of the neutrons

Preparation of the sample

Analysis of the data

Production corrections

Analysis of the scattered neutrons

Interpretation of the data

Models

Theories

Applications

Questions
Several instruments

**Triple Spectrometer**

**Time of Flight Spectrometer** *(based on energy transfer analysis)*
**Backscattering Spectrometer**

**Neutron spin Echo Spectrometer (NSE and NRSE)**
*Based on the Fourier time analysis of the scattered intensity*
Triple axis and Time of flight approaches

Create high continuous flux of monochromatic neutrons (ki fixed) continuous detection of monochromatic neutrons (kf fixed)

Create a high flux of pulsed monochromatic neutrons (ki fixed) Detection as a function of time (kf variable)

Create high flux white neutron beam (ki variable) Detection of monochromatic neutrons as function of time (kf fixed)

\[
\left( \frac{d^2\sigma}{d\Omega d\omega} \right) = \frac{k_f}{k_i} \frac{\sigma_{sc}}{4\pi} N S(\vec{Q},\omega)
\]
**Time focusing**
**IN6 (ILL) – Focus (PSI)**

Select a broad incident energy band

+ The « fastest » neutrons

Get to the detector at the same time that the « slowest » ones.

**Large flux** but resolution not triangular

**NB:**
Can focus in the inelastic region

---

**Inverted geometry**
**QENS (ANL/Intense Pulsed Neutron Source)**
**IRIS (ISIS)**

Use a white beam

+ analyzer in front of detectors.

**Concurrent measurement of elastic** \((S(Q))\)

**AND inelastic** \((S(Q,w))\)

On pulsed sources: measure the neutron energy loss side: Bose factor not a limiting factor

Can probe far in the inelastic even at low temperature

Resolution\(\pm\) fixed

High background
Fig. 3. FOCUS spectrometer at Paul-Scherrer Institut (PSI) [29]; FOCUS is a typical XTL-TOF spectrometer, i.e. a time-of-flight instrument with a Bragg monochromator and a time-of-flight analyzer. While the monochromator selects the incident neutron energy $E_0$, the energy of the scattered neutrons $E$ is determined by measuring the neutron flight time.
**The Fa# Project @ LLB**

J-M Zanotti, S Rodriguez

« Hybrid » Instrument:

- Time focusing (Soft matter / Biology)
- Energy focusing (Solid state physics)

**Scientific fields**
- Soft condensed matter
- Materials
- Magnetism
- Soft Matter
- Biology
- Electrolites
- ...
Measure the neutron energy after the sample

\[ V = \frac{D_{ED}}{\text{tof}} \quad \text{et} \quad E_f = \frac{1}{2} m V^2 \]
1- Avoid frame overlap
2- Avoid any harmonics
\[ 2 \frac{D\tau}{\tau} = \frac{DE}{E} \]
Time-of-flight: Theory vs reality

Conversion from \((2\theta, t_{\text{time of flight}})\) to \((Q, \omega)\)

To measure long correlation times:
Increase the chopper speed and/or \(I_0\).
But flux drops!

\[
S^{\text{Mesuré}}(Q, \omega) = S^{\text{Théo}}(Q, \omega) \otimes R(\omega)
\]

\(\text{NB: At } 298 \text{ K, } k_B T = 25 \text{ meV}\)

The maximum energy loss is \(E_0\)...
Find a good compromise between flux, energy resolution and wavelength.
Backscattering Spectrometers

monochromator and analyseur : perfect crystals in backscattering (2q=180°)
Bragg law differentiation

\[
\left( \frac{\Delta \lambda}{\lambda} \right)^2 = \left( \frac{\Delta d}{d} \right)^2 + \left( \frac{\Delta \theta}{\tan \theta} \right)^2
\]

analyseur
Large angles

analyseur
small angles

multi tube detecteur

deflecteur chopper

deflecteur

R=2

H53 cold neutron guid

background choppe
Be-filte

vertically focusin

Focusin guid

Moving spherica monochromato

B. Frick, IN16, ILL
doppler

détecteurs

Q=Qélastique

Résolution <1μeV
Gamme -15 à+15μeV

BASIS @SNS ± 300 μeV
Spin Echo Spectrometers

NSE measures the sample dynamics in the time domain, via the determination of the intermediate scattering functions $F(Q,t)$.

Neutron are polarised, pi/E rotation, then under magnetic field

Mezei 1972 NSE, Gähler 1987 NRSE

ILL, IN11A
The measured quantity: the scattered beam polarisation...

Neutron spin, precession...

\[
B_0 = 0
\]

\[
\frac{d\vec{S}}{dt} = \gamma \vec{S} \wedge \vec{B}_0
\]

\[
\omega = |\gamma_n|B_0
\]

\[
N = \gamma_n B_0 t / 2\pi
\]

\[
\gamma_n = -2913.2\pi (Gs)^{-1}
\]

\[
p_{|+\rangle} = \cos^2(\alpha/2)
\]

\[
p_{|->} = \sin^2(\alpha/2)
\]

\[
\langle P \rangle = \frac{n_+ - n_-}{n_+ + n_-} = \langle p_{|+\rangle} \rangle - \langle p_{|->} \rangle = \langle \cos^2(\alpha/2) \rangle - \langle \sin^2(\alpha/2) \rangle = \langle \cos(\alpha) \rangle
\]
to increase the energy resolution without drastic loss of intensity as in QENS-ToF energy transfer *is causing a phase shift of the neutron spin precession angle* for each scattered neutron.

\[ \int_{-\infty}^{+\infty} S(Q, \omega) \cos(\omega \tau_{NSE}) d\omega \]

\[ \lambda_2 > \lambda_1 \]

\[ \tau_{NSE} = \frac{m^2 \gamma \int Bdz}{2\pi h^2} (\lambda_0')^3 \]

Attention aux systèmes H/D !!!!
<table>
<thead>
<tr>
<th></th>
<th>IN11</th>
<th>IN15</th>
</tr>
</thead>
<tbody>
<tr>
<td>Instrument type</td>
<td>NSE</td>
<td>NSE</td>
</tr>
<tr>
<td>Beam wavel. [Å]</td>
<td>4–12</td>
<td>6–27</td>
</tr>
<tr>
<td>Beam energy [meV]</td>
<td>0.6 –5</td>
<td>0.1 –2.3</td>
</tr>
<tr>
<td>Time range [ns]</td>
<td>0.001 – 50</td>
<td>0.002 – 1000</td>
</tr>
<tr>
<td>Momentum transfer range [Å⁻¹]</td>
<td>0.02–2.7</td>
<td>0.01–1.8</td>
</tr>
<tr>
<td>Max. sample flux [cm⁻²s⁻¹]</td>
<td>2 × 10⁷</td>
<td>2 × 10⁸</td>
</tr>
<tr>
<td>Detector efficiency</td>
<td>~ 1</td>
<td>~ 1</td>
</tr>
<tr>
<td>Det. backgr. [Hz]</td>
<td>~ 1</td>
<td>~ 1</td>
</tr>
<tr>
<td>Det. solid angle [sr]</td>
<td>3 × 10⁻⁴</td>
<td>4 × 10⁻³</td>
</tr>
<tr>
<td></td>
<td>IN11A</td>
<td>IN11C</td>
</tr>
</tbody>
</table>
Comparaison TOF-BS et NSE

• Frequence measurement
• Scan at fixed $\omega$
• $S(Q,\omega)=S(Q,\omega)\otimes R(Q,\omega)$
• Self motions
• Shorter dynamical range
• Good $Q$ resolution
• Excitations, vibrations

• Time measurement
• Scan at fixed $t$
• $S(Q,t)=S(Q,t).R(Q,t)$
• Collective motions
• Large dynamical range (3 to 4 décades)
• $\Delta\lambda/\lambda \sim 15\% = \Delta Q/Q$
Recent Progress on Polymer Dynamics by Neutron Scattering: From Simple Polymers to Complex Materials

Juan Colmenero,1,2 Arantxa Arbe1

**TABLE 2** Currently operating high-level neutron facilities in the world and the available QENS spectrometers on them

<table>
<thead>
<tr>
<th>Facility</th>
<th>Instrument</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Institute Laue-Langevin (ILL)</td>
<td>IN10</td>
<td>BS</td>
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<tr>
<td>Grenoble, France</td>
<td>IN16</td>
<td>BS</td>
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<tr>
<td><a href="http://www.ill.eu">www.ill.eu</a></td>
<td>IN16a</td>
<td>BS</td>
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<tr>
<td></td>
<td>IN13b</td>
<td>BS</td>
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<tr>
<td></td>
<td>IN5</td>
<td>ToF</td>
</tr>
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<td></td>
<td>IN6</td>
<td>ToF</td>
</tr>
<tr>
<td></td>
<td>IN11</td>
<td>NSE</td>
</tr>
<tr>
<td>Helmholtz Zentrum Berlin (HZB)</td>
<td>NEATa</td>
<td>ToF</td>
</tr>
<tr>
<td>Berlin, Germany</td>
<td><a href="http://www.helmholtz-berlin.de">www.helmholtz-berlin.de</a></td>
<td></td>
</tr>
<tr>
<td>Paul Scherrer Institute (PSI)</td>
<td>MARS</td>
<td>BS-ToF</td>
</tr>
<tr>
<td>Villigen, Switzerland</td>
<td>FOCUS</td>
<td>ToF</td>
</tr>
<tr>
<td>sinq.web.psi.ch</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Laboratoire Léon-Brillouin (LLB)</td>
<td>FA#c</td>
<td>ToF</td>
</tr>
<tr>
<td>Saclay, France</td>
<td>MUSES</td>
<td>NSE</td>
</tr>
<tr>
<td>www-llb.cea.fr</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Spallation Neutron Source (SNS)</td>
<td>BASIS</td>
<td>BS-ToF</td>
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<tr>
<td>Oak Ridge, USA</td>
<td>CNCS</td>
<td>ToF</td>
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<tr>
<td>ISIS Facility, Rutherford</td>
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<tr>
<td>Appleton Laboratory</td>
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<td></td>
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<tr>
<td>Oxford, United Kingdom</td>
<td></td>
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<tr>
<td><a href="http://www.isis.stfc.ac.uk">www.isis.stfc.ac.uk</a></td>
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</tbody>
</table>

Quasielastic Neutron Scattering in Biology
Part I: Methods

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