mccluskey.scot/talks/panosc

QENS for battery materials: and QENS analysis

PaNOSC Summer School — 2022/09/14

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Outline

0. Neutron scattering
1. What is QENS?
2. How does QENS probe diffusion?
3. Why is it important for energy materials?
4. How do we analyse QENS data?
5. How else can we study diffusion?
6. What is the plan for the rest of the day?

Some ground rules

- I talk (quickly $*$ with an accent).
- Please shout if this convolution hinders understanding.
- Also shout if you have questions!

0. Neutron scattering

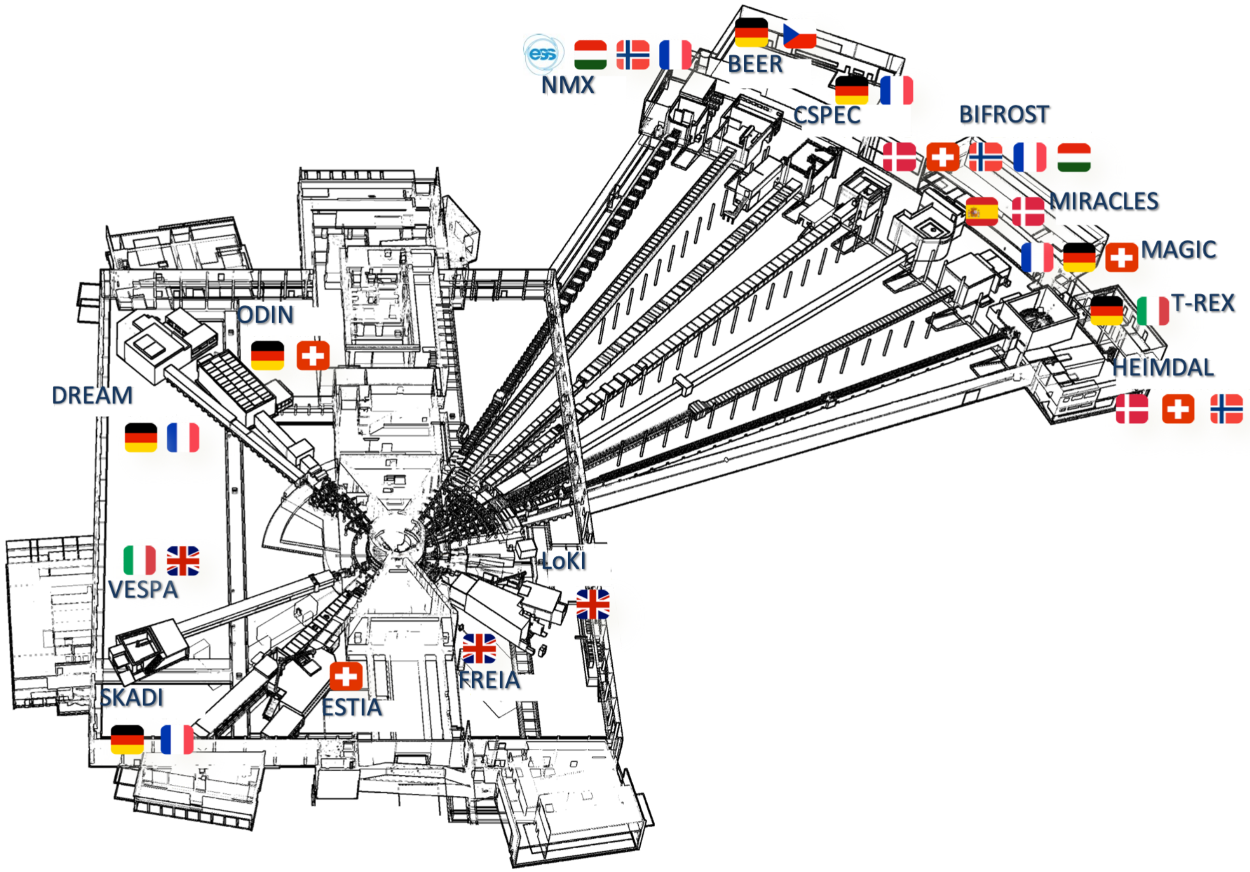
Neutron sources

- Neutrons for science can be produced at reactor or spallation sources (also low energy neutrons but these are less common).



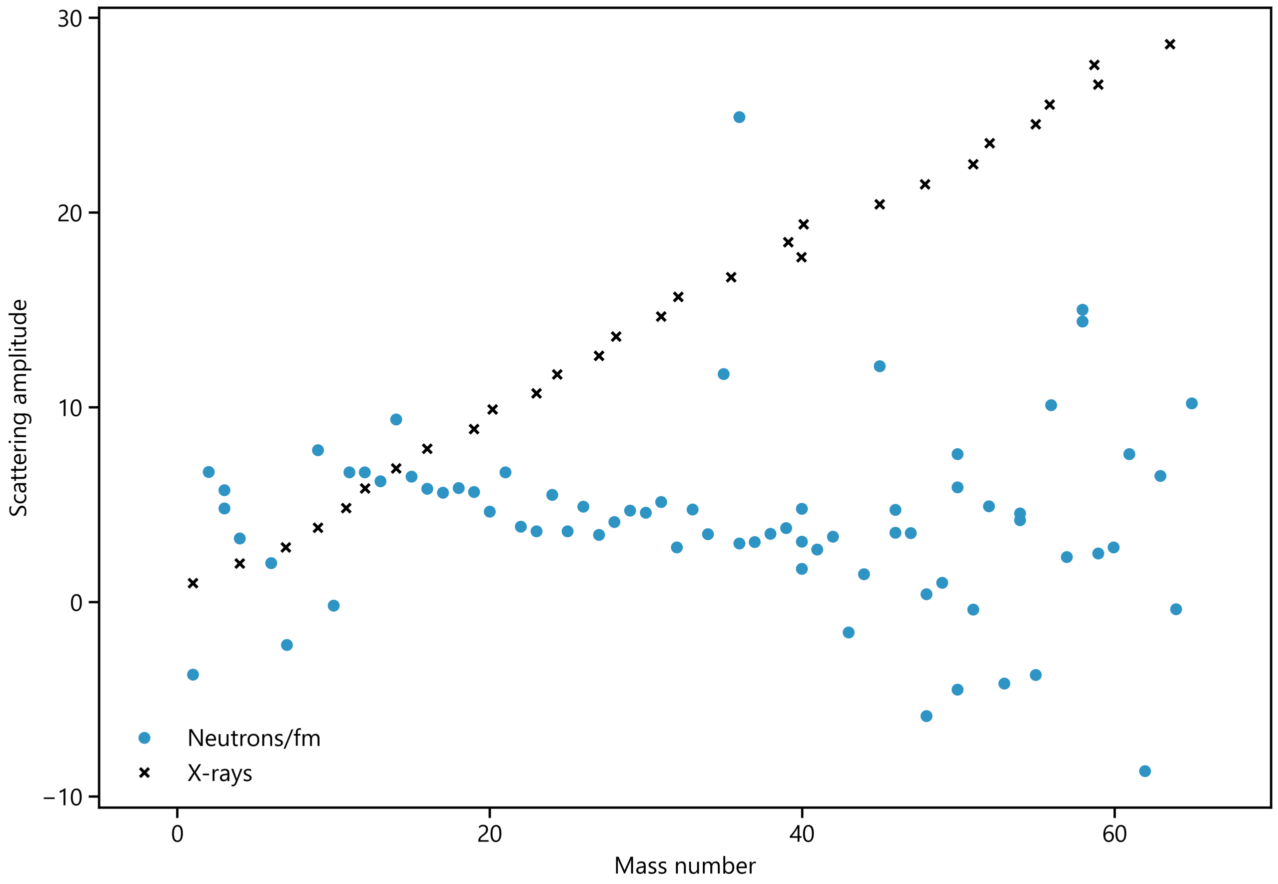
Neutron instruments

- The neutrons are then guided to instruments where they are scattered.
- The scattering gives information about structure and dynamics of a material.



Neutron scattering

- Neutron scattering gives different information to X-ray scattering.
- Neutrons interact with nuclei, X-rays with the electron cloud.



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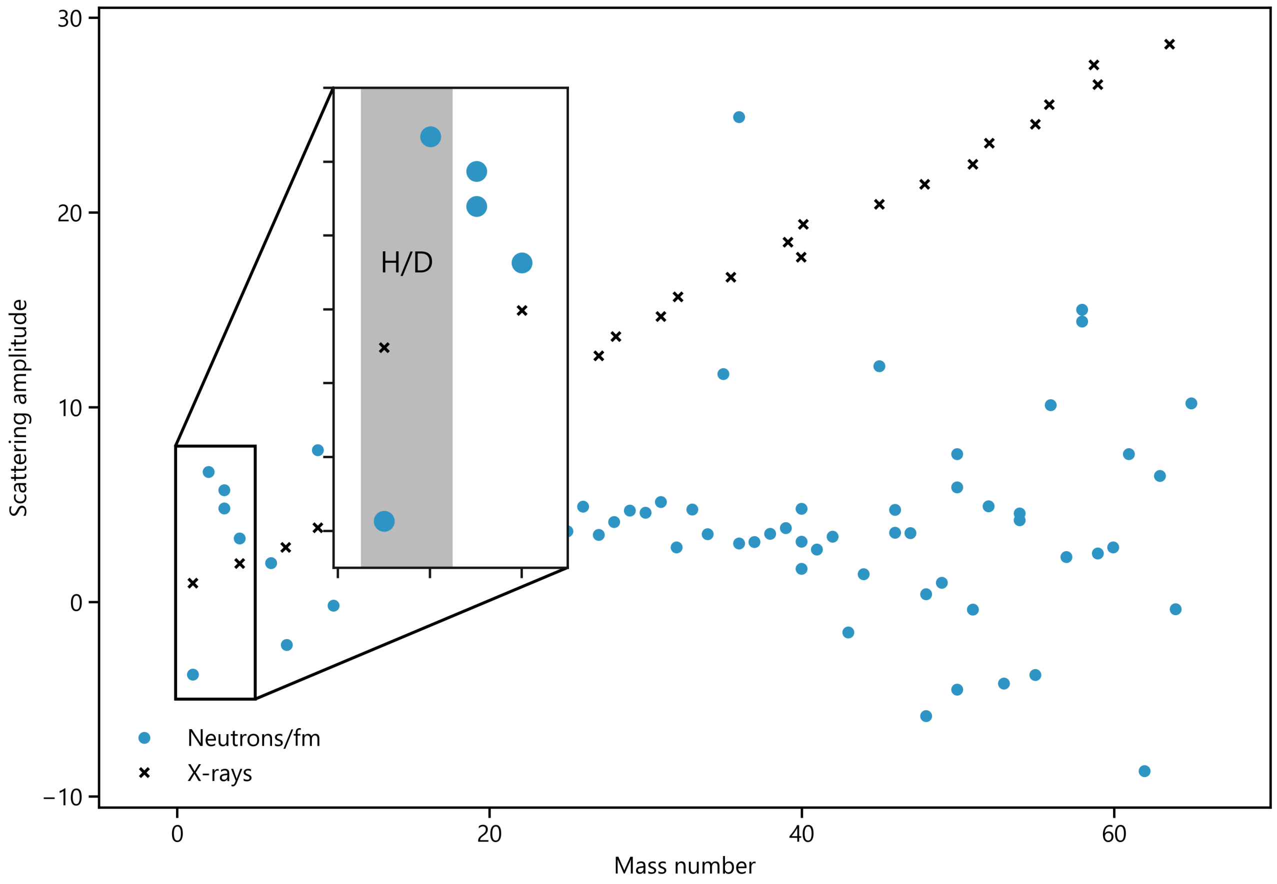
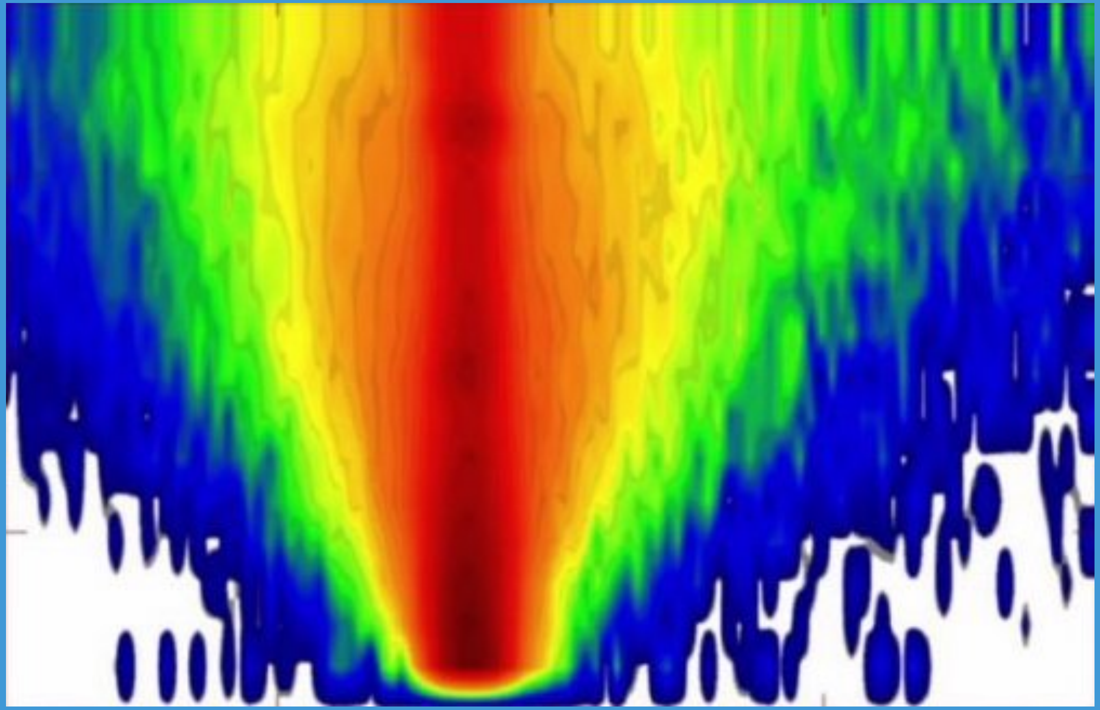
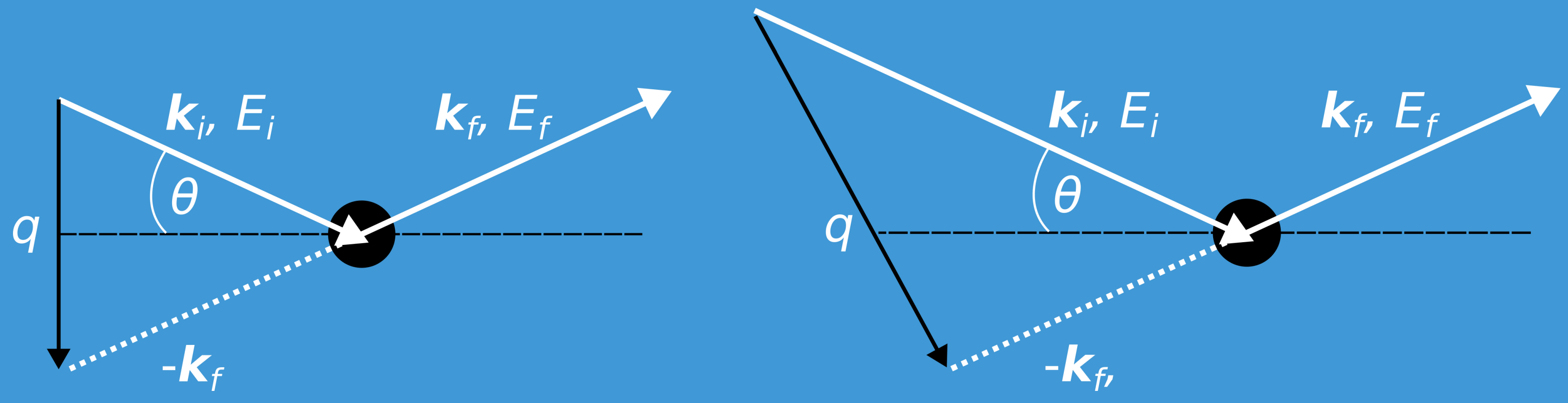


1. What is QENS?



Neutron scattering

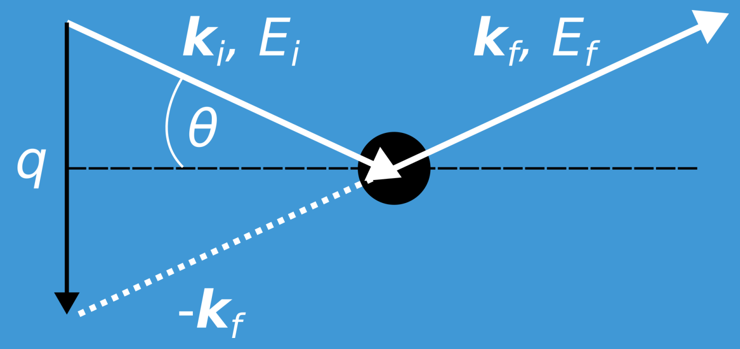
- Unlike light, neutrons interact with the nuclei atoms.
- In this interaction, the incident neutron is scattered by some angle and there may be energy transfer.
  - **Elastic scattering**: only scattered.
  - **Inelastic scattering**: scattering + energy transfer.



Elastic and inelastic scattering process diagrams

Elastic scattering

- No energy is transferred to or from the incident neutron.
    - This gives information about the structure of the material.
    - From, for example, Bragg diffraction or small angle scattering.



Elastic scattering process diagrams.

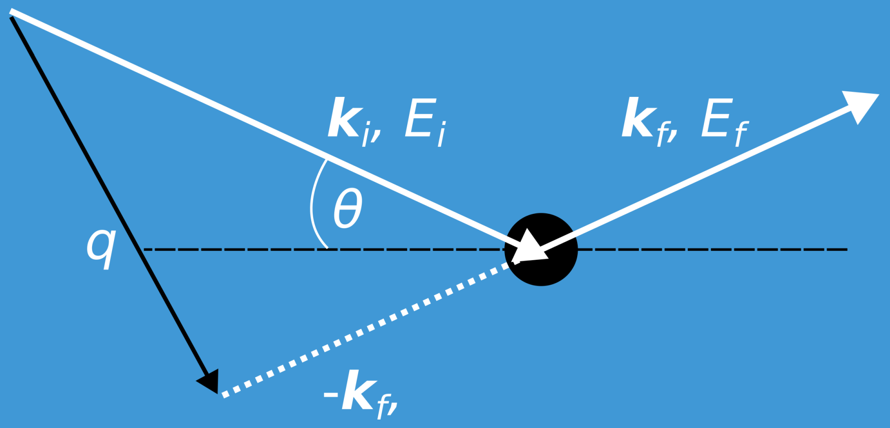
$$|\mathbf{k}_i| = |\mathbf{k}_f| = \frac{2\pi}{\lambda}$$

$$q = 2|\mathbf{k}|\sin\theta = \frac{4\pi}{\lambda}\sin\theta$$

$$\Delta E = 0$$

Inelastic scattering

- Energy is transferred, as momentum of the neutron.
  - The neutron can either gain or lose energy.
  - From this we gain information about the material dynamics.



Inelastic scattering process diagrams.

$$|\mathbf{k}_i| \neq |\mathbf{k}_f|$$

$$\hbar q = \hbar(\mathbf{k}_i - \mathbf{k}_f)$$

$$\Delta E = \hbar\omega = E_i - E_f$$

The scattering function

- The scattering function, $S(q, \omega)$, is the measurable in a neutron scattering experiment.
- This contains all the physics of the system, in space and time.

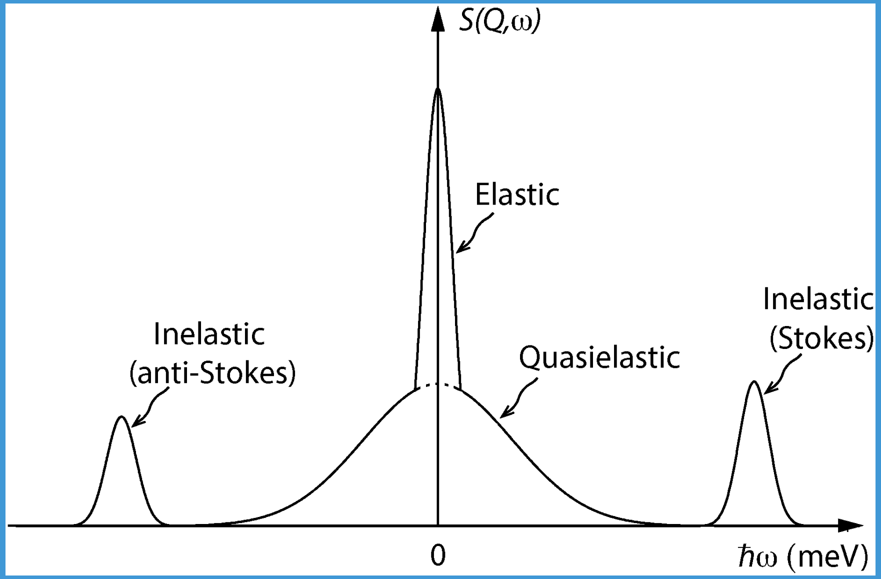
$$\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{k_f}{k_i} \frac{\sigma S(q, \omega)}{4\pi}$$

$d\Omega$ is the angular space, $d\omega$ is the energy space.

- The sum of the coherent and incoherent scattering.

$$\sigma S(q, \omega) = \sigma_{\text{inc}} S_{\text{inc}}(q, \omega) + \sigma_{\text{coh}} S_{\text{coh}}(q, \omega)$$

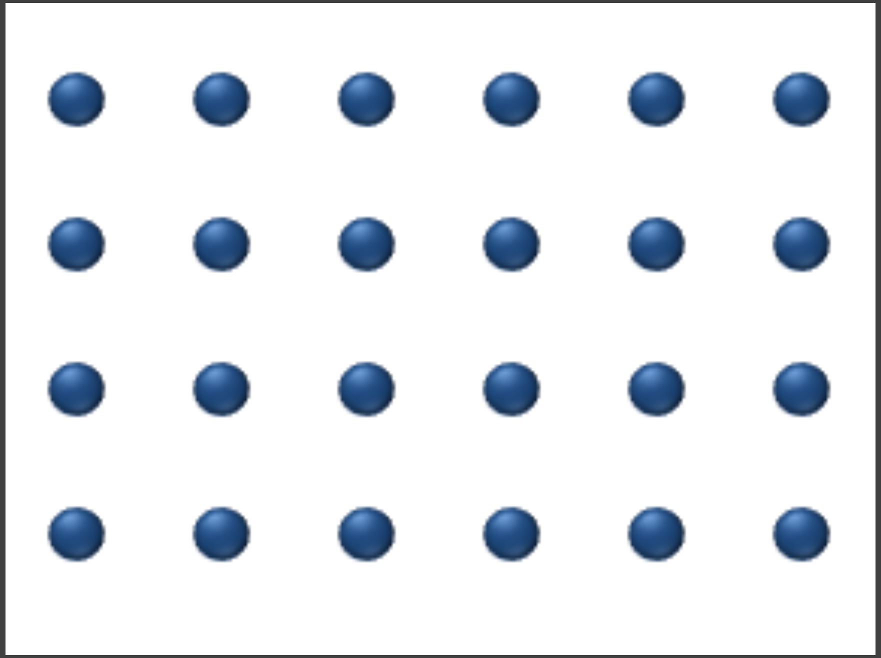
Neutron spectroscopy; energy spectra



M. Karlsson, *Phys. Chem. Chem. Phys.* 2015, **17**, 26-38. doi: 10.1039/C4CP04112G

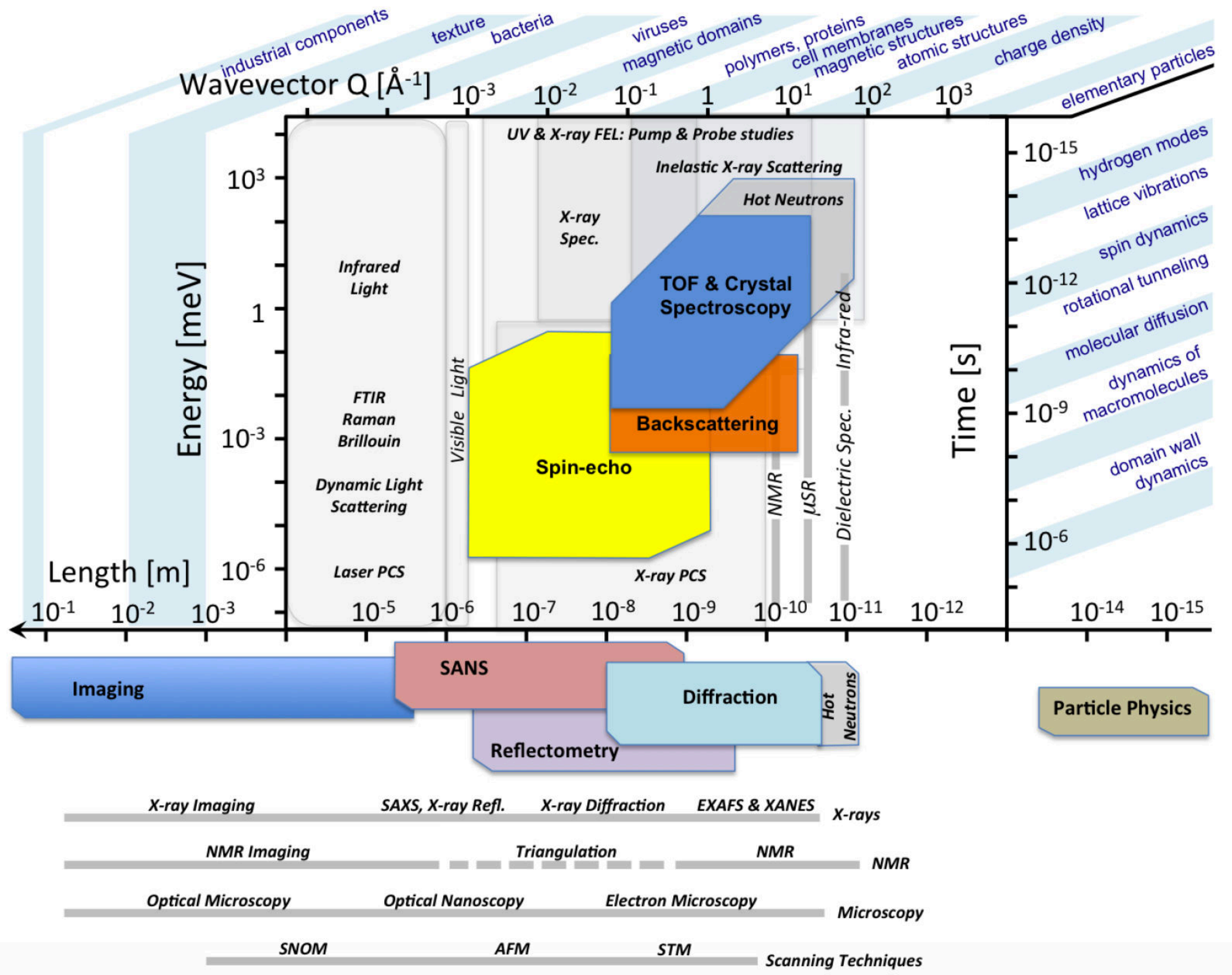
- Elastic scattering: no energy transfer, ideally a delta-function but in reality not.
- Inelastic scattering: energy transfer, $\hbar\omega \neq 0$. Processes with discrete energy steps (vibrational/stretching modes).
- Quasi-elastic scattering: small energy transfer, $\hbar\omega \neq 0 \approx$ neV or µeV. Processes occuring with a distribution of energies (rotations and **translations**).

2. How does QENS probe diffusion?



Runningamok19, Chemical_surface_diffusion_slow, Wikipedia, CC-BY 3.0. Accessed 2022-09-02.

What does QENS probe?



ESS: Technical Design Report 2013; Chapter 2

Incoherent scattering

- Describes dynamics of individual nuceli.

$$S_{\text{inc}}(q,\omega) \xrightarrow{\mathcal{F}} I_{\text{inc}}(q,t) \xrightarrow{\mathcal{F}^{-1}} G_{\text{self}}(r,t)$$

- $G_{\text{self}}(r,t)$ is the probability that the same nuclei will be found at position $\boldsymbol{r}$ at time $\mathbf{0}$ and time $\boldsymbol{t}$.

Coherent scattering

- Describes collective dynamics of nuclei.

$$S_{\mathrm{coh}}(q,\omega) \xrightarrow{\mathcal{F}} I_{\mathrm{coh}}(q,t) \xrightarrow{\mathcal{F}^{-1}} G(r,t)$$

- $G(r,t)$ is the probability that any nuclei will be found at position $\boldsymbol{r}$ at time $\mathbf{0}$ and time $\boldsymbol{t}$.

Some scattering cross sections

- Hydrogen is very popular for QENS studies; as $\sigma_{\mathrm{inc}} \gg \sigma_{\mathrm{coh}}$.
- Only a few examples of lithium investigations are found in the literature.

i.e. M. J. Klenk, *et al.*, *Solid State Ionics*, **312**, 1-7, 2017. & H. Nozaki, *et al.*, *Solid State Ionics*, **262**, 585-588, 2014.

Nuclei	σ_{coh} (barn)	σ_{inc} (barn)
Hydrogen	1.76	80.20
Deuterium	5.59	2.05
Lithium	**0.454**	**0.92**
Carbon	5.56	0.00

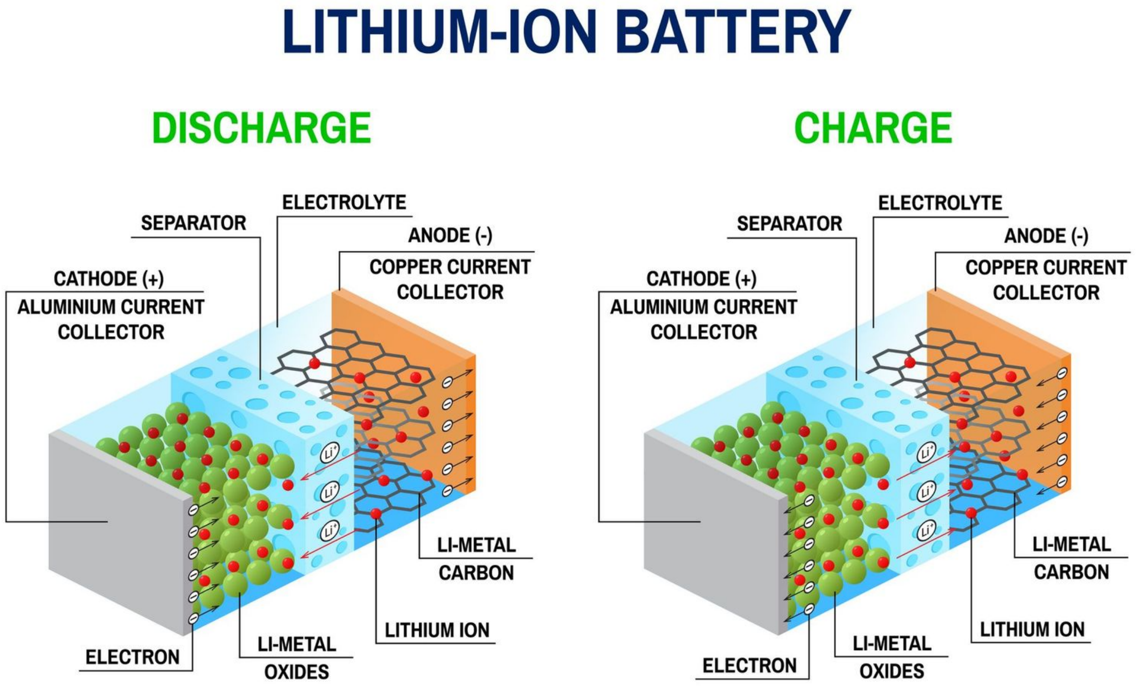
NIST Center for Neutron Research Database

3. Why is it important for energy materials?

Energy Materials | ANSTO

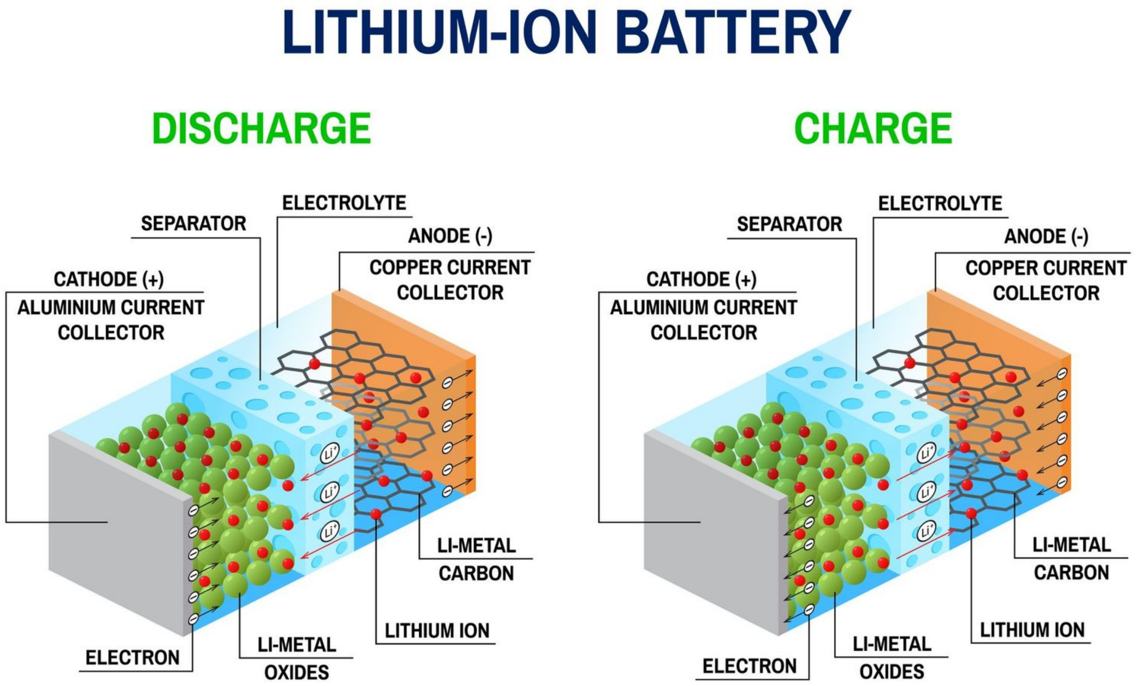
Diffusion in energy materials

- Energy materials covers a broad range of technologies; including photovoltaics and battery materials.
- Most modern devices are powered by a lithium-ion battery.
  - The function of lithium-ion batteries is dependent on the diffusion of lithium, leading to conductivity.



Diffusion in lithium ion batteries

- The faster the lithium can diffuse in the cathode material, the faster the battery can charge.

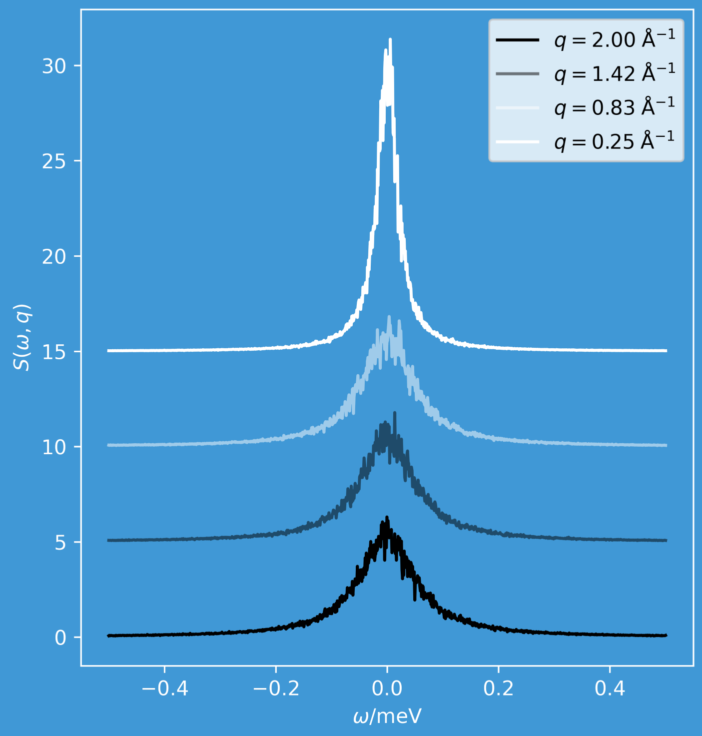


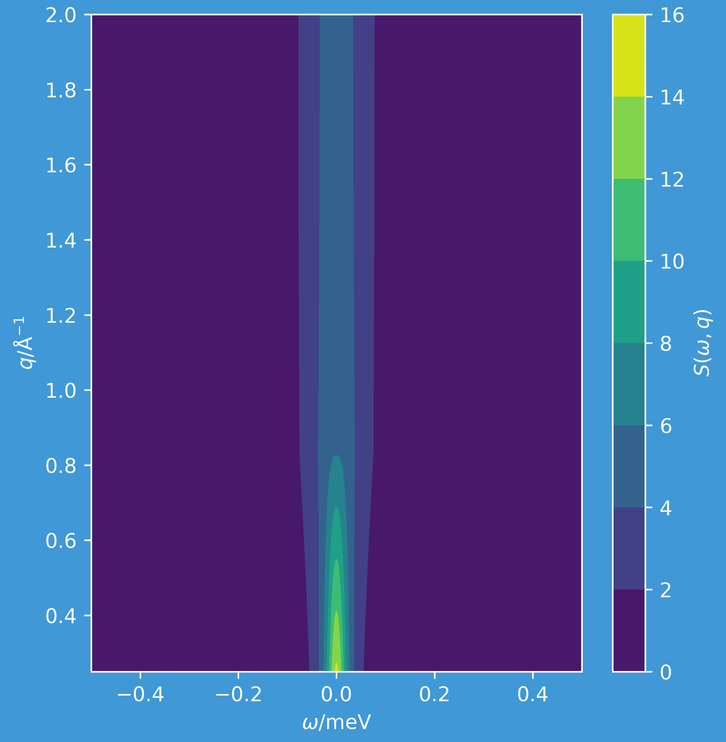
electricbee.co/lithium-ion-battery-how-does-it-work

4. How do we analyse QENS data?

Why your data analysis may be doomed from the start | TechRepublic

An example QENS dataset



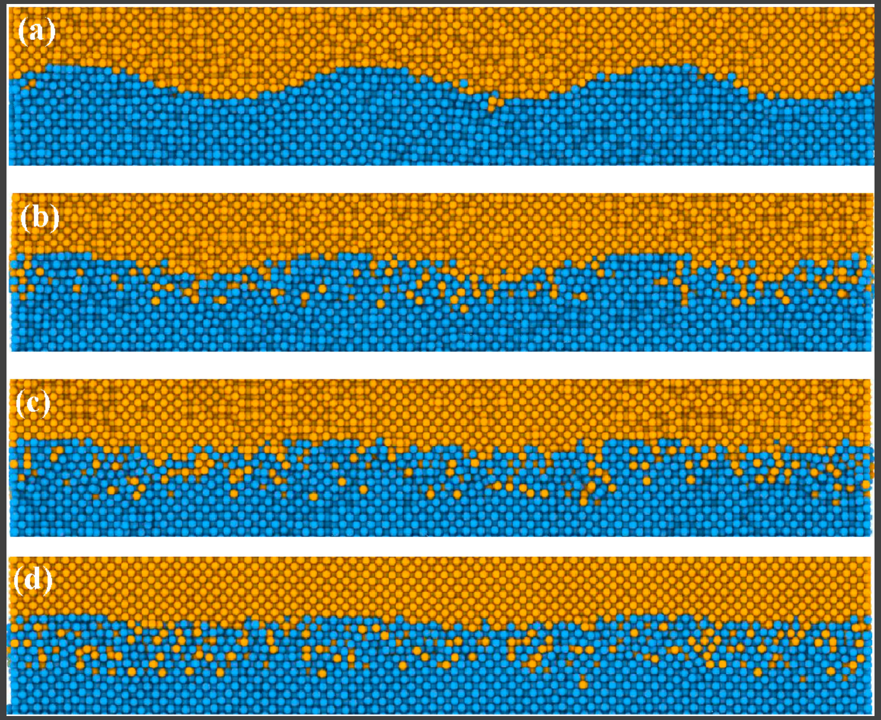


- Energy spectrum is measured at discrete $\boldsymbol{q}$ points.

Modelling

- The data is modelled using analytical functions describing diffusion.
- These functions have mathematical forms, and we fit the parameters to model best the observed data.
- These include (but are not limited to):
  - Singwi-Sjölander model;
  - Chudley-Elliot model; and
  - Random-jump model.
- In this workshop, we will focus on using the random-jump model.

5. How else can we study diffusion?



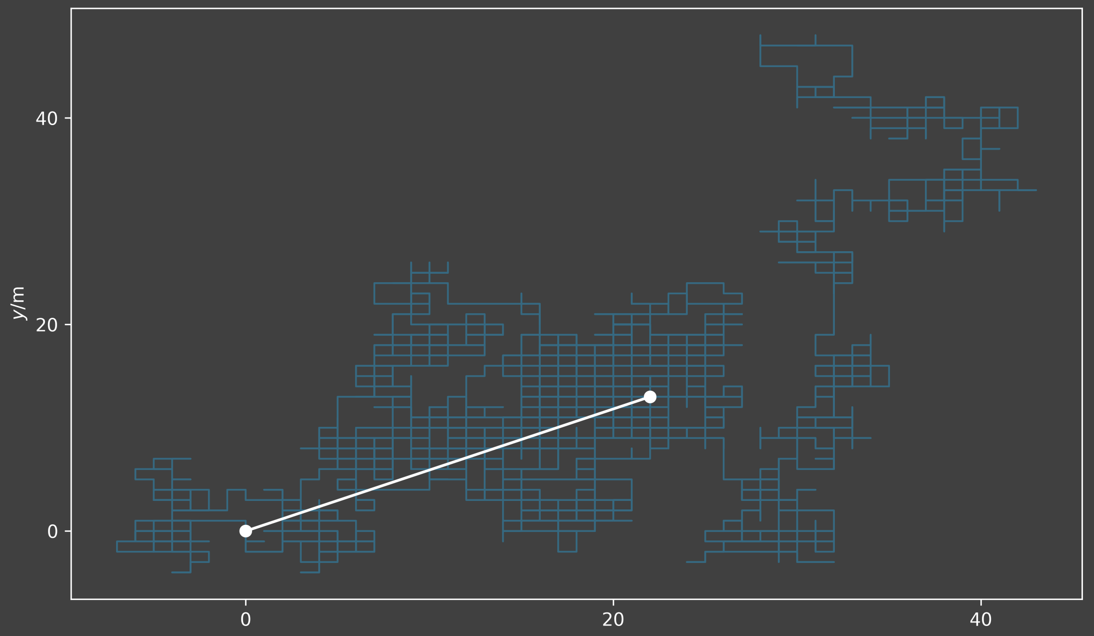
J. Wang, J. Zhang, & J. Qiao, *Materials*, 12(14), 2306, 2019.

Other experimental/simulation techniques

- Diffusion can also be studied with:
  - Nuclear magnetic resonance spectroscopy;
  - Muon-spin relaxation; and
  - Molecular dynamics simulations.

Molecular dynamics simulations

- Molecular dynamics simulations involve computing the forces on the atoms in a system and then using Netwonian mechanics to determine the motions of the atoms as a function of time.
- The forces can be computed using a range of different levels of theory; from potentials to quantum mechanics.
- This gives information about the trajectories (and displacements) of atoms as a function of time.

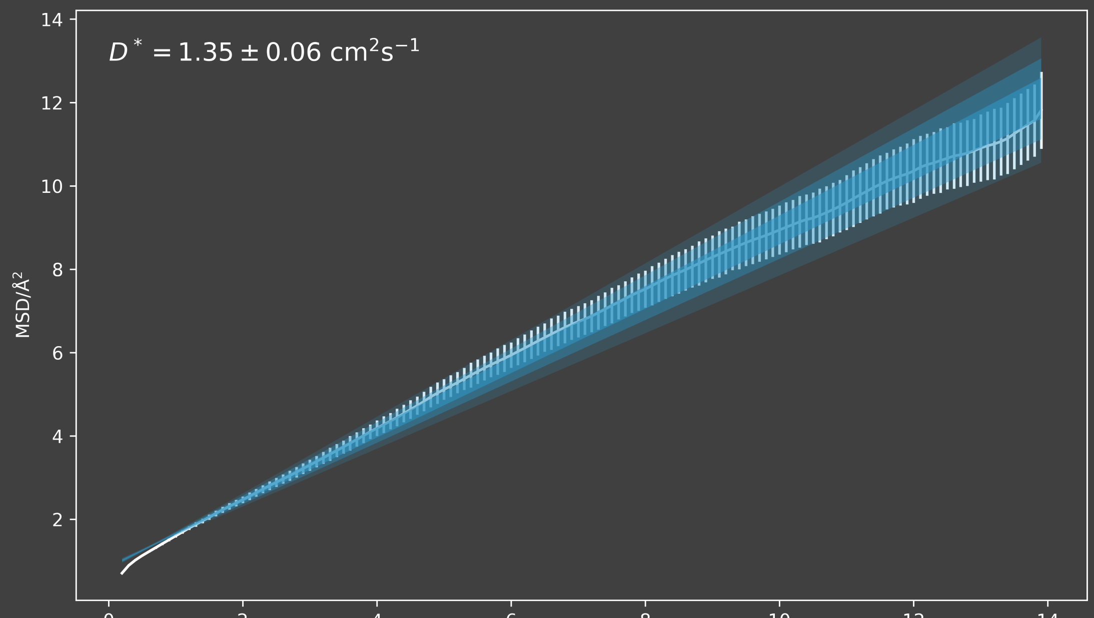


Diffusion *in-silico*

- The displacements of the atoms, as a function of time, can allow us to model the Einstein relation.

$$D^* = \frac{\langle \mathbf{s}^2(\Delta t) \rangle}{2\kappa \Delta t}$$

- This is only exact in the infinite time-limit, therefore, we fit a straight line where the gradient is $2\kappa D^*$.



6. What is the plan for the rest of the day?

The plan

Concept: You have been awarded beamtime to study the diffusion of lithium by QENS at the ISIS Neutron and Muon Source. You need to make sure that you are ready for the beamtime by studying the literature and producing some model data. After the beamtime you need to analyse your data. Throughout you will share your findings in an open and FAIR way.

- Now → 11:15: **Coffee Break** ☕
- 11:15 → 13:00: Literature survey of lithium diffusion
- 13:00 → 14:00: Lunch 🍽️
- 14:00 → 15:00: Feedback on FAIR data
- 15:00 → 15:15: Coffee Break ☕
- 15:15 → 16:30: Analysis of real QENS data