Magnetic Excitations I

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Plan:

- Basic tools
- Dynamic susceptibility
- Harmonic oscillators
- Calculating $S(\mathbf{Q}, \omega)$
  - Crystal Electric Field Levels
  - Spin Waves

Learn your Fourier transforms!

and

Learn and understand the convolution theorem!

$$f(r)\otimes g(r)=\int f(x)g(r-x)\cdot dx$$

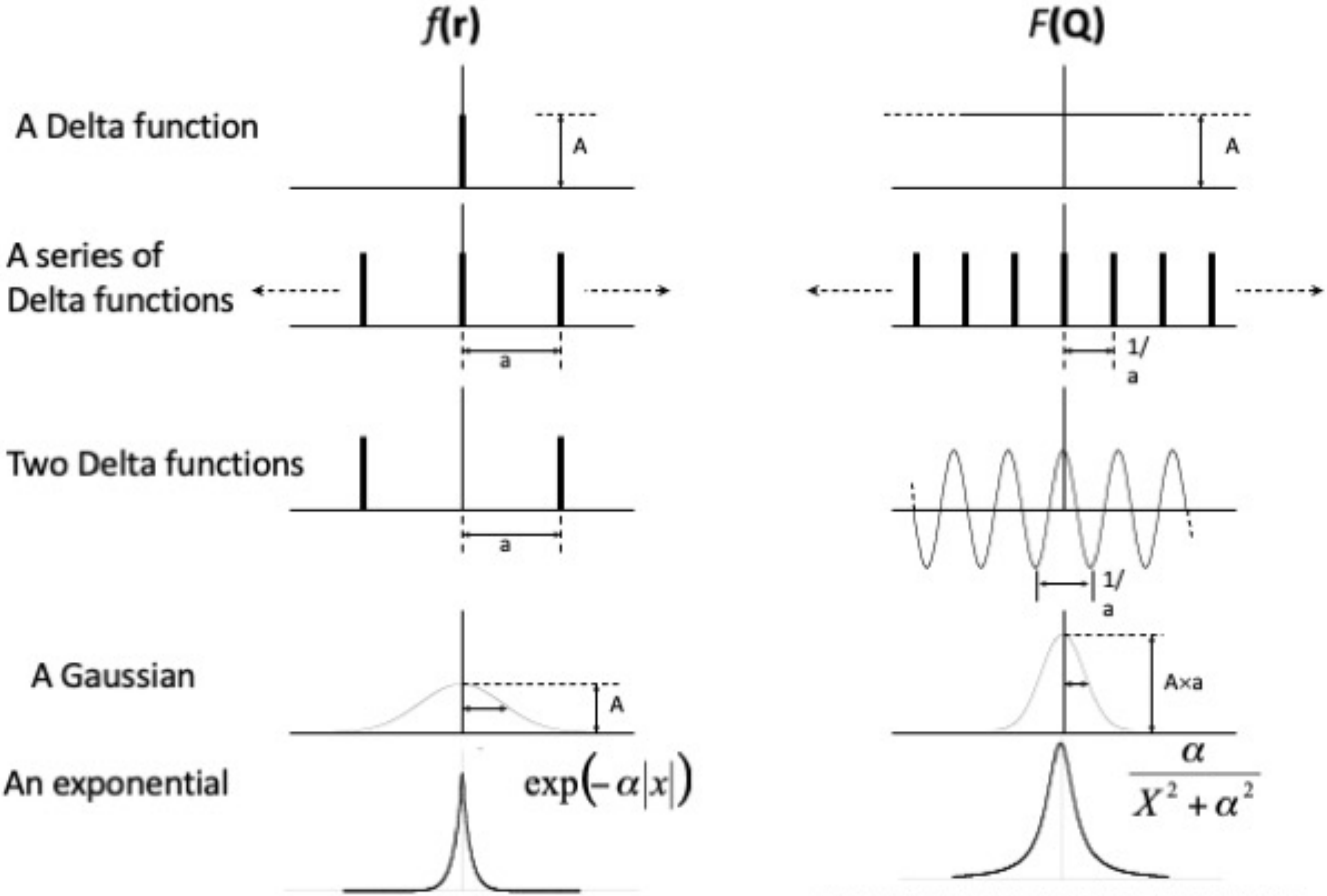
$$\Im\big(f(r)\big)=F(q)$$

$$\Im\big(g(r)\big)=G(q)$$

$$\Im\big(f(r)\otimes g(r)\big)=F(q)\times G(q)$$

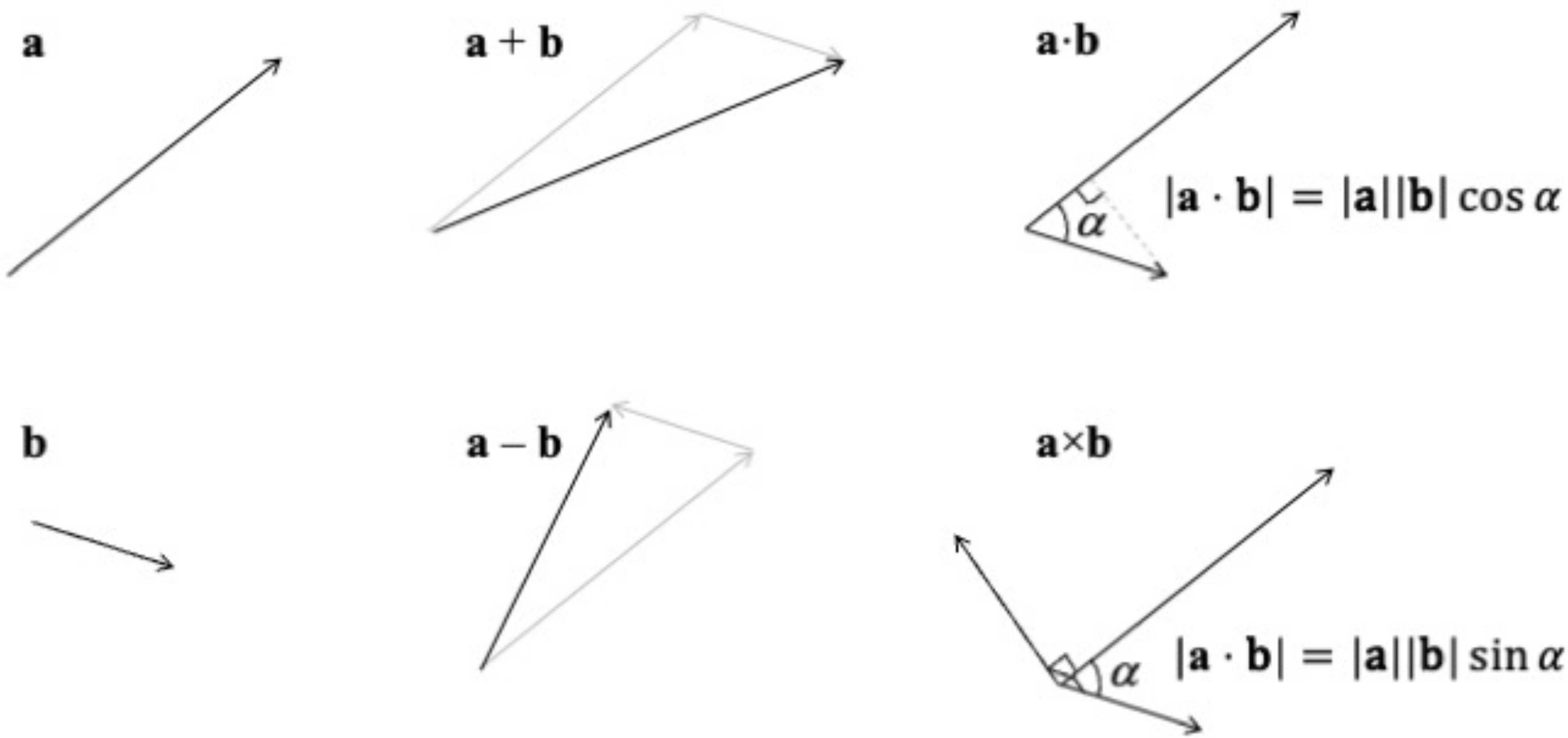
Fourier Transforms

$$F(\mathbf{Q}) = \int f(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot \mathrm{d}\mathbf{r}$$



Tools:

Learn to work with vectors



The fundamental rule of neutron magnetic scattering

Neutrons *only ever* see the components of the magnetization that are *perpendicular* to the scattering vector!

The fundamental rule of neutron magnetic scattering

Comes from the Fourier transform of $\hat{V}_m(\mathbf{r}) = -\gamma\mu_N\hat{\boldsymbol{\sigma}}\cdot\mathbf{B}(\mathbf{r})$

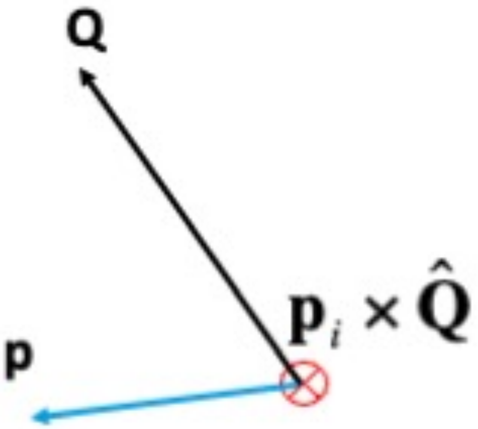
Magnetism is caused by unpaired electrons or movement of charge.
$\mathbf{B}(\mathbf{r})$ can be separated into two momentum contributions:

Linear / Orbital

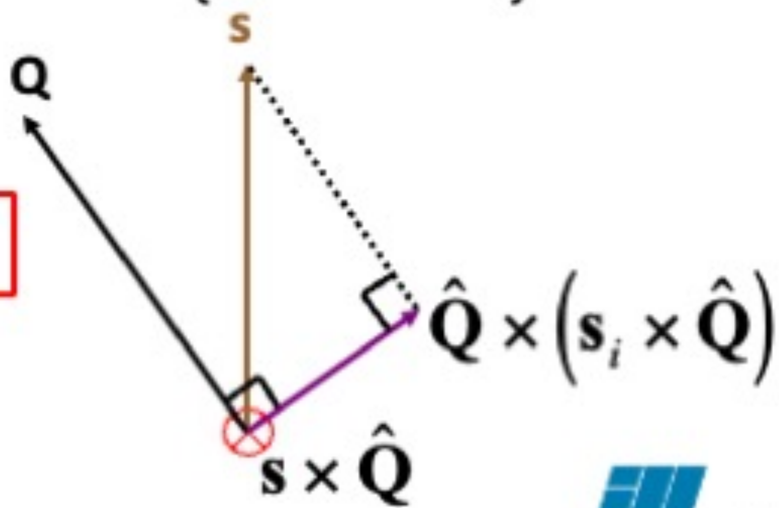
$$\frac{4\pi i}{\hbar Q}\exp\left(i\mathbf{Q}\cdot\mathbf{r}_i\right)\left(\mathbf{p}_i\times\hat{\mathbf{Q}}\right)$$

Spin:

$$4\pi\exp\left(i\mathbf{Q}\cdot\mathbf{r}_i\right)\left\{\hat{\mathbf{Q}}\times\left(\mathbf{s}_i\times\hat{\mathbf{Q}}\right)\right\}$$

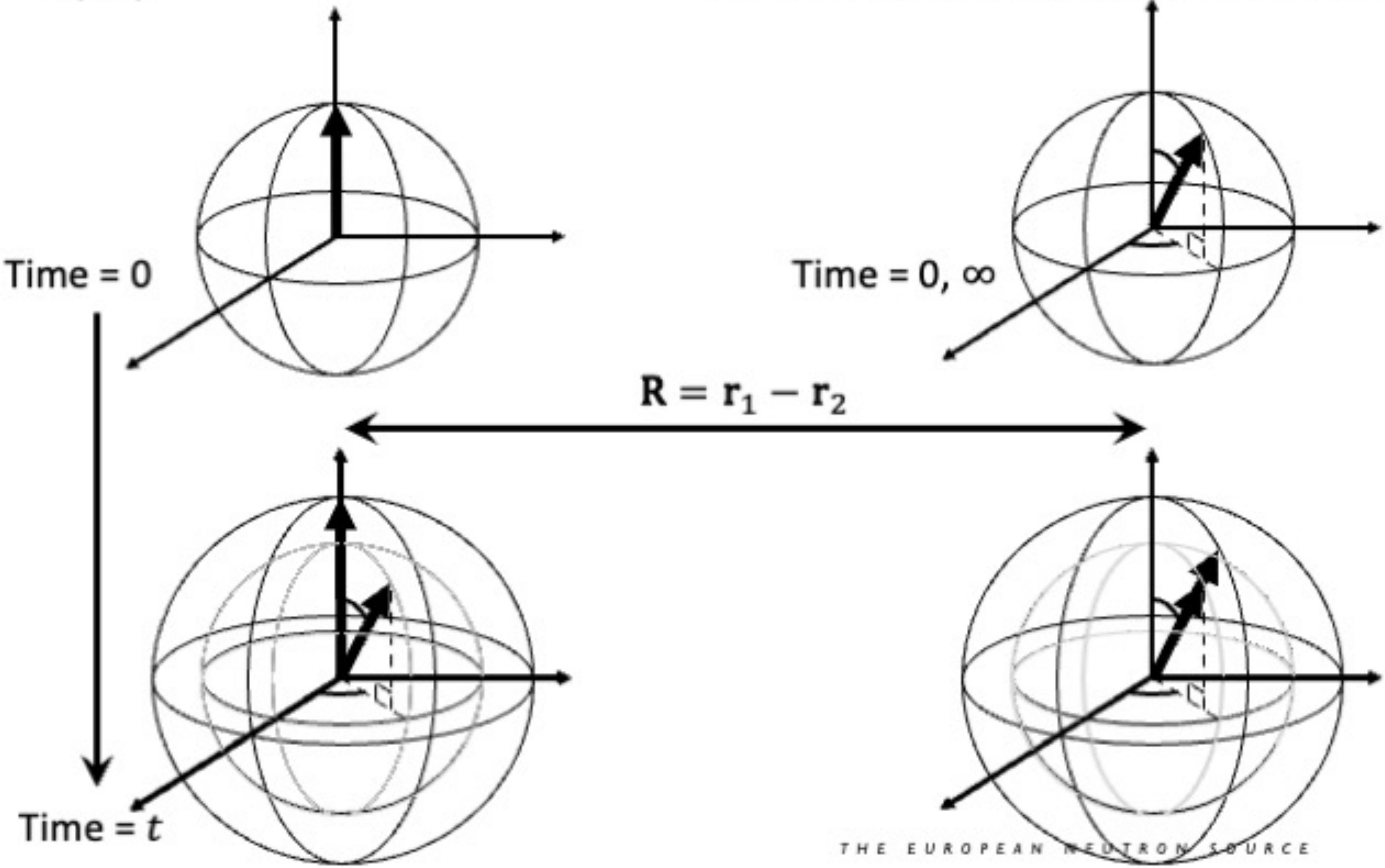


Perpendicular to **Q**



The cross-section and the dynamic structure factor

$G(\mathbf{r}, t)$ Time-dependent pair-correlation function



The cross-section and the dynamic structure factor

$G(\mathbf{r}, t)$ — Time-dependent pair-correlation function

$$I(\mathbf{Q}, t) = \int G(\mathbf{r}, t)\; e^{i\mathbf{Q}\cdot\mathbf{r}} \mathrm{d}\mathbf{r}$$

Intermediate scattering function

$$S(\mathbf{Q}, \omega) = \frac{1}{2\pi\hbar} \int I(\mathbf{Q}, t) e^{i\omega t} \mathrm{d}t$$

Response function (or dynamic structure factor)

$$= \frac{1}{2\pi\hbar} \int G(\mathbf{r}, t) e^{i(\mathbf{Q}\cdot\mathbf{r} - \omega t)} \mathrm{d}\mathbf{r}\mathrm{d}t$$

Condensed matter theorists love $S(\mathbf{Q}, \omega)$:

- The Fourier Transforms mean that:

  sums over enormous numbers of objects in real space (e.g. moles)

  become sums over a few objects in reciprocal space.

- It is expressed in variables appropriate for wave motion

  (i.e. fluctuations)

- $S(\mathbf{Q}, \omega)$ can be calculated directly from a Hamiltonian.

The cross-section and the dynamic structure factor

(Boothroyd, section 3.4)

$$\frac{d^2\sigma}{d\Omega dE_f} \propto \frac{k_f}{k_i} S(\mathbf{Q}, \omega)$$

For magnetic scattering,

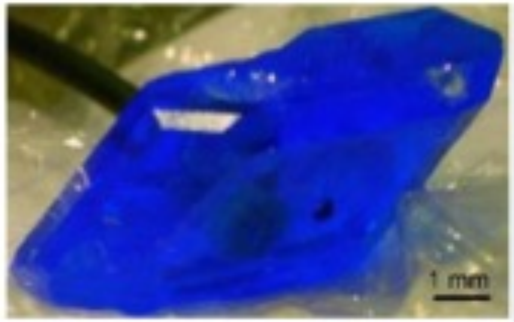
$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{\gamma r_0}{2\mu_B}\right)^2 \sum_{\alpha,\beta} (1 - \hat{Q}_\alpha \hat{Q}_\beta) S_{\alpha\beta}(\mathbf{Q}, \omega)$$

α and β are directions

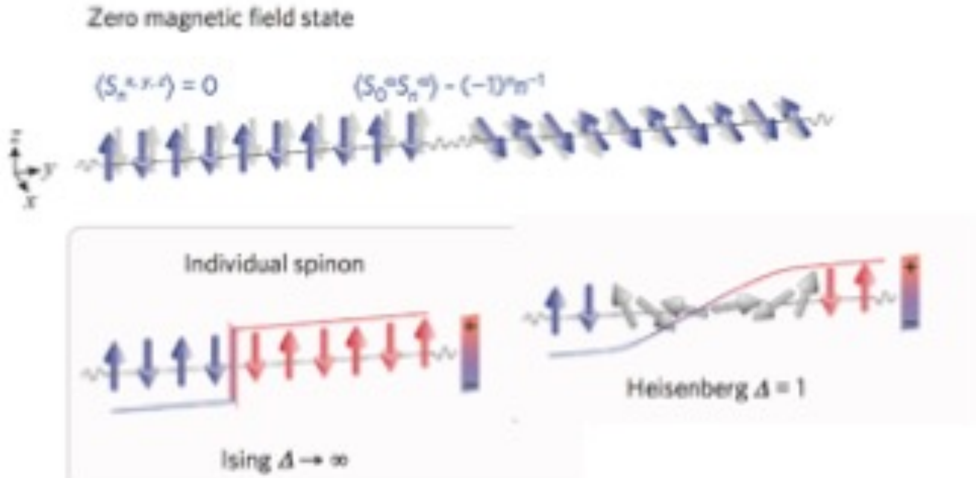
The neutron cross-section is *directly proportional* to the dynamic structure factor.

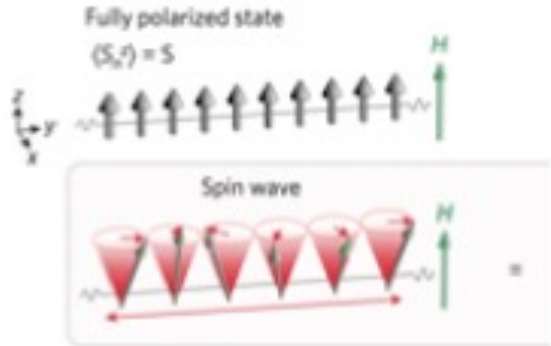
$S(\mathbf{Q}, \omega)$ can be calculated directly from a Hamiltonian.
Therefore, neutron scattering probes the Hamiltonian *directly* and *quantitatively*

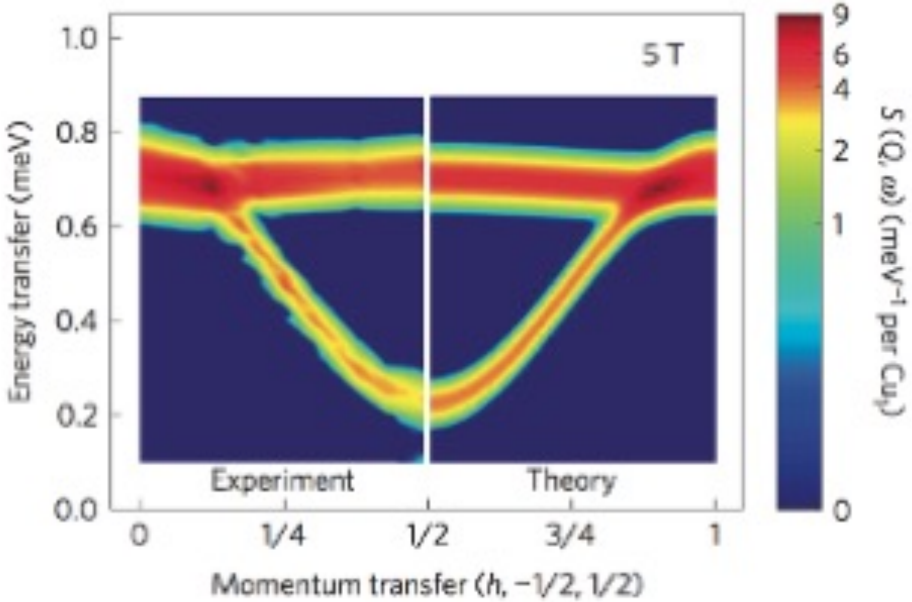
Magnetic excitations in $CuSO_4$

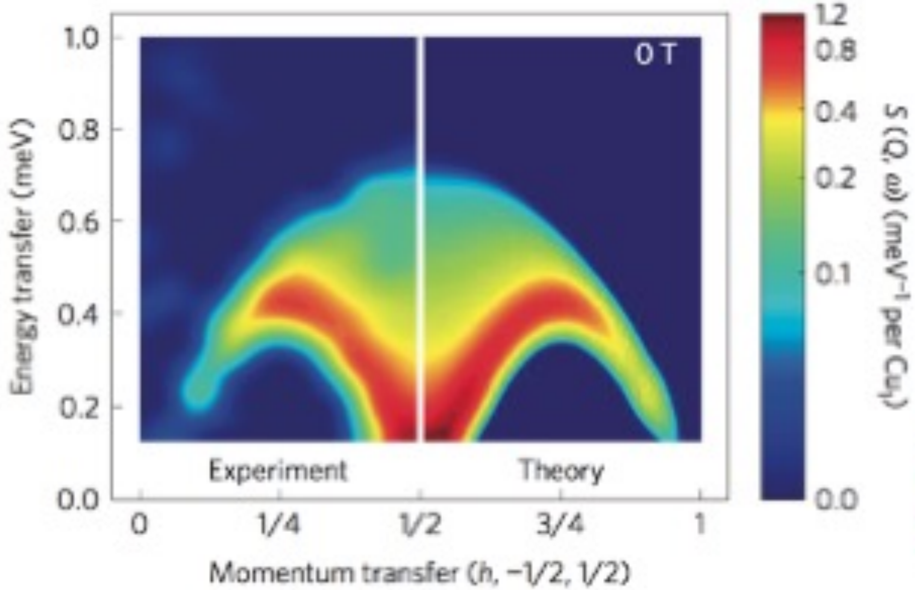


M. Mourigal *et al.*, Nature Phys. **9** (2013) 435









Neutron scattering and magnetic susceptibility

Magnetic susceptibility is a fundamental property of a material. It is defined as:

$$\chi = \frac{M}{H}$$

In a magnetic system, **M** is a vector which varies as a function of space, **r**, and (due to fluctuations) as a function of time, *t*.
The time is related to the susceptibility by:

$$M_\alpha(t) \propto \chi_{\alpha\alpha}(\omega) H_{0\alpha} e^{-i\omega t} + \chi^*_{\alpha\alpha}(\omega) H^*_{0\alpha} e^{i\omega t}$$

The susceptibility is a complex tensor:

$$\chi_{\alpha\alpha}(\omega) = \chi'_{\alpha\alpha}(\omega) + i\chi''_{\alpha\alpha}(\omega)$$

The rate of energy gain is given by:

$$\frac{d\bar{E}}{dt} = -M_\alpha \frac{dH}{dt} \propto \chi''_{\alpha\alpha}(\omega)$$

The dynamic structure factor and generalized susceptibility

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{\gamma r_0}{2\mu_B}\right)^2 \sum_{\alpha,\beta}(1 - \hat{Q}_\alpha \hat{Q}_\beta) S_{\alpha\beta}(\mathbf{Q}, \omega)$$

Through the Fluctuation-Dissipation Theorem

(Boothroyd, Appendix D)

$$S_{\alpha\beta}(\mathbf{Q}, \omega) = \frac{1 + n(\omega)}{\pi} \chi''_{\alpha\beta}(\mathbf{Q}, \omega)$$

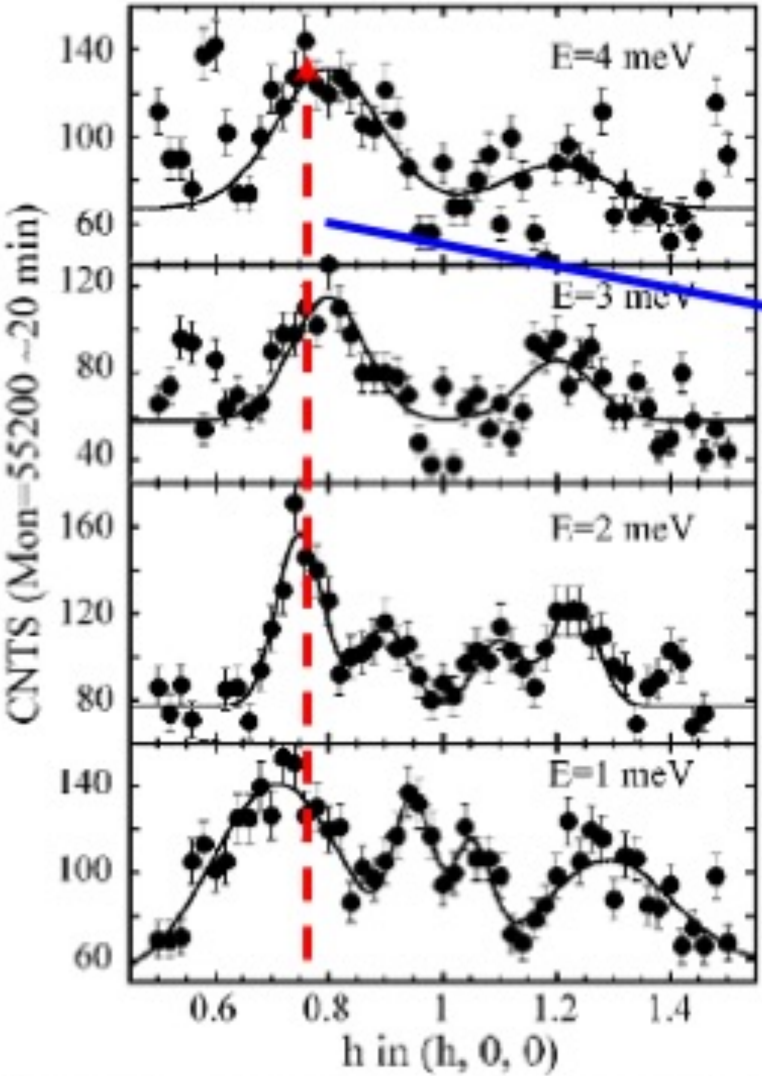
$$S(\mathbf{Q}, \omega) = \sum_{\alpha,\beta}(1 - \hat{Q}_\alpha \hat{Q}_\beta) \frac{1 + n(\omega)}{\pi} \chi''_{\alpha\beta}(\mathbf{Q}, \omega)$$

The inelastic cross-section is related to a *generalized* susceptibility

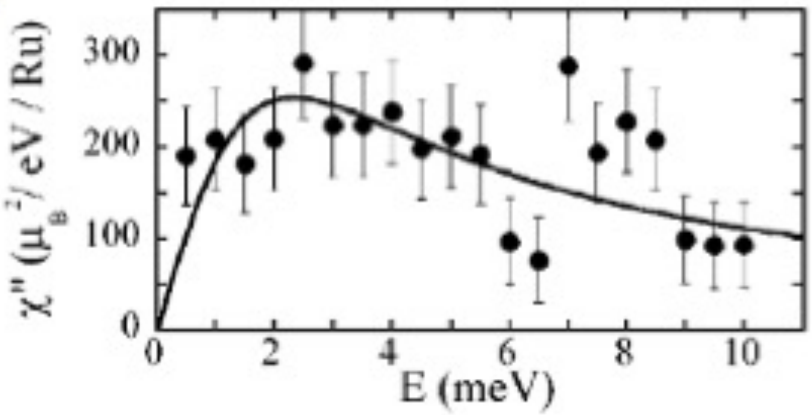
Spin excitations in $Sr_3Ru_2O_7$

L. Capogna *et al.*, Phys. Rev. B. **67** (2003) 012504

$Sr_3Ru_2O_7$ is from a family of materials that are low-dimensional magnetic, and superconductors



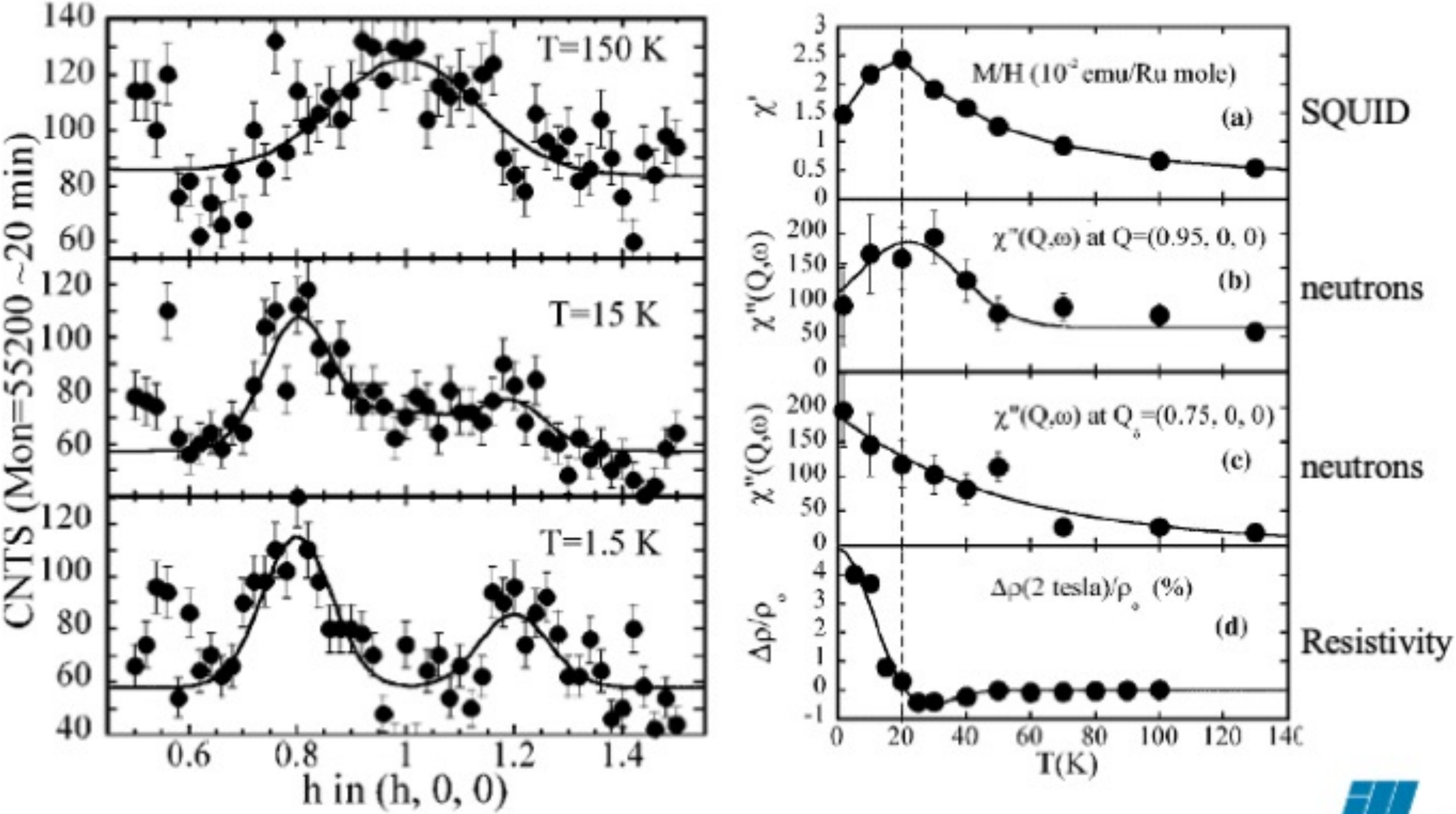
Inelastic neutron scattering at 1.5K



Susceptibility

Temperature dependence of the spin excitations in $Sr_3Ru_2O_7$

L. Capogna *et al.*, Phys. Rev. B. **67** (2003) 012504

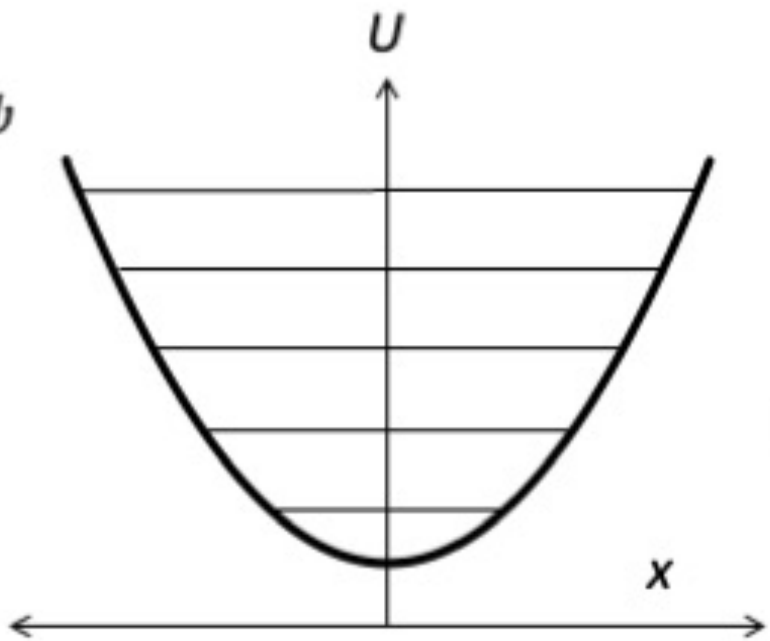


How do you calculate $S(\mathbf{Q}, \omega)$

Quantum harmonic oscillators

$$\mathcal{H}\psi = E\psi$$

$$-\frac{\hbar^2}{2m}\frac{d^2\psi}{dx^2} + U(x)\psi = E\psi$$



Raising ($a^\dagger$)
and
lowering (a) operators
move ψ up and down
one quantum state

$$U = \frac{1}{2}m\omega^2 x^2$$

$$E = \hbar\omega\left(n + \frac{1}{2}\right)$$

How do you calculate $S(\mathbf{Q}, \omega)$

Crystal Electric Fields: $$\mathcal{H} = \sum_{k=0}^{2l} \sum_{q=-k}^{k} B_q^k C_q^{(k)}$$

B_q^k = crystal field parameters

$C_q^{(k)}$ = Wybourne tensor operators

Heisenberg Hamiltonian: $$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

J_{ij} = magnetic exchange parameters

Crystal field levels

Crystal Electric Fields: $$\mathcal{H} = \sum_{k=0}^{2l} \sum_{q=-k}^{k} B_q^k C_q^{(k)}$$

B_q^k = crystal field parameters

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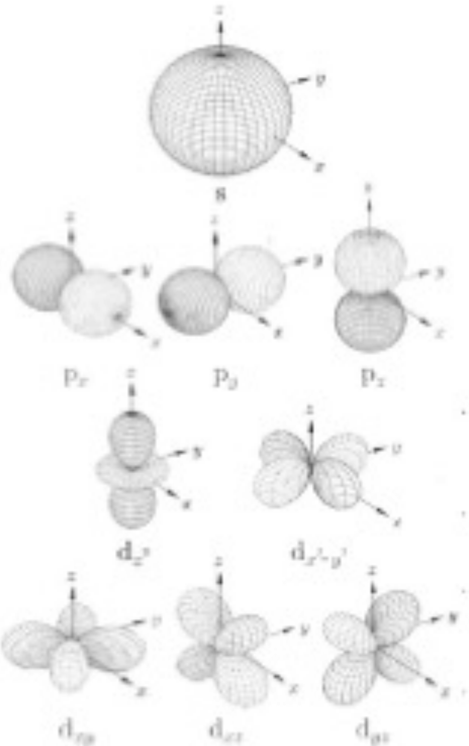
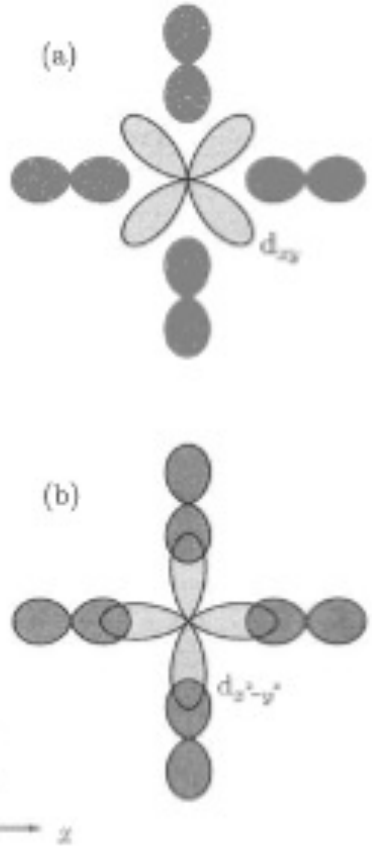
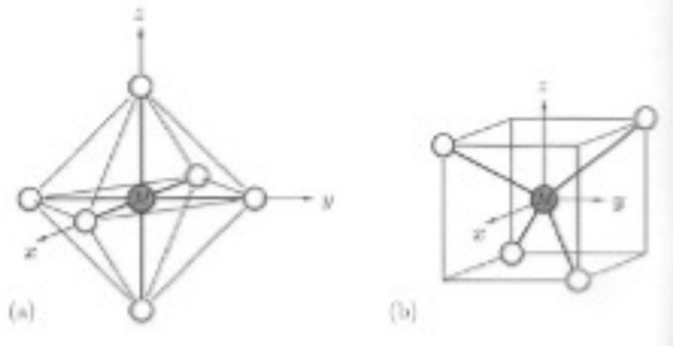
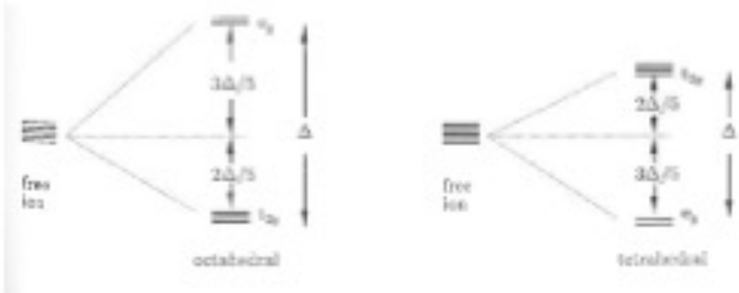


e_g

t_{2g}

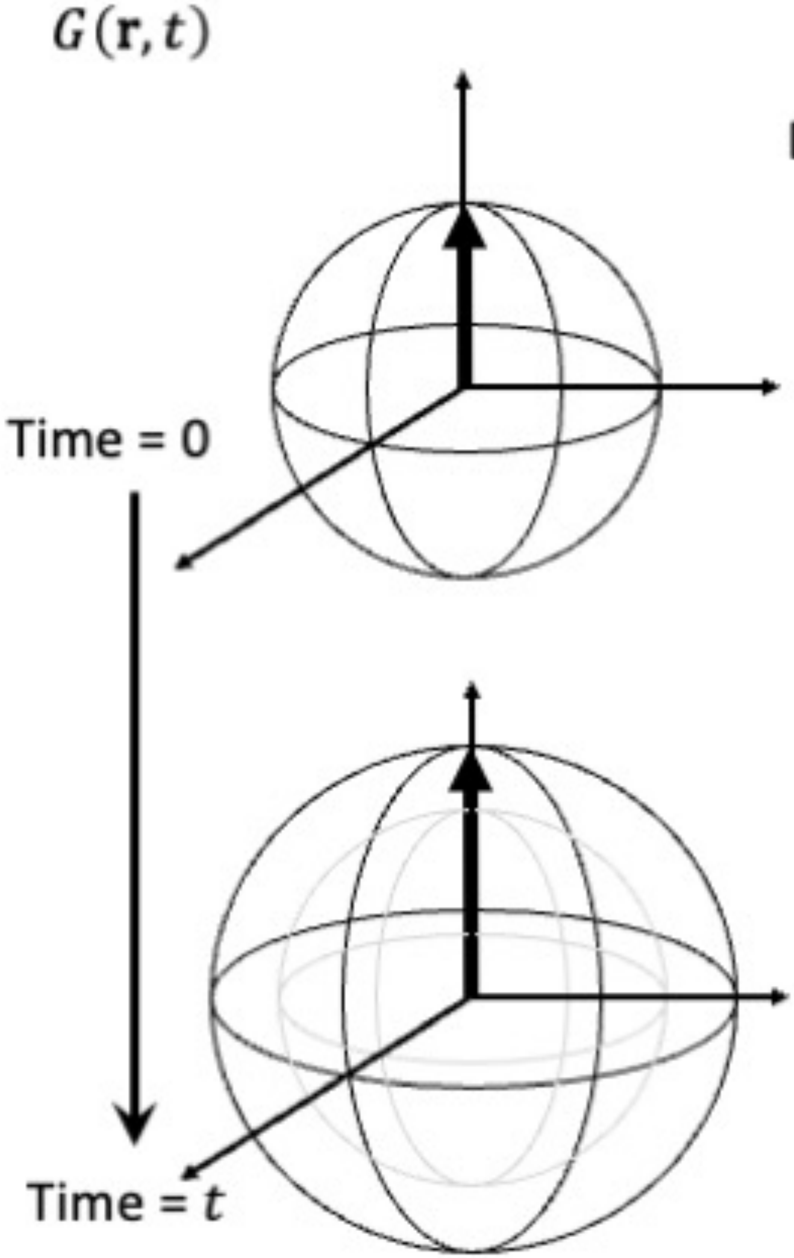






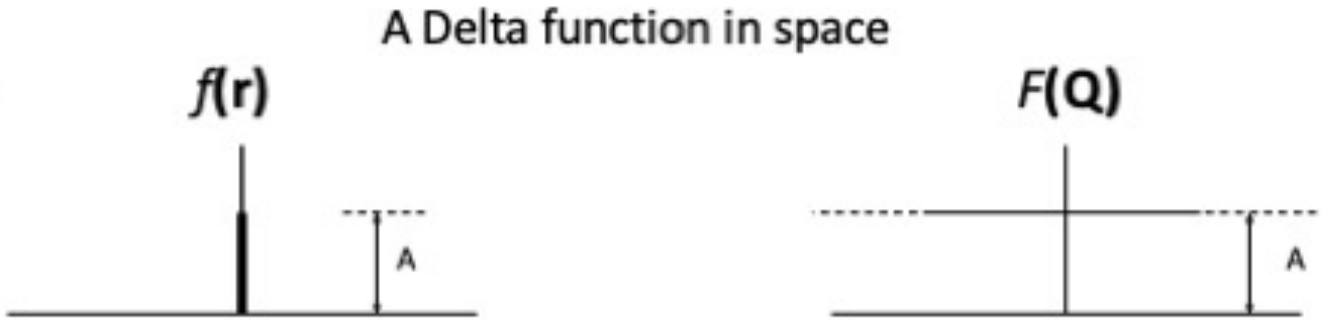
S. Blundell, *Magnetism in Condensed Matter* (2006) OUP (Oxford)

The cross-section and the dynamic structure factor



Moment size changes on individual atoms

Depends on both spin and orbital angular momentum



Q -independent

Crystal field levels

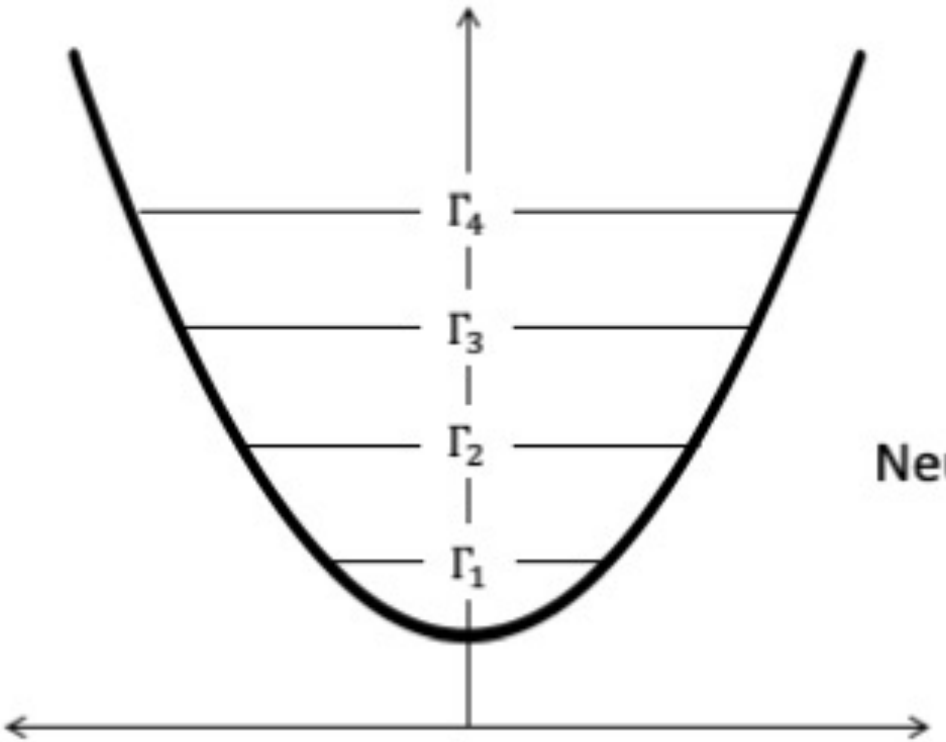
O. Moze., *Handbook of magnetic materials* vol. 11, 1998 Elsevier, Amsterdam, p.493

Crystal Electric Fields: $$\mathcal{H} = \sum_{k=0}^{2l} \sum_{q=-k}^{k} B_q^k C_q^{(k)}$$

B_q^k = crystal field parameters

$C_q^{(k)}$ = Wybourne tensor operators

The crystal field modifies the potential, and therefore the energy levels



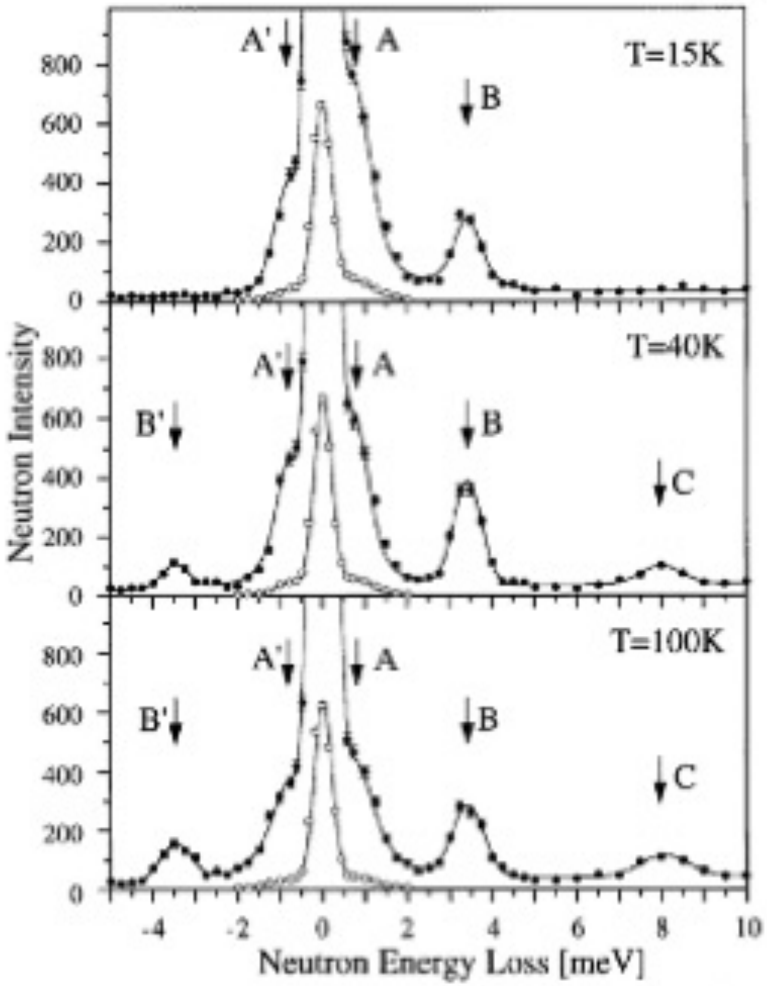
$$S(\hat{\mathbf{Q}}, \omega) \propto \sum_i p_i \sum_j \left|\langle \Gamma_i | \mathbf{M}_\perp | \Gamma_j \rangle\right|^2 \delta(E)$$

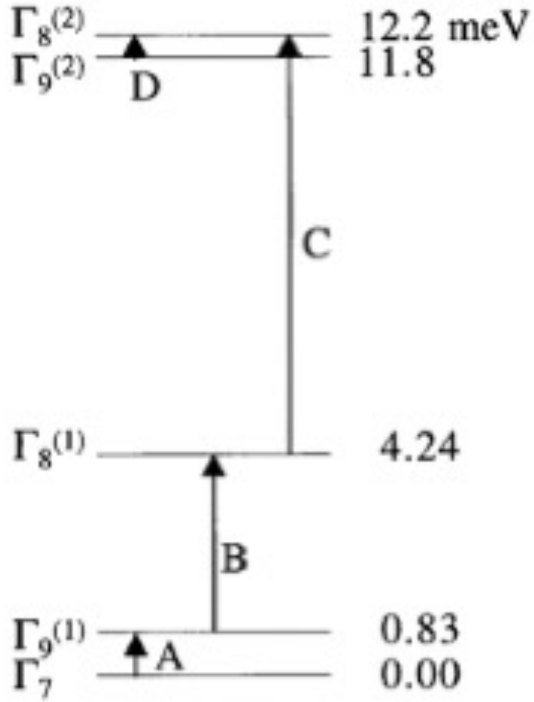
Neutrons see the *perpendicular* components of the magnetization to $\mathbf{Q}$

Crystal fields in $NdPd_2Al_3$

$$\mathcal{H} = \sum_{m,n} B_n^m O_n^m$$

O = Stevens parameters
(K. W. Stevens, Proc. Phys. Soc A**65** (1952) 209)
B = CF parameters, measured by neutrons





A. Dönni *et al.*, J. Phys.: Condens. Matter **9** (1997) 5921

O. Moze., *Handbook of magnetic materials* vol. 11, 1998 Elsevier, Amsterdam, p.493

Spin waves

D. C. Mattis, *The Theory of Magnetism I*, Springer-Verlag, Berlin, 1988

C. Kittel, *Quantum Theory of Solids*, Wiley, 1991

F. Keffer, *Handbuch der Physik* vol 18II, 1966 Springer-Verlag, Berlin

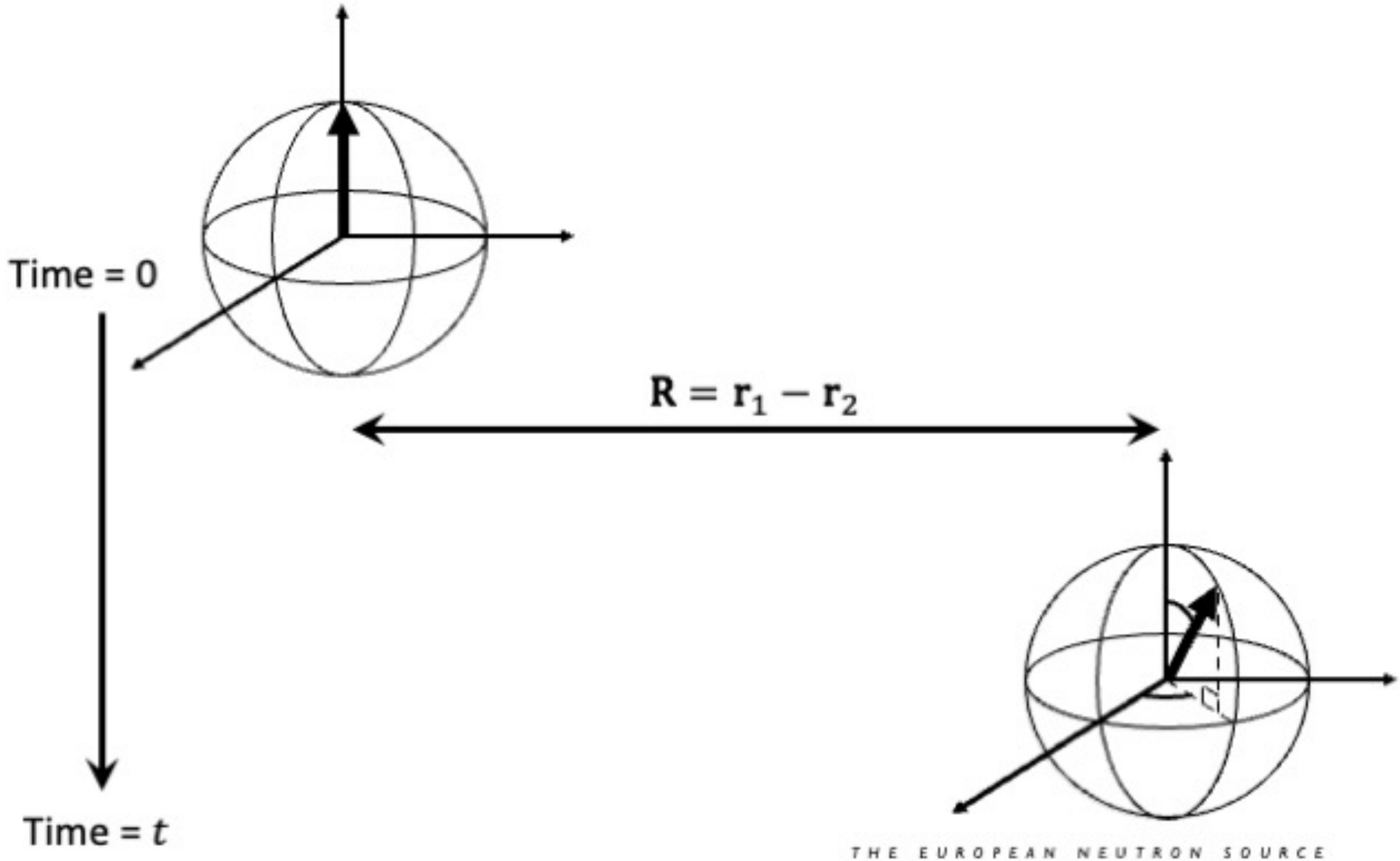
P. A. Lindgård *et al*., J. Phys. Chem. Solids **28** (1967) 1357

R. M. White *et al*., Phys. Rev. **139** (1965) A 450

C. Tsallis, J. Math. Phys. **19** (1978) 277

The cross-section and the dynamic structure factor

$G(\mathbf{r}, t)$ Time-dependent pair-correlation function

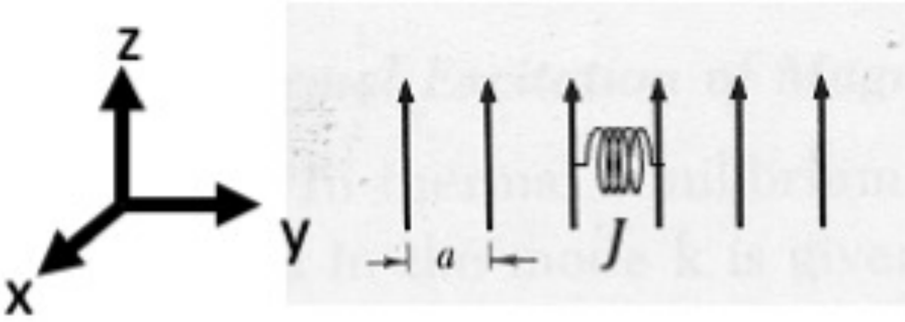


Spin waves

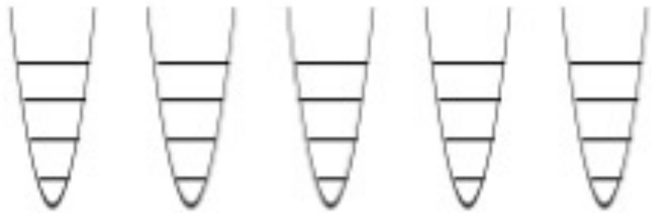
$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

J_{ij} = magnetic exchange parameters

Take a simple ferromagnet:



Change the energy by one quantum



$$\mathbf{S}_i \cdot \mathbf{S}_j = S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z$$

$S^+ = S^x + iS^y$ Raising operator

$S^- = S^x - iS^y$ Lowering operator

$$\mathbf{S}_i \cdot \mathbf{S}_j = \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right) + S_i^z S_j^z$$

The Holstein-Primakoff transformation

$$\mathbf{S}_i \cdot \mathbf{S}_j = \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right) + S_i^z S_j^z$$

$$\boxed{\begin{aligned} S^+ &= (2S)^{\frac{1}{2}} a \left(1 - \frac{a^\dagger a}{2S}\right)^{\frac{1}{2}} \approx (2S)^{\frac{1}{2}} a \\ S^- &= (2S)^{\frac{1}{2}} \left(1 - \frac{a^\dagger a}{2S}\right)^{\frac{1}{2}} a^\dagger \approx (2S)^{\frac{1}{2}} a^\dagger \\ S^z &= S - a^\dagger a \end{aligned}}$$

Linear spin-wave theory

Propagating spin waves

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

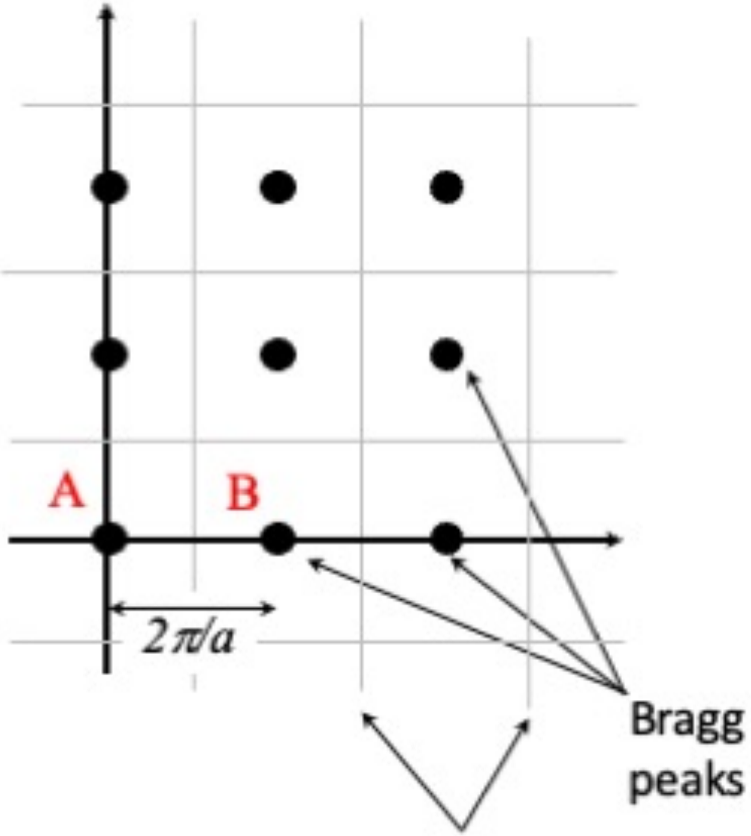
- Expand the Hamiltonian and group the terms
- Fourier-transform the raising and lowering operators, $a^\dagger$ and a
- Fourier transform $J(q) = \sum_{\mathbf{r}_i - \mathbf{r}_j} J_{ij} \exp\left(i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)\right)$
- Group the terms and respect the commutation relations

$$\mathcal{H} = \underbrace{\frac{1}{2} N S^2 J(0)}_{\text{Zero-point energy}} + \underbrace{S \sum_{\mathbf{q}} \left(J(\mathbf{q}) - J(0)\right) a^\dagger a}_{\boxed{\text{Propagating modes}}}$$

$$E_{\mathbf{q}} = \hbar\omega_{\mathbf{q}} = S \sum_{\mathbf{q}} \left(J(\mathbf{q}) - J(0)\right)$$

Magnons and reciprocal space

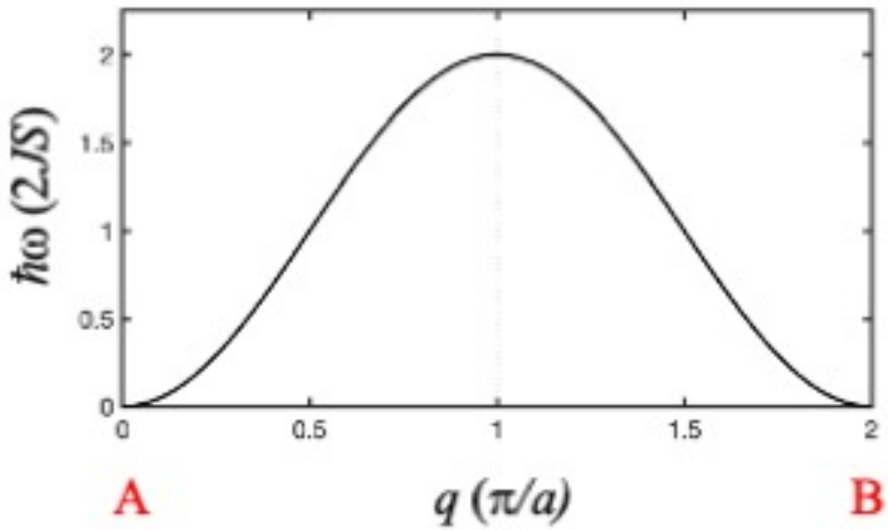
Square lattice, reciprocal space



nearest-neighbour exchange

$$S\sum_{\mathbf{q}}\big(J(\mathbf{q})-J(0)\big)=2SJ(2-\cos 2\pi h-\cos 2\pi k)$$

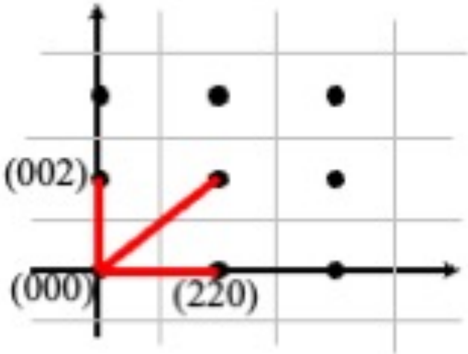
Spin wave dispersion

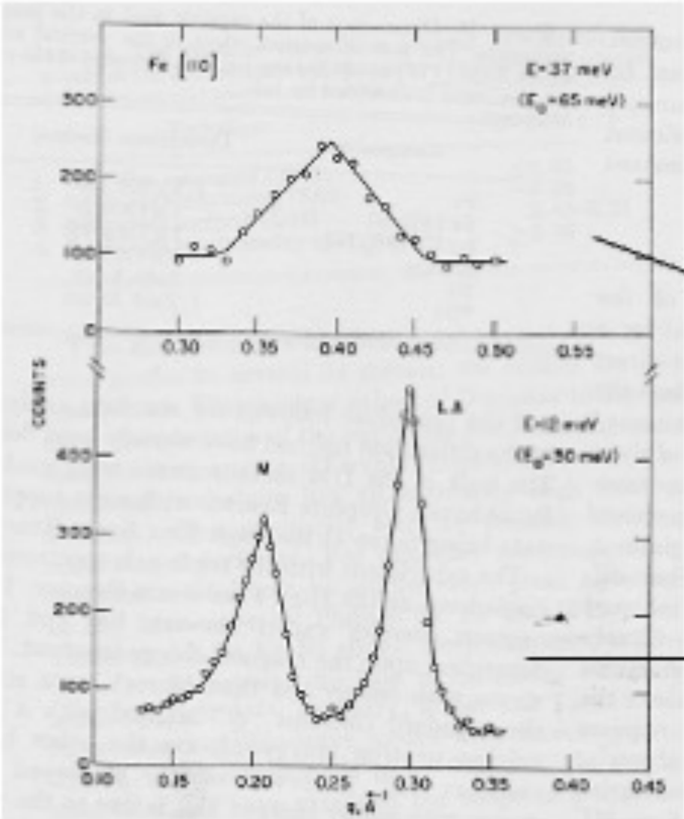


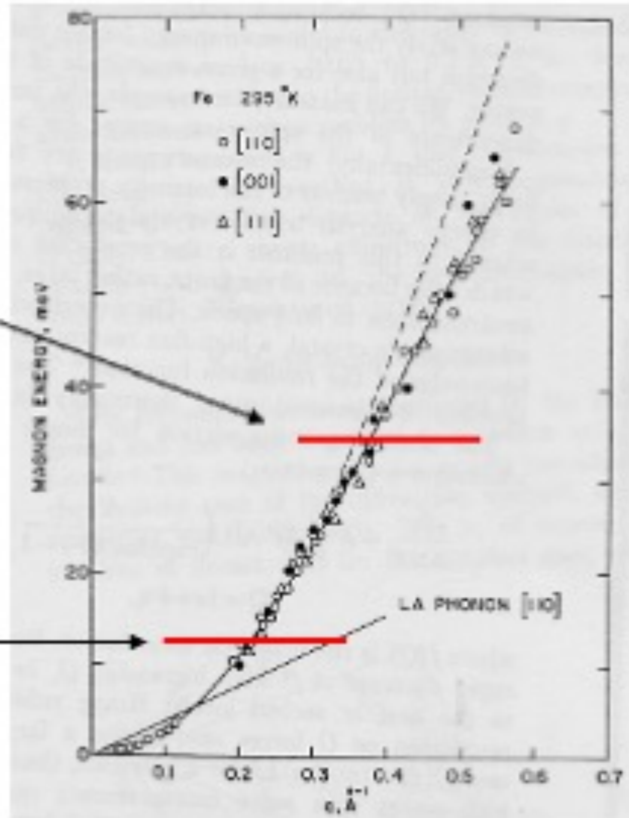
$\propto q^2$ for $qa \ll 1$

Magnons in crystalline iron

G. Shirane *et al.*, J. Appl. Phys. **39** (1968) 383





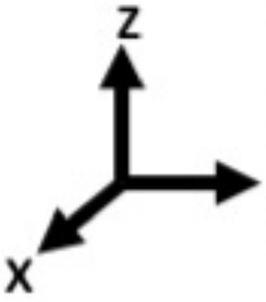


$\propto q^2$ for $qa \ll 1$

Spin waves and magnons

The classical picture of a spin wave

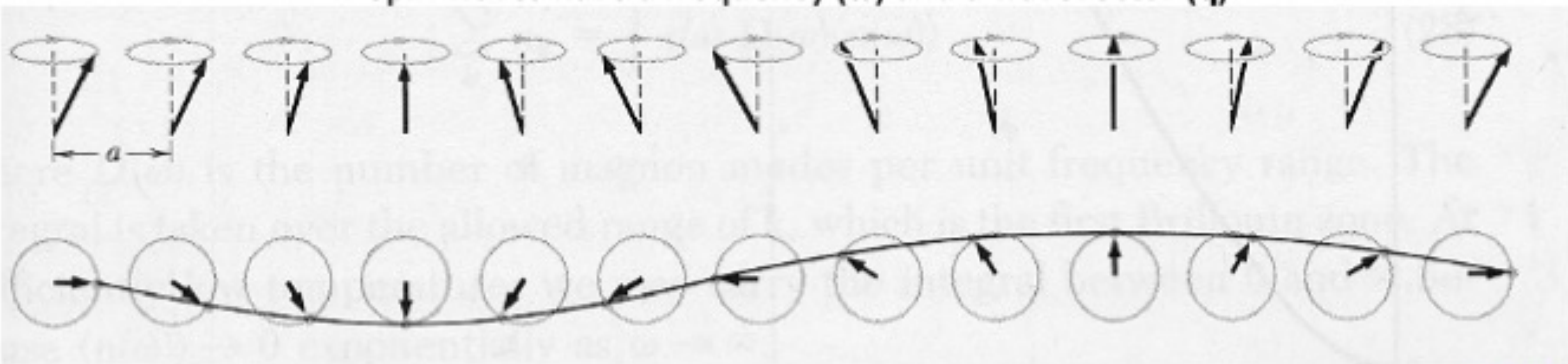
$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$



Take a simple ferromagnet:

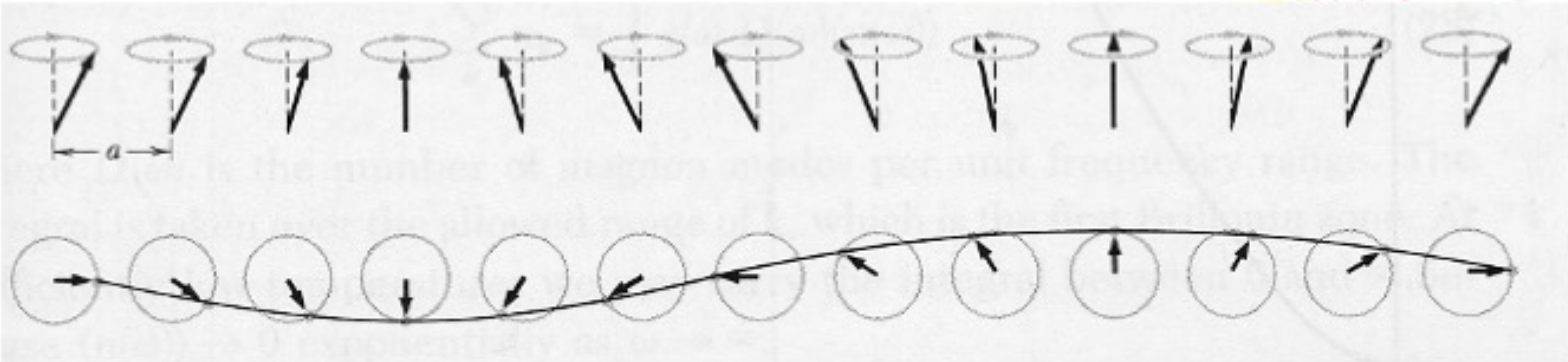
The spin waves might look like this:

Spin waves have a frequency (ω) and a wavevector (**q**)



The frequency and wavevector of the waves are *directly measurable* with neutrons

Magnons

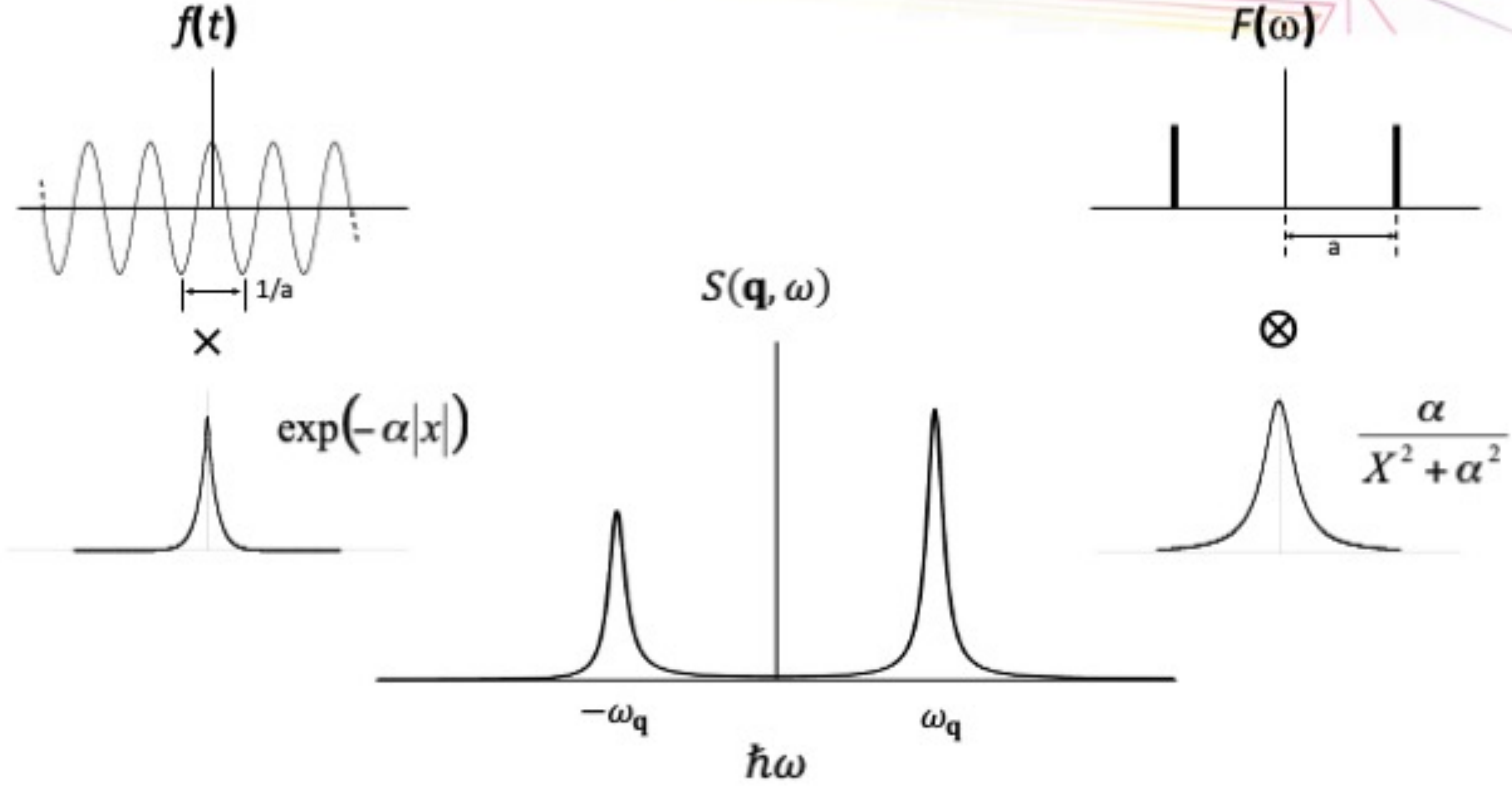


The Fourier Transform for a periodic function:

Each wavelength for the magnon has its own periodicity.

Each wavevector for the magnon has its own frequency (energy)

Modelling magnons

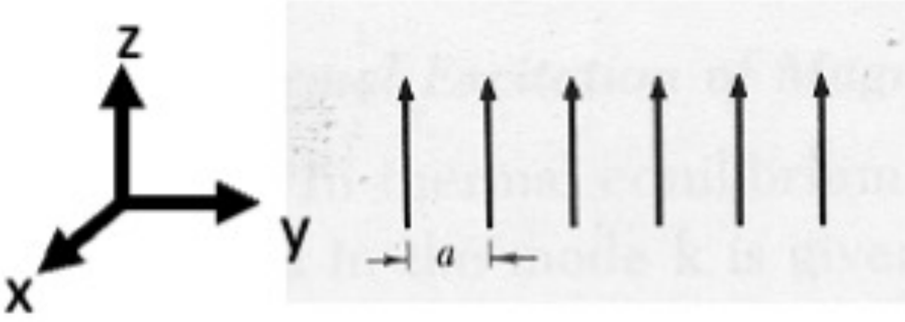


$$S(\mathbf{Q},\omega) = \sum_{\alpha,\beta} \left(1 - \hat{Q}_\alpha \hat{Q}_\beta\right) \frac{1+n(\omega)}{\pi} \chi''_{\alpha\beta}(\mathbf{Q},\omega)$$

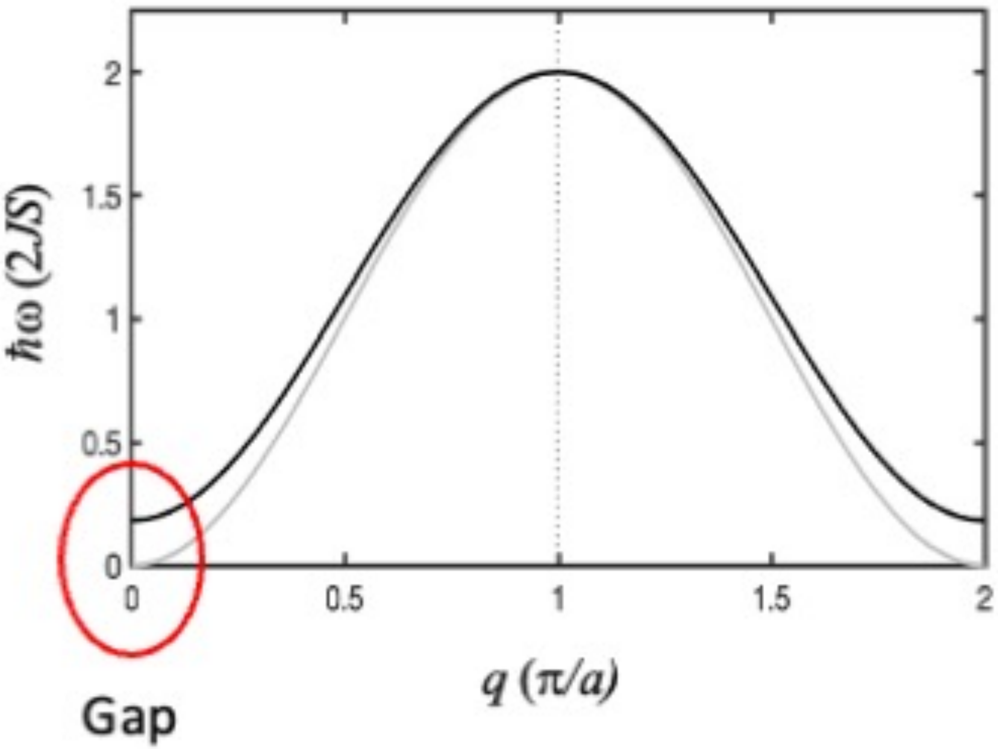
$$\chi''_{\alpha\beta}(\mathbf{Q},\omega) = \text{Lorentzian}(\omega_{\mathbf{q}}) - \text{Lorentzian}(-\omega_{\mathbf{q}})$$

Anisotropy

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \text{anisotropy}$$

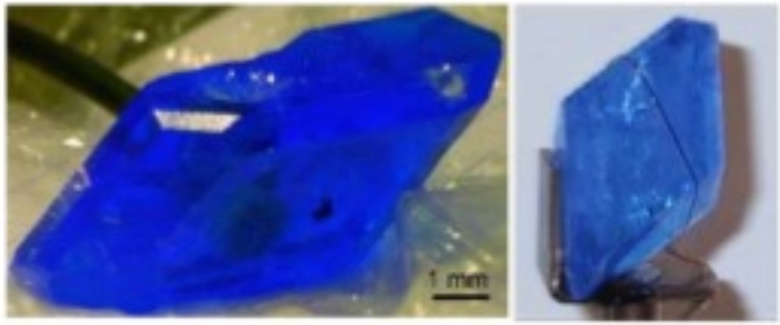


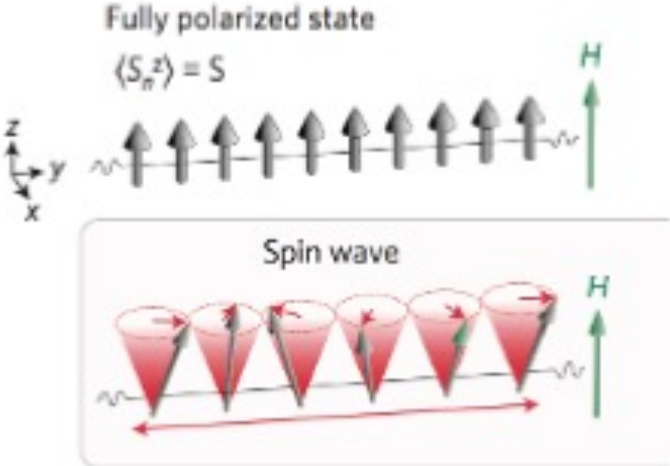
Spin wave dispersion

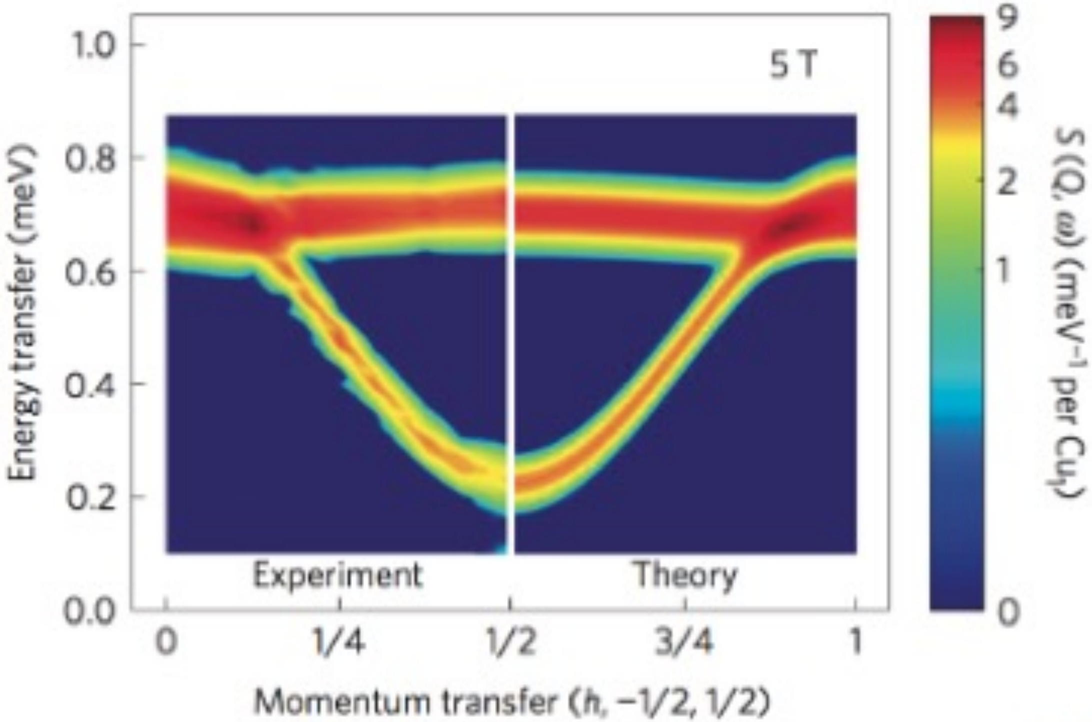


Magnetic excitations in $CuSO_4$

M. Mourigal *et al.*, Nature Phys. **9** (2013) 435



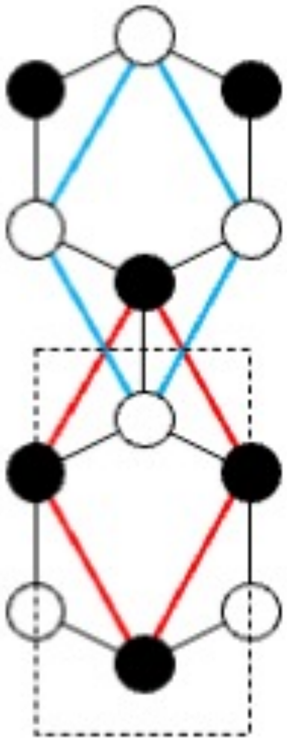




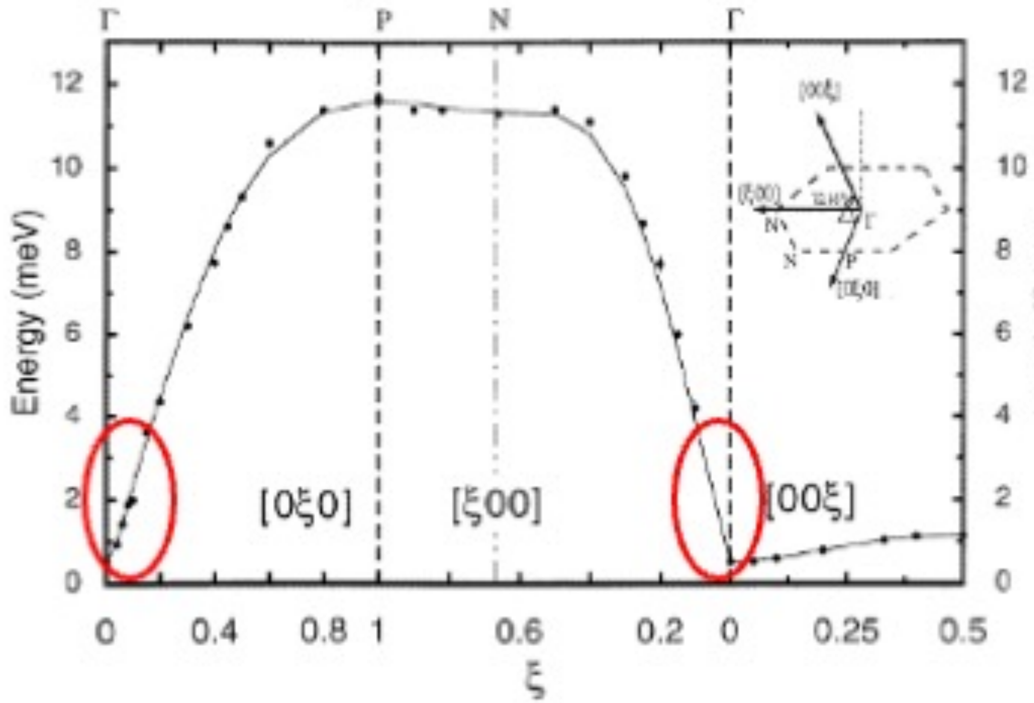
Antiferromagnets

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

The increase by a quantum on one sublattice
is equivalent to
the decrease by a quantum on the other sublattice



Magnons in $MnPS_3$



A. R. Wildes *et al.*, JPCM **10** (1998) 6417

$$\hbar\omega_q \propto q$$
for
$$qa \ll 1$$

Take home messages

- Neutron scattering gives a *quantitative measurement* of $S(\mathbf{Q},\omega)$ over *all* the Brillouin zone
- $S(\mathbf{Q},\omega)$ can be calculated from the Hamiltonian
- The inelastic cross-section is linked to the magnetic susceptibility