Neutron Scattering and Dynamics

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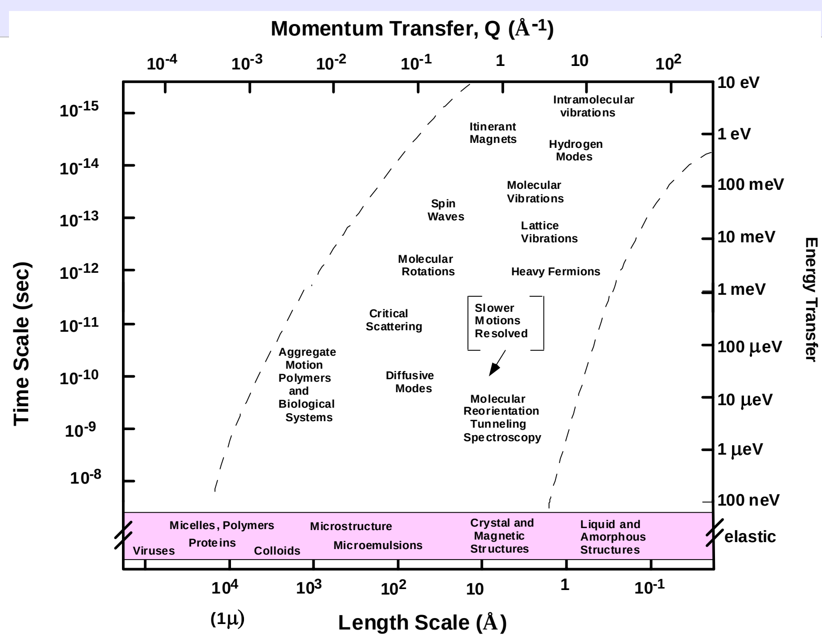
NCNR

CHRNS — Center for High Resolution Neutron Scattering — NSF — NIST

Dynamics of Solids and Liquids



Scattering Probes

How can we measure the atomic-scale properties of solids and liquids?

- ➢Use atomic-scale probes

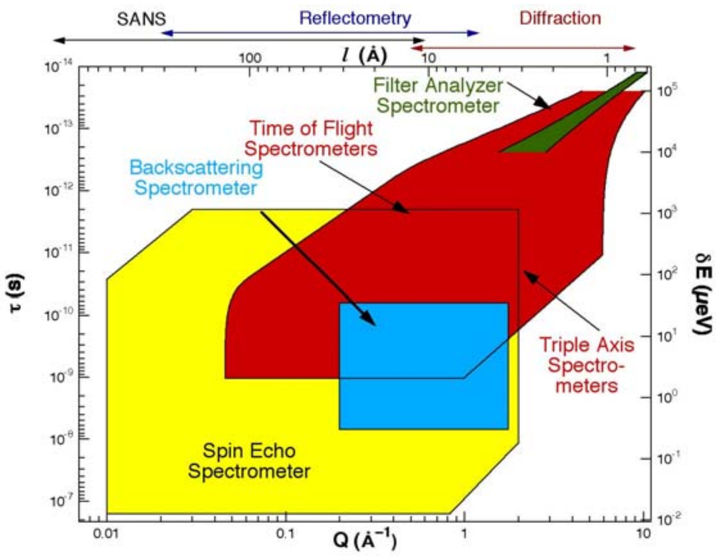
Scattering techniques:

- ➢ Measure how particles scatter off of a sample
- ➢ Scattering depends on interaction between sample and particles
- ➢ Different scattering probes have different characteristics
  - ✓Photons (X-ray, light)
  - ✓Electrons (RHEED, LEED)
  - ✓Helium atoms
  - ✓Neutrons
- ➢ Results can give us information on atomic-scale structure and dynamics

Why Neutrons?

- Wavelength: $\lambda = 9.044/\sqrt{E}$
  - At 10 meV, λ = 2.86 ***Å***
  - ***Similar length scales as structures of interest***
    - → ***Interference effects***
- Energy:
  - Thermal sources: ~ 5-100 meV
  - Cold sources: ~ 1-10 meV
  - Spallation sources: thermal + epithermal neutrons (> 100 meV)
  - Comparable to excitation energies in solids and liquids

Energy and Length Scales



NIST:

7 orders of magnitude range in energy.

3 orders of magnitude in length scale.

Neutron Interactions

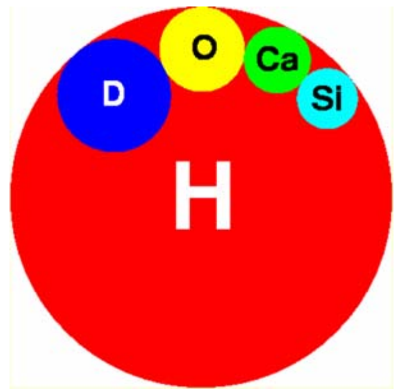
- Zero charge
  - No interaction with charge densities (electrons)
- Nuclear force
  - The interactions that bind neutrons to nuclei also scatter neutrons
- Magnetic dipole moment
  - $\mu_n = 1.04 \times 10^{-3}\ \mu_B$
  - Neutrons scatter from magnetic moments
- Interactions are weak
  - Neutrons penetrate deeply into samples
  - Samples can be enclosed during experiments
  - Sample size an important consideration

Nuclear Interactions

<u>Scattering cross section σ:</u>
Area which represents probability that a neutron will interact with a nucleus.

σ varies "randomly" from element to element and even isotope to isotope.

Typical $\sigma \sim 10^{-24}$ cm^2 for a single nucleus.



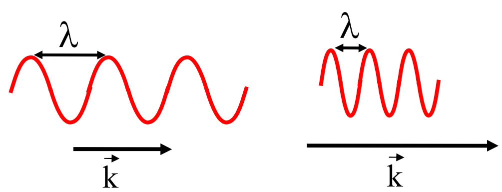
Total nuclear cross section for several isotopes

Neutron Wave Properties

Quantum mechanics: particles have wave properties

Momentum:

$$m\vec{v} = \vec{p} = \hbar\vec{k} = \hbar\frac{2\pi}{\lambda}$$



Energy:

$$E = \frac{1}{2}mv^2 = \frac{\hbar^2}{2m}k^2 = \hbar\omega$$

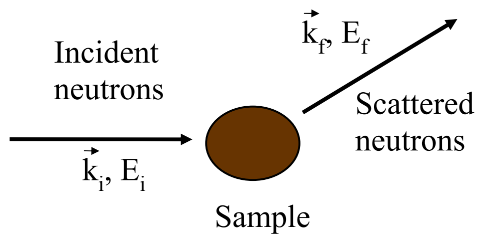
Inverse relationships:
length ~ 1/momentum
time ~ 1/frequency ~ 1/energy

Energy unit conversion:
1 meV ≅ 8 cm^{-1} ≅ 240 Ghz
≅ 12K ≅ 0.1 kJ/mol

Neutron Scattering Event

Scattering basics:

- incident neutron
- scattered neutron



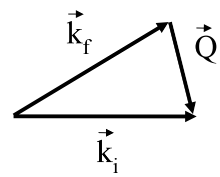
6 independent parameters:

- k_x, k_y, k_z for initial and final neutrons (E depends on k)

Momentum Conservation:

- $\vec{Q} = \vec{k}_i - \vec{k}_f$
- $\vec{Q}$ represents momentum transferred to sample

Scattering triangle:



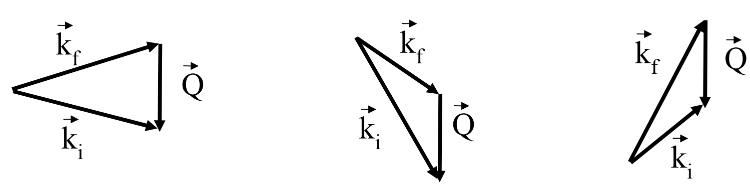
Neutron spectrometer must be able to determine k_i, k_f

Elastic vs. Inelastic

Energy conservation:

$$\Delta E_{neutron} = -\Delta E_{sample}$$

$$\Delta E_{sample} = E_i - E_f \equiv \hbar\omega = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$$

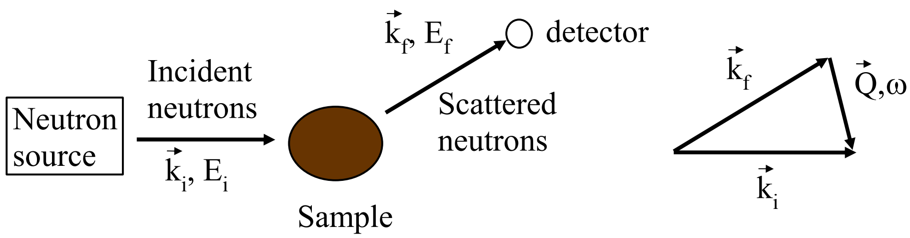


$k_i = k_f$	$k_i > k_f$	$k_i < k_f$
$\omega = 0$	$\omega > 0$	$\omega < 0$
Elastic scattering	Inelastic scattering	

Note: ω can vary indepdently of Q.

Neutron Scattering Measurement

- What is a neutron scattering measurement?
  - Neutron source sends neutrons to sample.
  - Some neutrons scatter from sample.
  - Scattered neutrons are detected.



- What are we measuring?
  - Number of scattered neutrons as a function of (**Q**,ω).
    - Our parameter space is 4-dimensional

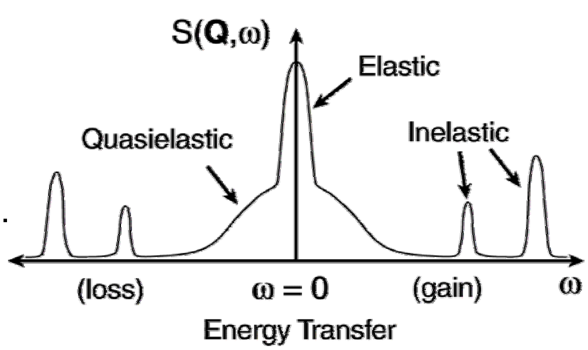
Scattering function **S(Q,ω)**

Intensity (number) of scattered neutrons is proportional to scattering function S(**Q**,ω).

- S(**Q**,ω) depends only on the sample, not on the neutron spectrometer.

What information does S(**Q**,ω) give us?

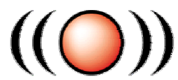
- **Q** gives information about structure.
- ω gives information about dynamics (motion).
  - –Elastic
  - –Quasielastic
  - –Inelastic



Scattering function **S(Q,ω)**

$S(\mathbf{Q},\omega)$ has contributions from single-particle scattering (incoherent) and from multiple-particle scattering (coherent):

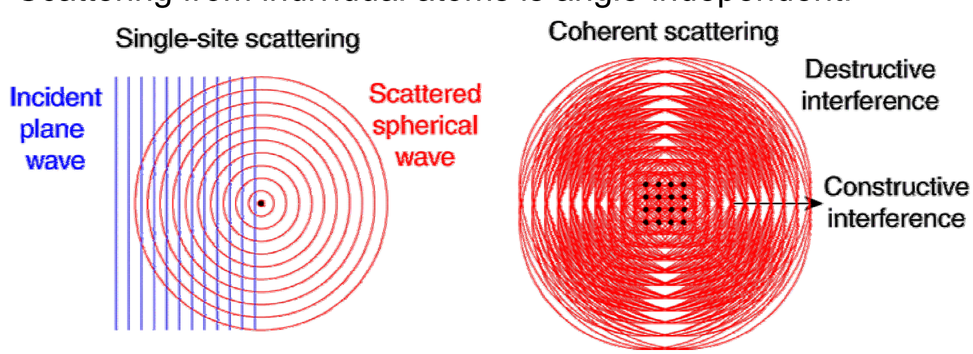
- Incoherent signal: $S_{inc}(\mathbf{Q},\omega)$ is the Fourier transform in space and time of the self correlation function.
  - How do individual atoms behave independent of other atoms?



- Coherent signal: $S_{coh}(\mathbf{Q},\omega)$ is the Fourier transform in space and time of the pair correlation function.
  - How do atoms behave in relation to other atoms?

Coherent Scattering

Scattering from individual atoms is angle-independent.

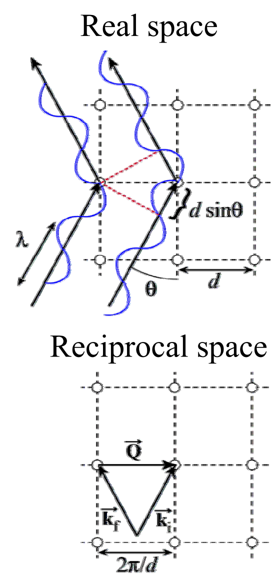


Interference effects:
- Scattering sites are spatially correlated
- Phase of scattered neutrons are correlated

Coherent scattering is angle-dependent
- Angular dependence reveals spatial correlations

Elastic Coherent Scattering

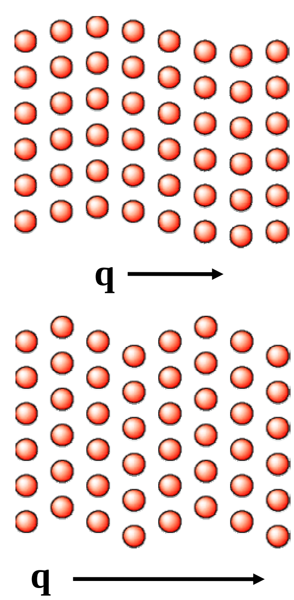
- If the time-averaged pair correlation function is periodic (such as for a crystal), then the Fourier transform will form a reciprocal lattice which is also periodic.
- S(**Q**,0) for a perfect crystal consists of delta functions at periodic **Q** positions. Each delta function corresponds to a Bragg reflection.
- Bragg's Law: $n\lambda = 2d \sin\theta$
  - Constructive interference when distance for two paths is multiple of wavelength
  - gives an intuitive picture of S(**Q**,0)



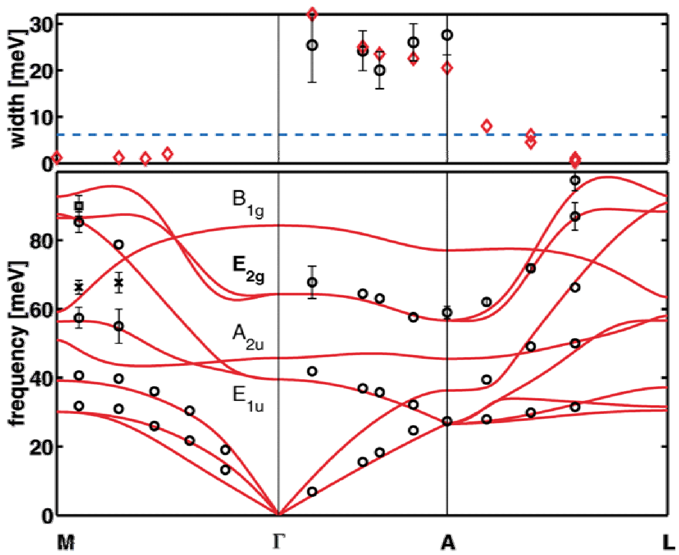
Inelastic Coherent Scattering

Phonons: quantized lattice vibrations
- Motion of atoms
  - → Inelastic scattering
- Correlated motion
  - → Interference effects (coherent scattering)

Phonon properties:
- Frequency depends on propagation vector **q**
  - ω(**q**) is dispersion relation
- Energy is quantized: $E = h\omega$
- Neutron scattering: S(**Q**=**q**,ω(**q**))



Phonon Dispersion: MgB_2

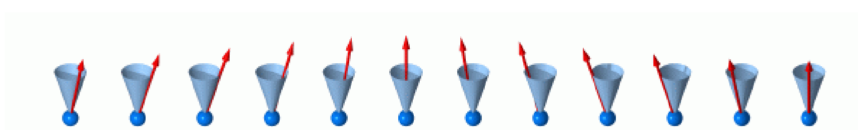


Shukla *et al.*, Phys. Rev. Lett. **90**, 095506 (2003)

Inelastic Magnetic Scattering

Magnetically ordered systems:

- Magnetic moments coupled to neighbors.
- Rotating one spin from equilibrium will exert torque on neighboring moments.
- Spin waves: excitations in magnetic order.

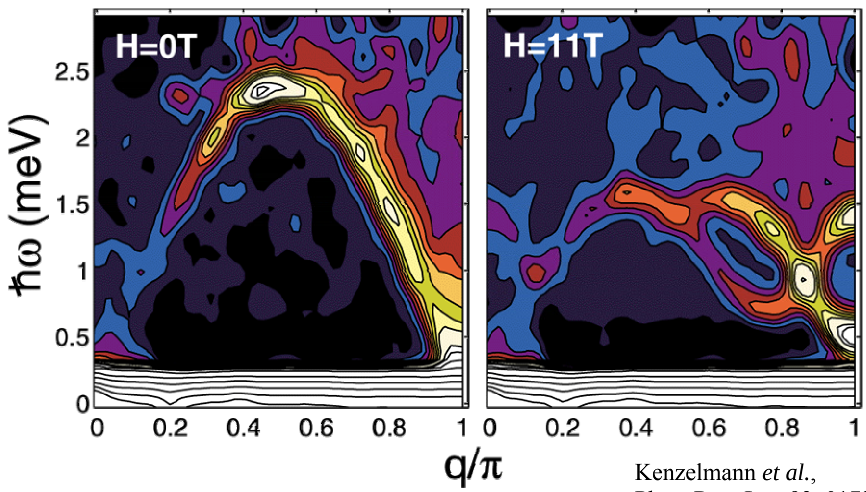


Animation courtesy of A. Zheludev

Inelastic neutron scattering can measure spin wave dispersion.

1D magnetic system: $CuCl_2$

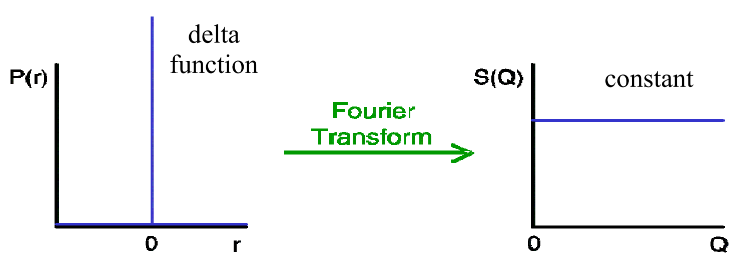
Intensity maps for magnetic scattering



Kenzelmann *et al.*,
Phys. Rev. Lett 93, 017204 (2004)

Incoherent Scattering

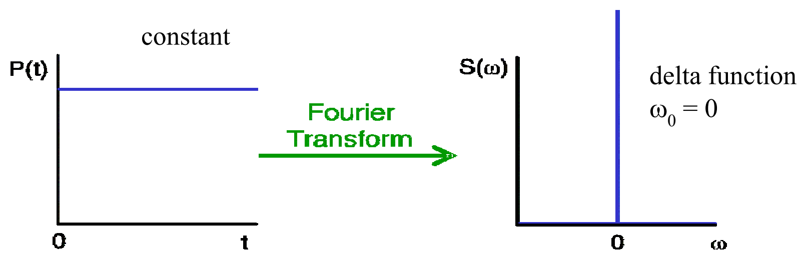
Every particle has perfect instantaneous correlation with itself. Its self-correlation function is therefore a delta function in space.



S_{inc}(**Q**) is independent of **Q. This contribution to the scattering at all momentum transfers is the primary source of background in many experiments.**

Elastic Incoherent Scattering

For a perfectly stationary particle, the self-correlation function is constant in time.

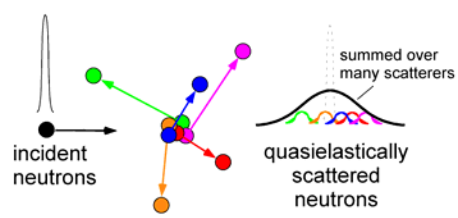


$S_{inc}(\mathbf{Q},\omega)$ for a stationary particle is a delta function in energy ω. **If atoms do not move, all incoherent scattering is elastic (ω = 0).**

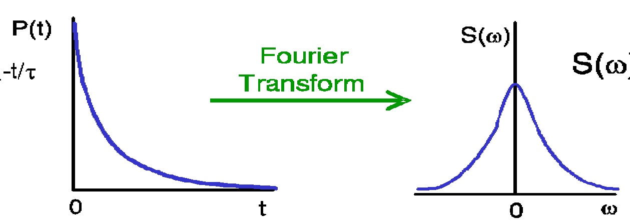
Quasielastic Incoherent Scattering

If an atom is moving, then neutrons which scatter from it may gain or lose energy.

Example: random diffusion



$$P(t) = e^{-t/\tau}$$



$$S(\omega) = \frac{1}{\pi}\frac{1}{1 + (\omega\tau)^2}$$

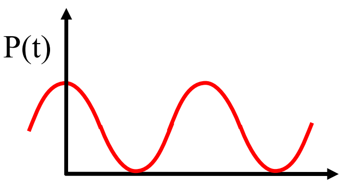
Quasielastic incoherent scattering (broad in energy but centered at ω=0) can contain useful information:

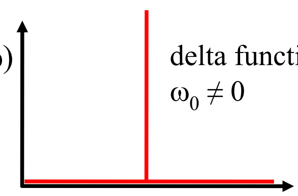
– Diffusion rates, molecular reorientations, relaxations

Inelastic Incoherent Scattering

Local excitations (no spatial correlations):
- Q-independent scattering
- Periodic correlations in time
- Examples: crystal field levels, molecular vibration

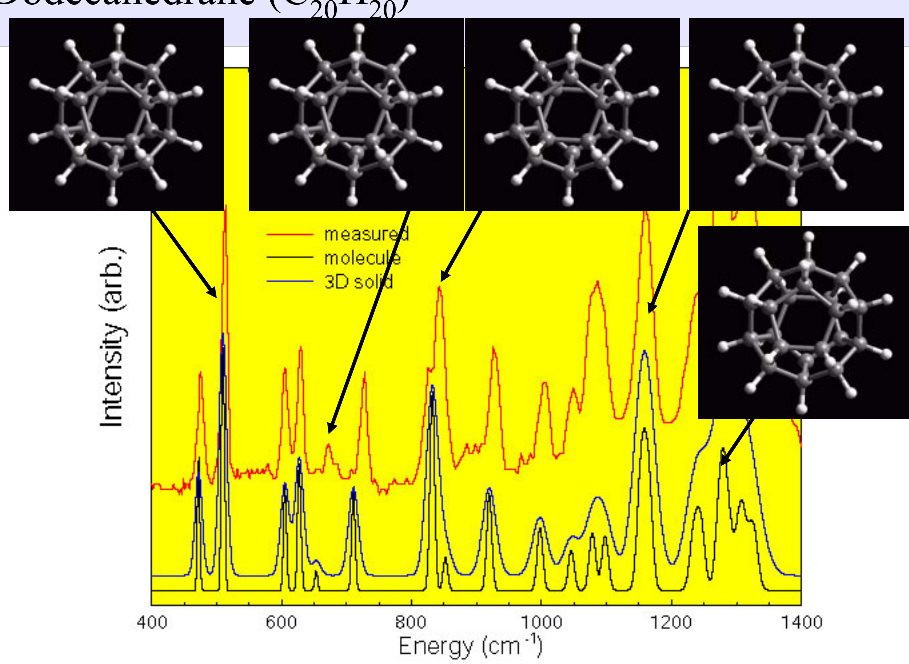
Inelastic incoherent scattering:
- scattered neutrons can gain or lose energy ω_0





Molecular Vibrations

Dodecahedrane ($C_{20}H_{20}$)



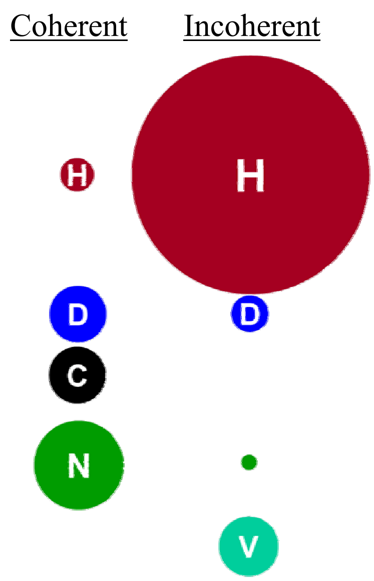
Coherent vs. Incoherent

Individual atomic scattering depend on isotope and nuclear spin state.

- Coherent cross section represents average scattering from that element.
- Incoherent cross section represents standard deviation in scattering.
- Deviations come from different isotopes of same element as well as nuclear spin state variations of single isotopes.

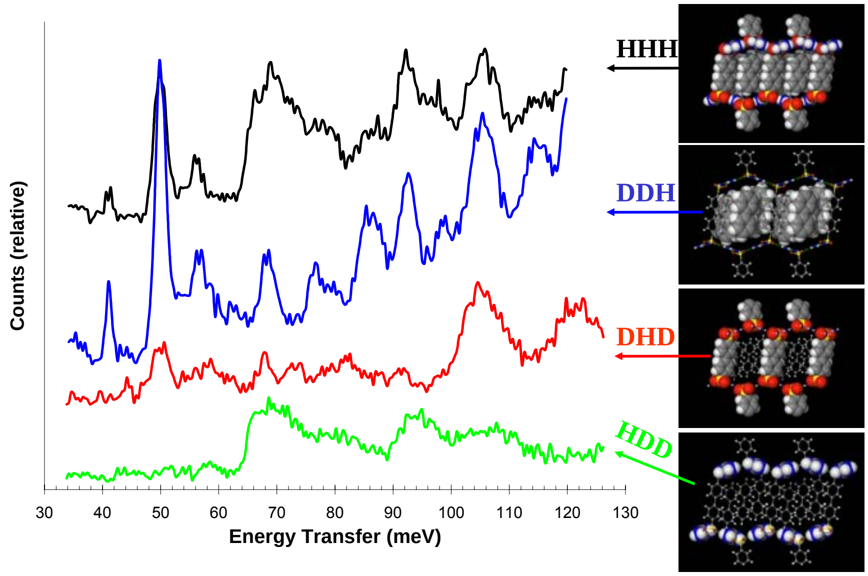
For most elements, scattering is primarily coherent.

- Hydrogen is a very significant exception.
- Isotope selection can change cross section significantly.



Isotope Specificity

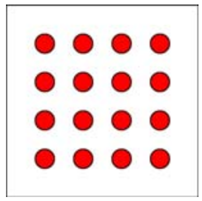
G_2[BPDS]*3(biphenyl)



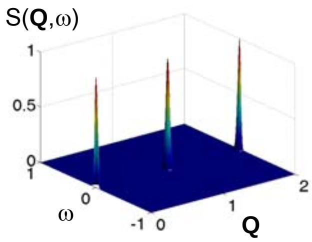
Scattering Review

Elastic
coherent

(**r**,t)

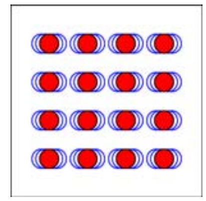


Crystal structures
Magnetic order

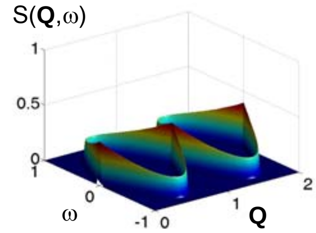


Inelastic
coherent

(**r**,t)



Phonons
Magnons

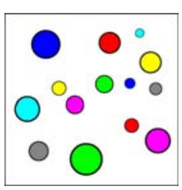


Scattering Review

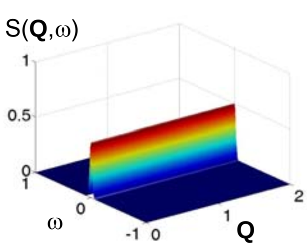
Elastic incoherent	Inelastic incoherent	Quasielastic incoherent

Elastic
incoherent

(**r**,t)

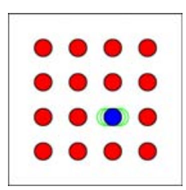


Isotope variation
Hydrogen

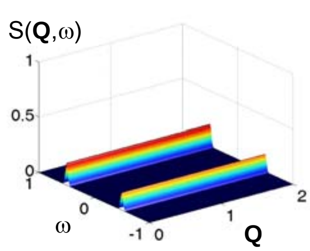


Inelastic
incoherent

(**r**,t)

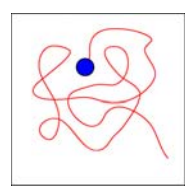


Local excitations
Molecular vibrations

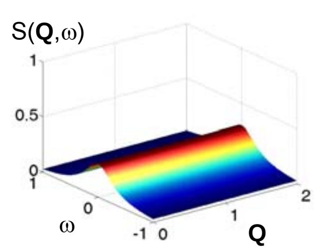


Quasielastic
incoherent

(**r**,t)



Diffusion
Relaxation



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