Neutrons for magnetism

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2011 School - Time-dependent phenomena in magnetism
Targoviste, August 2011

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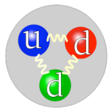
Neutrons



- Neutron discovered by James Chadwick in 1932
- Used in magnetic neutron diffraction in 1949 (Clifford Shull)
- Neutron mass close to proton mass
- Neutron decays in ~15 minutes
- Spin ½
- Magnetic moment $1.9\mu_{\mathrm{N}} \approx 0.001\mu_{\mathrm{B}}$

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Neutrons

- Neutron has **no electrical charge**
- Interaction is with *atomic nuclei* (strong force, short range) and *unpaired electrons* (EM)
- Neutron-matter interaction is **weak**: perturbation theory adequate (very roughly, probability of interaction in a solid ~10^{-8}, mean free path ~1 cm)
- Neutron interaction with nuclei and electrons similar in magnitude
- Scattering cross section with proton is huge: 82 barns. For deuterons, it is an order of magnitude smaller. [1 barn = 10^{-28} m^2]

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Neutrons

- Energy given by $E = \hbar^2 k^2/2m$
- Energies are often given in meV = 10^{-3} eV
- Wavelength λ given in Angstroms
- Useful results: $\dfrac{\hbar^2}{2m} = 2.08\,\mathrm{meV}\ \text{Å}^{-2}$

$$\lambda = (h^2/2mE)^{1/2} = \frac{9.04}{\sqrt{E}}$$

$$1\,\mathrm{meV} \equiv 0.24\,\mathrm{THz} \equiv 8.07\,\mathrm{cm}^{-1} \equiv 11.61\,\mathrm{K}$$

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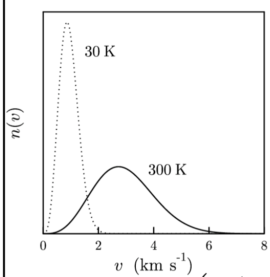
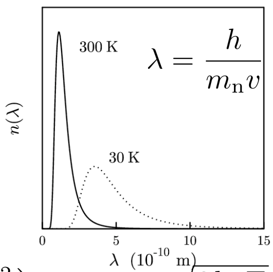
Moderator





$$\lambda = \frac{h}{m_{\mathrm{n}} v}$$

$$n(v) \propto v^3 \exp\left(-\frac{1}{2}\frac{m_{\mathrm{n}} v^2}{k_{\mathrm{B}} T}\right) \qquad v_{\max} = \sqrt{\frac{3k_{\mathrm{B}}T}{m_{\mathrm{n}}}}$$

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Neutrons

- Measure distribution of neutrons scattered from sample
- Interaction potential determines properties measured
- Scattering must be *coherent* for correlations to be measured
- Scattering of neutrons is very weak and so can use the *Born approximation*
- Means scattering depends on the Fourier transform of the interaction potential, and system responds linearly

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Neutrons

- The *Born approximation* assumes coherence and superposition
- Detected amplitude = $\phi_1 + \phi_2 + \cdots$
- $\Longrightarrow$ Detected intensity = $|\phi_1 + \phi_2 + \cdots|^2$

$$= |\phi_1|^2 + |\phi_2|^2 + \phi_1^*\phi_2 + \phi_2^*\phi_1 + \cdots$$

depends on relative positions of atoms 1 and 2

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Neutrons

- Measurement non-destructive (though they might activate the sample!)
- Bulk, not surface probe
- Samples have to be big (~1 mm^3 for diffraction experiments, ~1cm^3 for inelastic measurements)
- Technique expensive – need a nuclear reactor with holes in it (!) or a spallation source. These are not cheap. Fortunately, Europe has a number of excellent sources of neutrons.

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Kinematics

- Conservation of energy

$$\hbar\omega = E_{\rm i} - E_{\rm f} = \frac{\hbar^2}{2m}(k_{\rm i}^2 - k_{\rm f}^2)$$

- Conservation of momentum $\mathbf{Q} = \mathbf{k}_{\rm i} - \mathbf{k}_{\rm f}$
- Scattering event characterized by $(\mathbf{Q}, \omega)$
- (i) Elastic scattering $\hbar\omega = 0$
- (ii) Inelastic scattering $\hbar\omega \neq 0$
- Both processes occur in every experiment
- Can set $\mathbf{Q}$ and ω independently in the experiment

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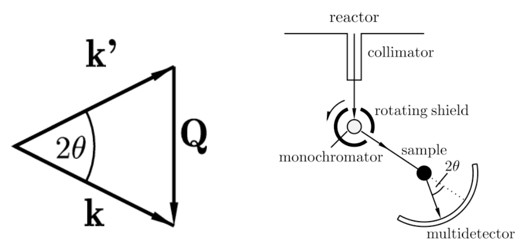
Elastic neutron scattering



$$\hbar\omega = E_{\rm i} - E_{\rm f} = 0$$
$$\mathbf{Q} = \mathbf{k}_{\rm i} - \mathbf{k}_{\rm f}$$

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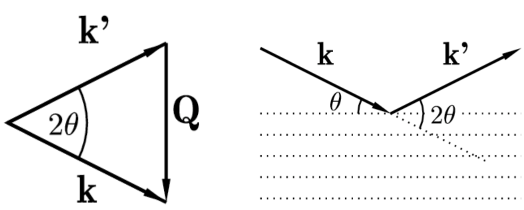
Elastic neutron scattering



$$\hbar\omega = E_{\rm i} - E_{\rm f} = 0$$
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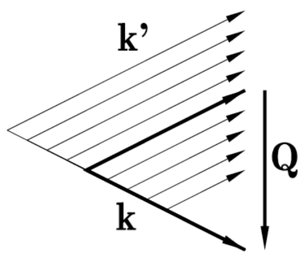
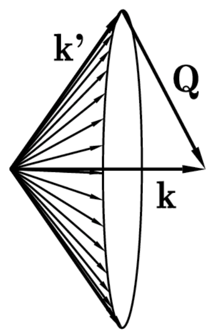
Elastic neutron scattering





$$\hbar\omega = E_{\rm i} - E_{\rm f} = 0$$
$$\mathbf{Q} = \mathbf{k}_{\rm i} - \mathbf{k}_{\rm f}$$

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Nuclear scattering

- Scalar potential, short-range: $V_{\mathrm{N}} = \frac{2\pi\hbar^2}{m_{\mathrm{n}}} b\delta(\mathbf{r})$
- Scattering length b~10^{-14} m
- Scattering function

$$S(\mathbf{Q}) = |\sum_j b_j \exp(i\mathbf{Q}\cdot\mathbf{r}_j)|^2$$

sum over nuclei in unit cell

- Rigid crystal

$$S(\mathbf{Q}) = N\frac{(2\pi)^3}{V_0}\sum_{\mathbf{G}} |F(\mathbf{G})|^2 \delta(\mathbf{Q}-\mathbf{G})$$

sum over reciprocal lattice vectors

- N=number of unit cells
- V_0=volume of unit cell

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Nuclear scattering

Rigid crystal

$$S(\mathbf{Q}) = N\frac{(2\pi)^3}{V_0}\sum_{\mathbf{G}} |F(\mathbf{G})|^2 \delta(\mathbf{Q}-\mathbf{G})$$

sum over reciprocal lattice vectors

- structure factor $F(\mathbf{G}) = \sum_j b_j \exp(i\mathbf{G}\cdot\mathbf{r}_j)$

sum over nuclei in unit cell

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Coherent and incoherent

- b varies with isotope and spin orientation
- separate b into an average value $\bar{b}$ and
- coherent scattering results from $\bar{b}$ and gives rise to diffraction peaks
- incoherent scattering results from $b-\bar{b}$ and gives rise to an incoherent background

$$S_{\mathrm{inc}}(\mathbf{Q}) = \sum_j \frac{(\sigma_{\mathrm{inc}})_j}{4\pi}$$

sum over nuclei in unit cell

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Magnetic neutron diffraction

- Magnetic interaction potential energy

$$V_{\mathrm{M}} = -\mu_{\mathrm{n}}\cdot\mathbf{B}(\mathbf{r})$$

- depends on spin and orbital currents
- depends on direction of neutron spin
- vector, not scalar, interaction
- anisotropic scattering
- derives from electronic states and so the *magnetic form factor* is important

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Magnetic neutron diffraction

- Magnetic interaction potential in Q-space

$$V_{\mathrm{M}}(\mathbf{Q}) = -\mu_{\mathrm{n}}\cdot\mathbf{B}(\mathbf{Q})$$

- Maxwell: $\nabla\cdot\mathbf{B} = 0 \Longrightarrow i\mathbf{Q}\cdot\mathbf{B}(\mathbf{Q}) = 0$

$\mathbf{B}(\mathbf{Q})$ is therefore perpendicular to $\mathbf{Q}$

- Neutrons scatter from $\mathbf{m}_\perp$, the component of the magnetic moment *perpendicular* to $\mathbf{Q}$

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Magnetic neutron diffraction

Magnetically ordered crystal

$$S_{\mathrm{M}}(\mathbf{Q}) = C\sum_{\mathbf{G}_{\mathrm{M}}} |F(\mathbf{G})|^2 \delta(\mathbf{Q}-\mathbf{G}_{\mathrm{M}})$$

sum over magnetic reciprocal lattice vectors

- magnetic structure factor

$$F_{\mathrm{M}}(\mathbf{G}) = \sum_j f_j(\mathbf{Q})\mathbf{m}_{\perp j}\exp(i\mathbf{G}\cdot\mathbf{r}_j)$$

sum over magnetically ordered nuclei in unit cell

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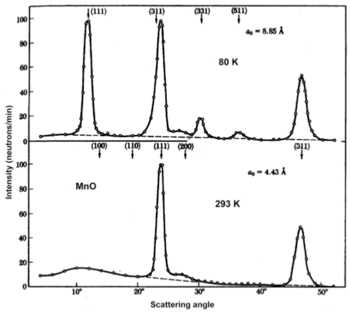
Magnetic neutron diffraction



MnO

C.G. Shull et al. (1951)

see also

A.L. Goodwin et al.
PRL **96,** 047209 (2006)

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Scattering cross section

Given the symbol σ

- Neutrons scattered into various final energies and also various bits of space (solid angle), so deal with the double differential cross section:

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_\mathrm{f}\,\mathrm{d}\Omega}$$

- This sums up to the total cross section

$$\sigma = \int\int \mathrm{d}E_\mathrm{f}\,\mathrm{d}\Omega\,\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_\mathrm{f}\,\mathrm{d}\Omega}$$

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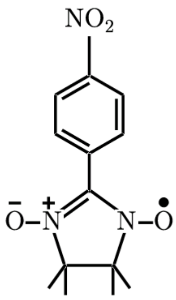
Example

para-nitro

-phenyl

nitronyl nitroxide



An organic ferromagnet with T_c=0.67 K

p-NPNN

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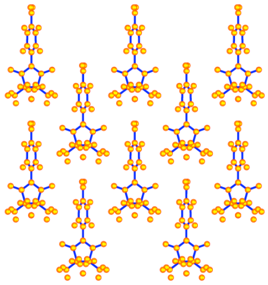
Example

Crystal structure strongly affects **magnetic coupling.**



Only the β-phase of *p*-NPNN is ferromagnetic.

One can use the polarized neutron diffraction maps to understand the important overlaps between regions of positive and negative spin density.

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Polarized neutron diffraction

The scattered intensity $I(\mathbf{k})$ for scattering vector $\mathbf{k}$ is given by

$$I(\mathbf{k}) = |F_\mathrm{N}(\mathbf{k})|^2 + \mathbf{P}\cdot(\mathbf{F}_\mathrm{M}^\perp(\mathbf{k})F_\mathrm{N}^*(\mathbf{k}) + \mathbf{F}_\mathrm{M}^{\perp *}(\mathbf{k})F_\mathrm{N}(\mathbf{k})) + |\mathbf{F}_\mathrm{M}^\perp(\mathbf{k})|^2$$

where $F_\mathrm{N}(\mathbf{k}) = \sum_j b_j \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}_j}\mathrm{e}^{-W_j}$ is the nuclear structure factor, $\mathbf{F}_\mathrm{M}^\perp(\mathbf{k})$ is the projection of the magnetic structure factor $\mathbf{F}_\mathrm{M}^\perp(\mathbf{k}) = \int_\mathrm{cell}\mathbf{M}(\mathbf{r})\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\,\mathrm{d}^3r$ on to the plane perpendicular to $\mathbf{k}$, $\mathbf{P}$ is the polarization of the beam, W_j is a Debye-Waller factor for the j^th atom in the unit cell at position r_j with scattering length b_j.

Measure *I(k)* for neutron polarization parallel or antiparallel with the magnetic field, and the sign of the interference term can be varied, allowing the magnetic structure factor to be deduced.

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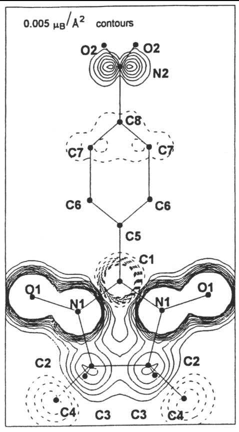
Example

spin density of p-NPNN



Zheludev et al
JMMM 135 147 (1994)

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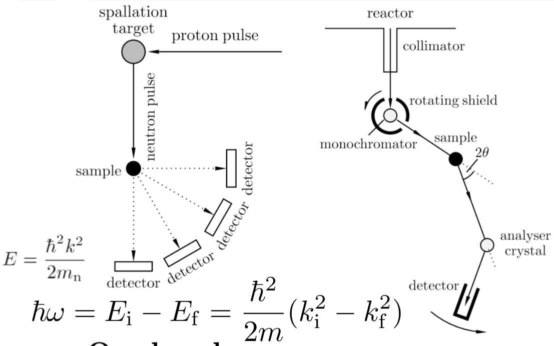
Inelastic neutron scattering



$$\hbar\omega = E_{\rm i} - E_{\rm f} = \frac{\hbar^2}{2m}(k_{\rm i}^2 - k_{\rm f}^2)$$

$$\mathbf{Q} = \mathbf{k}_{\rm i} - \mathbf{k}_{\rm f}$$

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Scattering cross section

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_{\rm f}\,\mathrm{d}\Omega} = \frac{\text{Number scattered s}^{-1}\text{ into d}\Omega,\ E_f \to E_f + \mathrm{d}E_f}{I_0\,\mathrm{d}\Omega\,\mathrm{d}E_{\rm f}}$$

- Numerator contains (i) $\mathrm{d}E_{\rm f}$ (ii) $\mathrm{d}\Omega$ (iii) speed of scattered neutrons $v_{\rm f} = \hbar k_{\rm f}/m$ (iv) density of incident neutrons ρ_i (v) transition probability in which the sample changes its momentum by $\mathbf{Q}$ and its energy by ω, i.e. $= S(\mathbf{Q},\omega)$
- Denominator contains (i) $I_0 = \rho_i v_i = \rho_i \hbar k_i/m$ and (ii)$\mathrm{d}E_{\rm f}$ (iii) $\mathrm{d}\Omega$

$$\Longrightarrow \quad \frac{\mathrm{d}^2\sigma}{\mathrm{d}E_{\rm f}\,\mathrm{d}\Omega} = \frac{k_{\rm f}}{k_{\rm i}} S(\mathbf{Q},\omega)$$

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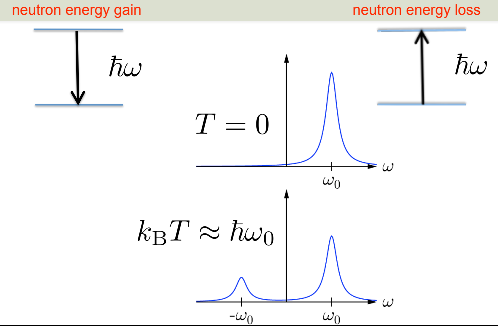
Detailed balance

$$S(\mathbf{Q},-\omega) = \exp(-\hbar\omega/k_{\rm B}T)S(\mathbf{Q},\omega)$$

neutron energy gain — neutron energy loss



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Calculation of $S(\mathbf{Q},\omega)$

$$S(\mathbf{Q},\omega) = \sum_i P_i \sum_j |M_{ij}|^2 \delta(\hbar\omega - E_j + E_i)$$

thermal occupation of initial state — matrix element

- Can look at local magnetic excitations, e.g. crystal field level spectroscopy

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Calculation of $S(\mathbf{Q},\omega)$

$$S(\mathbf{Q},\omega) = A\int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{-i\omega t} \sum_{\mathbf{R}} e^{i\mathbf{Q}\cdot\mathbf{R}} \langle S_{\mathbf{0}}^{\perp}(0) S_{\mathbf{R}}^{\perp}(t)\rangle$$

spin correlation function

- In spin waves, neutrons scatter from deviations perpendicular to ***Q***
- Intensity decreases with magnetic form factor
- Compare phonons, intensity ~ $b^2(\mathbf{Q}.\mathbf{e})^2$

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Calculation of $S(\mathbf{Q},\omega)$

$$S(\mathbf{Q},\omega) = (1 + n(\omega))\frac{1}{\pi}\chi''(\mathbf{Q},\omega)$$

spin correlation function

$$n(\omega) = \frac{1}{e^{-\hbar\omega/k_{\rm B}T} - 1}$$

$$M_\alpha(\mathbf{Q},\omega) = \chi_{\alpha\beta}(\mathbf{Q},\omega)H_\beta(\mathbf{Q},\omega)$$

$$\chi(\mathbf{Q},\omega) = \chi'(\mathbf{Q},\omega) + \mathrm{i}\chi''(\mathbf{Q},\omega)$$

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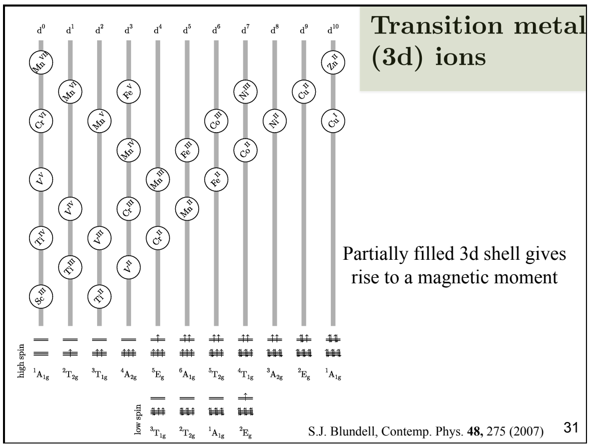
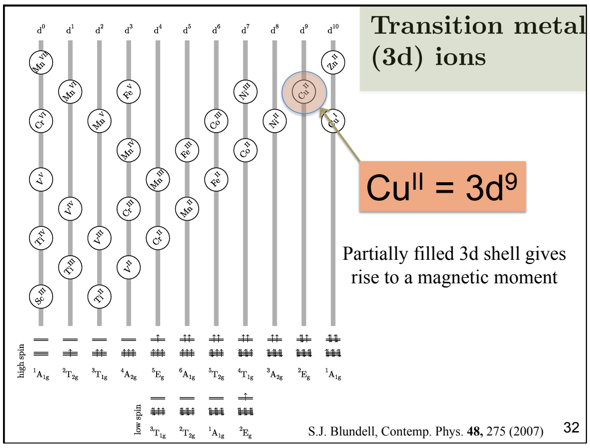
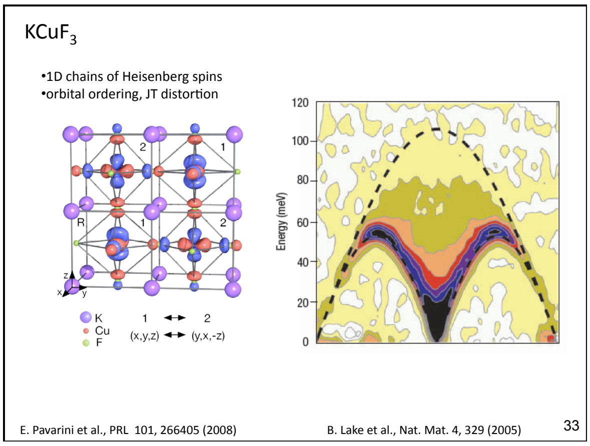
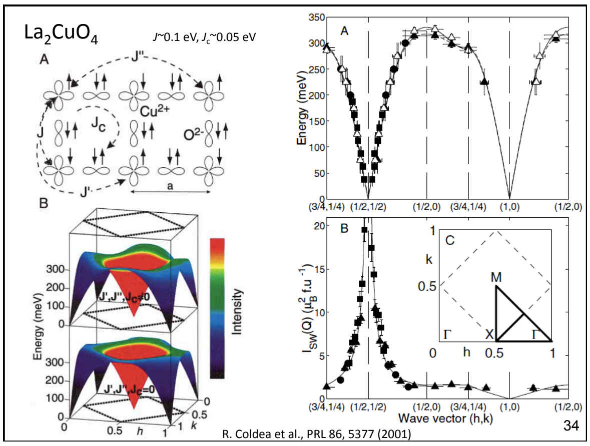
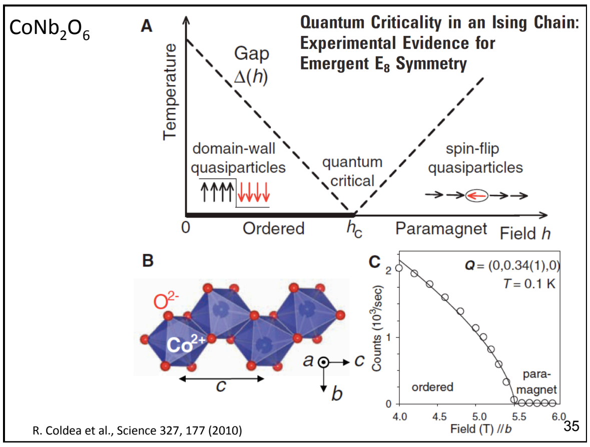
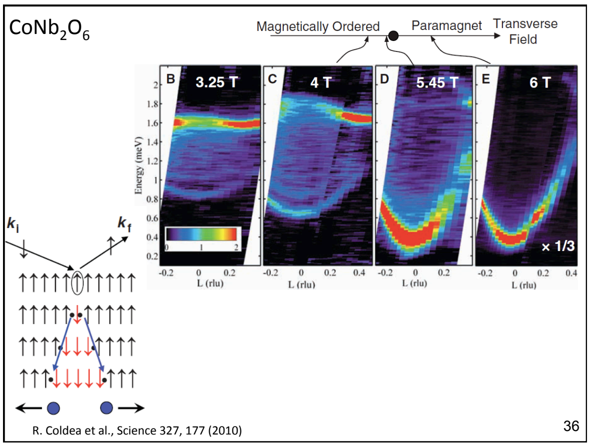

Transition metal (3d) ions
Partially filled 3d shell gives rise to a magnetic moment
high spin
low spin
S.J. Blundell, Contemp. Phys. 48, 275 (2007)
31


Transition metal (3d) ions
Cu^II = 3d^9
Partially filled 3d shell gives rise to a magnetic moment
high spin
low spin
S.J. Blundell, Contemp. Phys. 48, 275 (2007)
32


KCuF3
•1D chains of Heisenberg spins
•orbital ordering, JT distortion
K
Cu
F
1 ↔ 2
(x,y,z) ↔ (y,x,-z)
Energy (meV)
E. Pavarini et al., PRL 101, 266405 (2008)
B. Lake et al., Nat. Mat. 4, 329 (2005)
33


La2CuO4
J~0.1 eV, Jc~0.05 eV
Cu2+
O2-
Energy (meV)
Intensity
Wave vector (h,k)
R. Coldea et al., PRL 86, 5377 (2001)
34


CoNb2O6
Quantum Criticality in an Ising Chain: Experimental Evidence for Emergent E8 Symmetry
Temperature
Gap Δ(h)
domain-wall quasiparticles
quantum critical
spin-flip quasiparticles
Ordered
Paramagnet
Field h
O2-
Co2+
Q = (0,0.34(1),0)
T = 0.1 K
Counts (10^3/sec)
ordered
para-magnet
Field (T) // b
R. Coldea et al., Science 327, 177 (2010)
35


CoNb2O6
Magnetically Ordered
Paramagnet
Transverse Field
3.25 T
4 T
5.45 T
6 T
× 1/3
Energy (meV)
L (rlu)
R. Coldea et al., Science 327, 177 (2010)
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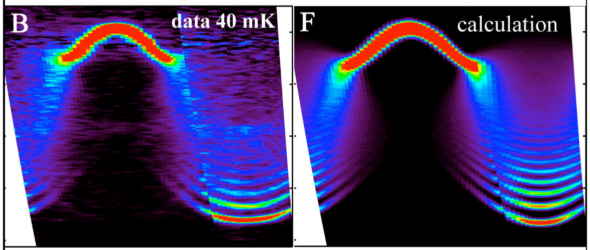
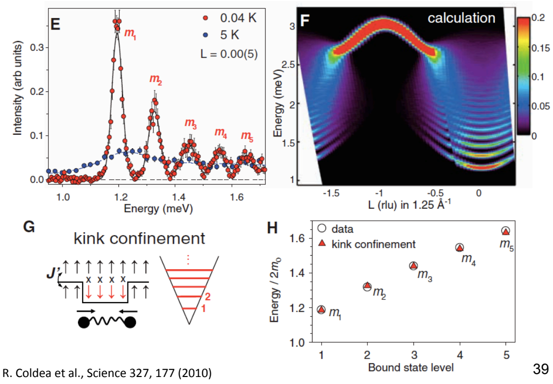
$CoNb_2O_6$



R. Coldea et al., Science 327, 177 (2010)

$CoNb_2O_6$

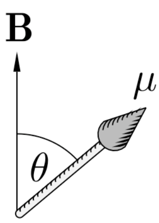


R. Coldea et al., Science 327, 177 (2010)

Magnetic moment μ in a field B

$$E = -\boldsymbol{\mu} \cdot \mathbf{B}$$

$$\boldsymbol{\mu} = \gamma \mathbf{L}$$

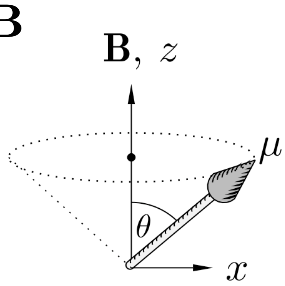


Spin precession classical

$$\frac{\mathrm{d}\boldsymbol{\mu}}{\mathrm{d}t} = \gamma \boldsymbol{\mu} \times \mathbf{B}$$

$$\begin{aligned} \dot{\mu}_x &= \gamma B \mu_y \\ \dot{\mu}_y &= -\gamma B \mu_x \\ \dot{\mu}_z &= 0, \end{aligned}$$

$$\begin{aligned} \mu_x(t) &= |\boldsymbol{\mu}| \sin\theta \cos(\gamma B t) \\ \mu_y(t) &= |\boldsymbol{\mu}| \sin\theta \sin(\gamma B t) \\ \mu_z(t) &= |\boldsymbol{\mu}| \cos\theta, \end{aligned}$$



Spin precession quantum

$$\hat{\mathcal{H}} = g\mu_{\mathrm{B}} \mathbf{B} \cdot \hat{\mathbf{S}} = g\mu_{\mathrm{B}} B \hat{S}_z.$$

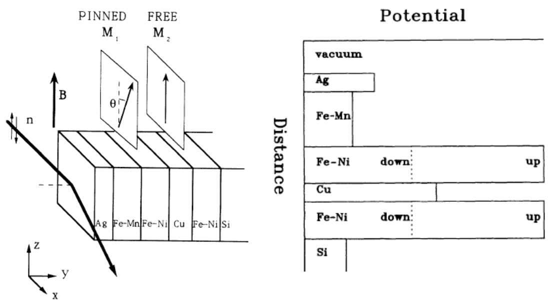
$[\hat{S}_x, \hat{S}_y] = \mathrm{i}\hat{S}_z$, or equivalently: $[\hat{\mathbf{S}} \cdot \mathbf{X}, \hat{\mathbf{S}}] = \mathrm{i}\hat{\mathbf{S}} \times \mathbf{X}$,

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \hat{\mathbf{S}} \rangle = \frac{1}{\mathrm{i}\hbar}\langle [\hat{\mathbf{S}}, \hat{\mathcal{H}}] \rangle = -\frac{g\mu_{\mathrm{B}}}{\hbar}\langle \hat{\mathbf{S}} \rangle \times \mathbf{B},$$

$$\gamma = -\frac{g\mu_{\mathrm{B}}}{\hbar} = -\frac{ge}{2m_{\mathrm{e}}}.$$

Polarized neutron reflection

$$V_\alpha = \frac{\hbar^2}{2\pi m_n} \rho_\alpha b_\alpha - \boldsymbol{\mu}_n \cdot \mathbf{B}_\alpha$$



S. J. Blundell and J. A. C. Bland, Phys. Rev. B 46, 3391 (1992)

Polarized neutron reflection

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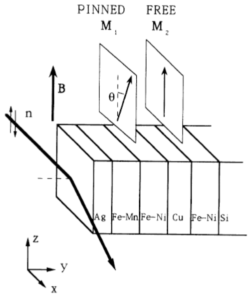

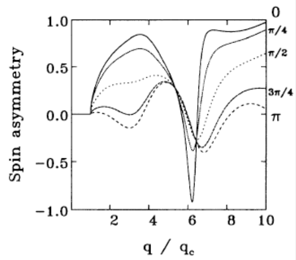

S. J. Blundell and J. A. C. Bland, Phys. Rev. B 46, 3391 (1992)

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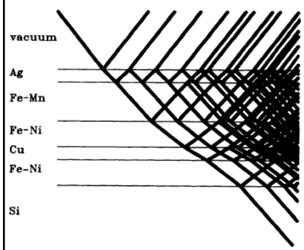
Polarized neutron reflection



Transfer matrix:

$$\begin{pmatrix} 1 \\ r \end{pmatrix} = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} \begin{pmatrix} t \\ 0 \end{pmatrix}$$

$$\underline{M} = \underbrace{\underline{D}^{-1}(q_1)}_{\text{interface}} \left\{ \prod_{j=2}^{N-1} [\underline{D}(q_j)\underbrace{\underline{P}(q_j,d_j)}_{\text{propagation}}\underline{D}^{-1}(q_j)] \right\} \underline{D}(q_N)$$

interface

propagation

S. J. Blundell and J. A. C. Bland, Phys. Rev. B 46, 3391 (1992)

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$$\underline{M} = \underline{D}^{-1}(q_1,q_1)\underline{R}(\theta_{1,2}) \left\{ \prod_{i=2}^{N-1} [\underline{D}(q_j^\uparrow,q_j^\downarrow)\underline{P}(q_j^\uparrow,q_j^\downarrow,d_j)\underline{D}^{-1}(q_j^\uparrow,q_j^\downarrow)\underline{R}(\theta_{j,j+1})] \right\} \underline{D}(q_N,q_N)$$

where

$$\underline{D}(q_j^\uparrow,q_j^\downarrow) = \begin{pmatrix} \underline{D}(q_j^\uparrow) & \underline{0} \\ \underline{0} & \underline{D}(q_j^\downarrow) \end{pmatrix}, \qquad (13)$$

is the 4×4 generalization of the *transmission matrices*,

$$\underline{P}(q_j^\uparrow,q_j^\downarrow,d_j) = \begin{pmatrix} \underline{P}(q_j^\uparrow,d_j) & \underline{0} \\ \underline{0} & \underline{P}(q_j^\downarrow,d_j) \end{pmatrix}, \qquad (14)$$

is the 4×4 generalization of the *propagation matrices*, and

$$\underline{R}(\theta_{j,j+1}) = \begin{pmatrix} \cos(\theta_{j,j+1}/2)\underline{I} & \sin(\theta_{j,j+1}/2)\underline{I} \\ -\sin(\theta_{j,j+1}/2)\underline{I} & \cos(\theta_{j,j+1}/2)\underline{I} \end{pmatrix}, \qquad (15)$$

Neutron Larmor precession in magnetic layers

are the matrices for rotating the quantization axis by $\theta_{j,j+1}$ at the j-$(j+1)$ interface.

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