Neutron spectroscopy

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Institut Laue-Langevin

Plan:

- Properties of the neutron
- Neutron spectroscopy
- Harmonic oscillators
- Atomic vibrations
  - Quantized energy levels
  - Tunnelling
- Magnetic vibrations
  - Crystal fields
  - Molecular magnets
- Propagating modes
  - Phonons
  - Magnons

The neutron

Forms part of the nucleus of the atom

Rest mass *(m_n)*	1.675×10^{-27} kg (c.f. mass of hydrogen = 1.674×10^{-27} kg)
Diameter	$\sim 10^{-15}$ m (c.f. diameter of an atom $\sim 10^{-10}$ m)
Charge	0
Spin	$-1/2$
Magnetic moment	0.966×10^{-26} JT^{-1} $-1.913\ \mu_N = -\gamma\mu_N$ $1.042 \times 10^{-3}\ \mu_B$ (c.f. moment on an electron = 1 μ_B)

Neutron beams

Through quantum mechanics, neutrons have a *wavelength*.

Energy of a neutron

$$E = h\upsilon = \hbar\omega$$

$$= \frac{1}{2} m_n v^2$$

h = Planck's constant
υ = frequency
ω = angular frequency
v = speed

Momentum of a neutron

$$p = h/\lambda = \hbar k$$

$$\mathbf{p} = \hbar\mathbf{k}$$

λ = wavelength
k = wavenumber

'Standard' thermal neutrons:

v = 2200 m/s
λ = 1.798 Å
T = 293 K

E = 25.3 meV = 6 THz
$p = 3.68 \times 10^{-24}$ kg m s^{-1} = 3.5 Å^{-1}

Neutron scattering – length and time scales

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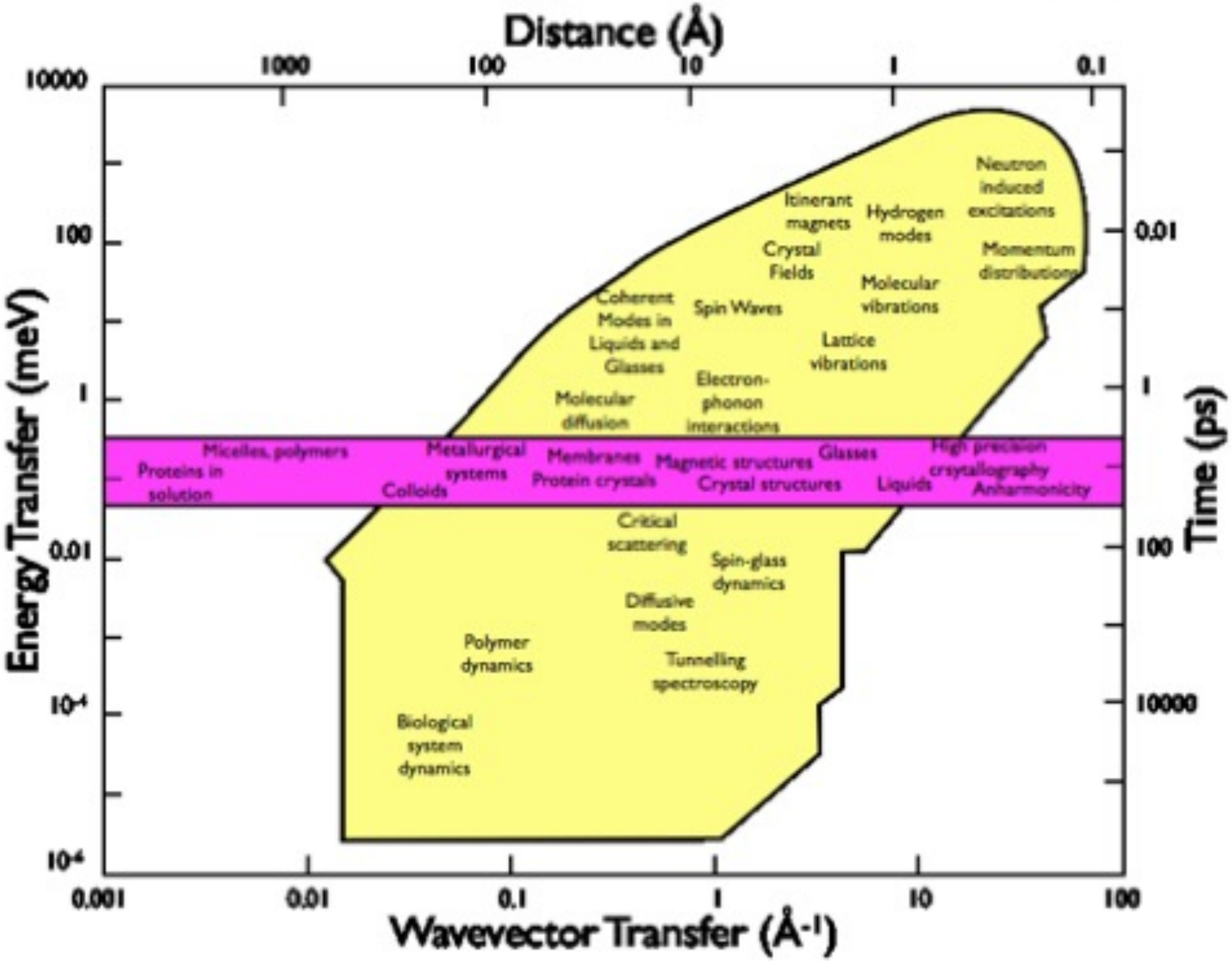
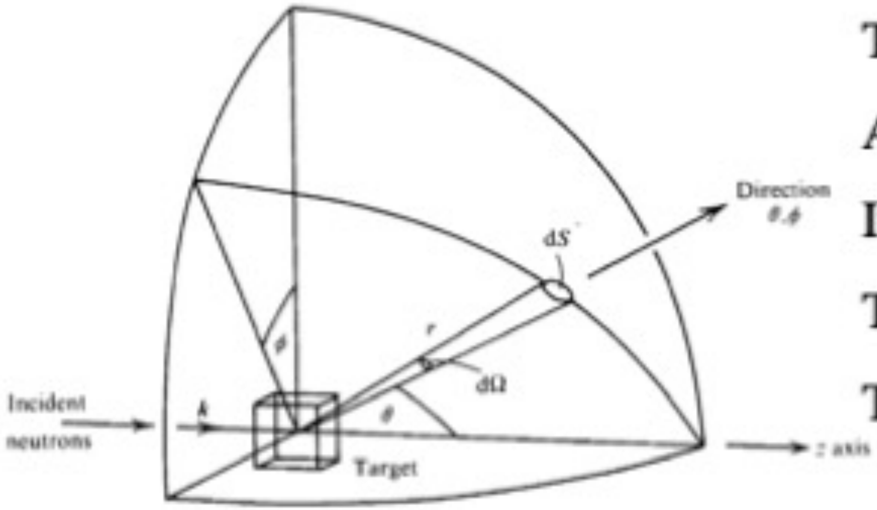


A neutron scattering experiment

Fig. 1.2 Geometry for scattering experiment.



The target volume is initially in state ζ.

A neutron enters with wave vector $\boldsymbol{k}$ and spin s

It interacts with the target.

The final neutron wave vector is $\boldsymbol{k}'$ and spin s'.

The final target state is ζ'.

We measure:

Momentum transfer: $\mathbf{Q} = \boldsymbol{k} - \boldsymbol{k}'$

$$Q^2 = k^2 + k'^2 - 2kk' \cos\Theta$$

$$\Delta s$$

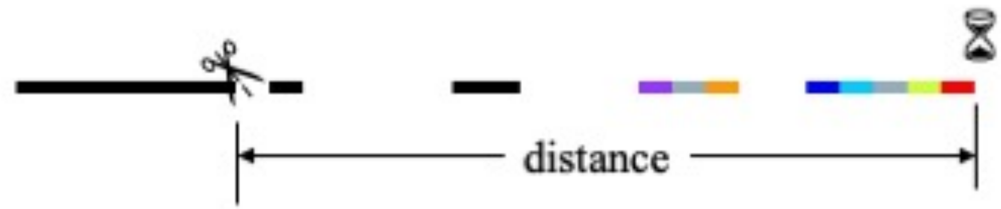
Energy transfer: $\Delta E = \hbar\omega = \frac{\hbar^2}{2m_n}\left(k^2 - k'^2\right)$

Knowing the neutron wavelength

You must know the wavelength to perform a scattering experiment

With neutrons, there are two ways of knowing the wavelength:

1. Use a monochromator
   – Bragg's law: $2d\sin\theta = \lambda$
2. Use time-of-flight
   – Neutron speed $\propto 1/\lambda$, 4Å ~ 1000 m/s
   – *chopper* to use *time-of-flight*

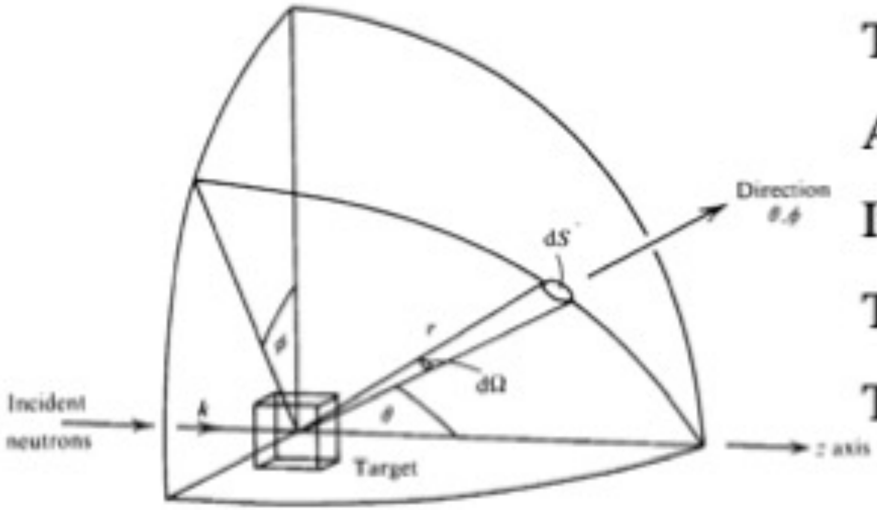


   – *velocity selector* to monochromate

The neutron spin precession can also give energy change information

A neutron scattering experiment

Fig. 1.2 Geometry for scattering experiment.



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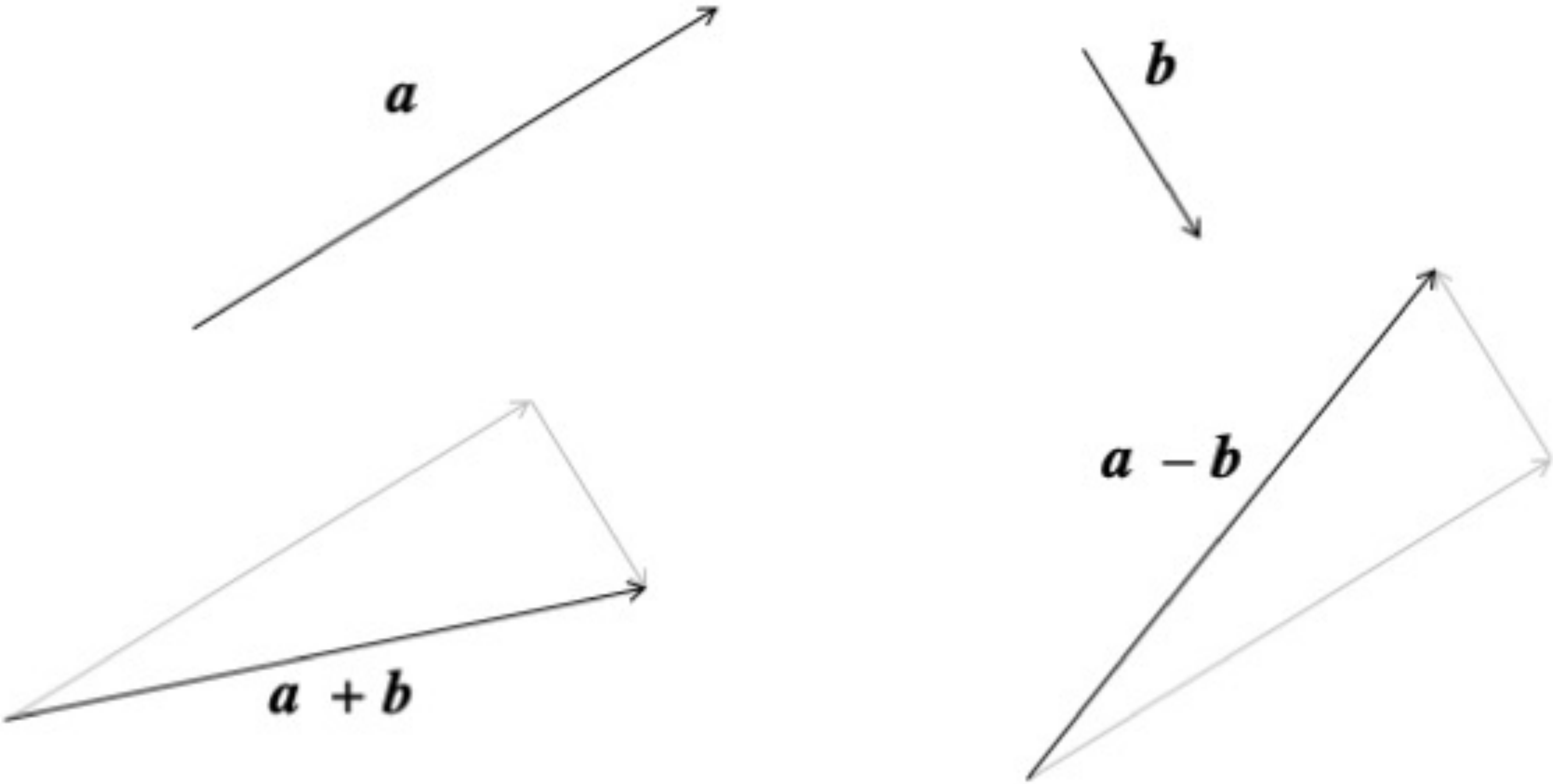
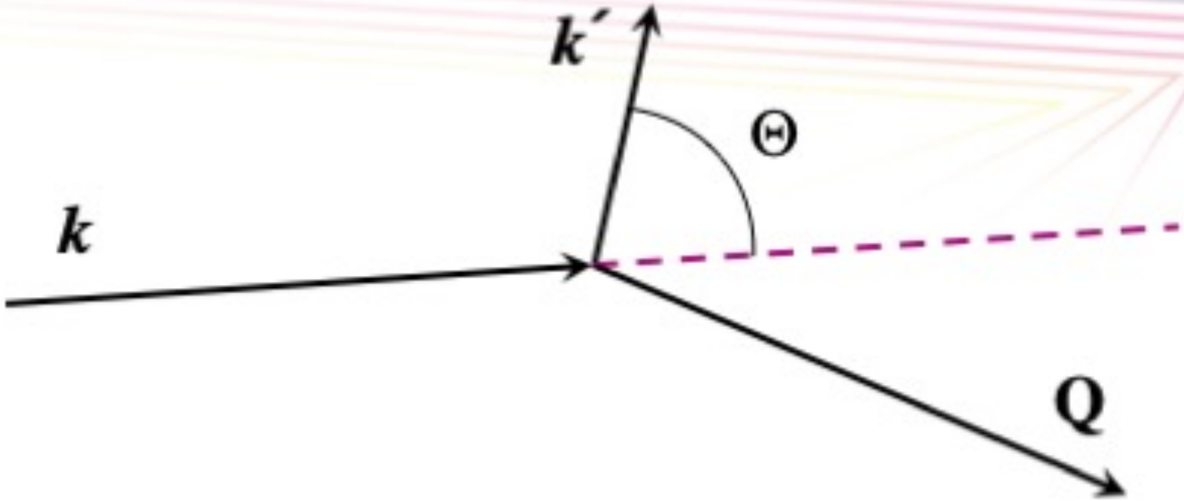
Learn to work with vectors



A neutron scattering experiment



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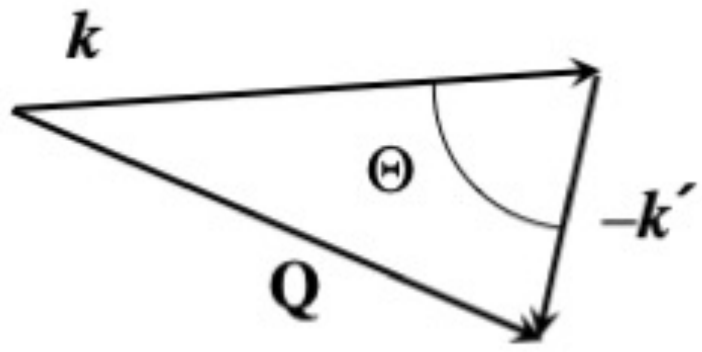
Closing the triangle



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A neutron scattering experiment

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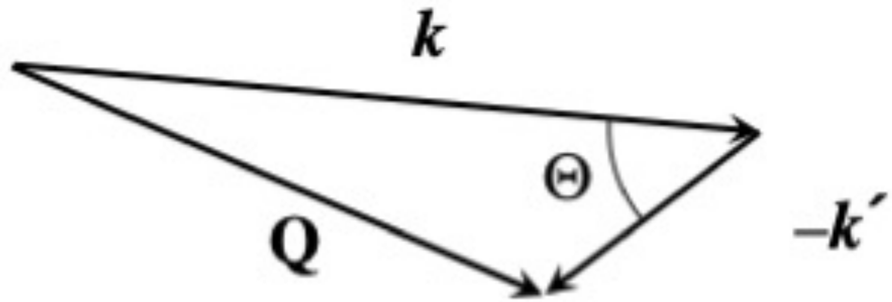

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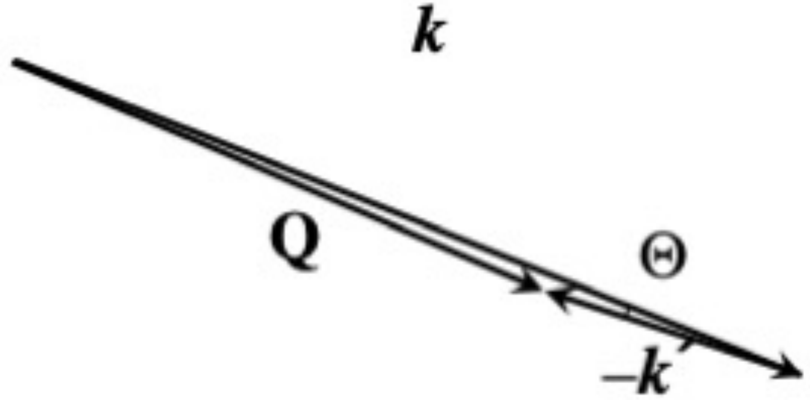

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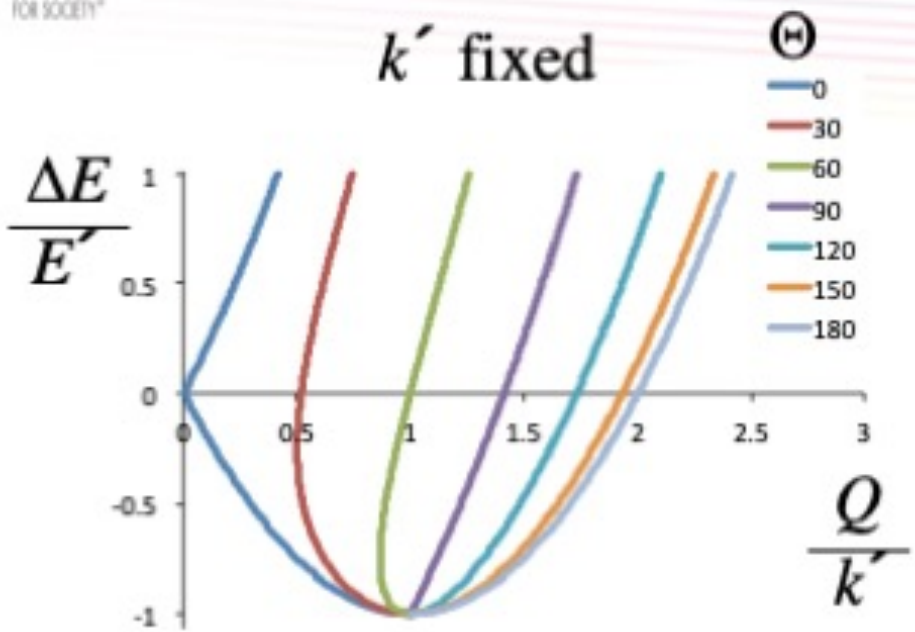

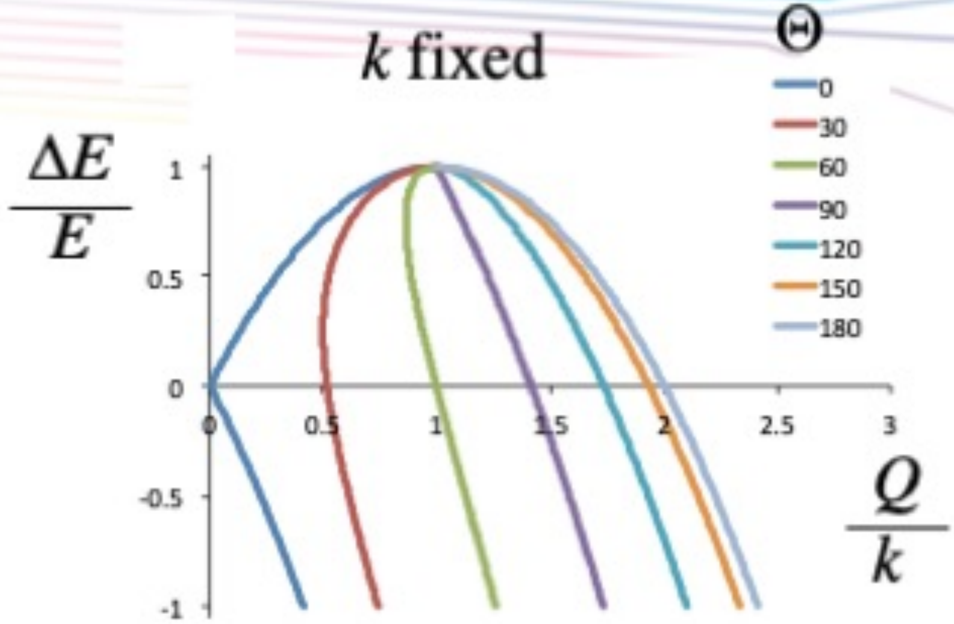

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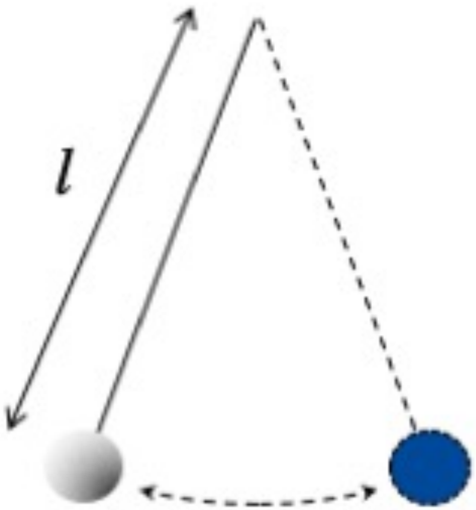
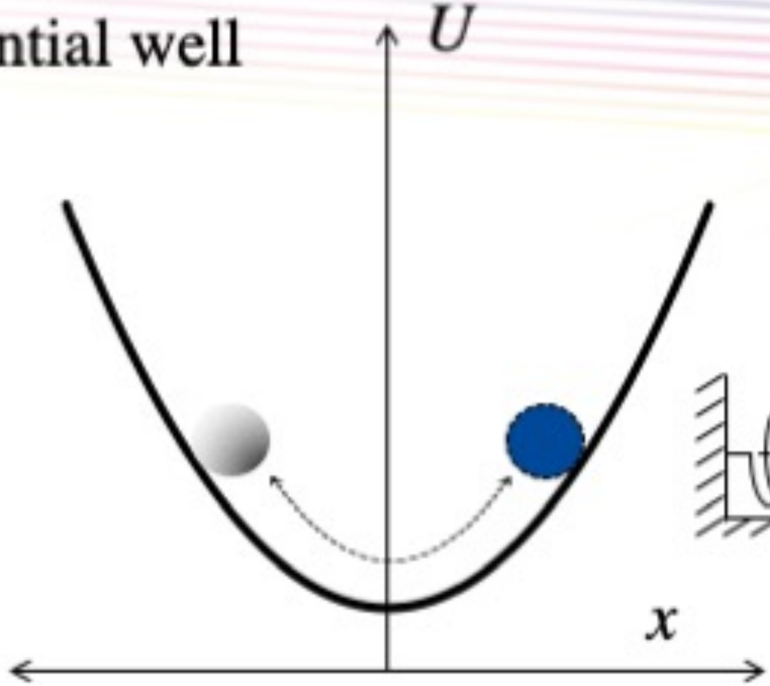
Classical harmonic oscillators

Pendulum



$$\upsilon = \frac{1}{2\pi}\sqrt{\frac{g}{l}}, \quad \omega = \sqrt{\frac{g}{l}}$$

Potential well



$$U = Ax^2$$

$$\upsilon = \frac{1}{2\pi}\sqrt{2Ag}, \quad \omega = \sqrt{2Ag}$$

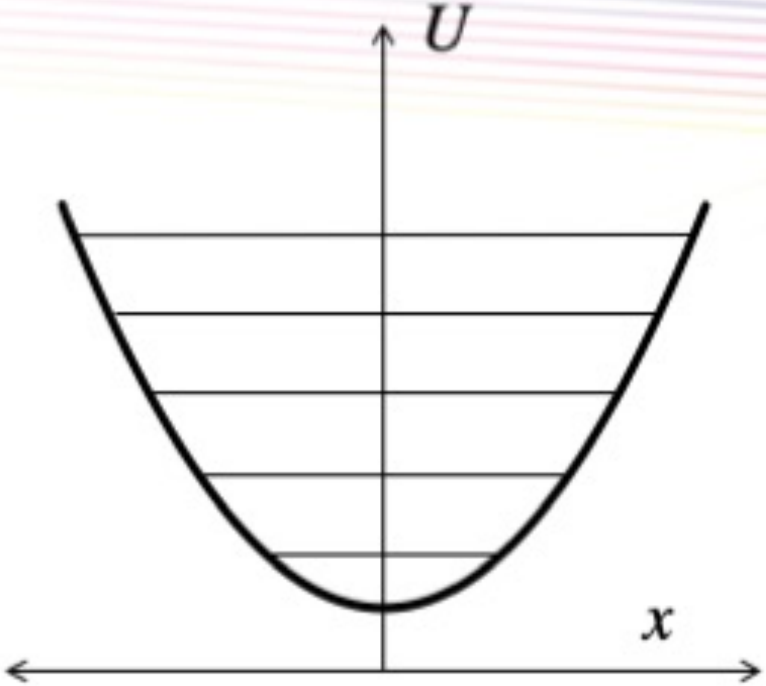
Spring (constant = C)

$$\upsilon = \frac{1}{2\pi}\sqrt{\frac{C}{m}}, \quad \omega = \sqrt{\frac{C}{m}}$$

Quantum harmonic oscillators

$$H\psi = E\psi$$

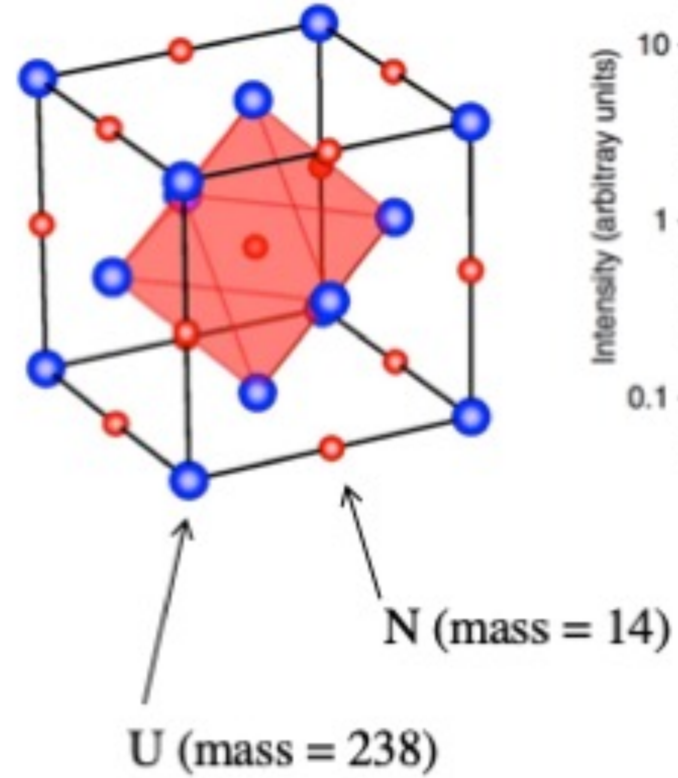
$$-\frac{\hbar^2}{2m}\frac{d^2\psi}{dx^2} + U(x)\psi = E\psi$$

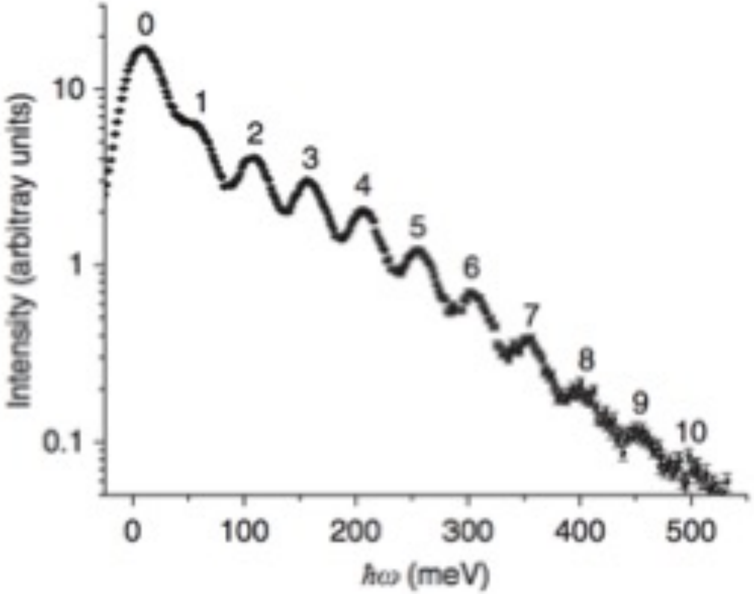


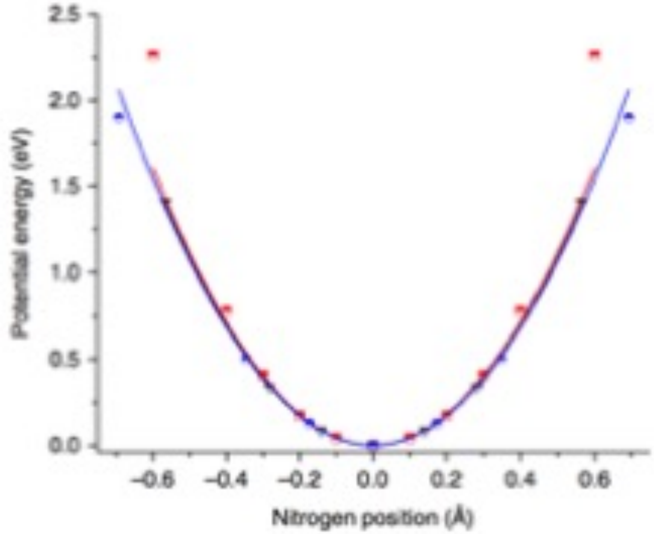
$$U = \frac{1}{2}m\omega^2 x^2$$

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

Nitrogen motion in UN



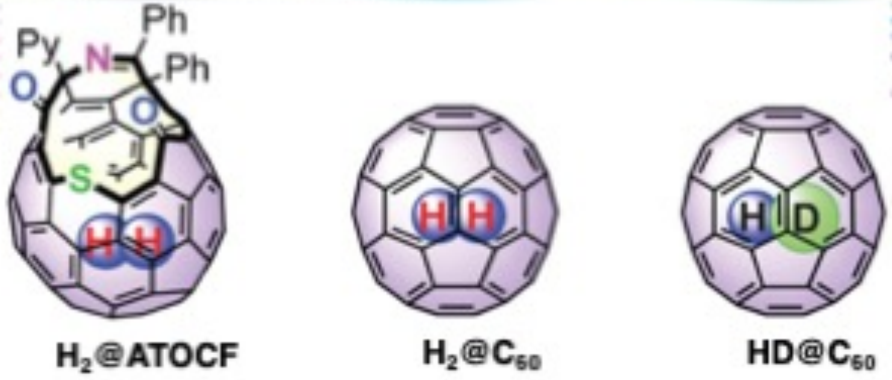




A. A. Aczel *et al*., Nat. Comm. **3** (2012) 1124

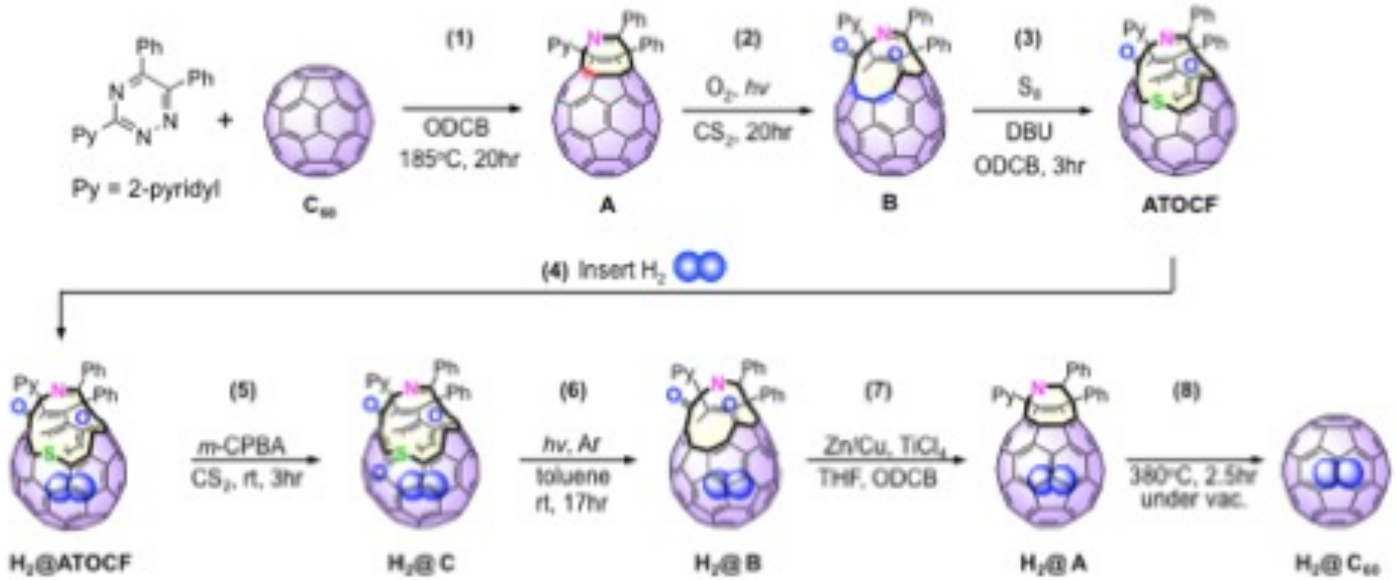
Endohedral fullerene

"Molecular surgery"



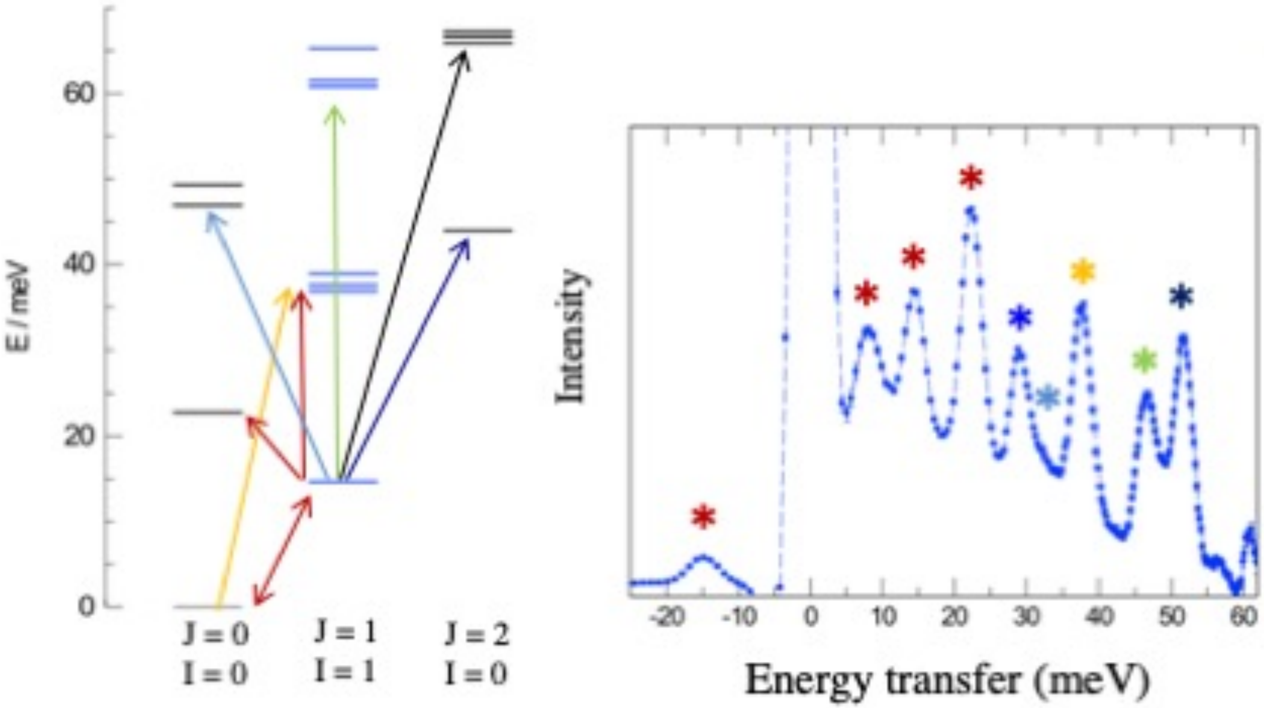
H_2, D_2, HD @C_{60}
H_2O @C_{60}

H_2, $(H_2)_2$ @C_{70}

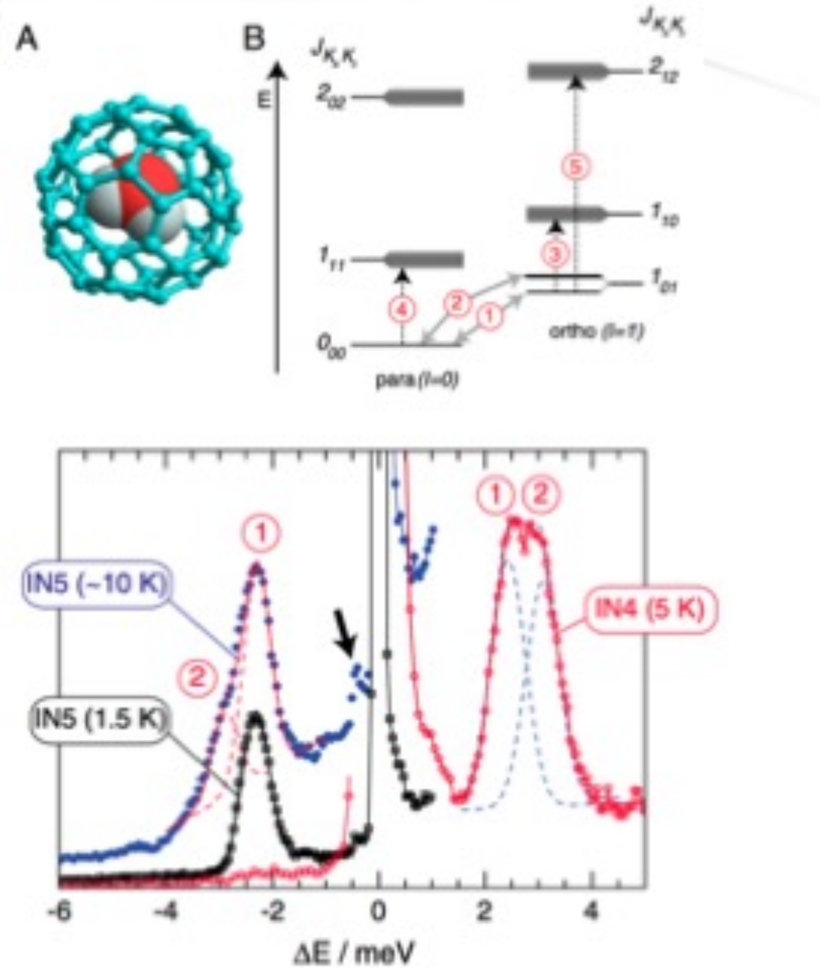


K.Komatsu, M.Murata and Y.Murata, Science 307, 238 (2005)

Endohedral fullerene

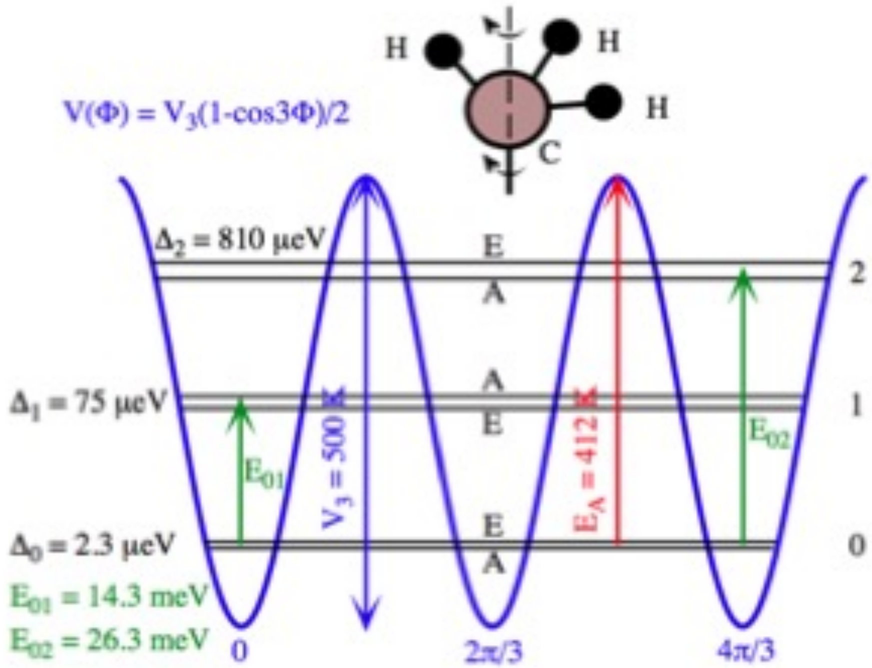


A. J. Horsewill *et al*., Proc. Trans. Roy. Soc A **371** (2013) 20110627

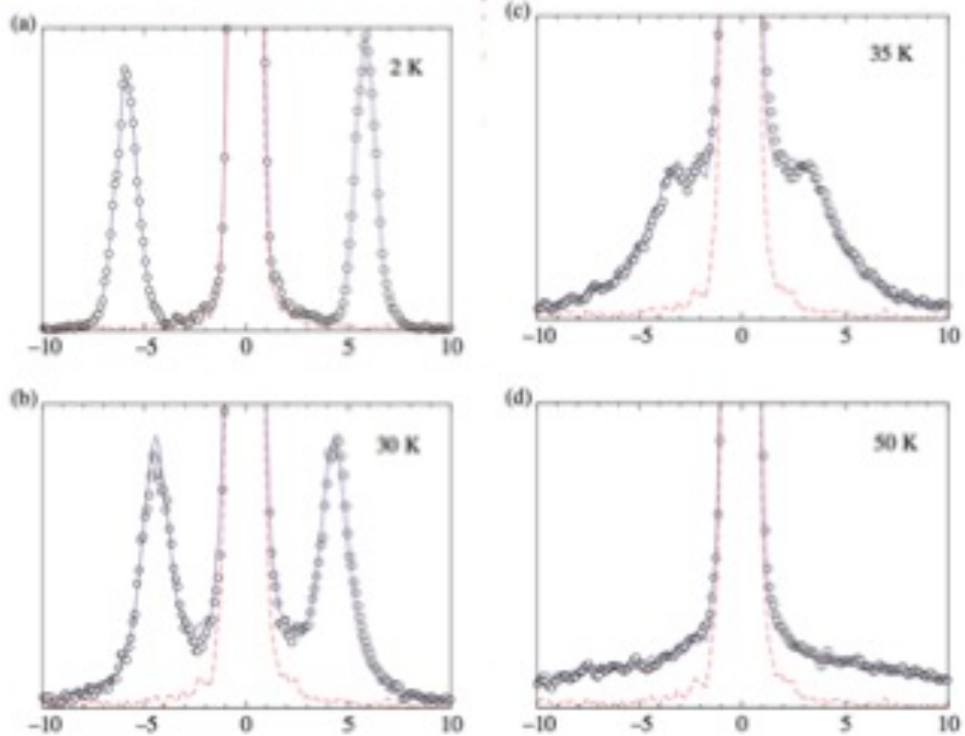


C. Beduz *et al*., PNAS **109** (2012) 12894

Methyl group motion



Tunnelling in Sodium Acetate Trihydride



Energy transfer (μeV)

J. Colmenero *et al*., Prog. Polym. Sci. **30** (2005) 1147

Tunnelling in (2,6)lutidine

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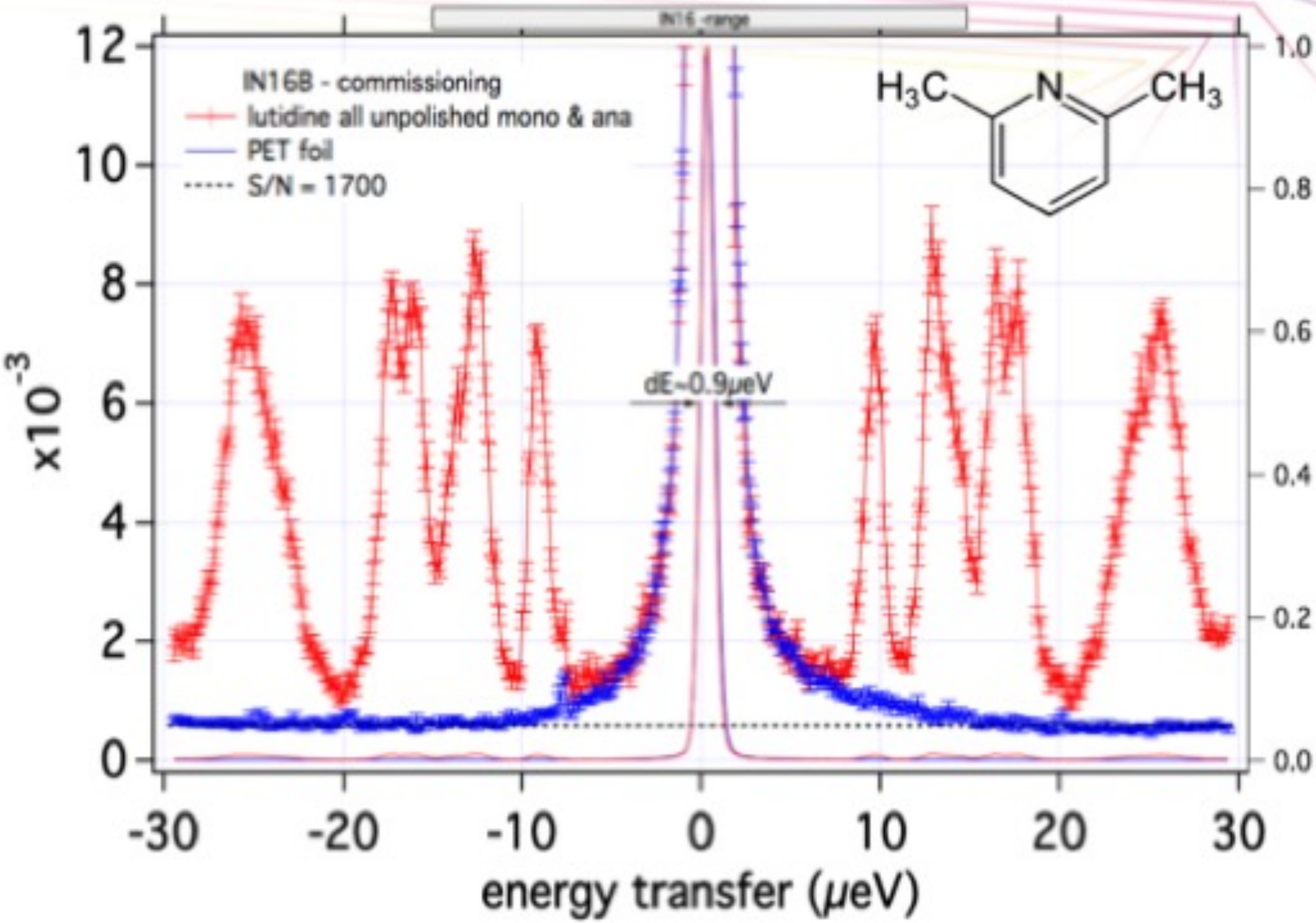
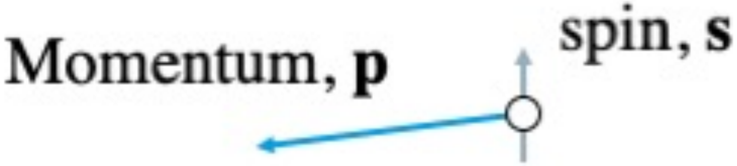


With thanks to B. Frick

Magnetism

Magnetism is caused by unpaired electrons or movement of charge.

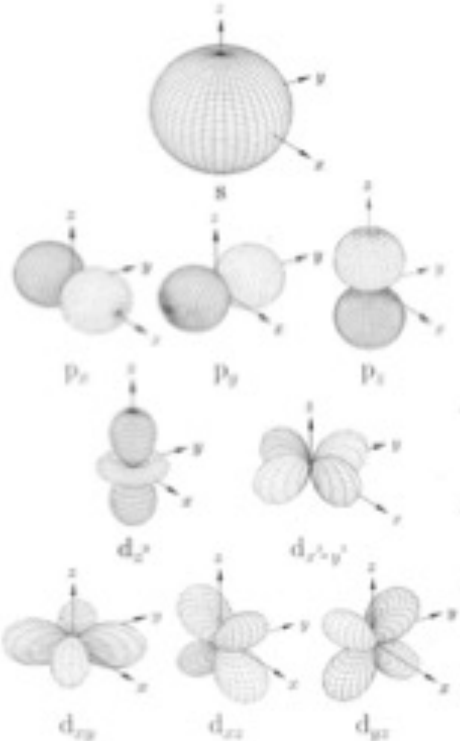


Magnetic spectroscopy requires a change of neutron *energy* and *angular momentum* (i.e. the neutron spin changes direction)

Magnetism can be classified as *localized* (i.e. confined to an atomic position) or *itinerant* (i.e. due to electrons that are moving through the sample)

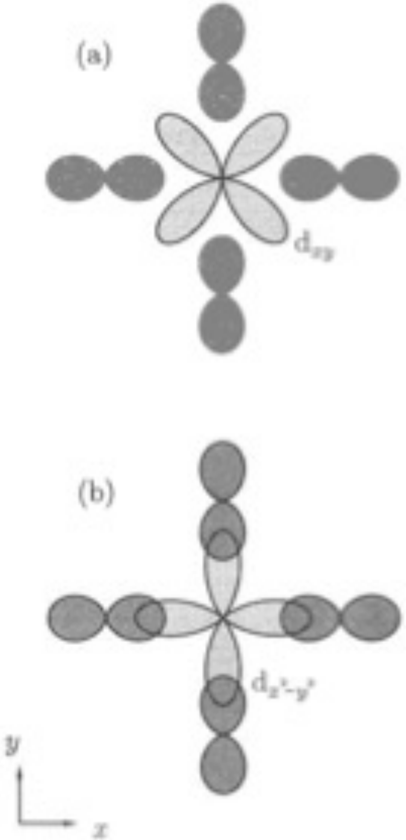
We'll only discuss localized magnetism today.

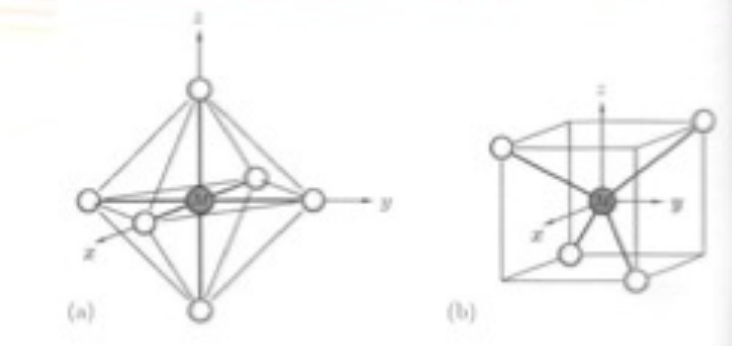
Crystal field levels

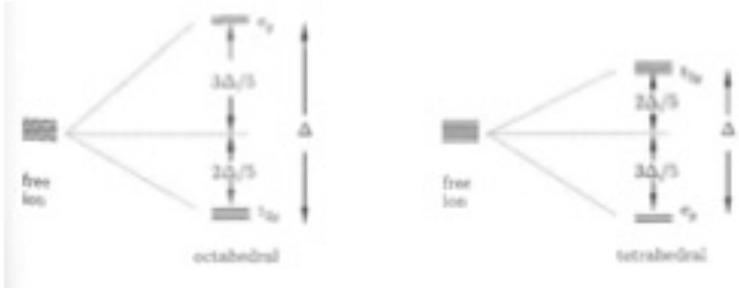


e_g

t_{2g}







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

Crystal fields in $NdPd_2Al_3$

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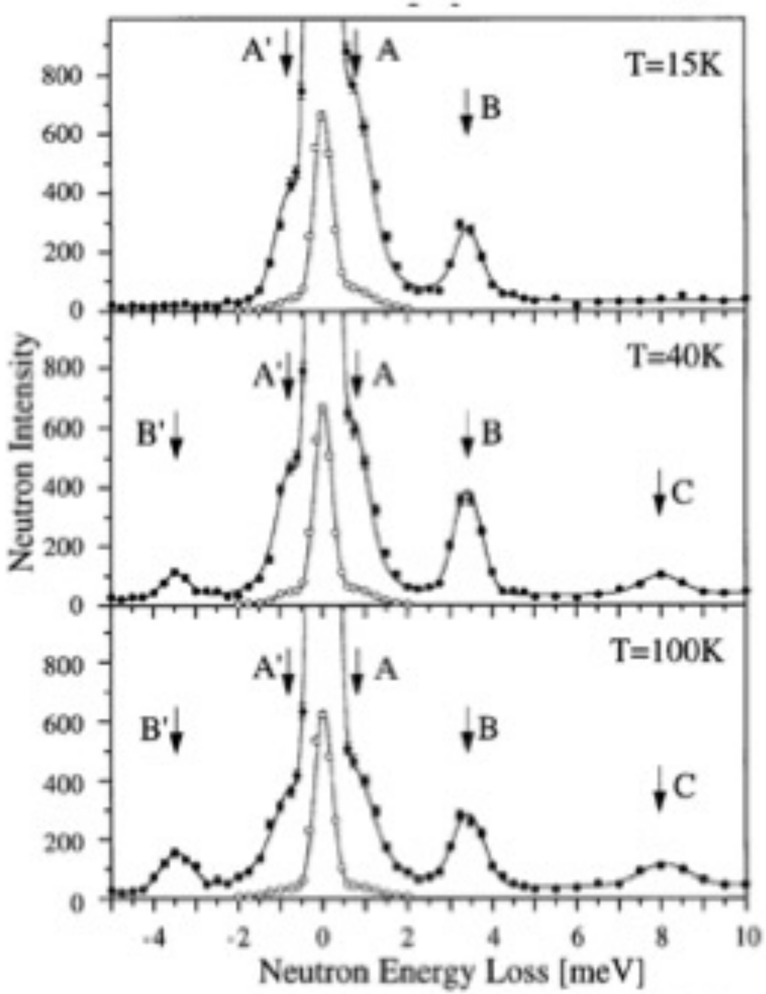
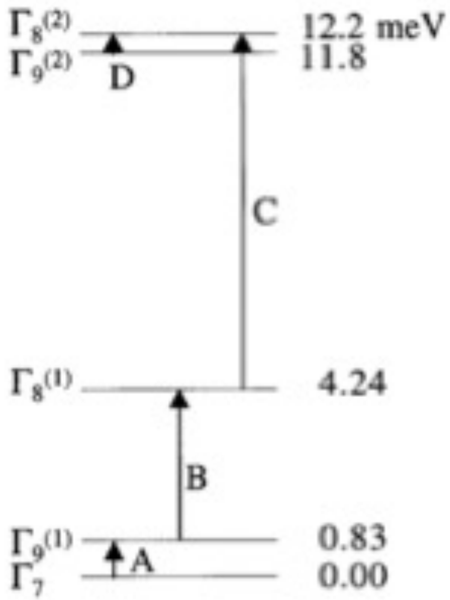
$$H = \sum_{l=2,4,6} B_l^0 O_l^0 + \sum_{l=2,4} B_l^4 O_l^4$$

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

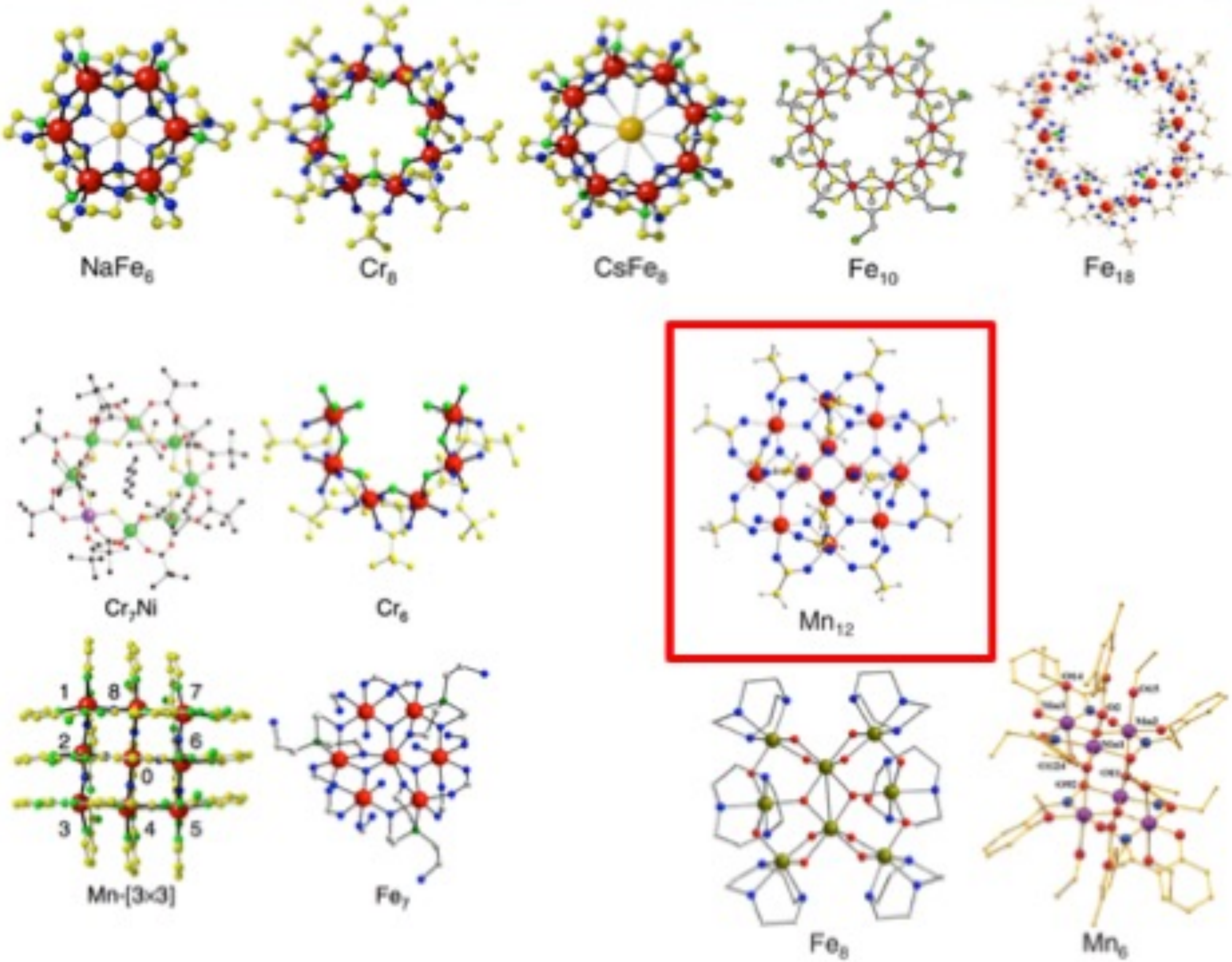




A. Dönni *et al*., J. Phys.: Condens. Matter **9** (1997) 5921

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

Molecular magnets



A. Furrer and O. Waldmann, Rev. Mod. Phys. **85** (2013) 367

Quantum tunneling in Mn_{12}-acetate

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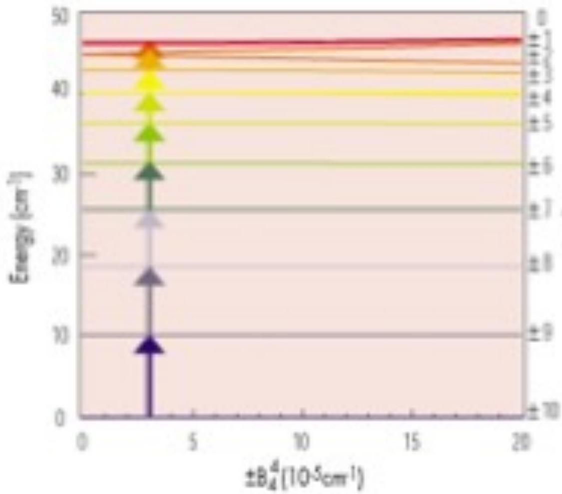
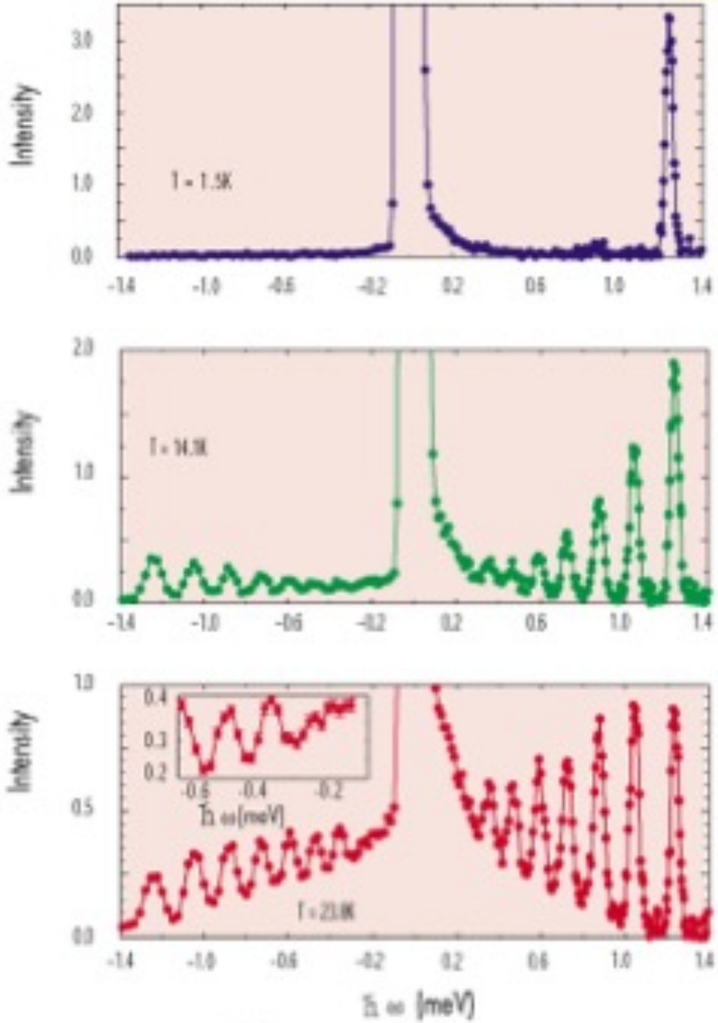
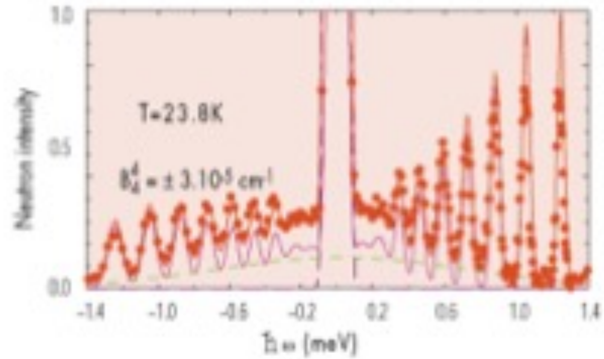
$$H = \sum_{l=2,4,6} B_l^0 O_l^0 + \sum_{l=2,4} B_l^4 O_l^4$$



Calculated energy terms



Neutron spectra



Fitted data with scattering from:

- energy levels
- elastic scattering
- incoherent background

I. Mirebeau *et al.*, Phys. Rev. Lett. **83** (1999) 628
R. Bircher *et al.*, Phys. Rev. B **70** (2004) 212413

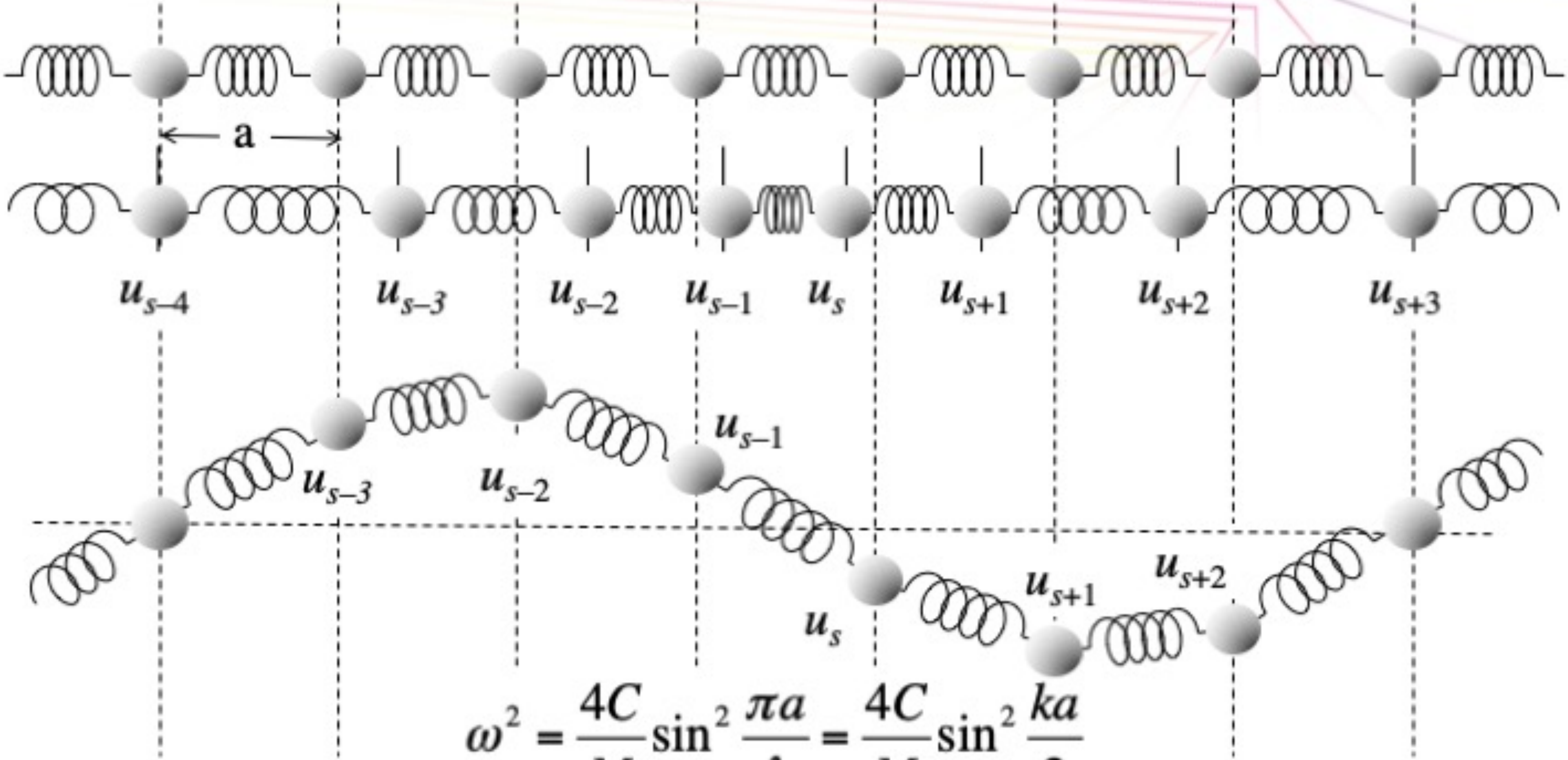
Classical harmonic oscillators

Spring (constant = C)

$$\upsilon = \frac{1}{2\pi}\sqrt{\frac{C}{m}}, \quad \omega = \sqrt{\frac{C}{m}}$$

Propagating lattice vibrations

Take a line of equal masses, M, joined by springs

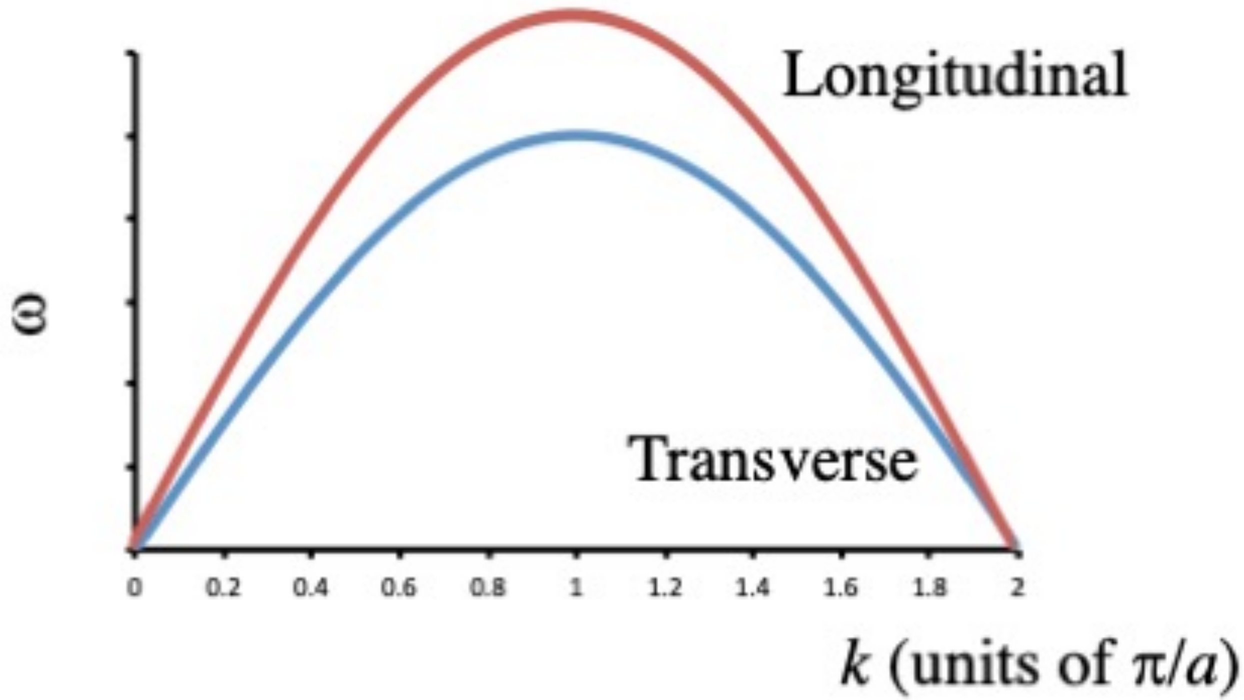


$$\omega^2 = \frac{4C}{M}\sin^2\frac{\pi a}{\lambda} = \frac{4C}{M}\sin^2\frac{ka}{2}$$

C. Kittel, *Introduction to Solid State Physics* (1996) Wiley, New York

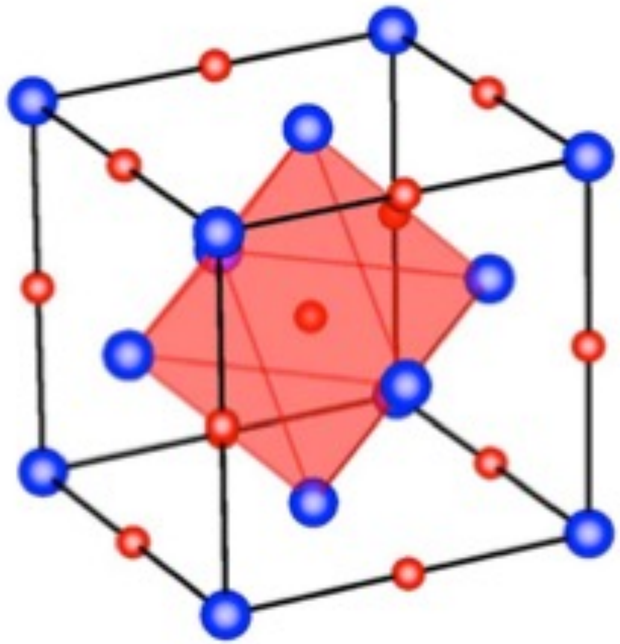
Phonon dispersion

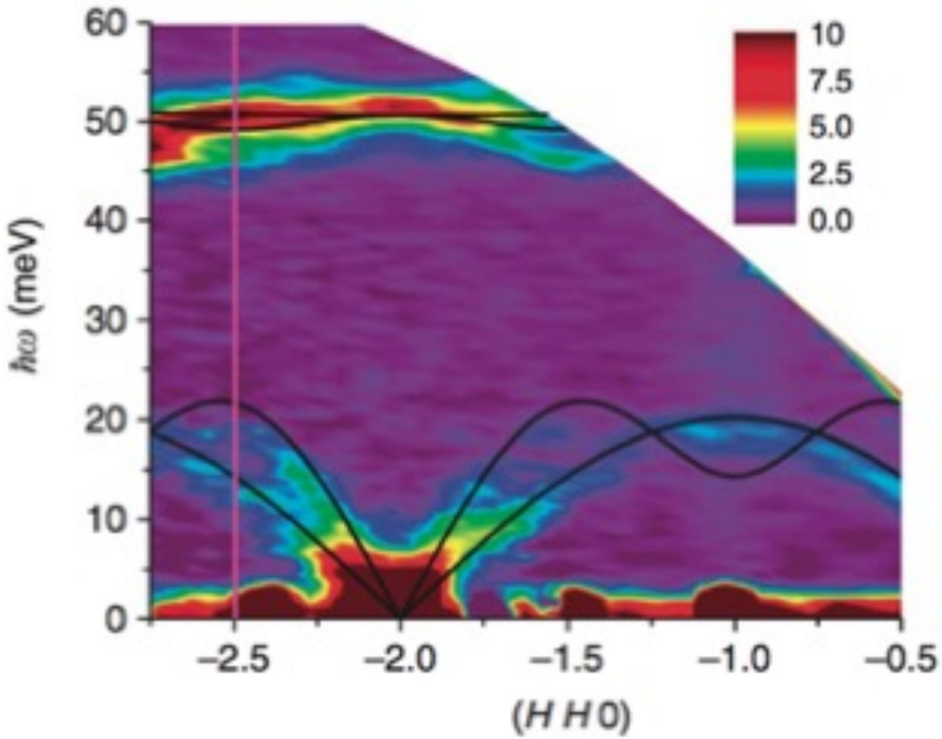
$$\omega^2 = \frac{4C}{M}\sin^2\frac{\pi a}{\lambda} = \frac{4C}{M}\sin^2\frac{ka}{2}$$



C. Kittel, *Introduction to Solid State Physics* (1996) Wiley, New York

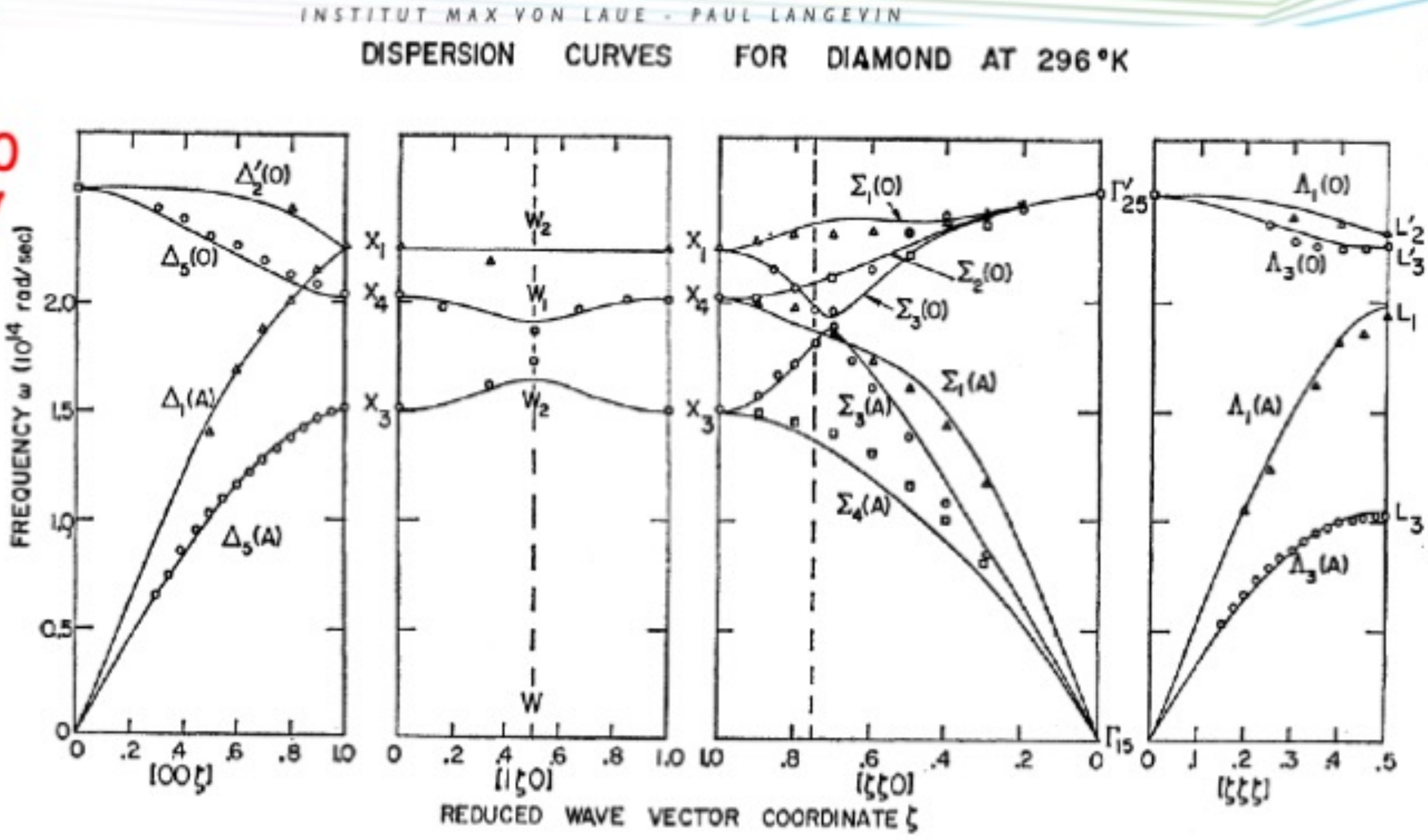
Phonons in UN





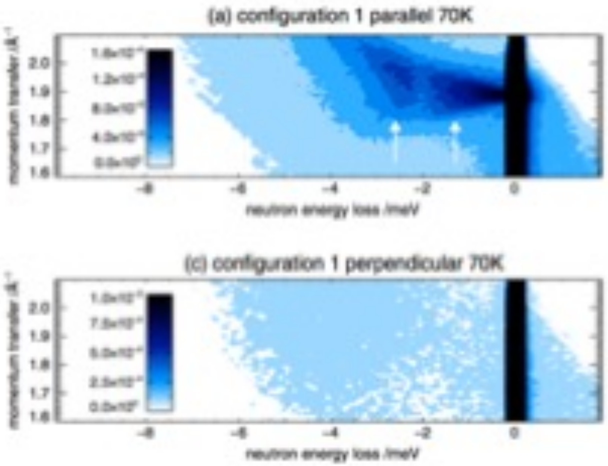
A. A. Aczel *et al*., Nat. Comm. **3** (2012) 1124

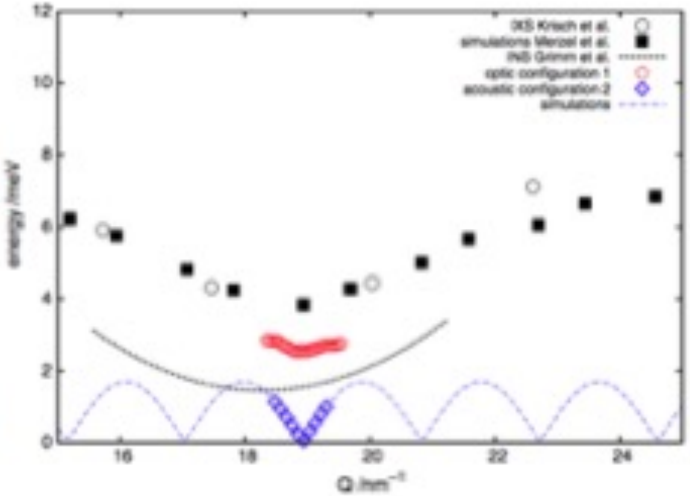
Phonons in Diamond

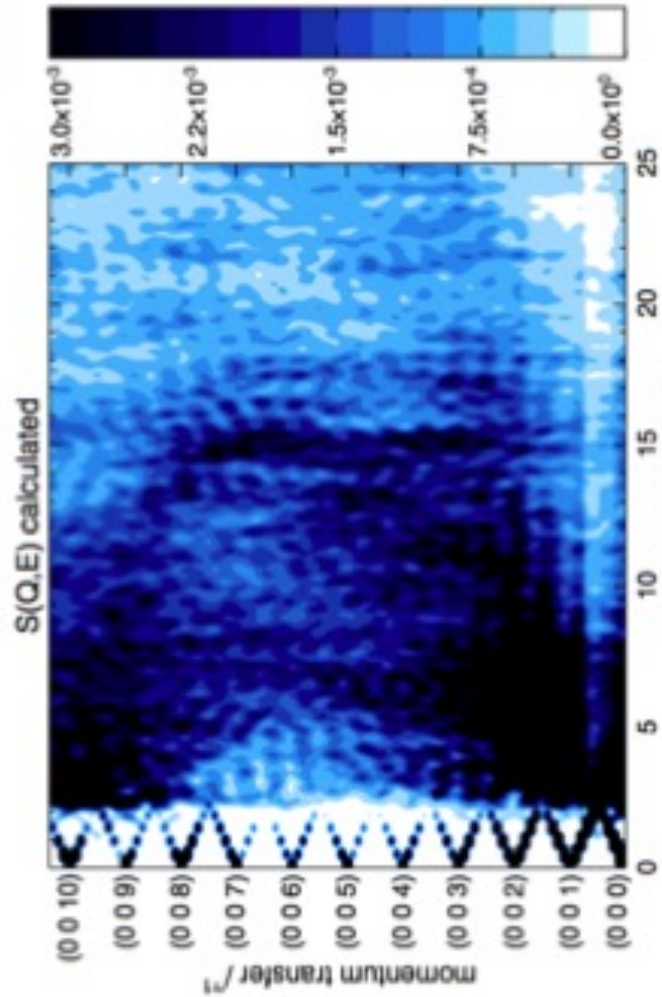


J.L. Warren et al. (1967) Phys.Rev. 158, 805

Phonons in fibre DNA

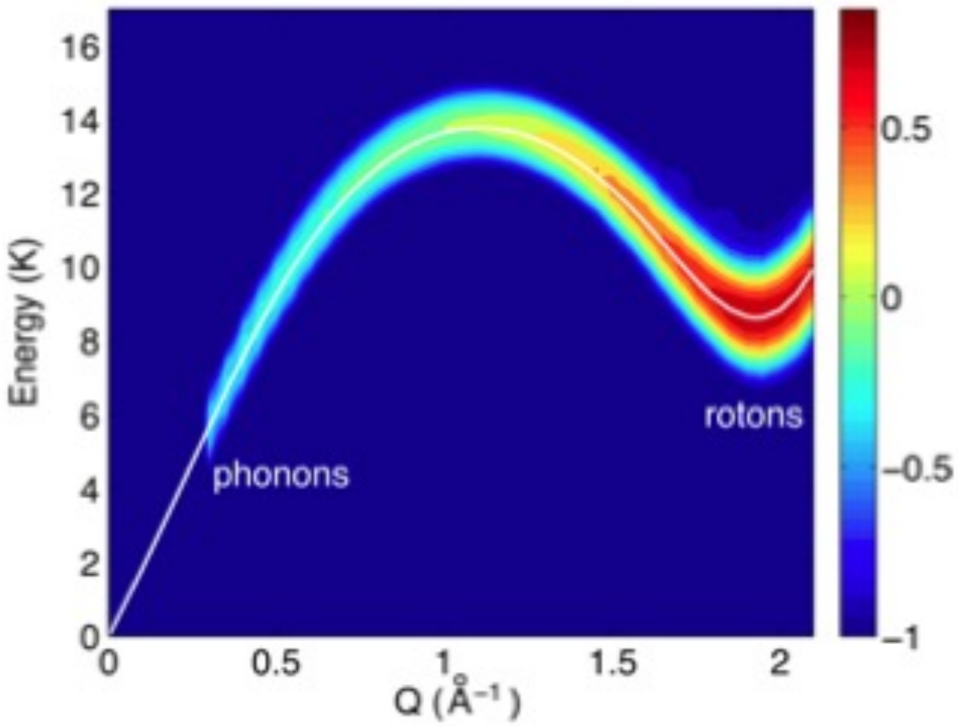




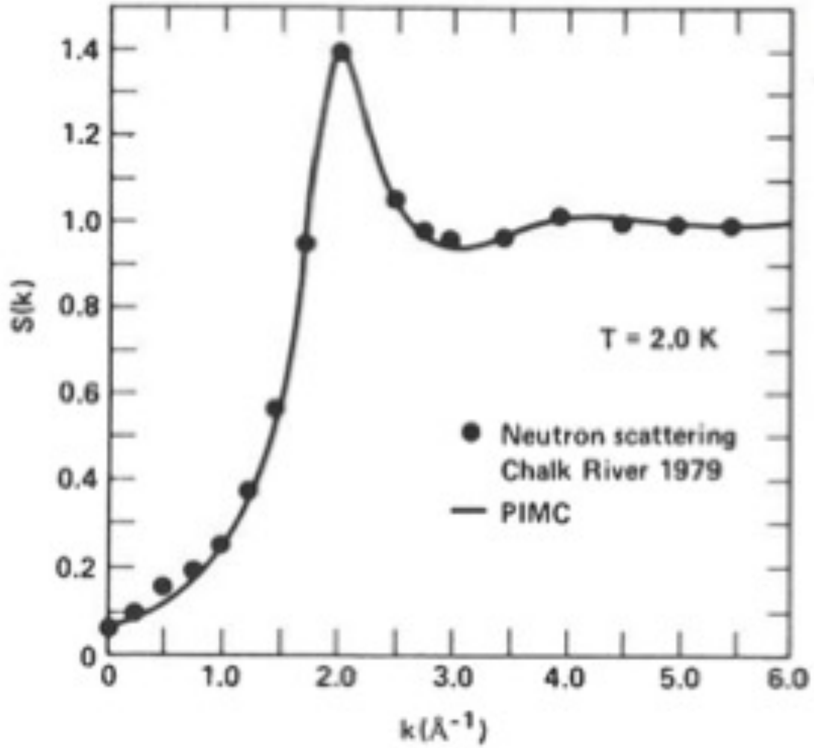


L. van Eijck *et al.*, PRL **107** (2011) 088102

Phonons in superfluid Helium



B. Fåk *et al*., PRL **109** (2012) 155305



E. C. Svensson *et al*., PRB **23** (1981) 4493
D. M. Ceperley and E. L. Pollock, Can. J. Phys. **65** (1987) 1416
H. R. Glyde, *Excitations in Liquid and Solid Helium*
(1994) Clarendon, Oxford

Spin waves and magnons

A simple Hamiltonian for spin waves is:

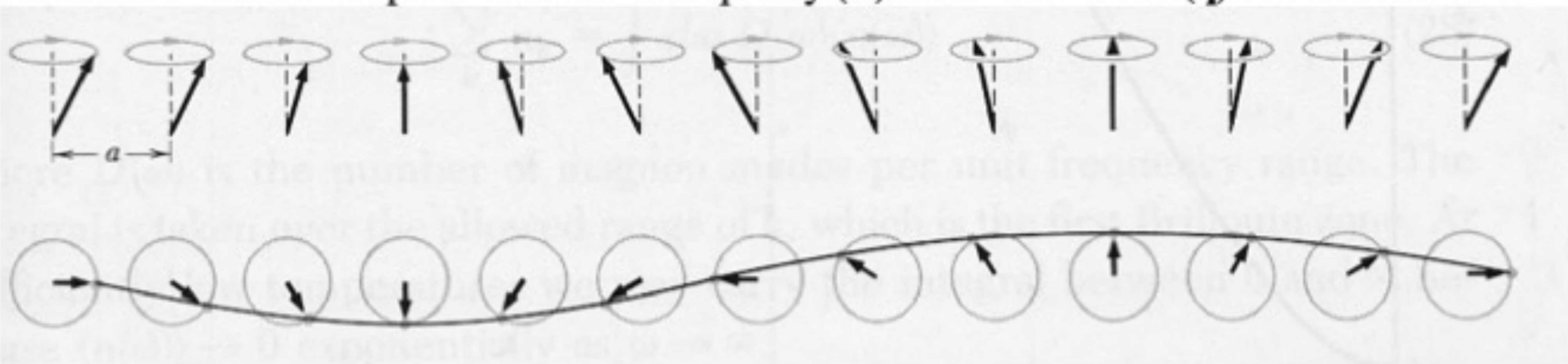
$$H = -J\sum_{i,j}\mathbf{s}_i\mathbf{s}_j$$

J is the magnetic exchange integral, which can be measured with neutrons.

Take a simple ferromagnet:

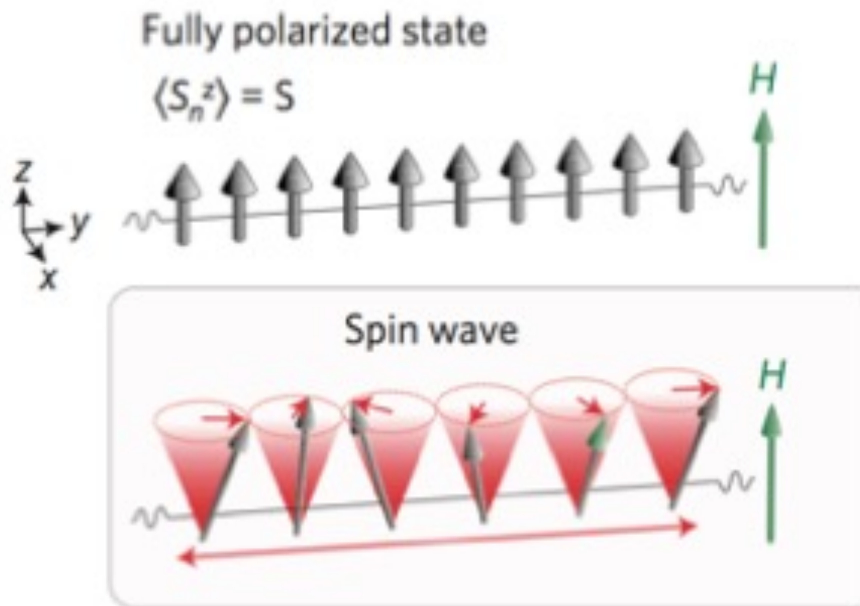
The spin waves might look like this:

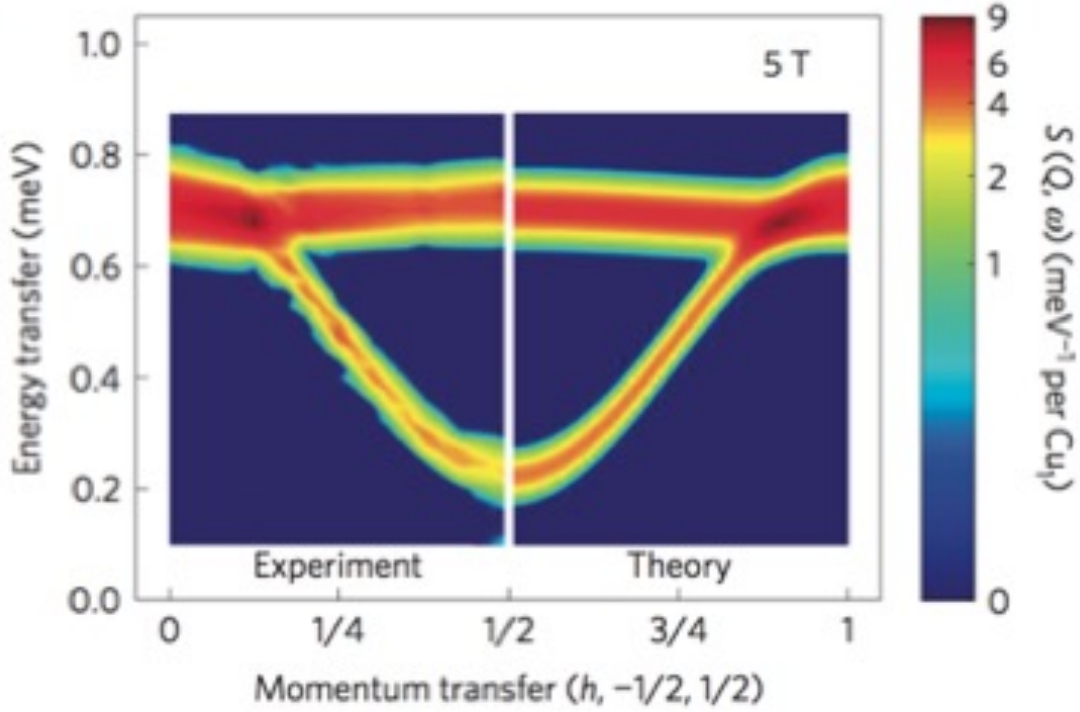
Spin waves have a frequency (ω) and a wavevector (**q**)



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

Magnetic excitations in $CuSO_4$





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

Conclusions

Neutron spectroscopy is about measuring quantum oscillations in solids and liquids

The neutron's momentum and energy are ideal for these

Neutron spectroscopy is a *quantitative* technique that *directly* probes the dynamics in the sample