Incoherent Quasielastic Neutron Scattering with NSE

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Pro vs Cons

Cons

- Depolarization of the beam.

    Best flipping ratio 0.5.
- Scattering is diffuse over 4π.

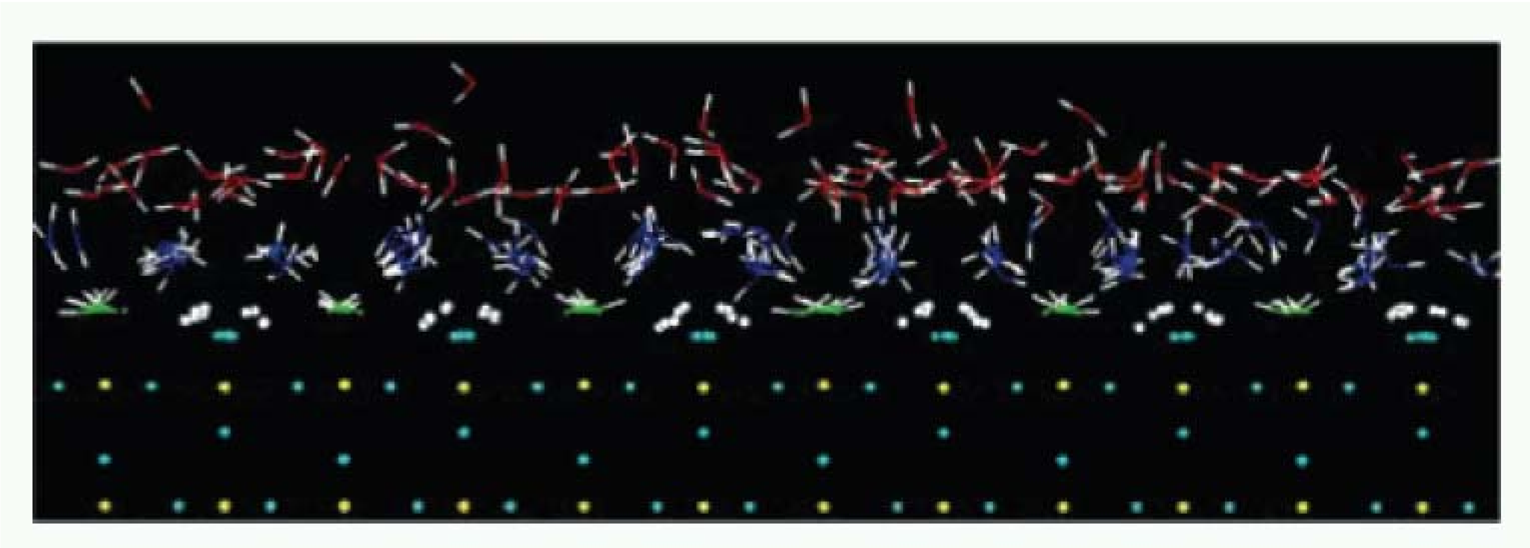
    NSE can only measure in a limited range.
- Long counting times.

Pros

- Best energy resolution available.
- Resolution effects can easily be taken into account.
- Can cover up to ~4 orders of magnitude in time.

Water Dynamics in ZrO_2

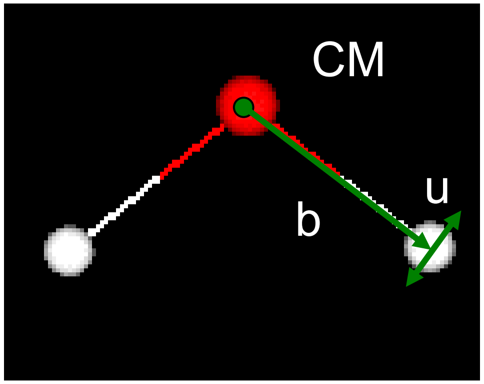
- Two layers of hydration water.



Mamontov E. et al., "Dynamics and Structure of Hydration Water on Rutile and Cassiterite Nanopowders Studied by Quasielastic Neutron Scattering and Molecular Dynamics Simulations", *J. Phys. Chem. C*, **111**, 4328 (2007).

Decoupling Approximation

$$I(Q,t) = \left\langle \exp\left[i\vec{Q}\left(\vec{r}_i(t) - \vec{r}_i(0) \right) \right] \right\rangle$$



$$I(Q,t) = \left\langle \exp\left[-i\vec{Q}\left(\vec{r}_{CM}(t) - \vec{r}_{CM}(0) + \vec{b}(t) - \vec{b}(0) + \vec{u}(t) - \vec{u}(0) \right) \right] \right\rangle$$

$$I(Q,t) = \left\langle \exp\left[-i\vec{Q}\left(\vec{r}_{CM}(t) - \vec{r}_{CM}(0) \right) \right] \right\rangle \left\langle \exp\left[-i\vec{Q}\left(\vec{b}(t) - \vec{b}(0) \right) \right] \right\rangle \left\langle \exp\left[-i\vec{Q}\left(\vec{u}(t) - \vec{u}(0) \right) \right] \right\rangle$$

Vibrational Dynamics

It is very fast… too fast to be seen by QENS.

The only effect is what is usually called the Debye-Waller factor.

$$I^V(Q,t) = \exp\left[-\frac{1}{3}Q^2u^2\right]$$

Rotational Dynamics

The exact Sears expansion holds:

$$I^R(Q,t) = \sum_{l=0} (2l+1)\, j_l^2(Qb) C_l(t)$$

$$C_l(t) = \langle P_l[b(t)\cdot b(0)]\rangle$$

Under the assumption of isotropic rotational diffusion:

$$C_l(t) = \exp[-l(l+1)D_R t]$$

Sears V.F., *Can. J. Phys.*, **44,** 1299 (1966).

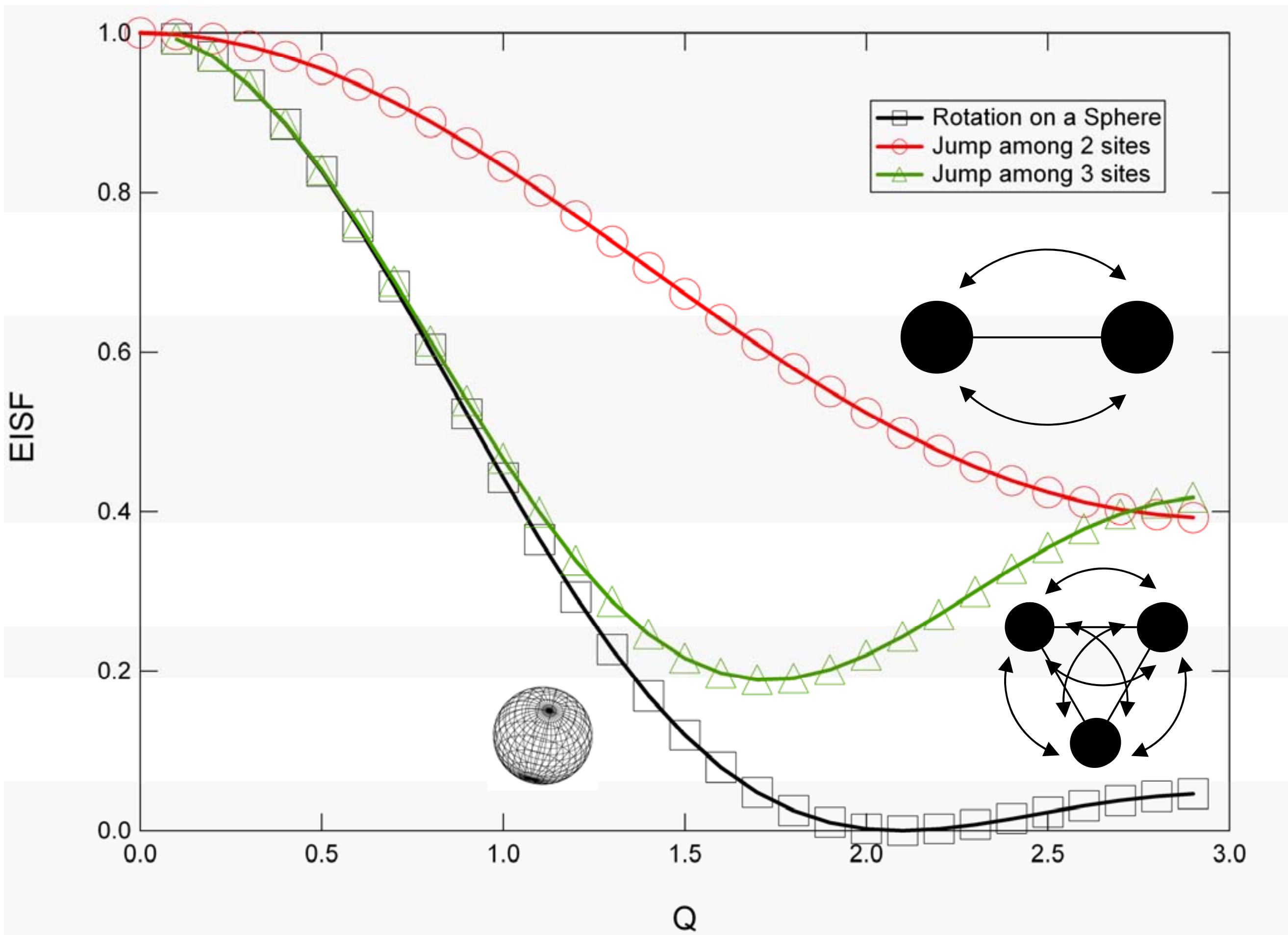
In general

$$I^R(Q,t) = A_0(Q) + \sum_{l=1} A_l(Q) F_l(t)$$

A_0 is the Elastic Incoherent Structure Factor (EISF)

The EISF is the form factor corresponding to the area explored by the scatterer in the limit of the time corresponding to the instrumental resolution.

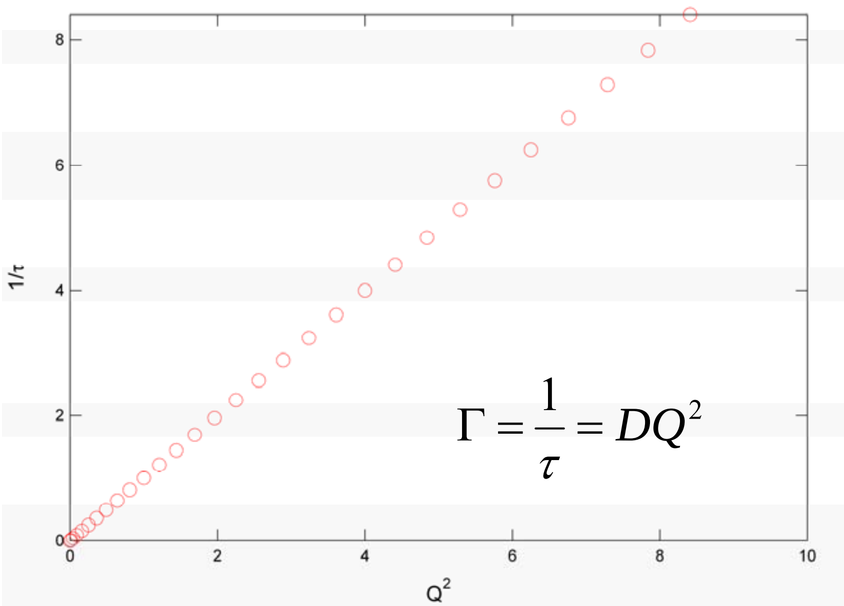
EISFs


Rotation on a Sphere
Jump among 2 sites
Jump among 3 sites
EISF
Q
1.0
0.8
0.6
0.4
0.2
0.0
0.0
0.5
1.0
1.5
2.0
2.5
3.0

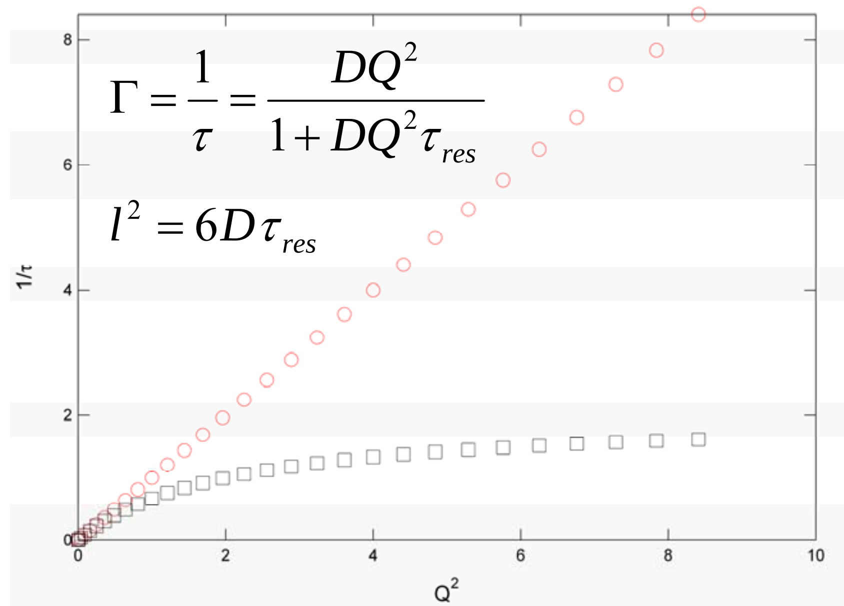
Translational dynamics

$$I^T(Q,t) = \exp(-\Gamma t)$$

Diffusion



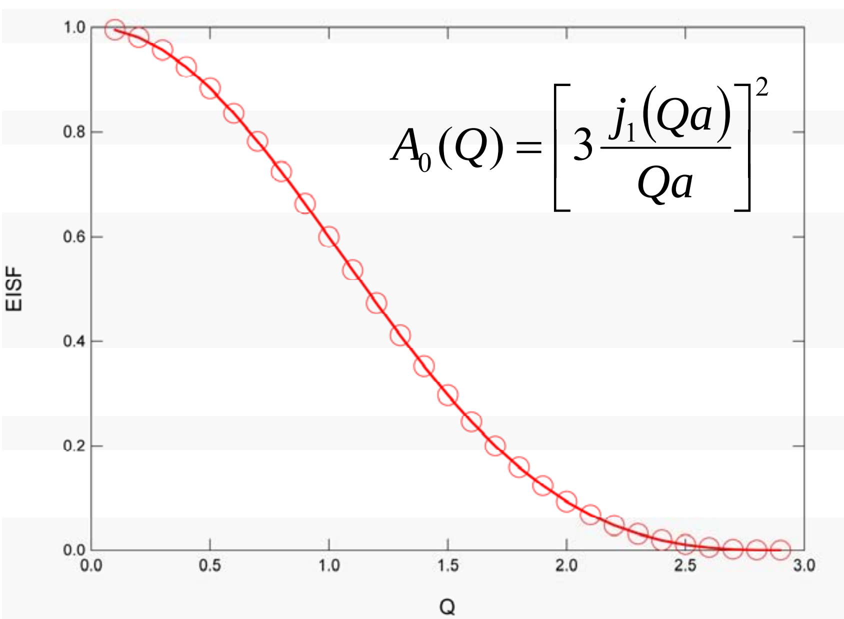
Random Jump Diffusion

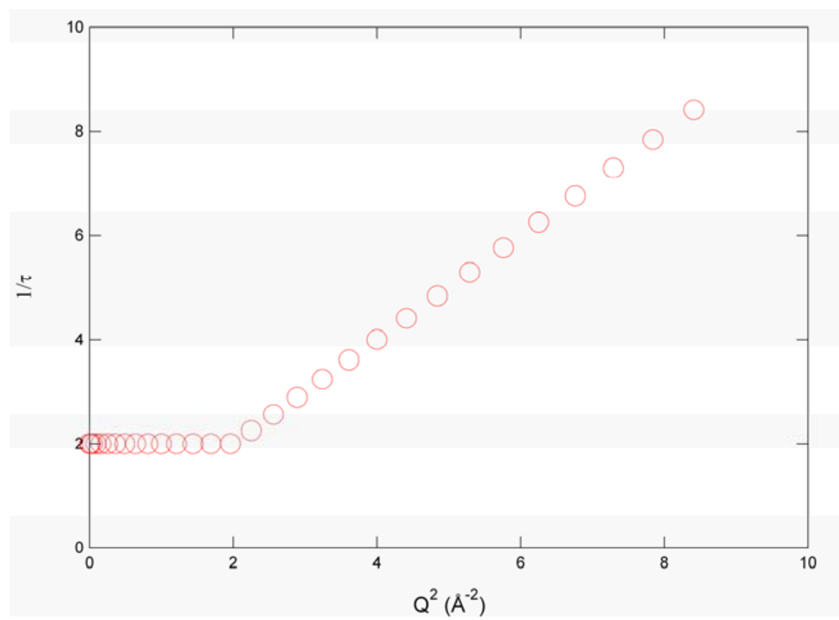


EISF for translational Dynamics

- Dynamics in Confinement:

$$I^{T}(Q,t) = A_0(Q) + \sum_{l=1} A_l(Q) F(Q,t)$$





Volino F. and Dianoux A.J., *Mol. Phys.*, **41,** 271 (1980).

Things can get complicated…

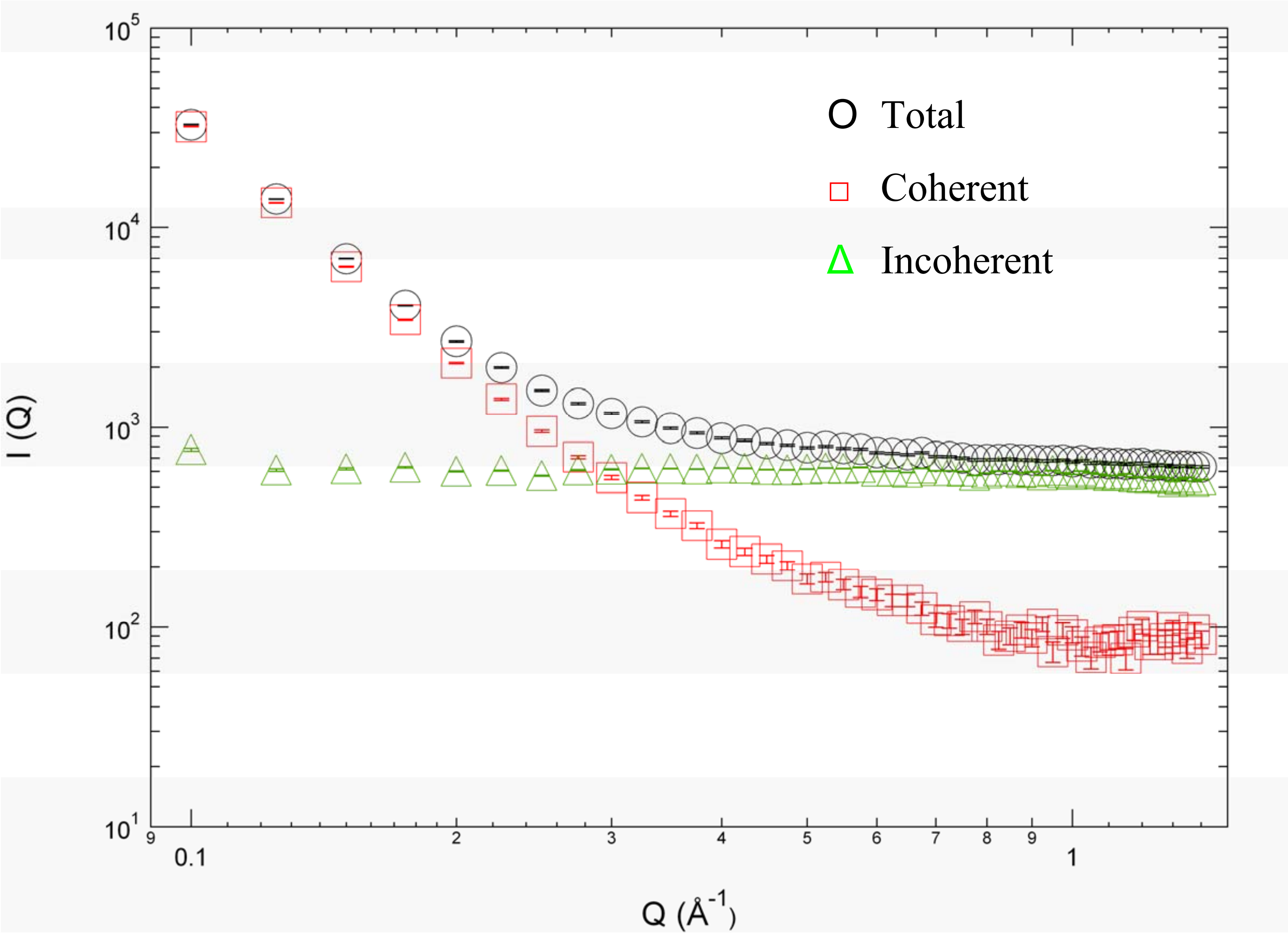
- Multiple relaxation.
- Non exponential decays.

Stretched exponential (KWW)

$$\exp\left[-\left(\frac{t}{\tau}\right)^{\beta}\right]$$

$$\langle\tau\rangle = \frac{\tau}{\beta}\Gamma\left(\frac{1}{\beta}\right)$$

Choose the Q range carefully



Conclusion

- Incoherent Dynamics on NSE is difficult.
- But… it can be worthy.